Doctoral Thesis

Time Crystals and Space Crystals in terms of Macroscopic Quantum Phenomena

by
Keiji Nakatsugawa

Examination Committee: Professor Satoshi Tanda (Supervisor)
Professor Yuji Hasegawa
Professor Kousuke Yakubo
Professor Asao Arai
Professor Toshiyuki Fujii

Department of Applied Physics,
Hokkaido University, Sapporo, Japan

March 2020

Acknowledgement

First, I would like to thank my academic supervisor, Professor Satoshi Tanda, for his help for my graduate studies at Hokkaido University. I especially appreciate his enthusiasm, education and contributions throughout this research.

Second, I would like to thank my research collaborators, namely Professor Toshiyuki Fujii (Asahikawa Medical University), Professor Avadh B. Saxena (Los Alamos National Laboratory), and Professor Tatsuhiko N. Ikeda (Institute for Solid State Physics, University of Tokyo). The completion of this research and a deep understanding of the subject would not have been possible without their support. I am indebted to their valuable comments and ideas.

Third, the development of this research was also stimulated by discussion with various researchers. Especially, I would like to thank Professor Asao Arai for lectures and discussions on time operators, as well as Professor John Michael Kosterlitz for important short discussion on topological operators.

Furthermore, the members of the Laboratory of Topological Science and Technology have contributed immensely to my personal and professional life at Hokkaido. The group has been a source of friendships as well as good advice and collaboration. I am especially grateful for the help of Professor Koichi Ichimura, Professor Toru Matsuura and Professor Masahito Sakoda for valuable discussions and precious suggestions.

Finally, I would like to thank my parents for their unfailing support and continuous encouragement throughout my years of study. This accomplishment would not have been possible without their support.

This thesis is based on the following work. Chapter 2 is based on

- K. Nakatsugawa, T. Fujii and S. Tanda, Quantum time crystal by decoherence: Proposal with an incommensurate charge density wave ring, *Phys. Rev. B* **96**, 094308 (2017).

Chapter 3 is based on

- K. Nakatsugawa, T. Fujii, Avadh B Saxena and S. Tanda, Time operators and time crystals: self-adjointness by topology change, *J. Phys. A: Math. Theor.* **53**, 025301(2020).

Chapter 4 is based on

- K. Nakatsugawa, S. Tanda and T. N. Ikeda, Multivalley Free Energy Landscape and the Origin of Stripe and Quasi-Stripe CDW Structures in Monolayer MX_2 Compounds, *Sci. Rep.* **10**, 1239 (2020)

which is a continuation of

- D. Sakabe, Z. Liu, K. Suenaga, K. Nakatsugawa and S. Tanda, Direct observation of mono-layer, bi-layer, and tri-layer charge density waves in $1T$-$TaS2_2$ by transmission electron microscopy without a substrate, *npj Quantum Materials* **2**, 22 (2017).

Keiji Nakatsugawa

Hokkaido University

March 2020

Abstract

Crystals are ubiquitous around us and they have been studied extensively. However, new kinds of crystals including fullerene, carbon nanotube and topological crystals have been discovered. On the other hands, a new type of state called time crystal has been proposed: In this state, a description with macroscopic wave functions is necessary. From these backgrounds, a reconsideration of crystalline orders becomes necessary. In this thesis, I study crystalline orders in terms of macroscopic wave functions as follows:

(1) Quantum time crystal by decoherence: Physical systems can possess continuous or discrete symmetries. For example, fluids like water have continuous spatial translational symmetry but crystals have discrete spatial translational symmetries. From the equivalence between space and time in relativity, a natural question is whether discrete time translation symmetry is possible. F. Wilczek considered this problem in terms of spontaneous breaking of time translation symmetry and proposed the idea of quantum time crystals. In this thesis, I show that time translation symmetry of a finite-size ring system with a macroscopic wave function is broken by decoherence, not by spontaneous symmetry breaking. I consider a ring-shaped incommensurate charge density wave (ICDW ring) threaded by a fluctuating magnetic flux: I extend the so-called Caldeira-Leggett model to ring systems to model the fluctuating flux as a bath of harmonic oscillators. The ground state of a quantized ICDW ring without environment is a superposition of ICDWs which possess continuous space/time translation symmetry. But, as the ICDW ring couples to environment, the charge density expectation value oscillates periodically with a radius-dependent period. This model forms a metastable quantum time crystal

with a finite length in space and in time.

(2) Time operators from time crystals: In quantum mechanics, position and momentum are observables (physical operators) and the position-momentum uncertainty relation is well-established. Meanwhile, uncertainty relations between time and energy are known to hold experimentally. Therefore, an important question is whether one can define time operators which can be used to derive energy-time uncertainty relations. The main difficulty is that Hermitian operators and self-adjoint operators are different objects. Real eigenvalues and orthonormal eigenstates are ensured only for self-adjoint operators, so observables are expected to be self-adjoint. However, most of the time operators proposed so far are non-self-adjoint Hermitian operators. So, time in quantum mechanics is still regarded as a parameter. One the other hand, time crystals seem to promote time from a parameter to a physical quantity. In this thesis I consider the problem of time operators in terms of time crystals in ring systems. The key ingredient is a consistent quantization of ring systems. For instance, wave functions in ring systems must satisfy periodic boundary condition, but the position operator or angle operator are multi-valued operators which break periodicity. I show that quantization of ring systems can be understood by periodic position operators via the generalized weak Weyl relation (GWWR). Then, self-adjoint time operators in ring systems are derived based on the GWWR. In addition, I study the relationship between self-adjoint time operators and non-Hermitian operators with space-time inversion ($\mathcal{P}T$) symmetry. These operators can describe the "temporal structure of quantum systems" and they are interpreted as time crystal order parameters.

(3) Origin of stripe and quasi-stripe CDW structures in monolayer space crystals: Layered compounds such as transition metal dichalcogenides exhibit various charge density wave (CDW) phases depending on temperature and crystal symmetry. In the conventional description of phase transition, symmetry breaking occurs as temperature decreases, whereas $1T$-TaS_2 ($2H$-$TaSe_2$) makes a phase transition to the triclinic phase (stripe phase) with anisotropic domain walls only when temperature is increased from the

ground state. The appearance of these anisotropic domain structures has been explained by inter-layer interaction. However, anisotropic domain walls were observed experimentally in monolayer $1T$-TaS_2. Therefore, a new model for the appearance of anisotropic phases is necessary. In this thesis, I use Landau theories of two-dimensional CDW phase transition to make a thorough numerical research of CDW free energy structure and discover a multivalley free-energy landscape with many local minima. In addition, I discover local minima which correspond to the triclinic phase and to the stripe phase. These results can be explained by topological defect theory (line defects in a two-dimensional plane) and interference between harmonics of macroscopic CDW wave functions, hence indicate the appearance of anisotropic CDW phases without inter-layer interaction (hence also in monolayer). Moreover, I discuss the temperature dependence of successive phase transition and predict the appearance of new CDW phases.

Contents

Chapter 1

GENERAL INTRODUCTION

1.1 CRYSTAL PHASES IN QUANTUM SYSTEMS

Physical systems can possess continuous or discrete symmetries. For example, fluids like water have continuous spatial translational symmetry but crystals have discrete spatial translational symmetries. Discrete symmetry is also apparent in quantum systems: Eigenvalues of an operator can be discrete in nature and most of the crystals want to form standing waves as a consequence of interference (Bragg reflection).

Recently, the concept of crystal is extended to other physical quantities. A new type of state called "time crystal" [1] has been proposed. Other generalizations such as "phase crystal" [2] and "phase space crystal" [3] have been considered as well. *Can these new type of crystals really exist?* To answer this question, a reconsideration of space crystals becomes necessary. In particular, I focus on the wave function of a crystal.

In this thesis, I study crystalline orders in terms of macroscopic wave functions. In particular, I study quantum time crystals and quantum space crystals as new orders in CDW systems. I will show that real-space topology and the phase of a macroscopic wave function play dominant roles for the description of quantum crystal phases. Like superconductors, the CDW order parameter is a macroscopic wave function. However, unlike

superconductors, CDW wave functions can appear in real-space as electronic spatial crystals. Moreover, the interplay between real-space topology and quantum phenomena such as the Aharonov-Bohm oscillation in CDW ring [4] have been observed experimentally. Therefore, CDWs are ideal to study the effect of quantum mechanics on crystal phases.

Some important concepts and ideas are introduced in the rest of this chapter. Spontaneous symmetry breaking and some of its subtleties in quantum systems are introduced in section 1.2. This allows us to define the concept of *quantum time crystals* which have off-diagonal long-range order in the time domain. A brief history of time crystals and recent developments are given in section 1.3. Next, the connection between operator domain and real-space topology is introduced in section 1.4. This connection is essential for a consistent quantization of topological systems and to understand the problem of time operators. Then, in section 1.5, I describe two-dimensional CDW phases in terms of CDW wave functions and CDW super-lattice structure. Finally, I explain the outline of this thesis.

1.2 SYMMETRY BREAKING IN QUANTUM SYSTEMS

Different phases of condensed matter systems can be characterized by an order parameter η. By Landau's definition with superconductivity, this is any parameter that is zero in the symmetric state and that has a non-zero average value uniquely specifying the state when the symmetry is broken [5, 6]. Spontaneous symmetry breaking occurs when a system or response from a system fails to preserve the symmetry of a Lagrangian or a Hamiltonian. For instance, as a snowflake crystallizes, it lowers its energy by breaking the homogeneity and isotropy of water. Such spontaneously broken symmetry permeates the world we live in. More specifically, broken symmetry manifests itself through the development of infinite-range correlations of the order parameter. As a collection of quantum particles becomes larger, the symmetry of a quantum system as a whole becomes more

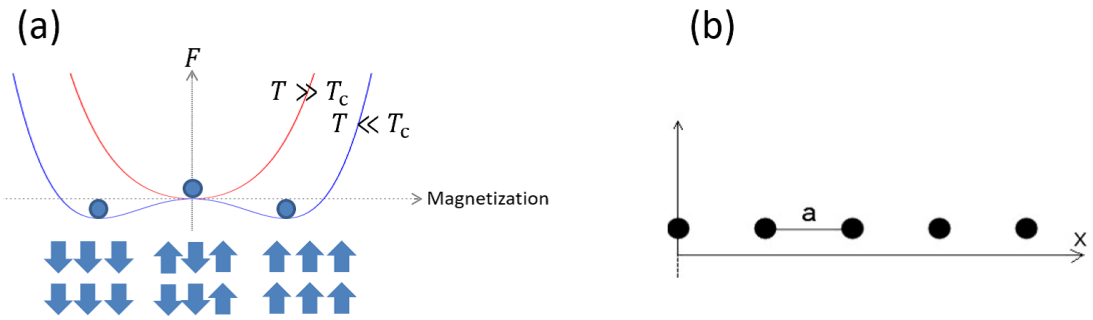

Figure 1.1: Typical examples of spontaneous symmetry breaking. (a) The Ising model (Eq. (1.1) with $\lambda \to \infty$): As temperature decreases, the free energy is minimized if all particles spins are oriented in the same direction and the system has a non-zero spontaneous magnetization. (b) Spontaneous formation of a lattice structure can be explained likewise: As temperature decreases, the inter-particle distance can be fixed and continuous spatial translation symmetry spontaneously reduces to a discrete subgroup.

unstable against small perturbations. In the limit of an infinite system, an infinitesimal perturbation is enough to cause the system to break the underlying symmetry of the Hamiltonian. For quantum systems with macroscopic wave functions, the development of these entangled, infinite-range spatial correlations in an otherwise spatially homogeneous wave function is called off-diagonal long-range order [7].

Consider ferromagnetic transition and crystallization (Fig. 1.1) to understand the effect of quantum mechanics on spontaneous symmetry breaking.

E.g. 1 Ising model: Consider a chain of spin 1/2 particles with a transverse magnetic field which is described by the following Hamiltonian [8, 9]:

$$H = -\sum_n \sigma_n^x - \lambda \sum_n \sigma_n^z \sigma_{n+1}^z. \tag{1.1}$$

Here, σ_n^x and σ_n^z are Pauli matrices which describe x-component and the z-component of the n^{th} spin, respectively. Let us consider the ground state of this system for $\lambda = 0$ and for $\lambda \to \infty$.

Case 1: $\lambda = 0$ Here, the ground state of the entire system with N spins is a coherent

state

$$|\Psi_0\rangle \propto \Big(|\uparrow\rangle_1 + |\downarrow\rangle_1\Big) \otimes \Big(|\uparrow\rangle_2 + |\downarrow\rangle_2\Big) \otimes \cdots \otimes \Big(|\uparrow\rangle_N + |\downarrow\rangle_N\Big) \tag{1.2}$$

where the measurement of a single spin does not affect the wave function of other spins.

Case 2: $\lambda \to \infty$ Here, the ground state is the entangled state

$$|\Psi_\infty\rangle \propto \Big(|\uparrow\rangle_1 \otimes |\uparrow\rangle_2 \otimes \cdots \otimes |\uparrow\rangle_N\Big) + \Big(|\downarrow\rangle_1 \otimes |\downarrow\rangle_2 \otimes \cdots \otimes |\downarrow\rangle_N\Big). \tag{1.3}$$

Eq. (1.3) is a Bell-like state with long-range entanglement. That is, the measurement of a single spin (or any other local interaction) will determine the state of the entire system. Consequently, all spins will point either up or down and symmetry is broken. The symmetry broken state is stable in the thermodynamic limit $N \to \infty$, and the system will have a finite magnetization. Although the Hamiltonian H has spatial inversion symmetry, the ground state after a measurement breaks this symmetry, hence we have spontaneous breaking of spatial inversion symmetry in the z-direction. Therefore, we see that spontaneous symmetry breaking is closely related to measurement and depends on the system size.

E.g. 2 Formation of crystalline lattice: Spontaneous formation of a lattice structure can be explained likewise. Let us consider a many-body system with N particles described by the Hamiltonian

$$H = \sum_i \frac{\mathbf{P}_i}{2m} + \frac{1}{2}\sum_{i\neq j} U_{ij}(|\mathbf{r}_i - \mathbf{r}_j|) \tag{1.4}$$

Here, $U_{ij}(|\mathbf{r}_i - \mathbf{r}_j|)$ is the potential between particle i and particle j. This system possesses continuous spatial translation symmetry $\mathbf{r}_i \to \mathbf{r}_i + \boldsymbol{\epsilon}$ for any $\boldsymbol{\epsilon}$. As temperature decreases, the inter-particle distance can be fixed and form a lattice structure. In this case, continuous spatial translation symmetry spontaneous reduces to its discrete subgroup.

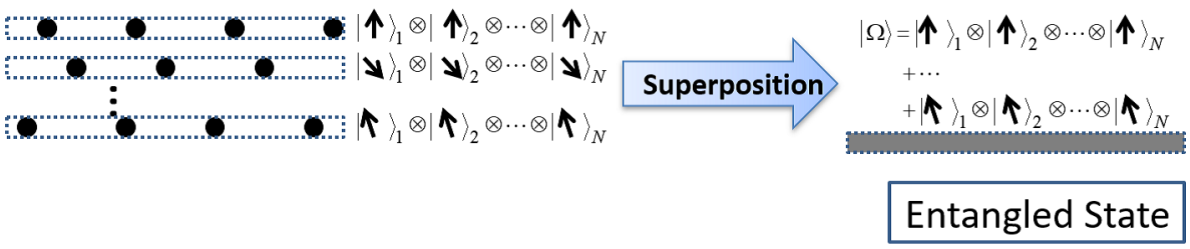

Figure 1.2: The true ground state of a quantum crystal with a macroscopic wave function is a superposition of lattices with different vertices. An analogy is a superposition of spin chains with $U(1)$ symmetry.

The full quantum description is more subtle. In quantum physics, broken symmetry is not immediately evident from a system's wave function because the vertices of the crystal are equally likely to occupy any point in space. The corresponding crystal wave function is a superposition of broken-symmetry states. The broken symmetry in a crystal is not evident until the position of one of its atomic constituents is measured. [1, 10, 11]

Let us explain this more analytically. At the ground state we can ignore relative degrees of freedom between individual particles and only consider the center of mass motion. In this case, this system can be regarded basically as a free particle.[1] The ground state of this systems has zero center-of-mass momentum $P = 0$ and zero momentum uncertainty $\Delta P = 0$ (otherwise, the average kinetic energy will increase). Then, from the the uncertainty relation $\Delta P \Delta X \sim \hbar$, the position uncertainty ΔX becomes infinite. This state can be interpreted as a superposition of lattices with different center of masses (different vertices) which preserves continuous spatial translation symmetry (Fig. 1.2). A state with broken symmetry, which can be obtained by symmetry-breaking perturbations, has energy higher than the symmetry-preserving true ground state. If the energy increase is negligible then the life-time of the symmetry-broken state diverges and we have an effective ground state (Fig. 1.3).

[1] If the interaction between particle is weak, then the Hamiltonian in (1.4) describes a system with coupled Harmonic oscillators. In this case, excitations are bosonic and we can indeed have a superposition of lattices.

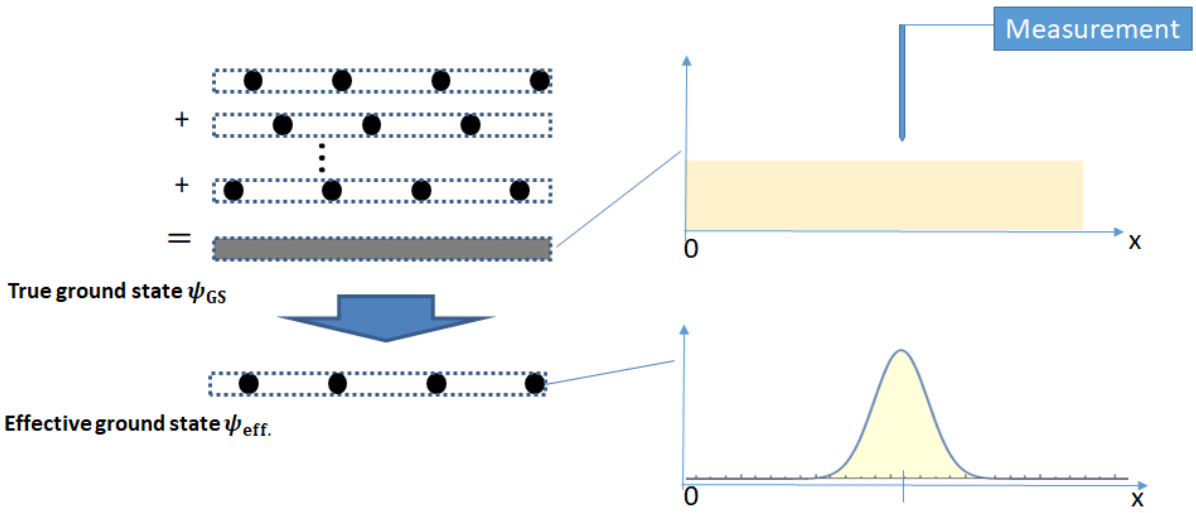

Figure 1.3: A state with broken symmetry can be obtained by symmetry-breaking perturbations such as measurements. If the measured state and the symmetric ground state are energetically degenerate, then the measured state can be regarded as an effective ground state. Conventionally, spontaneous symmetry breaking from the true ground state to the effective ground state is expected to occurs in the infinite-volume limit $V \to \infty$.

1.3 QUANTUM TIME CRYSTALS

In 2012, F. Wilczek proposed the idea of a "quantum time crystal" as a system which spontaneously breaks time translation symmetry at the ground state [1]. Wilczek argued that such a state would exhibit off-diagonal long-range order in the time domain. Naturally oscillating physical systems abound. But, the original idea behind a crystal in the time domain is that it requires no energy input. It will spontaneously form without a driving force.

Like its spatial analogue, the ground state of a QTC is determined not only by energy minimization but also by robust observation. There are two types of ground states that describe a QTC: a) the true ground state which possesses the time translation symmetry of the system's Hamiltonian; and b) the effective ground state which spontaneously breaks time translation symmetry.

Time crystals opened a new debate involving the connection between time and broken symmetry [12–18]. Conventionally, spontaneous symmetry breaking from the true

ground state to the effective ground state is expected to occurs in the infinite-volume limit $V \to \infty$. However, Watanabe-Oshikawa argued that spontaneous breaking of time translation symmetry does not occur in the infinite-volume limit, hence time crystal ground states cannot exist [17]. A workaround of this criticism was proposed with Greenberger–Horne–Zeilinger state (GHZ state) [19]. Volovik relaxed the condition of permanent oscillation and proposed the possibility of effective QTC, that is, a periodic oscillation in a metastable state such that the oscillation will persist for a finite duration $\tau_Q \gg P$ in the time domain and will eventually decay [20].

Spontaneous breaking of *discrete* time translation symmetry was also considered for periodically driven Floquet states [21–28]. However, Floquet time crystals will not be considered in this thesis (except in Section 3.3).

1.4 OPERATOR DOMAIN AND REAL-SPACE TOPOLOGY

Next, I introduce a few mathematical concepts which will be essential for developing the theory of space crystals, time crystals and time operators in topological systems.

1.4.1 Self-adjointness

A fact often glossed over in elementary introductions to quantum mechanics is that a Hermitian operator is not the same thing as a self-adjoint operator [29]. Overlooking this difference can lead to contradictions (especially in Chapter 3).

Let $\mathcal{H}$ be a Hilbert space. Define the domain of an operator $D(\hat{A}) \subseteq \mathcal{H}$, which is the set of functions upon which the operator $\hat{A}$ can act. Then,

- $\hat{A}$ is an Hermitian operator if $\langle \psi, \hat{A}\phi \rangle = \langle \hat{A}\psi, \phi \rangle$ for all $\psi, \phi \in D(A)$.
- $\hat{A}$ is a symmetric operator if it is an Hermitian operator and $D(\hat{A})$ is dense. In

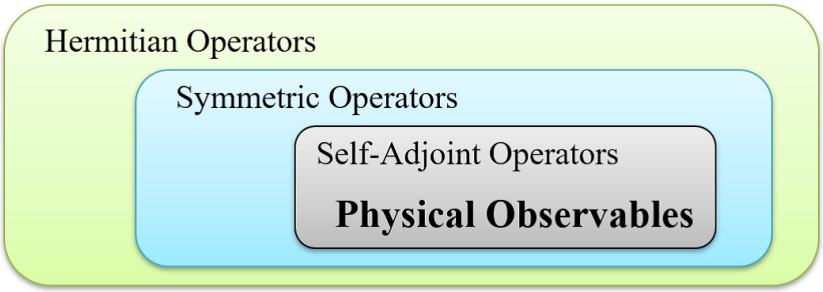

Figure 1.4: Relationship between Hermitian operators, symmetric operators and self-adjoint operators. Physical observables must be self-adjoint to ensure real eigenvalues and orthonormal eigenstates.

particular, we have $D(\hat{A}) \subset D(\hat{A}^\dagger)$.

- $\hat{A}$ is a self-adjoint operator if it is a symmetric operator and $D(\hat{A}) = D(\hat{A}^\dagger)$.

These operators are described in figure 1.4. Mathematically, one can prove that real eigenvalues and orthonormal eigenstates are ensured only for self-adjoint operators. Therefore, physical observables in quantum mechanics must be self-adjoint observables [2].

Let us investigate this difference with examples.

Example 1. Consider a single quantum particle confined in a finite range $[0, 1]$ with wave functions vanishing at the boundaries. Consider the momentum operator $\hat{p} = -i\hbar d/dx$. This operator is Hermitian, that is, for arbitrary differentiable wave functions $\varphi(x), \psi(x)$ we have

$$\langle \varphi, \hat{p}\psi \rangle = \int_0^1 dx\, \varphi^*(x)(-i\hbar\psi'(x)) = -i\hbar\varphi(x)\psi(x)\Big|_0^1 + \int_0^1 dx\, (-i\hbar\varphi'(x))^*\psi(x) = \langle \hat{p}\varphi, \psi \rangle. \tag{1.5}$$

However, from elementary quantum mechanics, we know that this momentum operator does not have eigenstates, hence it is not self-adjoint [3].

[2] But, self-adjointness is not the only possibility for physical observables. Another possibility is operators with space-time reversal symmetry. These operators are discussed in Chapter 3

[3] Proof: Assume that there exist eigenstates ψ_k such that $\hat{p}\psi_k = \hbar k\psi_k$. Moreover, using $[\hat{p}\psi_k](x) = -i\hbar d\psi_k(x)/dx$ we have $\psi_k(x) = A_k e^{ikx}$. However, from the boundary condition $\psi_k(0) = \psi_k(1) = 0$ we have $A_k = 0$ which implies $\psi_k = 0$. Therefore, the momentum operator here has an empty spectrum.

Example 2: Consider the same problem but with periodic boundary condition $\psi(0) = \psi(1)$. In this case, one can easily show that the momentum operator $\hat{p}$ is Hermitian and has a complete set of orthonormal eigenstates with discrete momentum eigenvalues, hence it is self-adjoint.

1.4.2 Quantization of ring systems

In contrast to quantum mechanics on Euclidean space ($\mathbb{R}^d$), quantum mechanics on constrained systems have infinite number of inequivalent representations [29–31]. Let us first consider the standard quantization scheme.

1. Replace real functions by self-adjoing operators.
2. Replace Poisson Brackets by Dirac Brackets.
3. Construct such irreducible representation of operator algebra on a Hilbert space.

$$\{A, B\} = C \quad \rightarrow \quad \frac{1}{i\hbar}[\hat{A}, \hat{B}] = \hat{C} \tag{1.6}$$

Note that this quantization scheme cannot be executed naively to any physical variables. In fact, for quantum mechanics on S^1, the canonical commutation relation between angle $\hat{\theta}$ and angular momentum $\hat{\pi}_\theta$

$$[\hat{\theta}, \hat{\pi}_\theta] = i\hbar \tag{1.7}$$

is not satisfactory because $\hat{\theta}$ is a multi-valued operator. Position operators are defined as a set of operators whose eigenvalues have one-to-one correspondence with points on a manifold. That is, the position of a particle should be somewhere between $0 \leq \theta < 2\pi$ but a wave function $\psi(\theta)$ for an S^1 system is periodic with $\theta \rightarrow \theta + 2\pi$. So, $\hat{\theta}$ is ill-defined because its multi-valuedness breaks the periodicity of the ring. For instance, defining $\phi(\theta) = [\hat{\theta}\psi](\theta) = \theta\psi(\theta)$ and using the periodicity of $\psi(\theta)$ we obtain

$$\phi(\theta + 2\pi) - \phi(\theta) = 2\pi\psi(\theta) \neq 0. \tag{1.8}$$

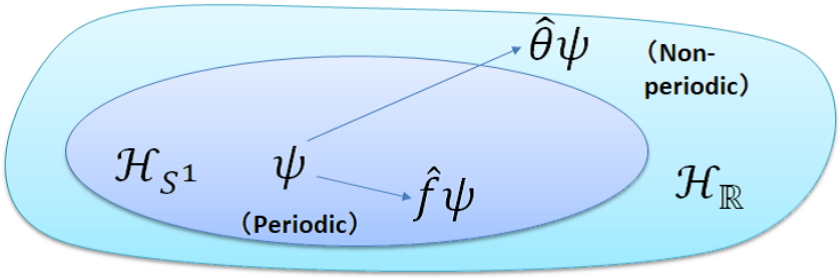

Figure 1.5: The angle operator $\hat{\theta}$ is a multi-valued operator and not suitable to represent the position of a particle on a ring. Instead, ring systems can be quantized with periodic angle operators $\hat{f}$. Here, $\mathcal{H}_{\mathbb{R}}$ is the Hilbert space for a line and $\mathcal{H}_{S^1}$ is the Hilbert space of a ring system.

Moreover, although the operator $[\hat{\theta}, \hat{\pi}_\theta] = i\hbar$ does not have this problem of periodicity (as it can be verified by direct computation) it leads to other problems. For example, the expectation value of the canonical commutation relation with angular momentum eigenstates ψ_l (including the ground state ψ_0) leads to two equivalent equations $\langle\psi_l, [\hat{\theta}, \hat{\pi}_\theta]\psi_l\rangle = l(\langle\psi_l, \hat{\theta}\psi_l\rangle - \langle\psi_l, \hat{\theta}\psi_l\rangle) = 0$ and $\langle\psi_l, [\hat{\theta}, \hat{\pi}_\theta]\psi_l\rangle = \langle\psi_l, i\hbar\psi_l\rangle = i\hbar$. These results lead to the contradiction $0 = i\hbar$. This facts suggest that the pair of operators $(\hat{\theta}, \hat{\pi}_\theta)$ is very sensitive to the domain on which their commutation relation is applied. In other words, the canonical commutation relation imposes a restriction on possible wave functions of a ring system [32]. Therefore, $\hat{\theta}$ is not suitable as a position operator. So, how can we quantize a ring system with general periodic wave functions? Previous studies to solve this problem include refs. [30, 31, 33]. In all cases, the solution was to use periodic angle operators $\hat{f}$ with $[\hat{f}\psi](\theta) = f(\theta)\psi(\theta)$ such as $f(\theta) = \theta \bmod 2\pi$, $f(\theta) = \cos\theta$, $f(\theta) = \sin\theta$, and $f(\theta) = e^{i\theta}$ (figure 1.5).

1.5 SUPER LATTICE STRUCTURES IN MX_2 COMPOUNDS

The effect of quantum mechanics (especially, the effect of macroscopic wave function) on crystal phases is also studied for two-dimensional CDW systems. Transition metal dichalcogenides (MX_2) belong to the group of van der Waals materials characterized

by their two-dimensional layered crystalline structures. These materials induce various two-dimensional charge density wave (CDW) phases depending on lattice symmetry, temperature, thickness of the crystal, and other external factors. Two dimensional CDW is gaining interests because of its rich physical content, potential applicability to nanoscale electromechanics, and due to its close relationship with high-T_C superconductivity.

Although CDWs have a macroscopic wave function and a complex scalar order parameter like superconductors, a CDW is an electronic crystal at the first place. So, different CDW phases are characterized by domain walls (topological defects): Such domain walls appear to minimize interaction between the CDW and the underlying lattice.

The MX_2 compounds $1T$-TaS_2 and $2H$-$TaSe_2$ (Figure 1 (a),(b)) exhibit various CDW phases which are characterized by domain walls (topological defects). Interesting CDW phases are the stripe phase with stripe domain walls and the triclinic (T) phase with quasi-stripe domain walls, which surprisingly show up only on heating from the C phase (Figure 1 (d) and (e)). These anisotropic domain structures were considered to appear only in bulk compounds. However, in my collaborated experimental work [34], T domain walls were also observed in monolayer $1T$-TaS_2. Therefore, a new mechanism is required to explain anisotropic phases.

1.6 OUTLINES OF THIS THESIS

In chapter 2, I show that time translation symmetry of a ring system with a macroscopic quantum ground state is broken by decoherence. In particular, I consider a ring-shaped incommensurate charge density wave (ICDW ring) threaded by a fluctuating magnetic flux: the Caldeira-Leggett model is used to model the fluctuating flux as a bath of harmonic oscillators. Then, I show that the charge density expectation value of a quantized ICDW ring coupled to its environment oscillates periodically. The Hamiltonians considered in this model are time independent unlike "Floquet time crystals" considered recently. This

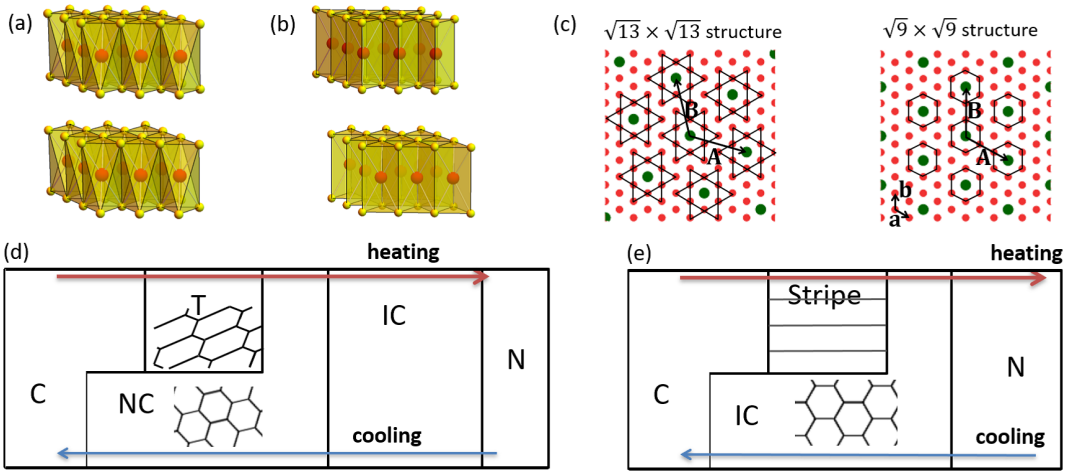

Figure 1.6: CDW structures in $1T$-TaS_2 and $2H$-$TaSe_2$. (a): Structure of $1T$-TaS_2. (b): Structure of $2H$-$TaSe_2$. These materials have two-dimensional layered crystalline structures like graphene. Here, the red spheres represent Ta atoms and the yellow spheres represent S or Se atoms. (c):The Ta atoms form a triangular lattice with superlattice vectors a and b. **A** and **B** are commensurate (C) CDW superlattice vectors ($\mathbf{A} = (\mu + \nu)\mathbf{a} + \nu\mathbf{b}, \mathbf{B} = -\nu\mathbf{a} + \mu\mathbf{b}, |\mathbf{A}| = |\mathbf{B}| = \sqrt{\mu^2 + \mu\nu + \nu^2}|\mathbf{a}|$). The CCDW superlattice for $1T$-TaS_2 ($\mu = 3, \nu = 1$) and $2H$-$TaSe_2$ ($\mu = 3, \nu = 0$) are shown with green spheres. The Ta atoms on the star-of-David or hexagonal lines are shifted toward the central green Ta atoms. (d) Temperature dependence of CDW phases in bulk $1T$-TaS_2. (e) Temperature dependence of CDW phases in bulk $2H$-$TaSe_2$. The blue arrows represent the cooling cycle. The red arrows represent the heating cycle. C: Commensurate phase, T: Triclinic phase with quasi-stripe domain walls (stretched honeycomb lattice). NC: Nearly commensurate phase, IC: Incommensurate phase, N: Normal metal phase. The domain walls of the NC, T, and stripe phases are depicted with solid lines.

model forms a metastable quantum time crystal with a finite length in space and in time.

In chapter 3, I investigate time operators in the context of quantum time crystals in ring systems. A generalized commutation relation called the *generalized weak Weyl relation* is used to derive a class of self-adjoint time operators for ring systems with a periodic time evolution: The conventional Aharonov-Bohm time operator is obtained by taking the infinite-radius limit. Then, I discuss the connection between time operators, time crystals and real-space topology. I also reveal the relationship between the above time operators and a $\mathcal{PT}$-symmetric time operator. These time operators are then used to derive several energy-time uncertainty relations.

In chapter 4, I use Landau theories of two-dimensional CDW phase transition to make a thorough numerical research of CDW free energy structure and discover a multivalley free-energy landscape with many local minima. In addition, I discover local minima which correspond to the T phase and to the stripe phase. These results can be explained by topological defect theory (line defects in a two-dimensional plane) and interference between harmonics of macroscopic CDW wave functions, hence indicate the appearance of anisotropic CDW phases without inter-layer interaction (hence also in monolayer). Moreover, I discussed the temperature dependence of successive phase transition, generalized the McMillan-Nakanishi-Shiba free energy theory for two-dimensional CDW phase transition, and predict the appearance of new CDW phases in $1T$-$TaSe_2$.

Finally, I conclude this thesis in chapter 5.

Chapter 2

QUANTUM TIME CRYSTAL BY DECOHERENCE: PROPOSAL WITH AN INCOMMENSURATE CHARGE DENSITY WAVE RING

2.1 INTRODUCTION

Wilczek's original proposal of a quantum time crystal (QTC) is a quantum mechanical ground state which spontaneously breaks time translation symmetry [1]. In this QTC ground state there exists an operator $\hat{Q}$ whose expectation value oscillates permanently with a well-defined "lattice constant" P, that is, with a well-defined period.

Volovik [20] relaxed the condition of permanent oscillation and proposed the possibility of effective QTC, that is, a periodic oscillation in a metastable state such that the oscillation will persist for a finite duration $\tau_Q \gg P$ in the time domain and will eventually decay.

In this chapter, I promote Volovik's line and consider the possibility of a metastable QTC state without spontaneous symmetry breaking: I consider symmetry breaking by

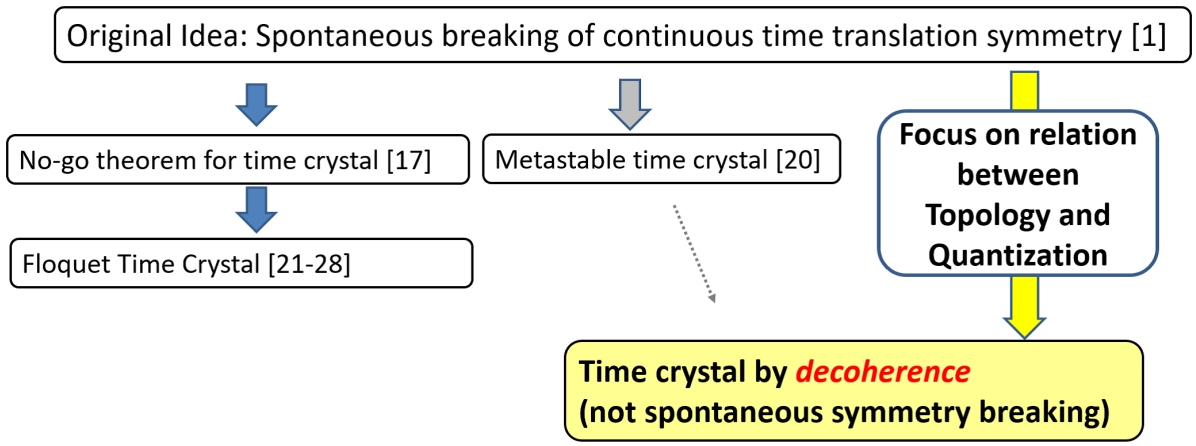

Figure 2.1: A brief history of time crystals and proposal of time crystal by decoherence

decoherence of a macroscopic quantum ground state (Fig. 2.1 and Fig. 2.2). Decoherence is defined as the loss of quantum coherence of a system coupled to its environment. Coupling to environment will inevitably introduce friction to the system such that the oscillation will eventually decay at $t = \tau_{\text{damp}}$. However, for $t < \tau_Q \ll \tau_{\text{damp}}$ the oscillation period P is well defined. If friction is sufficiently weak such that $P \ll \tau_Q$, then we have a model of effective QTC with life time τ_Q.

My model consists of a ring-shaped incommensurate charge density wave (ICDW ring) threaded by a fluctuating magnetic flux (Fig. 2.3(a)). A charge density wave (CDW) is a periodic (spatial) modulation of electric charge density which occurs in quasi-one-dimensional crystals [36–38]: the periodic modulation of the electric charge density occurs due to electron-phonon interaction. If the ratio between the CDW wavelength λ and the lattice constant a of the crystal is a simple fraction like 2, 5/2 etc, then the CDW is commensurate with the underlying lattice. A commensurate CDW cannot move freely because of commensurability pinning, i.e. the CDW phase is pinned by ions' position in the crystal. On the other hand, if λ/a is effectively an irrational number [37], then the CDW is incommensurate with the underlying lattice. An ICDW ring with a radius of 10μm, for instance, contains approximately 10^5 wavelengths [39], so it is possible that a/λ is very close to an irrational number. See the discussion section for an elaboration of this assumption. The sliding of an ICDW without pinning is described by a gapless

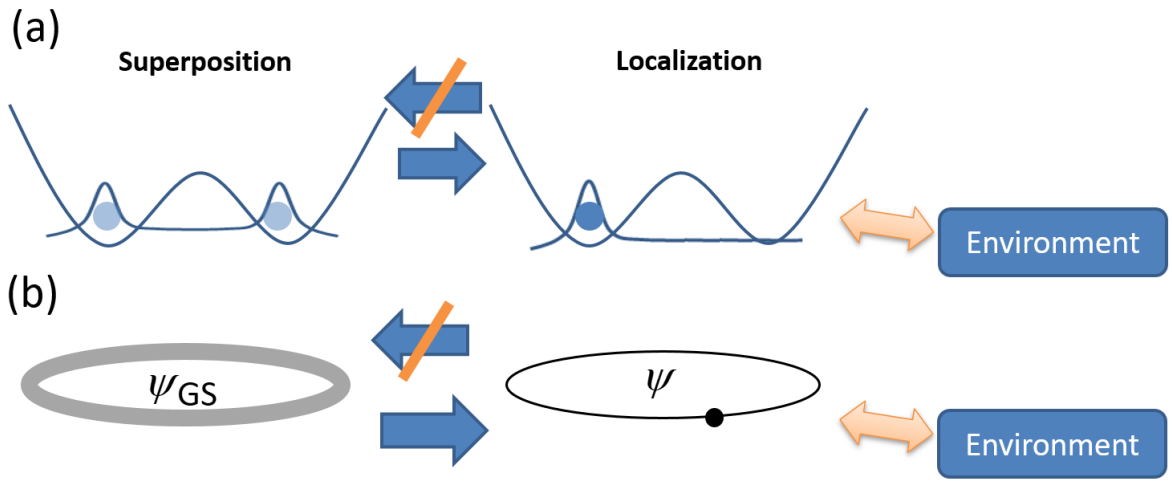

Figure 2.2: The concept of symmetry breaking by decoherence is illustrated. (a) A simplified version of the two-state system considered by Leggett *et al.* [35]. A particle can tunnel through a potential barrier and exist at two states simultaneously. However, if this system starts to interact with its surrounding environment, then the particle will localize at one of the states. (b) Similarly, the ground state of a free particle confined on a ring is a plane wave state. Coupling to environment will localize the particle and break rotational symmetry. Here, this "particle" corresponds to the phase of a charge density wave (FIG. 2.3).

Nambu-Goldstone (phason) mode [36] and the energy of an ICDW is independent of its phase (i.e. position), which implies that the expected ground state of an ICDW ring is a superposition of ICDWs with different phases. Ring-shaped crystals and ring-shaped (I)CDWs have been produced [40] (FIG. 2.3 (b)). The presence of circulating CDW current [41] and Aharonov-Bohm oscillation (evidence of macroscopic wave function) [4] are verified experimentally.

It is show in section 2.2 that the charge density expectation value of an isolated ICDW ring with moment of inertia I is periodic in time with period $P = 4\pi I/\hbar$. This periodicity is a consequence of the uncertainty relation on S^1 (ring). However, this oscillation becomes unobservable at ground state because the ground state of an isolated ICDW ring is a plane wave state, *i.e.* a coherent superposition of ICDWs with different phases. Yet, it is shown in section 2.3 that an isolated ICDW ring has off-diagonal long-range correlation in space and in time. Therefore, in section 2.4 I use the Caldeira-Leggett model [42,43] to show that

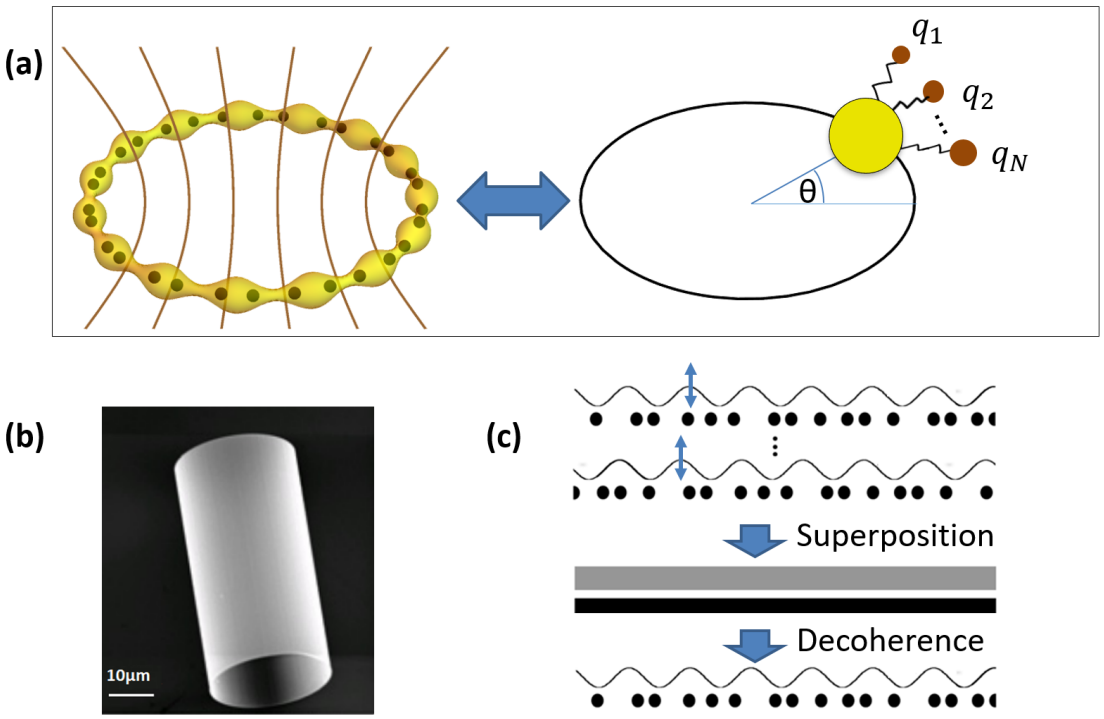

Figure 2.3: (a) I consider an incommensurate CDW ring threaded by a fluctuating magnetic flux. The wave (typically $\sim 10^5$ wavelengths) represents the charge density and the dots represent the atoms of a quasi-one dimensional crystal.(b) ICDW ring crystals such as monoclinic TaS_3 ring crystals and $NbSe_3$ ring crystals can be produced experimentally [4,40,41]. (c)The ground state of an isolated quantized ICDW ring is a superposition of periodically oscillating ICDWs, hence the oscillation is unobservable. Coupling to environment (fluctuating magnetic flux) will break the superposition and the oscillation becomes apparent.

time translation symmetry is broken by decoherence. More precisely, the superposition is broken by decoherence and the amplitude of the ICDW oscillates periodically (Fig. 2.3 (c)). If the ICDW ring weakly couples to its environment then this state is a metastable ground state. Therefore, this model forms an effective QTC with a finite length in space and in time. Finally, the results are summarized in section 2.5 and some experimental realizations are discussed.

2.2 GROUND STATE OF AN ISOLATED ICDW RING

2.2.1 Classical theory of ICDW ring

It is well known that the electric charge density of a quasi-one-dimensional crystal becomes periodic by opening a gap at the Fermi wave number k_{F} and form a charge density wave (CDW) ground state with a wavelength $\lambda = \pi/k_{\mathrm{F}}$ [36]. Consider a CDW formed on a ring-shaped quasi-one-dimensional crystal with radius R. The order parameter of this CDW ring is a complex scalar $\Delta(x,t) = |\Delta(x,t)| \exp[i\theta(x,t)]$, where $|\Delta(x,t)|$ is the size of the energy gap at $\pm k_{\mathrm{F}}$, $\theta(x,t)$ is the phase of the CDW, $x \in [0, 2\pi R)$ is the coordinate on the crystal, and t is the time coordinate. The charge density is given by

$$n(x,t) = n_0 + n_1 \cos[2k_{\mathrm{F}}x + \theta(x,t)] \tag{2.1}$$

where n_0 is the average charge density and n_1 is the amplitude of the wave. Bogachek *et al.* [44] derived the following Lagrangian density of the phase of a ring-shaped incommensurate CDW (ICDW ring) threaded by a magnetic flux

$$\mathscr{L}_0\left(\frac{\partial\theta}{\partial t}, \frac{\partial\theta}{\partial x}\right) = \frac{N_0}{2}\left[\left(\frac{\partial\theta}{\partial t}\right)^2 - c_0^2\left(\frac{\partial\theta}{\partial x}\right)^2\right] + \frac{eA}{\pi}\frac{\partial\theta}{\partial t}$$

where A is the magnetic vector potential, $N_0 = v_{\mathrm{F}}^2\hbar^2 N(\varepsilon_{\mathrm{F}})/(2c_0^2)$, $N(\varepsilon_{\mathrm{F}})$ is the density of states of electrons at the Fermi level per unit length and per spin direction, v_{F} is the Fermi

velocity of the crystal, $c_0 = \sqrt{m/m^*}v_{\rm F}$ is the phason velocity, m^* is the effective mass of electrons and $\hbar$ is the reduced Planck constant. We first consider an isolated ICDW ring with $A = 0$. Assuming that $N(\varepsilon_{\rm F})$ is equivalent to the density of states of electrons on a one dimensional line, that is, $N(\varepsilon_{\rm F}) = 1/(\pi\hbar v_{\rm F})$, we have $N_0 = \hbar v_{\rm F}/(2\pi c_0^2)$. In addition, assume the rigid-body model of ICDW, i.e. the ICDW ring does not deform locally and the phase $\theta(x,t) = \theta(t)$ is independent of position. Then, the Lagrangian, the canonical angular momentum and the Hamiltonian of the ICDW ring are, respectively

$$L_0(\dot{\theta}) = \int_0^{2\pi R} dx \mathscr{L}_0(\dot{\theta}) = \frac{I}{2}\dot{\theta}^2, \tag{2.2}$$

$$\pi_\theta(\dot{\theta}) = \frac{\partial L_0(\dot{\theta})}{\partial\dot{\theta}} = I\dot{\theta}, \tag{2.3}$$

$$H_0(\pi_\theta) = \pi_\theta\dot{\theta} - L_0(\dot{\theta}) = \frac{\pi_\theta^2}{2I} \tag{2.4}$$

where $\dot{\theta} = d\theta/dt$ and $I = \hbar R v_{\rm F}/c_0^2$ is the moment of inertia. We note that (2.2), (2.3), and (2.4) are time independent.

2.2.2 Quantization of an isolated ICDW ring

Next, I quantize the ICDW ring system. Let $\hat{H}_0 = \hat{\pi}_\theta^2/(2I)$ and $\hat{\pi}_\theta$ be the Hamiltonian operator and angular momentum operator of the ICDW ring, respectively. The macroscopic quantum state $\psi \in \mathscr{H}$ is defined in the Hilbert space $\mathscr{H}$ of positive square-integrable functions with the periodic boundary condition $\psi(\theta + 2\pi) = \psi(\theta)$. The canonical commutation relation $[\hat{\theta}, \hat{\pi}_\theta] = i\hbar$ is not satisfactory because $\hat{\theta}$ is a multi-valued operator and is not well-defined. Ohnuki and Kitakado [30] resolved this difficulty by using the unitary operator $\hat{W}$ and the self-adjoint angular momentum operator $\hat{\pi}_\theta$ defined by

$$[\hat{W}\psi](\theta) = e^{i\theta}\psi(\theta), \qquad [\hat{\pi}_\theta\psi](\theta) = -i\hbar\frac{\partial\psi(\theta)}{\partial\theta}$$

which satisfy the commutation relation on $\mathscr{H}$

$$[\hat{\pi}_\theta, \hat{W}] = \hbar\hat{W}. \tag{2.5}$$

$\hat{H}_0$ is a function of $\hat{\pi}_\theta$ only, hence the complete orthonormal set $\{\psi_l\}_{l=-\infty}^{\infty}$ of momentum eigenstates spans $\mathscr{H}$ and satisfy $\psi_l(\theta) = e^{il\theta}/\sqrt{2\pi}$. The eigenvalues of $\hat{\pi}_\theta$ are quantized with $\langle\psi_l|\hat{\pi}_\theta|\psi_l\rangle = l\hbar, l \in \mathbb{Z}$. $\hat{W}$ and $\hat{W}^\dagger$ are ladder operators which satisfy $\hat{W}\psi_l = \psi_{l+1}$ and $\hat{W}\psi_l = \psi_{l-1}$. Therefore, (2.5) is the one dimensional version of the well known angular momentum algebra [45]. Time evolution is introduced via the Heisenberg picture: $\hat{\pi}_\theta(t) = e^{i\hat{H}_0 t/\hbar}\hat{\pi}_\theta e^{-i\hat{H}_0 t/\hbar}$ and $\hat{W}(t) = e^{i\hat{H}_0 t/\hbar}\hat{W}e^{-i\hat{H}_0 t/\hbar}$. $\hat{\pi}_\theta$ commutes with $\hat{H}_0$, so $\hat{\pi}_\theta(t) = \hat{\pi}_\theta$. From the commutation relation (2.5) I obtain the following solutions of $\hat{W}(t)$:

$$\hat{W}(t) = e^{it\hat{\pi}_\theta/I}\hat{W}e^{-\frac{it}{2\mu}} = \hat{W}e^{it\hat{\pi}_\theta/I}e^{\frac{it}{2\mu}} \tag{2.6}$$

where $\mu = I/\hbar$. The two different expressions in (2.6) arise from the noncommutativity between $\hat{W}$ and $e^{it\hat{\pi}_\theta/I}$. For a QTC we need a periodic expectation value at the ground state. So, I define the time dependent charge density operator

$$\hat{n}(x,t) = n_0 + \frac{n_1}{2}\left(e^{2ik_{\mathrm{F}}x}\hat{W}(t) + \text{h.c.}\right) \tag{2.7}$$

and replace the classical charge density (2.1) by the expectation value

$$n(x,t) = \langle\hat{n}(x,t)\rangle\,. \tag{2.8}$$

Any states in $\mathscr{H}$ must be a linear superposition of $\{\psi_l\}$, that is $\psi = \sum_{l\in\mathbb{Z}} c_l\psi_l$ provided $\sum_l |c_l|^2 = 1$. Therefore, the expectation values $\langle W(t)\rangle$ and $\langle\hat{n}(x,t)\rangle$ are periodic with period $P = 4\pi\mu$ for any state ψ:

$$\begin{aligned}\langle\hat{W}(t)\rangle &= \frac{1}{2}\mathrm{tr}[\hat{W}(t)\hat{\rho} + \hat{\rho}\hat{W}(t)] \\ &= \frac{1}{2}\int_{-\pi}^{\pi} d\theta e^{i\theta}[e^{\frac{it}{2\mu}}\rho(\theta + t/\mu, \theta) + \rho(\theta, \theta - t/\mu)e^{-\frac{it}{2\mu}}].\end{aligned} \tag{2.9}$$

From the two different expressions of $W(t)$ in (2.6) we can define the Weyl form of the commutation relation [46]

$$\hat{W}e^{it\hat{\pi}_\theta/I} = e^{it\hat{\pi}_\theta/I}\hat{W}e^{-\frac{it}{\mu}}$$

hence the phase $\frac{t}{2\mu}$ and the periodic oscillation with period $4\pi\mu$ is a manifestation of the uncertainty principle. For an alternative explanation, let us consider an electron with effective mass m^* confined in a finite space with volume $L \sim 2R$. From the uncertainty principle, the momentum uncertainty of this particle is $\Delta p \sim \hbar/(4R)$. This means that the particle's wave packet expands with velocity $v = \Delta p/m^* \sim \hbar/(4m^*R)$. Then, because of the periodic boundary condition, the physical quantity $W = e^{ix/\lambda}$ is periodic with period $P = \lambda/v \sim 4\pi m^*R/(\hbar k_{\rm F}) = 4\pi m^*R/mv_{\rm F} = 4\pi Rv_{\rm F}/c_0^2 = 4\pi\mu$. So, the origin of the periodicity is (i) the macroscopic wave function of the ICDW ring expands due to the uncertainty principle then (ii) $W = e^{i\theta}$ oscillates periodically. However, the oscillation in (2.9) is not observable at the ground state $\hat{\rho}_0 \equiv |\psi_0\rangle\langle\psi_0|$ because $\langle\theta|\hat{\rho}_0|\phi\rangle = \frac{1}{2\pi}$ and the θ integral vanishes. Therefore, the ground state of an isolated ICDW ring is not yet a time crystal because of superposition. Despite of this results, I show in the next section that an ICDW ring has off-diagonal long-range correlation in space and in time.

2.3 SPACE-TIME CORRELATION OF ICDW RING

Consider the space-time correlation of an ICDW ring

$$C(x_1, x_2, t_1, t_2) = \langle\psi, \delta\hat{n}(x_1, t_1)\delta\hat{n}(x_2, t_2)\psi\rangle \tag{2.10}$$

Here, ψ is the state of the ICDW ring. $\delta\hat{n}(x,t)$ is the charge density modulation defined by

$$\delta\hat{n}(x,t) = \hat{n}(x,t) - n_0 = \frac{n_1}{2}\left(e^{2ik_{\rm F}x}\hat{W}(t) + e^{-2ik_{\rm F}x}\hat{W}^\dagger(t)\right). \tag{2.11}$$

From Eq. (2.6) I obtain

$$\begin{aligned}\delta n(x_1,t_1)\delta n(x_2,t_2) = \frac{n_1^2}{4}\Big(&e^{2ik_{\rm F}(x_1+x_2)}\hat{W}(t_1)\hat{W}(t_2) + e^{2ik_{\rm F}(x_1-x_2)}\hat{W}(t_1)\hat{W}^\dagger(t_2)\\ &+ e^{-2ik_{\rm F}(x_1-x_2)}\hat{W}^\dagger(t_1)\hat{W}(t_2) + e^{-2ik_{\rm F}(x_1+x_2)}\hat{W}^\dagger(t_1)\hat{W}^\dagger(t_2)\Big)\\ = \frac{n_1^2}{4}\Big(&e^{2ik_{\rm f}(x_1+x_2)}\hat{W}e^{\frac{it_1}{2I}(2\pi_\theta+\hbar)}\hat{W}e^{\frac{it_2}{2I}(2\hat{\pi}_\theta+\hbar)} + e^{2ik_{\rm F}(x_1-x_2)}e^{\frac{i(t_1-t_2)}{2I}(2\hat{\pi}_\theta-\hbar)}\\ + e^{-2ik_{\rm F}(x_1-x_2)}&e^{\frac{i(t_2-t_1)}{2I}(2\hat{\pi}_\theta+\hbar)} + e^{-2ik_{\rm F}(x_1+x_2)}e^{-\frac{it_1}{2I}(2\hat{\pi}_\theta+1)}\hat{W}^\dagger e^{-\frac{it_2}{2I}(2\hat{\pi}_\theta+1)}\hat{W}^\dagger\Big)\end{aligned}$$

Our interest is the space-time correlation at the ground state. The ground state of an ICDW ring is the zero angular momentum state ψ_0. In this case, I obtain

$$C_0(x_1,x_2,t_1,t_2) = \langle\psi_0,\delta\hat{n}(x_1,t_1)\delta\hat{n}(x_2,t_2)\psi_0\rangle \tag{2.12}$$

$$= \frac{n_1^2}{4}\left(e^{2ik_{\rm F}(x_1-x_2)}e^{\frac{i(t_1-t_2)}{2I}(0-\hbar)} + e^{-2ik_{\rm F}(x_1-x_2)}e^{\frac{i(t_2-t_1)}{2I}(0+\hbar)}\right) \tag{2.13}$$

$$= \frac{n_1^2}{2}e^{-\frac{i\hbar(t_1-t_2)}{2I}}\cos[2k_{\rm F}(x_1-x_2)] \tag{2.14}$$

Therefore, an ICDW ring has correlation in space and in time even at the ground state(Figure (2.4)) This space-time modulation can be measured with a projection measurement, but the measured state will immediately return to the superimposed state due to strong quantum fluctuation (see Appendix A). Therefore, one needs a continuous measurement to break space-time translation symmetry. I use interaction with environment to model this continuous measurement process.

2.4 COUPLING TO ENVIRONMENT

[1]Now, suppose that the ICDW ring starts to interact with its surrounding enviromnent at $t = 0$. Then, we expect decoherence of the phase θ. This interaction is modeled using the Caldeira-Leggett model [42] which is a model quantum Brownian motion. It describes a particle coupled to its environment. This environment is described as an set of non-interacting harmonic oscillators. First, the classical solution $\theta(t)$ is calculated to

[1]add a colorbar to the figure

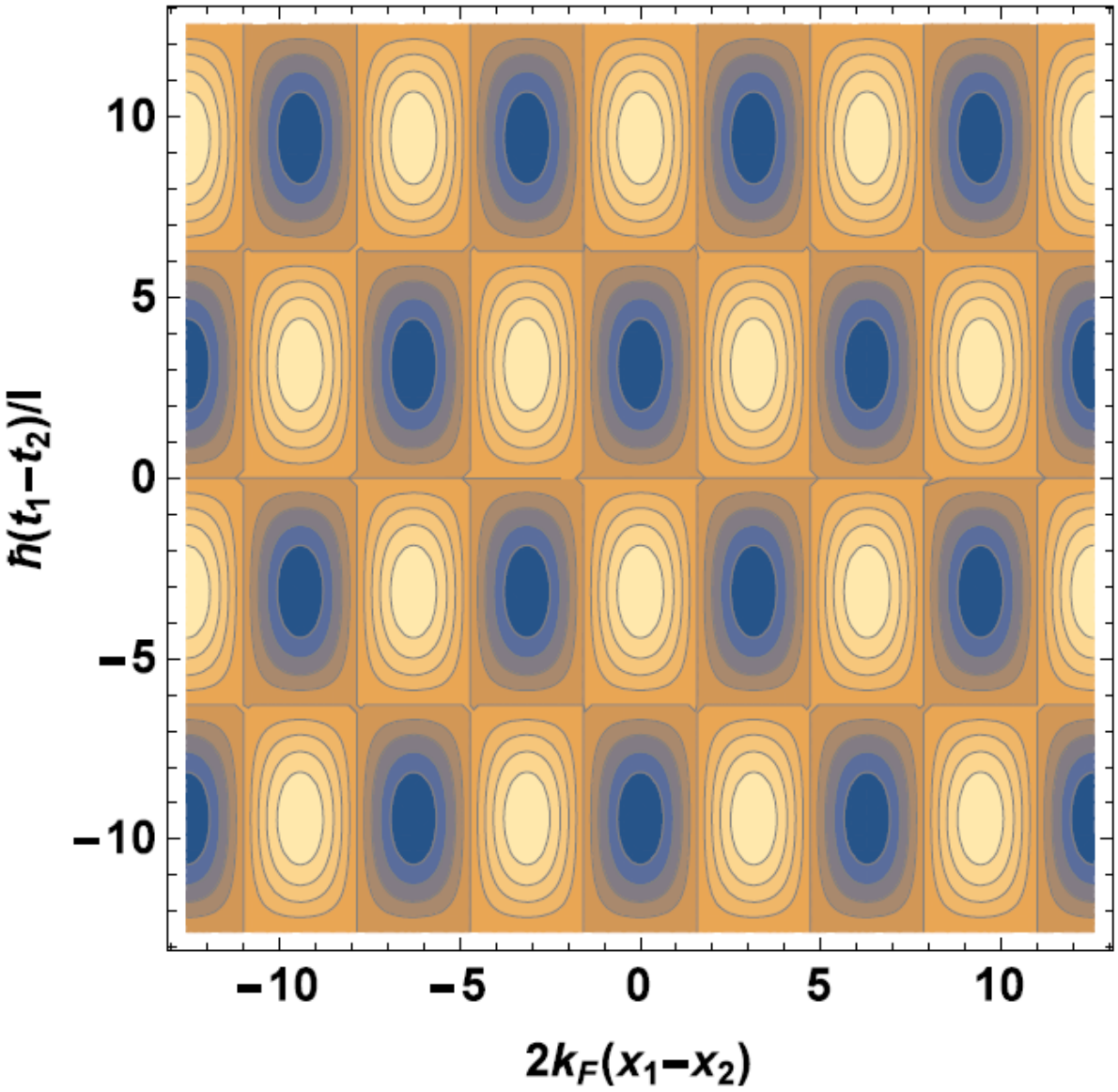

Figure 2.4: Contour plot of correlation function $\mathrm{Im}[C(x_1, x_2, t_1, t_2)]$

study the dynamics of the ICDW ring. Next, this system is quantized to calculate the amplitude of the charge density expectation value $\langle \hat{n}(x,t) \rangle$.

2.4.1 Lagrangian of ICDW ring with environment

Let us consider the following Lagrangian of an ICDW ring threaded by a fluctuating magnetic flux

$$\tilde{L}(\dot{\theta}, \mathbf{q}, \dot{\mathbf{q}}) = \frac{1}{2} I \dot{\theta}^2 + A(\mathbf{q})\dot{\theta} + \sum_{j=1}^{\mathcal{N}} \left(\frac{1}{2} m_j \dot{q}_j^2 - \frac{1}{2} m_j \omega_j^2 q_j^2 \right) \tag{2.15}$$

where q_j are the normal coordinates of the fluctuation with mass m_j and angular frequency ω_j. The fluctuating magnetic flux is given by

$$A(\mathbf{q}) = \sum_{j=1}^{\mathcal{N}} c_j q_j.$$

Classically, this magnetic flux will randomly changes the phase and the *mechanical* angular momentum $I\dot{\theta}$ of the ICDW ring due to electromotive force. Now, the principle of least action $\delta \int L dt = 0$ ensures that adding a total time derivative to a Lagrangian does not change the equation of motion. One can show that an equivalent Lagrangian obtained by the transformation

$$L = \tilde{L} - \frac{d}{dt} \sum_{j=1}^{\mathcal{N}} m_j q_j \dot{q}_j \tag{2.16}$$

is

$$L(\dot{\theta}, \mathbf{R}, \dot{\mathbf{R}}) = \frac{1}{2} I \dot{\theta}^2 + \sum_{j}^{\mathcal{N}} \left[\frac{m_j}{2} \dot{R}_j^2(\theta) - \frac{m_j \omega_j^2}{2} \left(R_j(\theta) - \frac{C_j \theta}{m_j \omega_j^2} \right)^2 \right] \tag{2.17}$$

which is the Lagrangian of a bath of field particles R_j coupled to the phase θ by springs. Here, we have $R_j(p_j, \theta) = -\frac{p_j + c_j \theta}{m_j \omega_j}$, $P_j(q_j) = m_j \omega_j q_j$ and $C_j = -c_j \omega_j$. Note that translation invariance is conserved by this transformation. Eq. (2.15) and Eq. (2.17) are precisely the kinds of Lagrangian considered by Caldeira and Leggett, so we can use the results in [42] but with slight modifications due to periodicity.

2.4.2 Equation of motion

The equations of motion from the Lagrangian (2.17) are

$$\frac{d}{dt}\frac{dL}{d\dot{\theta}} = \frac{dL}{d\theta} \quad \Rightarrow \quad I\ddot{\theta} = \sum_{j=1}^{\mathcal{N}} C_j \left(R_j(\theta) - \frac{C_j\theta}{m_j\omega_j^2} \right), \tag{2.18}$$

$$\frac{d}{dt}\frac{dL}{d\dot{R}_j} = \frac{dL}{dR_j} \quad \Rightarrow \quad m_j\ddot{R}_j(\theta) = -m_j\omega_j^2 R_j(\theta) + C_j\theta \tag{2.19}$$

Eq. (2.19) represents a harmonic oscillator with an external force $C_j\theta$: This equation has a well-known solution

$$R_j(\theta) = R_{j,0}\cos\omega_j t + \frac{\dot{R}_{j,0}}{\omega_j}\sin\omega_j t + \int_0^t \frac{C_j\theta(\tau)}{m_j\omega_j}\sin\omega_j(t-\tau)d\tau \tag{2.20}$$

where $R_{j,0}$ and $\dot{R}_{j,0}$ are the values of $R_j(\theta)$ and $\dot{R}_j(\theta)$ at $t = 0$, respectively. Upon inserting Eq. (2.20) into Eq. (2.18) we find the generalized Langevin equation [47]

$$I\ddot{\theta}(t) + 2\int_0^t d\tau\alpha_{\mathrm{I}}(t-\tau)\theta(\tau) = \xi(t) \tag{2.21}$$

with the dissipation kernel $\alpha_{\mathrm{I}}(t-\tau)$, the memory function $\gamma(t-\tau)$ and the classical fluctuating force $\xi(t)$ defined by

$$\begin{aligned}
\alpha_{\mathrm{I}}(t-\tau) &= I\gamma(0)\delta(t-\tau) + \frac{I}{2}\frac{d}{dt}\gamma(t-\tau), \\
\gamma(t-\tau) &= \sum_{j=1}^{\mathcal{N}} \frac{C_j^2}{Im_j\omega_j^2}\cos\omega_j(t-\tau), \\
\xi(t) &= \sum_{j=1}^{\mathcal{N}} C_j \left[R_{j,0}(0)\cos\omega_j t + \frac{\dot{R}_{j,0}}{\omega_j}\sin\omega_j t \right].
\end{aligned}$$

The correlation function of the classical force is given by the noise kernel $\alpha_R(t-\tau)$

$$\begin{aligned}
\langle\xi(t)\xi(\tau)\rangle_{\mathrm{env}} &= \hbar\alpha_{\mathrm{R}}(t-\tau) \\
\alpha_{\mathrm{R}}(t-\tau) &= \sum_j^{\mathcal{N}} \frac{C_j^2}{2m_j\omega_j}\coth\left(\frac{\hbar\omega_j}{2k_{\mathrm{B}}T}\right)\cos\omega_j(t-\tau)
\end{aligned}$$

where the average $\langle\cdot\rangle_{\rm env}$ is taken with respect to the environment coordinate at equilibrium. It is convenient to define the spectral density function

$$\mathcal{J}(\omega) = \frac{\pi}{2}\sum_j^{\mathcal{N}} \frac{C_j^2}{m_j\omega_j}\delta(\omega-\omega_j) \tag{2.22}$$

and assume the power law spectrum $\mathcal{J}(\omega) = Ig_s\omega^s$ [48,49] with a cutoff frequency Ω and $0 < s < 2$. Then, $\alpha_{\rm R}(t-\tau)$ can be written

$$\alpha_{\rm R}(t-\tau) = \frac{Ig_s}{\pi}\int_0^\Omega \omega^s \coth\left(\frac{\hbar\omega}{2k_{\rm B}T}\right)\cos\omega(t-\tau)d\omega.$$

2.4.3 Classical solution

Classical solution of the generalized Langevin equation Eq. (2.21) is obtained by a Laplace transform:

$$\theta(t) = G(t)\dot\theta(0) + \dot G(t)\theta(0) + \frac{1}{I}\int_0^t d\tau G(t-\tau)\xi(t) \tag{2.23}$$

with the fundamental solution

$$G(t) = \mathcal{L}^{-1}\left[\frac{1}{z^2+z\hat\gamma(z)}\right](t).$$

where $\mathcal{L}^{-1}$ is the inverse Laplace transform. The Laplace transform of the memory function $\gamma(t)$ can be written [43]

$$\hat\gamma(z) = \omega_s^{2-s}z^{s-1}, \qquad \omega_s = \left(\frac{g_s}{\sin\frac{\pi s}{2}}\right)^{1/(2-s)}$$

and $G(t)$ takes the form of a generalized Mittag-Leffler function $E_{\alpha,\beta}(x) = \sum_{k=0}^\infty \frac{x^k}{\Gamma(\alpha k+\beta)}$:

$$G(t) = tE_{2-s,s}[-(\omega_s t)^{2-s}].$$

For ohmic damping with $s = 1$ and $\hat\gamma(z) = g_1 \equiv 2\gamma$, we obtain

$$G(t) = \frac{1-e^{-2\gamma t}}{2\gamma}. \quad \text{(Ohmic)} \tag{2.24}$$

$G(t)$ and $\dot{G}(t)$ are shown in FIG. 2.5. We note that $\dot{G}(t) \approx 1$ for t less than some damping time scale $\tau_{\mathrm{damp},s}$. In other words, the fluctuating magnetic flux does not affect the dynamics of an ICDW ring for $t < \tau_{\mathrm{damp},s}$ and $e^{i\theta(t)}$ oscillates periodically with period $P \approx 4\pi\mu$. Next, we quantize the ICDW ring + environment system to show that this oscillation is observable for a finite time τ_Q and form an effective QTC as a metastable state.

2.4.4 Quantization of ICDW ring coupled to environment

The ICDW ring + environment system is quantized using the commutation relations $[\hat{\pi}_\theta, \hat{W}] = \hbar\hat{W}$ and $[\hat{q}_j, \hat{p}_k] = i\hbar\delta_{jk}$.

$\hat{W}$ is independent of the environmental coordinate. So, the expectation value of $\hat{W}$ is

$$\langle \hat{W}(t) \rangle = \int_{-\pi}^{\pi} d\theta_{\mathrm{f}} \int_{-\pi}^{\pi} d\phi_{\mathrm{f}} e^{i\theta_{\mathrm{f}}} \rho(\theta_{\mathrm{f}}, \phi_{\mathrm{f}}, t) \delta(\theta_{\mathrm{f}} - \phi_{\mathrm{f}}) \tag{2.25}$$

where the delta function $\delta(\theta_{\mathrm{f}} - \phi_{\mathrm{f}})$ is included for later convenience and the reduced density matrix of the ICDW ring is (see *Appendix* B)

$$\rho(\theta_{\mathrm{f}}, \phi_{\mathrm{f}}, t) = \int_{-\pi}^{\pi} d\theta_{\mathrm{i}} \int_{-\pi}^{\pi} d\phi_{\mathrm{i}} \sum_{l_1, l_2 \in \mathbb{Z}} \rho(\theta_{\mathrm{i}}, \phi_{\mathrm{i}}, 0) J(\theta_{\mathrm{f}} + 2\pi l_1, \phi_{\mathrm{f}} + 2\pi l_2, t; \theta_{\mathrm{i}}, \phi_{\mathrm{i}}, 0). \tag{2.26}$$

The effect of environment is included in density matrix propagator J. Here, environmental degrees of freedom are traced out using the path integral formalism. The path integral is evaluated by expanding CDW coordinate and environment coordinates around classical paths with initial conditions which is presumed to determine the initial CDW phase. Other initial conditions can be obtained by a global gauge transformation. The exact form of $J(\theta_{\mathrm{f}}, \phi_{\mathrm{f}}, t; \theta_{\mathrm{i}}, \phi_{\mathrm{i}}, 0)$ for ohmic dissipation $s = 1$ was calculated in [42]. Calculation of the reduced density matrix for general damping with arbitrary s is essentially equivalent

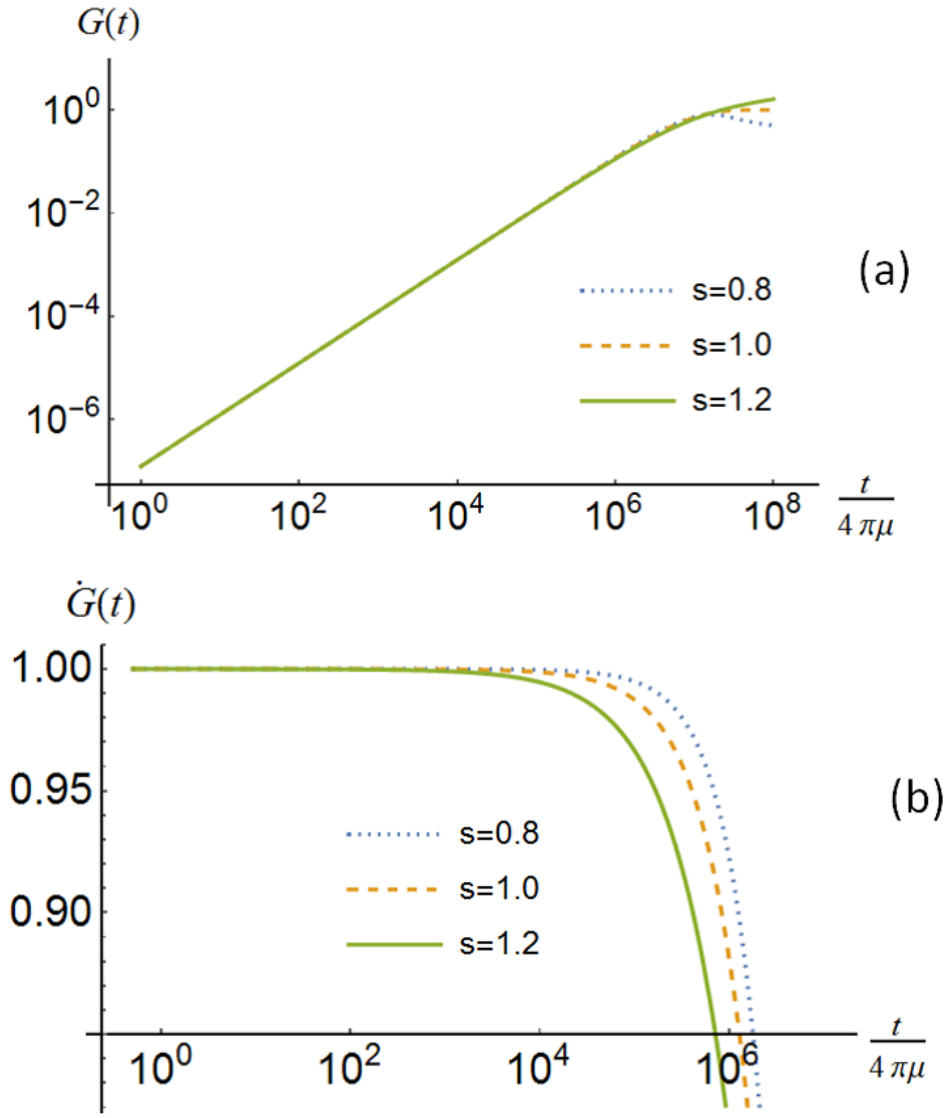

Figure 2.5: These plots are shown with $g_s = 1/\text{sec.}$ and $\mu = 10^{-8}$ sec. (a)The fundamental solution $G(t)$ of a classical ICDW ring coupled to its environment is shown for sub-ohmic ($s < 1$), ohmic ($s = 1$) and super-ohmic ($s > 1$) damping. Note that $G(t) \approx t$ for t less than some time scale $\tau_{\text{damp},s}$. The time t is normalized by $4\pi\mu$, so the horizontal axis gives the number of "lattice points". (b)The derivative of the fundamental solution, $\dot{G}(t)$, heuristically describes the velocity of the ICDW ring. Note that $\dot{G}(t) \approx 1$ for $t \ll \tau_{\text{damp,s}}$.

to [42] and I obtain

$$J(\theta_{\rm f},\phi_{\rm f},t;\theta_{\rm i},\phi_{\rm i},0)=F^2(t)\exp\left(\frac{i}{\hbar}S[\varphi^+_{\rm cl},\varphi^-_{\rm cl}]-\Gamma[\varphi^-_{\rm cl}]\right). \tag{2.27}$$

$\varphi^+_{\rm cl}=\frac{1}{2}(\theta_{\rm cl}+\phi_{\rm cl})$ and $\varphi^-_{\rm cl}=\phi_{\rm cl}-\theta_{\rm cl}$ are the classical coordinates obtained from the Euler-Lagrange equation (see Appendix C)

$$I\ddot{\varphi}^-_{\rm cl}(u)+2\int_u^t d\tau\varphi^-_{\rm cl}(\tau)\alpha_{\rm I}(\tau-u)=0, \tag{2.28}$$

$$I\ddot{\varphi}^+_{\rm cl}(u)+2\int_0^u d\tau\varphi^+_{\rm cl}(\tau)\alpha_{\rm I}(u-\tau)=0, \tag{2.29}$$

whose solution are given in terms of boundary conditions $\varphi^\pm_{\rm i}=\varphi^\pm_{\rm cl}(0),\varphi^\pm_{\rm f}=\varphi^\pm_{\rm cl}(t)$:

$$\varphi^+_{\rm cl}(u)=\kappa_i(u;t)\varphi^+_{\rm i}+\kappa_f(u;t)\varphi^+_{\rm f}, \tag{2.30}$$

$$\varphi^-_{\rm cl}(u)=\kappa_i(t-u;t)\varphi^-_{\rm f}+\kappa_f(t-u;t)\varphi^-_{\rm i}, \tag{2.31}$$

$$\kappa_i(u;t)=\dot{G}(u)-\frac{\dot{G}(t)}{G(t)}G(u), \tag{2.32}$$

$$\kappa_f(u;t)=\frac{G(u)}{G(t)}. \tag{2.33}$$

The classical action and the noise action are given by

$$S[\varphi^+_{\rm cl},\varphi^-_{\rm cl}]=S_{\rm cl}(\varphi^+_{\rm f},\varphi^-_{\rm f},t;\varphi^+_{\rm i},\varphi^-_{\rm i},0)=-I[\dot{\varphi}^+_{\rm cl}(t)\varphi^-_{\rm f}-\dot{\varphi}^+_{\rm cl}(0)\varphi^-_{\rm i}],$$
$$\Gamma[\varphi^-_{\rm cl}]=\Gamma_{\rm cl}(\varphi^-_f,t;\varphi^-_i,0)=\frac{1}{2\hbar}\int_0^t d\tau\int_0^t d\tau'\varphi^-(\tau)\alpha_{\rm R}(\tau-\tau')\varphi^-(s).$$

$F^2(t)=\left|\frac{I}{\hbar G(t)}\right|$ is a normalization function such that ${\rm tr}[\rho(t)]=\langle 1\rangle=1$. The winding numbers l_1 and l_2 can be absorbed into the $\theta_{\rm f}$ and $\phi_{\rm f}$ integrals, respectively, by changing the domain of $\theta_{\rm f}$ and $\phi_{\rm f}$ from S^1 to $\mathbb{R}^1$. The charge density expectation can be calculated

and I obtain (see Appendix C)

$$\langle \hat{W}(t) \rangle = \frac{1}{2} \int_{-\pi}^{\pi} d\theta_i \exp \left\{ i\dot{G}(t)\theta_i - i\frac{G(t)\dot{G}(t)}{2\mu} - \Gamma_{\rm cl}(2\pi n, t; -G(t)/\mu, 0) \right\} \rho(\theta_{\rm i}, \theta_{\rm i} - G(t)/2\mu, 0)$$
$$+ \frac{1}{2} \int_{-\pi}^{\pi} d\theta_i \exp \left\{ i\dot{G}(t)\theta_i + i\frac{G(t)\dot{G}(t)}{2\mu} - \Gamma_{\rm cl}(2\pi n, t; -G(t)/\mu, 0) \right\} \rho(\theta_{\rm i} + G(t)/2\mu, \theta_{\rm i}, 0). \tag{2.34}$$

This is the most general form of the expectation value of $\hat{W}$ for an ICDW ring coupled to its environment.

2.4.5 Early time approximation

Let us consider the classical solution (B.54) with $\varphi_{\rm cl}^{+} = \theta_{\rm f}$ and ohmic damping (2.24), then we see immediately that $\dot{\theta}(t) \sim \dot{\theta}(0)e^{-2\gamma t}$. Therefore, we are interested in the range $t \ll 1/(2\gamma) \equiv \tau_{\rm damp,1}$. For general dissipation, we can see from Fig. 2.5 that there exist a time scale $\tau_{{\rm damp},s}$ such that $G(t) \approx t$, $\dot{G}(t) \approx 1$ for $t \ll \tau_{{\rm damp},s}$. For $t \ll \tau_{{\rm damp},s}$ I obtain $\varphi_{\rm cl}^{-}(\tau) \approx \tau/\mu$ and the noise action can be written

$$\Gamma_{\rm cl}(2\pi n, t; -G(t)/\mu, 0) \approx \Gamma_{T,s}(t) = \frac{g_s}{2\pi\mu} \int_0^{\Omega} d\omega \coth\left(\frac{\hbar\omega}{2k_{\rm B}T}\right) \Upsilon(\omega),$$
$$\Upsilon(\omega) = \omega^{s-4}(2 + \omega^2 t^2 - 2\cos\omega t - 2\omega t \sin\omega t).$$

Taking the low temperature limit $\frac{\hbar\Omega}{2k_{\rm B}T} \to \infty$ such that $\coth\frac{\hbar\Omega}{2k_{\rm B}T} \to 1$ I obtain

$$\Gamma_{T,s}(t) - \frac{g_s}{\pi\mu} \frac{\Omega^{s-3}\left({}_1F_2\left(\frac{s-3}{2}; \frac{1}{2}, \frac{s-1}{2}; -\frac{1}{4}t^2\Omega^2\right) - 1\right)}{s-3} - \frac{g_s t^2}{2\pi\mu} \frac{\Omega^{s-1}\left({}_1F_2\left(\frac{s-1}{2}; \frac{1}{2}, \frac{s+1}{2}; -\frac{1}{4}t^2\Omega^2\right) - 1\right)}{s-1}$$

where ${}_1F_2\left(a_1; b_1, b_2; z\right)$ is the generalized hypergeometric function. Define the decoherence time $\tau_{{\rm decoh},s}$ such that $\Gamma_{T,s}(\tau_{{\rm decoh},s}) = 1$. Numerical analysis shows that the order of $\tau_{{\rm decoh},s}$ does not change with $\Omega < 1/(2\mu)$ and decreases rapidly for $\Omega > 1/(2\mu)$. If we set $\Omega \sim 1/\mu$

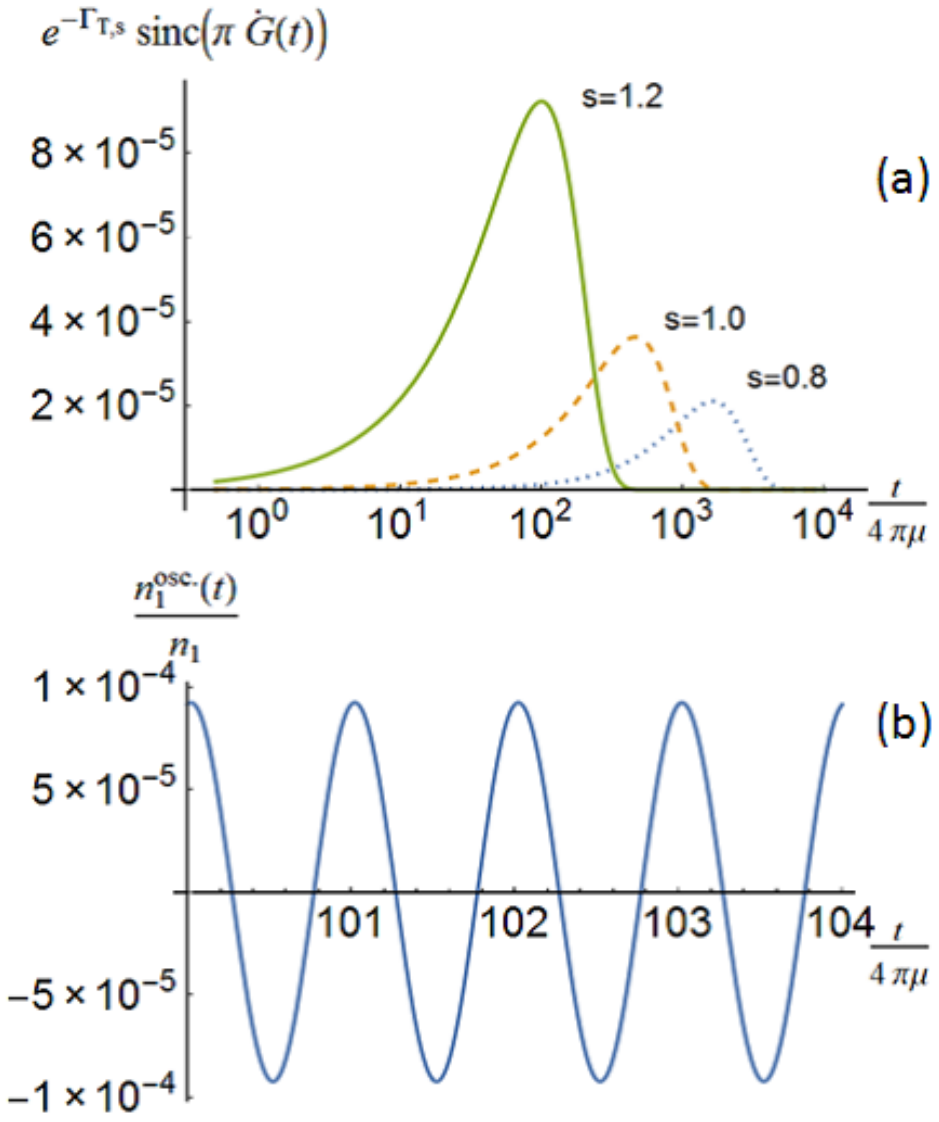

Figure 2.6: (a) The amplitude of the charge density oscillation is shown for $g_s = 1$Hz, $\mu = 10^{-8}$sec. and $\Omega = 1/\mu$. This oscillation is an effective QTC. In general, the oscillation amplitude is larger for super-ohmic($s > 1$) damping but has a small life time τ_Q. On the other hand, the oscillation amplitude is small for sub-ohmic ($s < 1$) damping but has a long τ_Q. (b) The charge density oscillation is shown for super-ohmic damping with $s = 1.2$, $g_{1.2} = 1$/sec., $\mu = 10^{-8}$sec. and $\Omega = 1/\mu$. The time t is normalized such that the horizontal axis gives the number of "lattice points".

and use the ground state ρ_0, then for $t \ll \tau_Q \equiv \min\{\tau_{\mathrm{damp,s}}, \tau_{\mathrm{decoh},s}\}$ we have

$$n(x,t) \approx n_0 + n_1^{\mathrm{osc.}}(t)\cos(2k_{\mathrm{F}}x), \tag{2.35}$$

$$n_1^{\mathrm{osc.}}(t) = n_1 \mathrm{sinc}[\pi\dot{G}(t)]\cos\left[\frac{\dot{G}(t)G(t)}{2\mu}\right] e^{-\Gamma_{T,s}(t)}. \tag{2.36}$$

Equation (2.35) is the main result of this chapter. It shows that the amplitude of an ICDW ring threaded by a (time independent) fluctuating magnetic flux oscillates for a finite time τ_Q and form an effective QTC. In the no-damping limit $\hat{\gamma}(z) \to 0$ we have $G(t) \to t, \dot{G}(t) \to 1$ and recover $\langle \hat{W}(t) \rangle = 0$. This charge density oscillation is shown in FIG. 2.6.

2.5 DISCUSSION

First, I elaborate the assumtion of ICDW ring. A mathematical definition of ICDW is that λ/a is an irrational number. We note that a CDW formed on a macroscopic crystal is basically incommensurate because the wavelength of a CDW is given by $\lambda = \pi/k_{\mathrm{F}}$, where the Fermi wave number k_{F} is usually an irrational number for an arbitrary band filling. However, strictly speaking, λ/a of a finite-size system can never be irrational. A physical condition is that the commensurability pinning energy is negligible, which is possible if λ/a cannot be expressed as a simple fraction like 2, 5/2, etc. In order to explain this, suppose that Ma/λ is an integer for some integer $M \geq 2$. In other words, the same atom-electron configuration is obtained if we move the CDW by M wavelengths and $\epsilon_{k+2Mk_{\mathrm{F}}} = \epsilon_k$ (ϵ_k is the energy of an electron with momentum $\hbar k$). Then, the energy required to move a CDW by a small phase ϕ from its equilibrium is [50]

$$\epsilon(\phi; M) = \frac{|\Delta|^2}{\epsilon_{\mathrm{F}}}\left(\frac{e|\Delta|}{W}\right)^{M-2}\frac{M\phi^2}{2}$$

where ϵ_{F} is the Fermi energy, W is the band width, $|\Delta|$ is the CDW gap width and e is the elementary charge in natural units. The M-dependence of this energy is shown in Fig. 2.7. We see that $\epsilon(\phi; M)$ approaches zero rapidly for large M as the distinction between

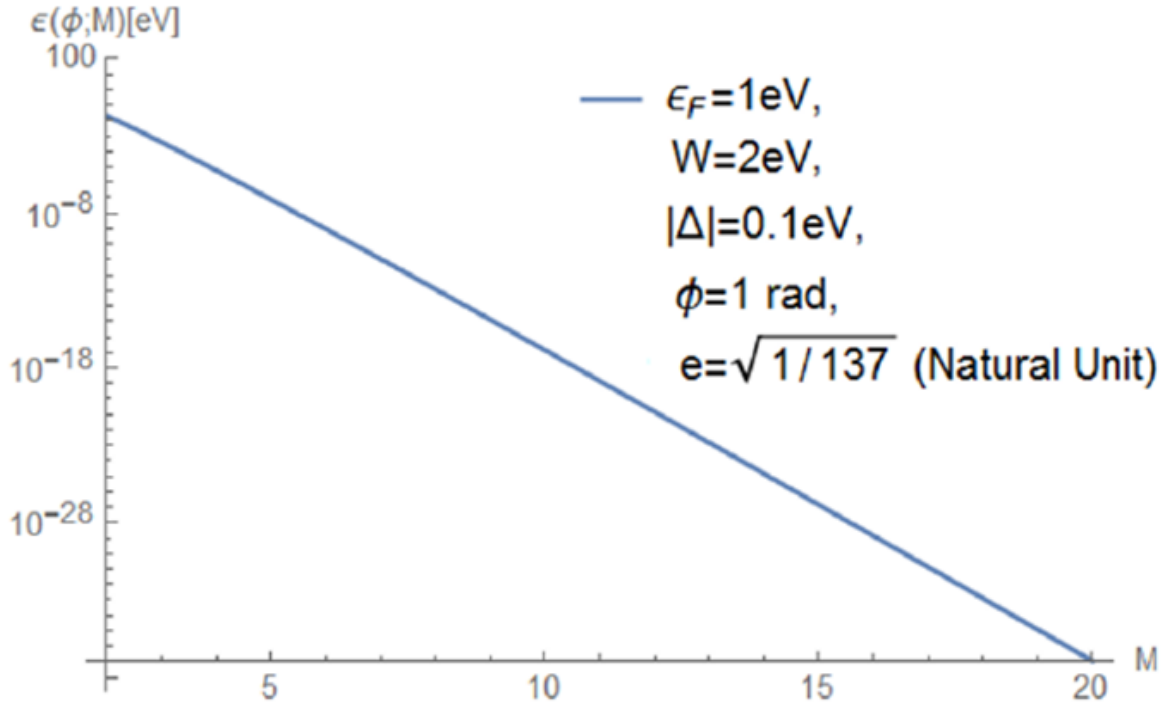

Figure 2.7: M-dependence of commensurability energy

rational and irrational numbers becomes academic. For example, for a ring crystal with N_a atoms and N_λ CDW wavelengths, we obtain $\lambda/a = N_a/N_\lambda$. Therefore, for a large N_a and a large N_λ, M can always be arbitrary large, hence the commensurability energy is completely negligible.

Usually, superposition of ICDWs with different phases is not observed because of impurity pinning. However, the probability of impurity decreases with decreasing radius. Commensurability effect may become significant for small radius (more precisely, for some small M). But, the origin of the "time crystal periodicity" in our model is the uncertainty principle, which appears as a collective fluctuation of the ICDW phase. If commensurability effect becomes significant, then the ICDW phase is expected to fluctuate periodically around some phase $\theta = \theta_0$ determined by the ions' position.

Next, I discuss the presence of an upper bound and a lower bound for the radius of the ICDW ring in this model. Decoherence induced breaking of time translation symmetry

occurs, in principle, only in mesoscopic systems: There is an upper bound for the CDW radius determined by the coupling strength $\gamma = \omega_s/2$ and a lower bound given by the CDW wavelength λ. I will first calculate the upper bound by replacing the environment with an equivalent LC circuit. Here, I focus on Ohmic damping because an approximate form of γ can be calculated explicitly. In general, the coupling strength γ depends on the CDW radius R. In order to explicitly see the R dependence, suppose that the fluctuating magnetic flux comes from an external coil connected to a series of parallel capacitors. Then, the Lagrangian of the CDW + environment system is

$$L_{\text{circuit}} = L_0 + MI_{CDW}\sum_{j=1}^{\mathcal{N}} I_j + \sum_{j=1}^{\mathcal{N}} \frac{m}{2}(I_j^2 - \omega_j^2 Q_j^2).$$

L_0 is the Lagrangian of an isolated ICDW ring, $I_{CDW} = e\dot{\theta}/\pi$ is the CDW current induced by the fluctuating flux, Q_j is the net charge on the capacitor j with capacitance $\mathscr{C}_j$, m is the inductance of the coil, $\omega_j = 1/\sqrt{m\mathscr{C}_j}$, and M is the mutual inductance between the coil and the CDW. We immediately see that L_{circuit} is equivalent to the Lagrangian in equation (2.15) but with the interaction Lagrangian replaced by $\dot{\theta}\sum_{j=1}^{\mathcal{N}} \frac{Me}{\pi}\dot{Q}$. Therefore, define $p_j = mI_j$, $C_j = Me\omega_j^2/\pi$ and we obtain the Lagrangian in equation (2.17) after the canonical transformation

$$L = L_{\text{circuit}} - \frac{d}{dt}\sum_{j}^{\mathcal{N}}(MI_{CDW} + p_j)Q_j$$

with $P_j = m\dot{R}_j = m\omega_j Q_j$ and $R_j = -(p_j - MI_{CDW})/(m\omega_j)$. Assume that the radius of the coil r_{coil} is much larger than the CDW radius R and that the coil and the CDW are concentric. Then, we obtain $M = (\mu_0\pi R^2)/(2r_{\text{coil}})$. Next, integrate (2.22) with respect to ω from $\omega = 0$ to $\omega = \Omega = 1/\mu$ and obtain $\gamma = \beta R$, $\beta = (\pi\mu_0^2 e^2 \rho c_0^6)/(32\hbar r_{\text{coil}}^2 m v_F^3)$. Here, μ_0 is the permeability of free space and ρ is the density of states defined by $\sum_j \to \int d\omega\rho$. If we assume that $\rho \sim \Omega$ like in the Caldeira-Leggett model, then the order of β may change depending on the parameters of the CDW and the parameters of the coil, but $\gamma = \beta R$ is usually smaller than 1Hz. Now, if the radius of the CDW ring is too large, then

periodicity does not appear because the oscillation period exceeds the lifetime τ_Q of the time crystal. The upper bound of the CDW radius R is given by the condition $N > 1$, where $N = \min\{\tau_{\mathrm{damp},s}, \tau_{\mathrm{deph},s}\}/(4\pi\mu)$ is the number of oscillations. For Ohmic damping ($s = 1$) we have the approximate form $\tau_{\mathrm{damp},1} = \gamma^{-1}$ and $\tau_{\mathrm{deph},1} = \sqrt{\mu\gamma^{-1}} = \sqrt{\mu\gamma}\tau_{\mathrm{damp},1}$. Let $\mu\gamma < 1$, i.e. $\tau_{\mathrm{damp},1} > \tau_{\mathrm{deph},1}$ (which is a valid assumption because a typical value for μ with radius 10^{-6}m is 10^{-6}s and γ is usually smaller than 1Hz), then, the upper bound to observe more than one oscillation is $R < \frac{c_0}{4\pi\sqrt{v_F\beta}}$ which is typically 1mm. There is also a lower bound determined by the CDW wavelength λ: The radius should be large enough to define a Fermi surface. This condition is given by $p_{\mathrm{F}} = \hbar\pi/\lambda \gg \hbar/R$. In other words, R should be much larger than λ. [2]

The origin of friction in the Caldeira-Leggett model is that the system energy dissipates into the surrounding environment and, because the Caldeira-Leggett model assumes an environment with an infinite degrees of freedom, the dissipated energy never comes back. Therefore, I expect that friction can be reduced for an environment with finite degrees of freedom. Ideally, one would like to construct a model of time crystal with an infinite lifetime using dephasing, that is decoherence without dissipation. Dephasing is possible with a spin environment, and the possibility of time crystal by dephasing is left as future study.

Finally, I discuss how this model can be tested experimentally and discuss how my results may be applicable to other physical systems. Ring-shaped crystals and ICDW ring crystals (such as monoclinic TaS_3 ring crystals and $NbSe_3$ ring crystals) have been produced and studied by the Hokkaido group [4, 40, 41]. Therefore, this model can be tested provided ring crystals with almost no defects and almost no impurities can be produced. The oscillation in (2.35) implies that the local charge density of the ICDW ring oscillates with frequency $\omega = \hbar/2I$. For a ring with diameter $2R = 1\mu$m, $v_{\mathrm{F}}/c_0 = 10^3$

[2] For another reason, suppose that the de-Broglie wavelength of the ICDW ring $\lambda_{dB} = 2\pi\hbar/p$ can be defined consistently. Then, superposition of ICDWs with different phases is expected to be broken for $\lambda_{dB} < \lambda$. The mass $m^*$of an ICDW is a constant determined by the electron-phonon interaction strength. So, using $p = (m^*\lambda)/\mu$ we obtain $\lambda_{dB} = 8\pi R$. Therefore, the condition $\lambda_{dB} > \lambda$ implies $R > \lambda/8\pi$, which is always true here.

and $v_{\rm F} = 10^5$m/s, we have $\omega = 10^8 Hz$. The time dependence of the charge density modulation can be measured using scanning tunneling microscopy (STM) [51] and/or using narrow-band noise with vanishing threshhold voltage [52].

Recall that the origin of the quantum oscillation in this model is the uncertainty principle. If we were to consider a single particle with mass m^* confined on a ring with radius R, then the particle's wave packet expands with a velocity $v \sim \hbar/(4m^*R)$ and $e^{i\theta}$ oscillates with period $P = 2\pi R/v$. However, a charge density wave has an internal periodicity given by the wavelength λ. Then the period of oscillation is $P \sim \lambda/v$. Therefore, because an ICDW ring is described by a macroscopic wave function with internal periodicity, the number of lattice points N in the present model is numerous. And yet, the periodicity of $\hat{W}(t)$ seems to be universal for any wave functions on S^1 (ring system). Therefore, the analysis and results in this section may be applicable to earlier models of QTC such as [1,53,54] and annular Josephson junctions [55]. Moreover, it was shown in ref. [56] that the ground state of a $^{40}Ca^+$ ring trap possesses rotational symmetry as the number of ions is decreased. Our results predict that quantum oscillations may appear in such ring traps with the appropriate set up. We also recall that Volovik's proposal of metastable effective QTC [20] is not restricted to ring systems. Therefore, time translation symmetry breaking by *decoherence* may occur in other systems coupled to time-independent environment, and without a periodic driving field. I also expect that many other incommensurate systems such as incommensurate spin density waves [36], incommensurate mass density waves [57–59], or possibly some dielectrics that exhibit incommensurate phases [60], may be used to test time crystal by decoherence and to model QTC without spontaneous symmetry breaking.

Chapter 3

TIME OPERATORS AND TIME CRYSTALS

3.1 INTRODUCTION

In quantum mechanics and quantum field theory, position operators, momentum operators and the position-momentum uncertainty relation are well-established. For a one dimensional system, the position-momentum uncertainty relation $\Delta x \Delta p \geq \hbar/2$ can be derived mathematically from the canonical commutation relation $[\hat{x}, \hat{p}] = i\hbar$. The energy-time uncertainty relation $\Delta E \Delta T \sim \hbar$ is also known *experimentally*. In analogy with position and momentum, one exepts that the energy-time uncertainty relation can be obtained from a commutation relation as shown in figure 3.1. However, how to define a corresponding time operator is still an open problem: One of the reasons is the difficulty to define a self-adjoint time operator $\hat{T}$ conjugate to a Hamiltonian operator $\hat{H}$ which

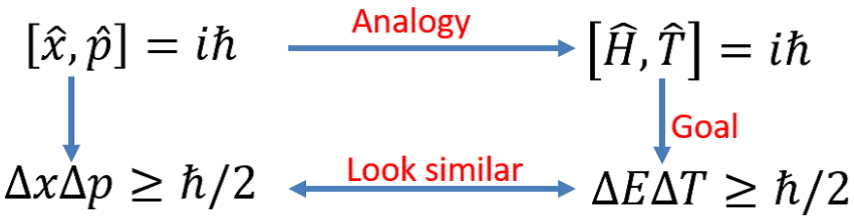

Figure 3.1: A motivation to define time operators.

satisfies the canonical commutation relation [61, 62][1]

$$[\hat{H}, \hat{T}] = i\hbar. \tag{3.1}$$

The difficulty to define time operators lies in the difference between self-adjoint operators and symmetric operators even though these are both Hermitian operators (Fig. 3.2). The existence of orthogonal eigenstates and real eigenvalues is ensured for self-adjoint operators but not ensured for symmetric operators. So, observables in standard quantum mechanics have to be represented by self-adjoint operators [61] but most of the time operators which satisfy Eq. (3.1) are symmetric non-self-adjoint operators [62]. For example, the Aharonov-Bohm time operator [63]

$$\hat{T}_{\mathbb{R}} = -\frac{m}{2}(\hat{x}\hat{p}^{-1} + \hat{p}^{-1}\hat{x}) \tag{3.2}$$

which describes the arrival time of a free particle on a line ($\mathbb{R}$) is a symmetric operator without orthonormal eigenstates [62]. Such objection was first raised by W. Pauli's well-known theorem [64]. E. Galapon disproved Pauli's theorem by specifying Pauli's implicit assumptions, then he proved that self-adjoint time operators which satisfy Eq. (3.1) can be defined in confined systems [65, 66]. The mathematical structure of time operators for a Hamiltonian with discrete eigenvalues was studied by Galapon [65, 67], Arai and Matsuzawa [32], and Arai [68].

In this chapter, I consider the above problem in the context of quantum time crystals (QTC) in chapter 2. The periodic oscillation of a QTC seems to promote time from a parameter to a physical observable. So, I propose that QTC are candidates for constructing time operators.

In section 3.2 I use a generalized commutation relation called the *generalized weak*

[1]More precisely, if the eigenvalues of a Hamiltonian are continuous and bounded below, then there is no self-adjoint time operators which satisfy the canonical commutation relation (1).

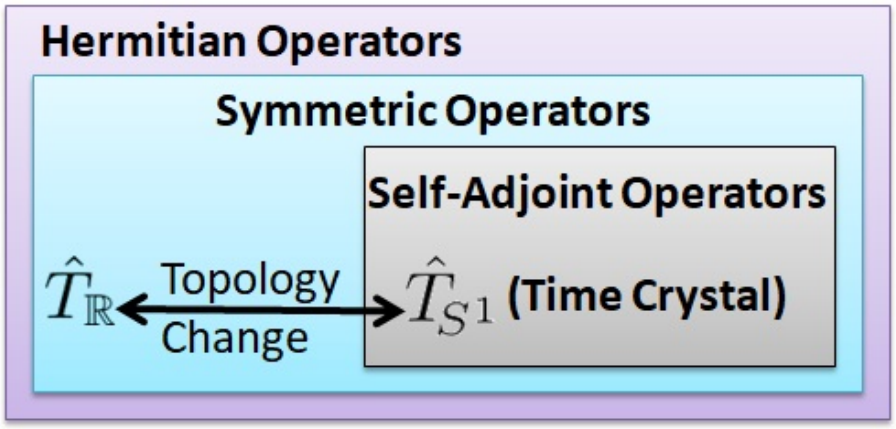

Figure 3.2: Conventionally, physical observables should be represented by self-adjoint operators. The Aharonov-Bohm time operator $\hat{T}_{\mathbb{R}}$ [Eq. (3.2)] is a symmetric operator which describes the arrival time of a free particle on a line $\mathbb{R}$ [63]. I derive the self-adjoint operator $\hat{T}_{S^1}$ [Eq. (3.18)] for a free particle on a ring system S^1. $\hat{T}_{\mathbb{R}}$ is obtained by taking the infinite radius limit of $\hat{T}_{S^1}$.

Weyl relation (GWWR) [69] to derive a class of self-adjoint time operators for a free particle in a ring system. The GWWR is required for a consistent quantization of ring systems. These time operators show a periodic time evolution intrinsic to ring systems. The conventional Aharonov-Bohm time operator is obtained by taking the infinite-radius limit. Then I discuss the connection between the GWWR, time operators, time crystals, and real-space topology of the system. Moreover, operators obeying $\mathcal{PT}$-symmetry have real eigenvalues if their eigenstates are $\mathcal{PT}$-symmetric as well [70–72], but the possibility of $\mathcal{PT}$-symmetric time operators has not been considered in literature. So, in section 3.4 I reveal the relationship between the self-adjoint time operators in section 3.2 and a $\mathcal{PT}$-symmetric time operator with orthogonal eigenstates and real eigenvalues. This $\mathcal{PT}$-symmetric operator reduces to a non-Hermitian time operator [73] in the infinite-radius limit. Then, in section 3.5, I derive several energy-time uncertainty relations for self-adjoint and $\mathcal{PT}$-symmetric time operators.

3.2 SELF-ADJOINT OPERATOR IN S^1

3.2.1 Generalized weak Weyl relation and real-space topology

There is a hierarchy of operators which satisfy stronger and weaker versions of the canonical commutation relation [30, 31, 33, 61, 69, 74–76]. Here, I consider the *generalized weak Weyl relation* (GWWR) introduced by A. Arai [69]. As I exemplify below, the GWWR is necessary for a consistent quantization of ring systems. Let $\hat{A}$ be a symmetric operator on a Hilbert space $\mathcal{H}$, let $\hat{B}$ be a self-adjoint operator on $\mathcal{H}$ and $\hat{K}(s)$ ($s \in \mathbb{R}$, s may denote time, position, or some other variable) be a bounded self-adjoint operator on $\mathcal{H}$ with $D(\hat{K}(s)) = \mathcal{H}$ ($D(\cdot)$ denotes operator domain), $\forall s \in \mathbb{R}$. We say that $(\hat{A}, \hat{B}, \hat{K})$ obeys the GWWR in $\mathcal{H}$ if for all $|\psi\rangle \in D(\hat{A})$ and for all $s \in \mathbb{R}$ we have $e^{-is\hat{B}/\hbar}|\psi\rangle \in D(\hat{A})$ and

$$\hat{A}e^{-is\hat{B}/\hbar}|\psi\rangle = e^{-is\hat{B}/\hbar}(\hat{A} + \hat{K}(s))|\psi\rangle\,. \tag{3.3}$$

$\hat{K}(s)$ is called the commutation factor of the GWWR. $\hat{A}$ and $\hat{B}$ may represent position, momentum, angular momentum, Hamiltonian, time operator, etc. Moreover, if $\hat{K}(s)$ is differentiable with respect to s, then one can differentiate both sides of Eq. (3.3) and set $s = 0$ to obtain the *generalized canonical commutation relation* (GCCR) for all $|\psi\rangle \in D(\hat{A}\hat{B}) \cap D(\hat{B}\hat{A})$

$$[\hat{A}, \hat{B}]|\psi\rangle = i\hbar\hat{K}'(0)|\psi\rangle\,, \tag{3.4}$$

where $'$ denotes derivative with respect to s. For $\hat{K}(s) = \pm s$ Eq. (3.3) reduces to the *weak Weyl relation* [74, 75] and Eq. (3.4) reduces to the canonical commutation relation$[\hat{A}, \hat{B}]|\psi\rangle = \pm i\hbar|\psi\rangle$. These relations are summarized in Fig. (3.3) I conjecture that the commutation factor $\hat{K}(s)$, in general, depends on the real-space topology of a quantum system. Several examples are given below.

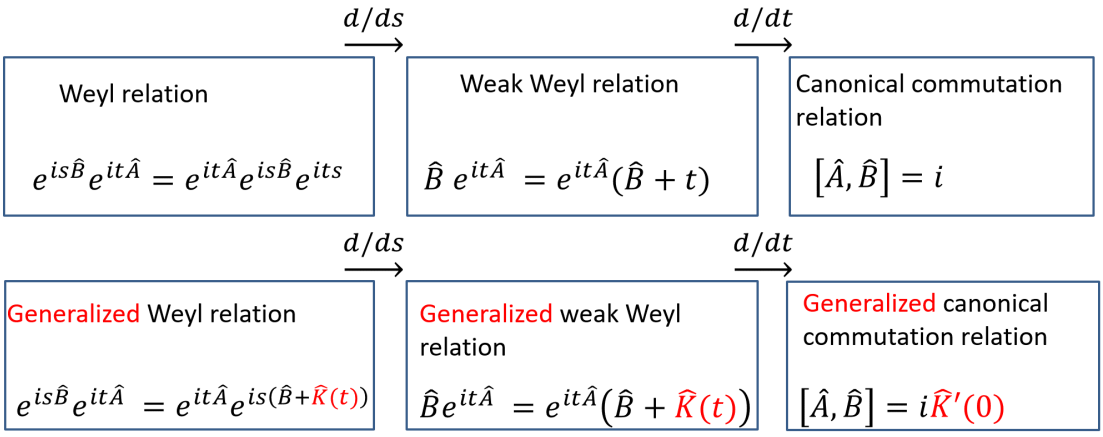

Figure 3.3: Relationship between the Weyl relation, weak Weyl relation, canonical commutation relation, and their generalization.

One-dimensional system

The Hilbert space of a one-dimensional system is $\mathcal{H} = L^2(\mathbb{R})$. The position operator $\hat{x}$ and the momentum operator $\hat{p}$ are both self-adjoint. For $(\hat{A}, \hat{B}, \hat{K}(s)) = (\hat{x}, \hat{p}, s)$ we have $[\hat{x}, \hat{p}] = i\hbar$. For $(\hat{A}, \hat{B}, \hat{K}(t)) = (\hat{T}_{\mathbb{R}}, \hat{H}, -t)$ we have $[\hat{H}, \hat{T}_{\mathbb{R}}] = i\hbar$, where $\hat{H} = \hat{p}^2/2m$ and $\hat{T}_{\mathbb{R}}$ is the Aharonov-Bohm time operator defined in Eq. (3.2).

Confined system

Galapon [65] and Galapon, Caballar, and Bahague [66] considered time operators in confined systems with the Hilbert space $\mathcal{H} = L^2([-l, l])$ and with the boundary condition $\phi(-l) = e^{-2i\gamma}\phi(l)$, $\phi \in \mathcal{H}$, $|\gamma| < \pi$. Let us consider a free particle with the Hamiltonian $\hat{H}_\gamma = \hat{p}_\gamma^2/2m$. If $\gamma \neq 0$ then $p = 0$ is *not* an eigenvalue of $\hat{p}_\gamma$, so $\hat{p}_\gamma^{-1}$ is a bounded self-adjoint operator. If $\gamma = 0$ (periodic boundary condition) then $p = 0$ is an eigenvalue of $\hat{p}_\gamma$, so $\hat{p}_\gamma^{-1}$ is well-defined only if the zero-momentum state $|p = 0\rangle$ is removed using projection operators [65,66]. In both cases, it is possible to define a self-adjoint time operator $\hat{T}_\gamma = -\frac{m}{2}(\hat{x}\hat{p}_\gamma^{-1} + \hat{p}_\gamma^{-1}\hat{x})$ which satisfies the canonical commutation relation $[\hat{H}_\gamma, \hat{T}_\gamma] = i\hbar$. This commutation relation is well-defined within a restricted domain.

My main concern is as follows. Position operators are defined as a set of operators whose eigenvalues have one-to-one correspondence with points on a manifold. But in the

case $\gamma = 0$ (periodic boundary condition) $\hat{x}$ is a multivalued operator, hence it is not suitable as a position operator [30, 31, 33]. For instance, any continuous wave function ψ of a ring system must satisfy the periodic boundary condition but $\hat{x}\psi$ is not periodic. Moreover, if $\hat{x}$ is not used with care, then many contradictions occur. For example, the expectation value of the canonical commutation relation with momentum eigenstates ψ_p (including the ground state ψ_0) leads to two equivalent equations $\langle\psi_p|[\hat{x}, \hat{p}_0]|\psi_p\rangle = p(\langle\psi_p|\hat{x}|\psi_p\rangle - \langle\psi_p|\hat{x}|\psi_p\rangle) = 0$ and $\langle\psi_p|[\hat{x}, \hat{p}_0]|\psi_p\rangle = \langle\psi_p|i\hbar|\psi_p\rangle = i\hbar$. These results lead to the contradiction $0 = i\hbar$. Similarly, one can verify that $\langle\psi_p|[\hat{H}_0, \hat{T}_0]|\psi_p\rangle = 0$ for $p \neq 0$. These facts suggest that the pairs $(\hat{x}, \hat{p}_0)$ and $(\hat{T}_0, \hat{H}_0)$ are very sensitive to the domain on which their commutation relation is applied. In other words, the canonical commutation relation imposes a restriction on possible wave functions of a ring system [32, 65]. So, how can we obtain position operators, time operators and commutation relations for general periodic functions? This problem can be solved with periodic position operators as explained below.

Ring systems

Let $\mathcal{H} = L^2([-\pi, \pi])$ be the Hilbert space of ring systems with the periodic boundary condition $\psi(\theta) = \psi(\theta + 2\pi)$, where $\theta = x/R$ is the angular coordinate. As I exemplified above, how to quantize ring systems has been an important problem in the formulation of quantum mechanics. Previous studies to solve this problem include refs. [30, 31, 33]. In all cases, the solution was to use periodic angle variables such as $f(\theta) = \theta \bmod 2\pi$, $f(\theta) = \cos\theta$, $f(\theta) = \sin\theta$, and $f(\theta) = e^{i\theta}$. In fact, these different formulations have the same mathematical structure, namely the GWWR. Instead of the multivalued position operator $\hat{x}$ or the angle operator $\hat{\theta} = \hat{x}/R$, I use a periodic position operator $\hat{f}$ such that $\hat{f}\,|\theta\rangle = f(\theta)\,|\theta\rangle$ with $f(\theta + 2\pi) = f(\theta)$. I also define $\hat{f}_s = e^{i\hat{\pi}_\theta s/\hbar}\hat{f}e^{-i\hat{\pi}_\theta s/\hbar}$ with $\hat{f}_s\,|\theta\rangle = f(\theta + s)\,|\theta\rangle$, $\hat{\pi}_\theta = R\hat{p}$ is the canonical angular momentum operator. Then, from the GWWR with $\hat{A} = \hat{f}$ and $\hat{B} = \hat{\pi}_\theta$ I obtain $\hat{K}(s) = \hat{f}_s - \hat{f}$, $\hat{K}'(s) = \hat{f}'_s = d\hat{f}_s/ds$, and

these operators satisfy the GCCR

$$[\hat{\pi}_\theta, \hat{f}] = -i\hbar \hat{f}'_s\big|_{s=0}. \tag{3.5}$$

Any physical periodic operators can be constructed using Fourier series

$$\hat{f}\,|\theta\rangle = f(\theta)\,|\theta\rangle = \sum_{n=-\infty}^{\infty} c_n e^{in\theta}\,|\theta\rangle = \sum_{n=-\infty}^{\infty} c_n \hat{W}^n\,|\theta\rangle$$

where $\hat{W} = \widehat{e^{i\theta}}$, $\hat{W}^\dagger = \hat{W}^{-1}$ [30, 31]. $\hat{\pi}_\theta$ and $\hat{W}$ satisfy the following eigenvalue equations and commutation relation in $\mathcal{H}$

$$\hat{W}^n\,|\theta\rangle = e^{in\theta}\,|\theta\rangle\,, \tag{3.6}$$

$$\hat{\pi}_\theta\,|l\rangle = l\hbar\,|l\rangle\,, \tag{3.7}$$

$$\hat{W}^n\,|l\rangle = |l+n\rangle \tag{3.8}$$

$$[\hat{\pi}_\theta, \hat{W}^n] = n\hbar\hat{W}^n. \tag{3.9}$$

Note that l and n are integers and one can verify that $\hat{W}^{-n} = \hat{W}^{\dagger n}$. From this definition with Eq. (3.5) and Eq. (3.9) I obtain

$$\hat{f}'_s\big|_{s=0} = \sum_{n=-\infty}^{\infty} c_n \frac{i}{\hbar}[\hat{\pi}_\theta, \hat{W}^n] = \sum_{n=-\infty}^{\infty} c_n in\hat{W}^n.$$

Besides, it is a known fact from spectral analysis that the real and imaginary parts of a bounded operator (i.e. an operator whose spectrum is bounded from above and below) are self-adjoint operators. $\hat{W}^n$s are bounded operators, so $\hat{f}$ is a bounded self-adjoint operator if $c_n = c^*_{-n}$. As specific examples of $\hat{f}$ we may take the self-adjoint sine operator $\hat{S}$ and the self-adjoint cosine operator $\hat{C}$ to specify a point on a ring [31, 33].

$$\hat{C} = \frac{\hat{W} + \hat{W}^\dagger}{2} \quad (c_n = c^*_{-n} = \frac{1}{2}\delta_{n,1}), \tag{3.10}$$

$$\hat{S} = \frac{\hat{W} - \hat{W}^\dagger}{2i} \quad (c_n = c^*_{-n} = \frac{i}{2}\delta_{n,1}), \tag{3.11}$$

Another important operator is the single-valued periodic angle operator $\hat{\Theta}\,|\theta\rangle = (\theta \bmod 2\pi)\,|\theta\rangle$ [33]

$$\hat{\Theta} = \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{in}(\hat{W}^n - \hat{W}^{-n}) = -i\mathrm{Log}(\hat{W}) \tag{3.12}$$

which is the operator version of the "sawtooth function" $\Theta(\theta) = 2\sum_{n=1}^{\infty}(-1)^{n+1}n^{-1}\sin(n\theta) = -i\mathrm{Log}(e^{i\theta})$, i.e. the angle θ restricted in the region $[-\pi, \pi]$. These operators satisfy the commutation relations

$$[\hat{\pi}_\theta, \hat{C}] = i\hbar\hat{S}, \tag{3.13}$$

$$[\hat{\pi}_\theta, \hat{S}] = -i\hbar\hat{C}, \tag{3.14}$$

$$[\hat{\Theta}, \hat{\pi}_\theta] = i\hbar\{1 - \delta(\hat{\Theta} + \pi)\} \tag{3.15}$$

Now, one of the requirements to define time operators is that they satisfy a commutation relation similar to that of position and momentum: This requirement is necessary to derive time-energy uncertainty relations as well as for the unification of space and time in quantum mechanics. In this context, I propose that the quantization of physical time observables in ring systems, including time intervals and events, should be done based on the GWWR. One can either use $[\hat{H}, \hat{T}] = i\hbar$ with a restricted domain or consider a generalized time operator with a larger domain (i.e. many other physical states) such that $[\hat{H}, \hat{T}] = i\hbar\hat{K}'(0)$. There is no obvious reason that a time operator must satisfy the former case, especially when boundary conditions play an important role. If periodic position operators $\hat{f}$ are used instead of $\hat{\theta}$, then time operators are expected to obey a commutation relation similar to Eq. (3.5).

3.2.2 Derivation of generalized self-adjoint time operators

Using the quantization of ring systems based on the GWWR and the GCCR, I show that there exists a class of generalized self-adjoint time operators in ring systems with $\hat{K}'(0) = \hat{f}'_s\big|_{s=0}$. I take Eq. (3.5) as a starting point to derive these operators. Consider a

free particle with moment of inertia I and Hamiltonian $\hat{H} = \frac{\hat{\pi}_\theta^2}{2I}$. Consider the following commutation relation

$$\begin{aligned}[\hat{H}, \hat{W}^n] |\psi\rangle &= [\hat{H}, \hat{W}^n] \sum_{l=-\infty}^{\infty} |l\rangle \langle l|\psi\rangle \\ &= \sum_{l=-\infty}^{\infty} (E_{l+n} - E_l) |l+n\rangle \langle l|\psi\rangle ,\end{aligned}$$

where I used Eq. (3.8), $\hat{H}|l\rangle = E_l |l\rangle$ and the identity operator $\mathbf{1} = \sum_{l=-\infty}^{\infty} |l\rangle\langle l|$. The desired time operator is obtained if we can get rid of $E_{l+n} - E_l = \frac{(2l+n)n\hbar^2}{2I}$. If n is an odd integer then $l = -\frac{n}{2}$ is not an eigenvalue of $\hat{\pi}_\theta$, so $1/(2l+n)$ is bounded. On the other hand, if n is an even integer, we can use the projection operator $\mathscr{P}_{-n/2}$ to remove the state $|-\frac{n}{2}\rangle$. In both cases, I define the time operator $\hat{T}$ as

$$\begin{aligned}\hat{T}|\psi\rangle &= -\sum_{n,l=-\infty}^{\infty} c_n n\hbar \frac{(1-\delta_{l,-\frac{n}{2}})}{E_{l+n} - E_l} |l+n\rangle \langle l|\psi\rangle , \\ |\psi\rangle \in D(\hat{T}) &= \left\{ \sum_{l=-\infty}^{\infty} a_{nl} |l\rangle : \sum_{l=-\infty}^{\infty} |a_{nl}|^2 < \infty \right\}.\end{aligned}$$

This kind of time operator has been studied in refs. [32,67,68]. The commutation relation between this time operator and the Hamiltonian gives

$$\begin{aligned}[\hat{H}, \hat{T}] |\psi\rangle &= -\sum_{n,l=-\infty}^{\infty} c_n n\hbar (1-\delta_{l,-\frac{n}{2}}) |l+n\rangle \langle l|\psi\rangle \\ &= -\sum_{n=-\infty}^{\infty} c_n n\hbar \hat{W}^n |\psi\rangle + \sum_{n=-\infty}^{\infty} c_{2n} 2n\hbar |n\rangle \langle -n|\psi\rangle \\ &= i\hbar \hat{f}_s'\big|_{s=0} |\psi\rangle + \sum_{n=-\infty}^{\infty} c_{2n} 2n\hbar |n\rangle \langle -n|\psi\rangle , \\ |\psi\rangle \in D(\hat{H}\hat{T}) &\cap D(\hat{T}\hat{H}).\end{aligned}$$

Here, $|n\rangle$ are linearly independent, hence the last term with c_{2n} vanishes only if $c_{2n}\langle -n|\psi\rangle = 0$ for all n. Finally, I define the bounded self-adjoint operator which has the dimension of

time

$$\hat{\mu}_n = \begin{cases} \frac{2I}{2\hat{\pi}_\theta + n\hbar} & , \ n = \text{odd} \\ \mathscr{P}_{-n/2} \frac{2I}{2\hat{\pi}_\theta + n\hbar} \mathscr{P}_{-n/2} & , \ n = \text{even} \end{cases}$$

and write our time operator as

$$\begin{aligned} \hat{T} &= -\sum_n c_n \hat{W}^n \hat{\mu}_n \\ &= -\sum_n \frac{(c_n \hat{W}^n \hat{\mu}_n + c_n^* \hat{\mu}_n \hat{W}^{-n})}{2}, \end{aligned} \tag{3.16}$$

$$[\hat{H}, \hat{T}] \left|\psi\right\rangle = i\hbar \hat{f}_s' \big|_{s=0} \left|\psi\right\rangle, \tag{3.17}$$

$$\left|\psi\right\rangle \in \left\{ \left|\psi\right\rangle \in D(\hat{T}) : c_{2n} \left\langle -n|\psi\right\rangle = 0, \forall n \right\}.$$

The symmetrized form in the second line is obtained from the fact that $\hat{W}^n \hat{\mu}_n = \hat{\mu}_{-n} \hat{W}^n$ and $c_n^* = c_{-n}$. This time operator has the following properties:

1. $\hat{T}$ is a bounded symmetric operator, hence it is self-adjoint. Another reason that this operator is self-adjoint is that the Hamiltonian has discrete energy spectrum [32,65,67,68]. Discreteness of the energy spectrum is very important but, as we have seen, it is not the only factor that determines the time operators.

2. $\hat{T}$ satisfies the GWWR (3.3) with $(\hat{A}, \hat{B}, \hat{K}(t)) = (\hat{T}, \hat{H}, \hat{T}(t) - \hat{T})$, $\hat{T}(t) = e^{i\hat{H}t/\hbar} \hat{T} e^{-i\hat{H}t/\hbar}$. One can readily show that $d\hat{K}(t)/dt|_{t=0} = -\hat{f}_s' \big|_{s=0}$ and obtain the correct generalized commutation relation.

3. Note that $\hat{T}$ is not unique because, for any self-adjoint operator $\hat{F}$ which commutes with the Hamiltonian, $\hat{T}$ and the new operator $\hat{T} + \hat{F}$ satisfy the same commutation relation with $\hat{H}$. Such operators can be added if necessary. However, if $\hat{f}$ is prescribed then $\hat{T}$ is unique up to addition of $\hat{F}$. Which $\hat{f}$ we choose depends on physical basis, i.e. on the phenomena (such as time of arrival, time crystal or other events) that we want to observe.

4. If we choose $\hat{f}$ such that $R\hat{f} \to \hat{x}$ in the infinite-volume limit $R \to \infty$, then the com-

mutation relation Eq. (3.17) reduces to the canonical commutation relation (3.1). This limit must be taken for $|\psi\rangle$ in the domain $\mathcal{D} \subset D(\hat{H}\hat{T}) \cap D(\hat{T}\hat{H})$ such that $|\psi\rangle \in \mathcal{D}$ is also square-integrable for $R \to \infty$. This domain includes the Gaussian-like minimum-uncertainty state considered by S. Tanimura [31].

5. $\hat{T}$ is defined as a linear combination of non-Hermitian operators $\hat{W}^n\mu_n$. These operators have space-time reversal ($\mathcal{PT}$) symmetry. The significance of $\mathcal{PT}$-symmetric time operators is discussed in section 3.4.

Now, let us consider some special cases and discuss their physical significance. If $c_n = c^*_{-n} = \frac{i}{2}\delta_{n,1}$ then obtain

$$\hat{T}_{S^1} = \frac{1}{2i}[\hat{\mu}_1\hat{W}^\dagger - \hat{W}\hat{\mu}_1] = \mathrm{Im}[\hat{\mu}_1\hat{W}^\dagger], \tag{3.18}$$

which satisfies the commutation relation

$$[\hat{H},\hat{T}_{S^1}] = i\hbar\hat{C}. \tag{3.19}$$

Note that $\hat{T}_{S^1}$ reduces to $\hat{T}_{\mathbb{R}}$ in the infinite-radius limit $R \to \infty$. Using $x = R\theta$, $k = l/R$, $p = \hbar k$, $\langle\theta|l\rangle = \langle x|k\rangle$, and $I = mR^2$, one can verify that

$$\begin{aligned}
\langle\theta|\hat{T}_{S^1}|l\rangle &= \frac{1}{2i}\langle\theta|(\hat{\mu}_1\hat{W}^\dagger - \hat{W}\hat{\mu}_1)|l\rangle \\
&= \frac{1}{2i}\left(\frac{2mR^2}{(2l-1)\hbar}e^{-i\theta} - \frac{2mR^2}{(2l+1)\hbar}e^{i\theta}\right)\langle\theta|l\rangle \\
&= -\frac{m}{2}\left(\frac{i}{k^2\hbar} + \frac{2x}{k\hbar}\right)\langle x|k\rangle + O(R^{-2}).
\end{aligned}$$

Then, from the commutation relation $[\hat{x},\hat{p}^{-1}] = -i\hbar/\hat{p}^2$ and equation (3.2) I obtain

$$\langle\theta|\hat{T}_{S^1}|l\rangle = \langle x|\hat{T}_{\mathbb{R}}|k\rangle + O(R^{-2}).$$

Similarly, one can show that $R\hat{S} \to \hat{x}$ and $\hat{C} \to 1$ as $R \to \infty$, hence

$$\begin{aligned}
&\hat{T}_{S^1} \to \hat{T}_{\mathbb{R}}, \\
&[\hat{S}, \hat{\pi}_\theta] = i\hbar\hat{C} \to [\hat{x}, \hat{p}] = i\hbar, \\
&[\hat{H}, \hat{T}_{S^1}] = i\hbar\hat{C} \to [\hat{H}, \hat{T}_{\mathbb{R}}] = i\hbar,
\end{aligned}$$

as $R \to \infty$. Therefore, I conclude that $\hat{T}_{S^1}$ is a self-adjoint analogue of the Aharonov-Bohm time operator $\hat{T}_{\mathbb{R}}$ in S^1.

If $c_n = c^*_{-n} = \frac{1}{2}\delta_{n,1}$, then I obtain the time operator $\hat{T}^{\mathrm{Re}}_{S^1}$

$$\hat{T}^{\mathrm{Re}}_{S^1} = \hat{\mu}_1 - \frac{1}{2}[\hat{W}\hat{\mu}_1 + \hat{\mu}_1\hat{W}^\dagger] \tag{3.20}$$

which satisfies the commutation relation

$$[\hat{H}, \hat{T}^{\mathrm{Re}}_{S^1}] = -i\hbar\hat{S}. \tag{3.21}$$

Note that $\hat{\mu}_1$ in Eq. (3.20), which commutes with the Hamiltonian $\hat{H}$, was included so that the matrix elements of $\hat{T}^{\mathrm{Re}}_{S^1}$ do not diverge in the limit $R \to \infty$. The physical significance of $\hat{T}^{\mathrm{Re}}_{S^1}$ can be understood by taking the large radius limit. Using $I = mR^2$, $l = Rk = Rp/\hbar$ and $\theta = x/R$, I find that $\hat{T}^{\mathrm{Re}}_{S^1}$ has matrix elements

$$\begin{aligned}
\langle\theta|\hat{T}^{\mathrm{Re}}_{S^1}|l\rangle &= \frac{2I}{\hbar}\frac{(1 - 2l + 2l\cos\theta - i\sin\theta)}{1 - 4l^2}\langle\theta|l\rangle \\
&= -\frac{1}{4\pi}\frac{\lambda_{\mathrm{dB}}}{v}\langle\theta|l\rangle + O(R^{-1}).
\end{aligned}$$

In the second line I did a Taylor expansion for $\theta = x/R \ll 1$. Here $\lambda_{\mathrm{dB}} = 2\pi\hbar/p$ is the de Broglie wavelength and $v = p/m$ is the group velocity of the particle. The term λ_{dB}/v describes a "matter wave clock", i.e. because of the periodicity of de Broglie wavelength, a particle moving with a fixed velocity v has an internal clock with period λ_{dB}/v [77, 78].

Note that $\hat{S}$ in Eq. (3.21) vanishes as $R \to \infty$. So, although $T_{S^1}^{\mathrm{Re}}$ is a time operator which satisfies the GWWR, it is not a time operator in the sense of Eq. (3.1).

The third example is for the periodic angle operator $\hat{\Theta}$ defined in Eq. (3.12). The corresponding time operator is defined as

$$\hat{T}_\Theta = -\sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{in}(\hat{W}^n \hat{\mu}_n - \hat{\mu}_n \hat{W}^{-n}) \tag{3.22}$$

which satisfies the commutation relation

$$[\hat{H}, \hat{T}_\Theta] = -i\hbar\{1 - \delta(\hat{\Theta} + \pi)\}. \tag{3.23}$$

The delta function $\delta(\hat{\Theta} + \pi)$ has a contribution only if $\theta + (2n+1)\pi = 0$. Otherwise $\hat{T}_\Theta$ satisfies the canonical commutation relation and it is equivalent to Galapon's time operator $\hat{T}_0$ for periodic system. Therefore, $\hat{T}_\Theta$ is interpreted as a time-of-arrival operator.

3.3 CONNECTION TO TIME CRYSTALS

How are these time operators connected to QTC? For the previous QTC model in Chapter 2, the Heisenberg operator $\hat{C}(t) = e^{i\hat{H}t/\hbar}\hat{C}e^{-i\hat{H}t/\hbar}$ describes the local oscillation of an incommensurate charge density wave: this oscillation is intrinsic to a ring system and it is used to model a QTC [79]. Other periodic operators $\hat{f}$ can also be used to model QTC in ring systems: Note that $\hat{W}^n$, as a momentum raising operator, can be written as $\hat{W}^n = \sum_l |l+n\rangle\langle l|$. Then, because $(E_{l+n} - E_l)/(E_{0+n} - E_0) = 2l + n$ is an integer, the Heisenberg operators $\hat{W}(t) = e^{i\hat{H}t/\hbar}\hat{W}e^{-i\hat{H}t/\hbar}$, $\hat{f}(t) = e^{i\hat{H}t/\hbar}\hat{f}e^{-i\hat{H}t/\hbar}$ and $\hat{T}(t) = e^{i\hat{H}t/\hbar}\hat{T}e^{-i\hat{H}t/\hbar}$ have a

periodic time evolution with period $P = 2\pi(E_1 - E_0)^{-1} = 4\pi I/\hbar$:

$$\begin{aligned}\hat{W} &= \sum_{l=-\infty}^{\infty} |l+n\rangle \langle l| \, e^{it(E_{l+n}-E_l)/\hbar} \\ &= \sum_{l=-\infty}^{\infty} |l+n\rangle \langle l| \, e^{i(t+P)(E_{l+n}-E_l)/\hbar} \\ &= \hat{W}(t+P),\end{aligned}$$

$$\Rightarrow \hat{T}(t) = \hat{T}(t+P) \text{ and } \hat{f}(t) = \hat{f}(t+P).$$

The period P diverges as $R \to \infty$, so this periodicity is intrinsic to ring systems. In fact, for a one-dimensional free particle, it is clear from $\hat{x}(t) = \hat{x} + \hat{p}t/m$ that

$$\hat{T}_{\mathbb{R}}(t) = \hat{T}_{\mathbb{R}} + t \tag{3.24}$$

is not periodic. The commutation factor $\hat{K}(t) = \hat{T}(t) - \hat{T}$ of a ring system is also periodic with a radius-dependent period. On the other hand, for a one-dimensional system ($\mathbb{R}$) we have $\hat{K}(t) = t$ which implies Eq. (3.1), hence time for a one-dimensional system is not necessarily periodic. Therefore, $\hat{K}(t)$ may be interpreted as a function which gives the "temporal structure" of a quantum system.

Because I am using a general mathematical structure (Eq. (3.3)) to construct time operators, the method in this chapter is not limited to ring systems but applies to other QTC models as well. I surmise $\hat{K}(t)$ characterizes the time-periodic evolution of a QTC. For the QTC model in chapter 2, this quantity is the charge density operator $\hat{C}(t)$. QTC states can also be realized in excited Floquet states [21–28]. If a Floquet system were driven with a period P, a Floquet time crystal (FTC) would return to its initial state after period nP (n is an integer), hence time translation symmetry is spontaneously broken.

So, I conjecture the following set of operators for a FTC

$$\begin{aligned} [\hat{H}(t), \hat{T}(t)] &= \dot{\hat{K}}(t), \\ \hat{H}(t+P) &= \hat{H}(t), \\ \hat{T}(t+nP) &= \hat{T}(t), \\ \hat{K}(t+nP) &= \hat{K}(t). \end{aligned} \tag{3.25}$$

For instance, the Hamiltonians $\hat{H}(t)$ for a typical FTC model is composed of Pauli matrices σ_i. Let $\hat{H}(t+P) = \hat{H}(t) = \begin{cases} \sigma_1, & 0 \leq t < \pi, \\ 0, & \pi \leq t < P \end{cases}$ be a prototype of a Floquet Hamiltonian, then I can identify $\hat{T}(t) = U^\dagger(t)\sigma_2 U(t), \hat{K}(t) = \hat{T}(t) - T(0)$, and $\dot{\hat{K}}(t) = 2U^\dagger(t)\sigma_3 U(t)$ with the unitary time evolution $U(t) = e^{-\frac{i}{\hbar}\int_0^t \hat{H}(t')dt'}$. In this case, $\hat{T}$ and $\hat{K}(t)$ are periodic with period $2P$. As a GCCR I obtain the $SU(2)$ commutation relation $[\sigma_1, \sigma_2] = 2i\sigma_3$. Further investigation of this conjecture is left for future study.

Therefore, it seems like time operators and time crystals are interrelated. The periods of a QTC seems to promote time from a parameter to a physical quantity. So, QTC are promising systems to define time operators. In spite of recently proposed models of QTC in excited states [21–28], the original idea of a QTC is to define a dynamical *ground state* which breaks time translation symmetry [1]. How to construct a QTC ground state is a very important open problem in quantum mechanics, quantum field theory, condensed matter physics, and related fields, because it concerns the question of what a ground state is. The existence of QTC ground states have been criticized by showing that spontaneous braking of time translation symmetry at ground state does not occur in the infinite-volume limit [15, 17]. However, if the periodic boundary condition is not negligible, the results in previous sections suggest how a time crystal can be defined in ring systems. We will have a QTC ground state if $\hat{K}(t) = \hat{T}(t) - \hat{T}$ and $d\hat{K}(t)/dt$ have periodic expectation value at ground state. Instead of a ring system we can also consider other systems with non-trivial real-space topologies. Once we have the appropriate mathematical structure to define a time operator (such as Eq. (3.3)), then we can apply it to models of time

crystals, possibly including ground states.

3.4 EXTENSION TO $\mathcal{PT}$-SYMMETRIC TIME OPERATORS

In the previous sections, I focused on self-adjoint time operators because real eigenvalues and orthogonal eigenstates are not ensured for non-self-adjoint symmetric operators. Here, I extend time operators to $\mathcal{PT}$-symmetric operators for the following reasons. First, if $\mathcal{PT}$ symmetry is not spontaneously broken, then $\mathcal{PT}$-symmetric operators have real eigenvalues and bi-orthogonal eigenstates [70–72, 80]. In other words, time operators do not have to be self-adjoint to have real-eigenvalues and orthogonal eigenstates. Second, one cannot completely distinguish from the canonical commutation relation and from the time-energy uncertainty relation if a time operator is self-adjoint or $\mathcal{PT}$-symmetric. This argument is elaborated in section 3.5. Therefore, it is possible that many time-operators are not self-adjoint (not even Hermitian) but $\mathcal{PT}$-symmetric. The third reason is of academic interest: So far, the main emphasis of $\mathcal{PT}$-symmetric quantum mechanics is on the spectral properties of $\mathcal{PT}$-symmetric Hamiltonians. So, $\mathcal{PT}$-symmetric time operators open a door to study other non-Hermitian operators, which can give us real eigenvalues and (bi-)orthonormal eigenstates. Therefore, in this section I define $\mathcal{PT}$ symmetry in ring systems. Then, I derive a $\mathcal{PT}$-symmetric time operator. For simplicity I will focus only on the $\mathcal{PT}$-symmetric extension of (3.18). Other $\mathcal{PT}$-symmetric operators can be defined likewise.

Before I define $\mathcal{PT}$-symmetric time operators it may be appropriate to discuss their validity as observables. Observables in quantum mechanics, by definition, are operators which can be measured experimentally. Self-adjoint operators are observables because real eigenvalues and orthogonal eigenstates are ensured. But, this does not mean that observables must be self-adjoint. In fact, $\mathcal{PT}$-symmetric operators can be measured [72]. In addition, nowadays non-Hermitian operators can also be measured by methods like weak

measurement. Therefore, in my opinion, $\mathcal{PT}$-symmetric operators are genuine observables which can be measured experimentally.

3.4.1 $\mathcal{PT}$-symmetry for ring systems

For quantum mechanics in one-dimension $\mathbb{R}$, the parity operator $\mathcal{P}$ changes the sign of the momentum operator $\hat{p}$ and the sign of position operator $\hat{x}$: $\hat{p} \to -\hat{p}$ and $\hat{x} \to -\hat{x}$. Time reversal $\mathcal{T}$ has the effect $\hat{p} \to -\hat{p}$, $\hat{x} \to \hat{x}$, and $i \to -i$. $\mathcal{T}$ changes the sign of i because $\mathcal{T}$ is required to preserve the fundamental commutation relation $[\hat{x}, \hat{p}] = i\hbar$ [70]. For quantum mechanics in S^1, the parity operator $\mathcal{P}$ satisfies the following properties [31]: $\mathcal{P}^\dagger\mathcal{P} = \mathcal{P}^2 = 1$, $\mathcal{P}\hat{W}\mathcal{P}^{-1} = \hat{W}^\dagger$, $\mathcal{P}\hat{\pi}_\theta\mathcal{P}^{-1} = -\hat{\pi}_\theta$, and $\mathcal{P}\,|l\rangle = |-l\rangle$. I show that the time reversal operator $\mathcal{T}$ shares the same properties. The time reversal operator for a bosonic particle is an anti-unitary operator which satisfies $\mathcal{T}^\dagger\mathcal{T} = \mathcal{T}^2 = 1$ [81]. Time reversal changes the direction of motion of a particle in S^1, i.e. $\hat{\pi}_\theta\mathcal{T}\,|l\rangle = -l\hbar\mathcal{T}\,|l\rangle$ should be satisfied. Therefore, we have $\mathcal{T}\,|l\rangle = a_l\,|-l\rangle$ with a coefficient a_l. Anti-unitarity of $\mathcal{T}$ implies $a_l = 1$. Then, using Eq. (3.9) we readily obtain

$$
\begin{aligned}
&\mathcal{P}^\dagger\mathcal{P} = \mathcal{P}^2 = 1,\ \mathcal{T}^\dagger\mathcal{T} = \mathcal{T}^2 = 1,\\
&\mathcal{P}\hat{\pi}_\theta\mathcal{P}^{-1} = -\hat{\pi}_\theta,\ \mathcal{P}\hat{W}\mathcal{P}^{-1} = \hat{W}^\dagger,\ \mathcal{P}\,|l\rangle = |-l\rangle\,,\\
&\mathcal{T}\hat{\pi}_\theta\mathcal{T}^{-1} = -\hat{\pi}_\theta,\ \mathcal{T}\hat{W}\mathcal{T}^{-1} = \hat{W}^\dagger,\ \mathcal{T}\,|l\rangle = |-l\rangle\,.
\end{aligned}
$$

So, $\hat{\pi}_\theta$, $\hat{W}$ and $|l\rangle$ are $\mathcal{PT}$-symmetric:

$$
\begin{aligned}
(\mathcal{PT})\hat{\pi}_\theta(\mathcal{PT})^{-1} &= \hat{\pi}_\theta,\\
(\mathcal{PT})\hat{W}(\mathcal{PT})^{-1} &= \hat{W},\\
(\mathcal{PT})\,|l\rangle &= |l\rangle\,.
\end{aligned}
$$

Note that the Hermitian time operators in Eq. (3.2) and Eq. (3.18) change their signs under $\mathcal{PT}$, in agreement with their interpretation as quantization for the time variable t. (For quantum mechanics on S^1, $\mathcal{T}\hat{W}\mathcal{T}^{-1} = \hat{W}^\dagger$ is possible only if $\mathcal{T}$ changes the sign of

i.)

3.4.2 $\mathcal{PT}$-symmetric time operator

Now, I define a $\mathcal{PT}$-symmetric time operator by

$$\hat{T}_{S^1}^{\mathcal{PT}} = \hat{F} - \hat{W}\hat{\mu}_1,$$

where $\hat{F}$ is a real $\mathcal{PT}$-symmetric operator which commutes with $\hat{H}$. This operator satisfies the commutation relation

$$[\hat{H}, \hat{T}_{S^1}^{\mathcal{PT}}] \, |l\rangle = -\hbar \hat{W} \, |l\rangle \, . \tag{3.26}$$

Formally, the large radius limit is given by

$$\hat{T}_{S^1}^{\mathcal{PT}} = i\hat{x}\hat{p}^{-1} + (\hat{F} - \hat{\mu}_1) + O(R^{-1}),$$

so the matrix elements of $\hat{T}_{S^1}^{\mathcal{PT}}$ diverge as $R \to \infty$ unless $\hat{F}$ is chosen carefully. The simplest form is $\hat{F} = \hat{\mu}_1$. Thus, I obtain the *$\mathcal{PT}$-symmetric time operator in S^1*

$$\hat{T}_{S^1}^{\mathcal{PT}} = (1 - \hat{W})\hat{\mu}_1 = \hat{T}_{S^1}^{\mathrm{Re}} + i\hat{T}_{S^1}. \tag{3.27}$$

$\hat{T}_{S^1}$ and $\hat{T}_{S^1}^{\mathrm{Re}}$ are defined by Eq. (3.18) and Eq. (3.20), respectively. This $\mathcal{PT}$-symmetric operator is interpreted as an imaginary time operator $i\hat{T}_{S^1}$ (i.e. quantization of the imaginary time $\tau = it$) with the addition of a quantum correction. $\mathcal{PT}$ changes the sign of i as well as the sign of the time operator, hence an imaginary time operator is invariant under $\mathcal{PT}$. Conversely, it is also reasonable to define the time operator in Eq. (3.18) as the imaginary part of (3.27).

The eigenstates and eigenvalues of $\hat{T}_{S^1}^{\mathcal{PT}}$ are calculated using biorthogonal quantum mechanics [80]: Suppose that the time operator $\hat{T}_{S^1}^{\mathcal{PT}}$ and its Hermitian conjugate $(\hat{T}_{S^1}^{\mathcal{PT}})^\dagger$

satisfy the eigenvalue equations

$$\hat{T}_{S^1}^{\mathcal{PT}} |\phi_l\rangle = \tau_l |\phi_l\rangle , \tag{3.28}$$

$$(\hat{T}_{S^1}^{\mathcal{PT}})^\dagger |\chi_l\rangle = \tau_l^* |\chi_l\rangle . \tag{3.29}$$

Let us adopt the position representation $\hat{\pi}_\theta \to -i\hbar\frac{\partial}{\partial\theta}$ which implies $\hat{\mu}_1^{-1} \to \frac{\hbar}{I}\left(-i\frac{\partial}{\partial\theta} + \frac{1}{2}\right)$. Then, Eq. (3.28) and Eq. (3.29) are equivalent to the following differential equations:

$$\begin{aligned}
\phi_l(\theta) &= \frac{\hbar}{I}\left(-i\frac{\partial}{\partial\theta} + \frac{1}{2}\right)\Phi_l(\theta), \\
(1 - e^{i\theta})\Phi_l(\theta) &= \frac{\tau_l\hbar}{I}\left(-i\frac{\partial}{\partial\theta} + \frac{1}{2}\right)\Phi_l(\theta), \\
(1 - e^{-i\theta})\chi_l(\theta) &= \frac{\tau_l^*\hbar}{I}\left(-i\frac{\partial}{\partial\theta} + \frac{1}{2}\right)\chi_l(\theta),
\end{aligned}$$

which have the orthonormal solutions

$$\begin{aligned}
&\phi_l(\theta) = \phi_{l0}\left(1 - e^{i\theta}\right)e^{i\theta\nu_l}e^{-(\nu_l+\frac{1}{2})e^{i\theta}}, \\
&\chi_l(\theta) = \chi_{l0}e^{i\theta\nu_l^*}e^{(\nu_l^*+\frac{1}{2})e^{-i\theta}}, \\
&\int_{-\pi}^{\pi} d\theta\chi_l^*(\theta)\phi_m(\theta) = 2\pi\chi_{l0}^*\phi_{m0}\delta_{l,m} = \delta_{l,m},
\end{aligned}$$

where $\nu_l = \frac{I}{\tau_l\hbar} - \frac{1}{2}$ was introduced for brevity. The periodic boundary condition requires ν_l to be an integer. Therefore, I obtain the eigenvalues

$$\tau_l = \tau_l^* = \frac{2I}{(2\nu_l + 1)\hbar}. \tag{3.30}$$

These eigenvalues are interpreted as the time required for a free particle with velocity $v = (\nu_l + \frac{1}{2})\frac{\hbar}{mR}$ to move a distance R on the ring; i.e. the time required to make a full rotation is $2\pi\tau_l = 2\pi R/v$ (Fig. 3.4). The validity of this interpretation is supported by the generalized commutation relation. The Aharonov-Bohm time operator, for instance, is defined as an operator whose eigenvalue t corresponds to the time required for a particle to move a finite distance d. Although d/t gives the average ve-

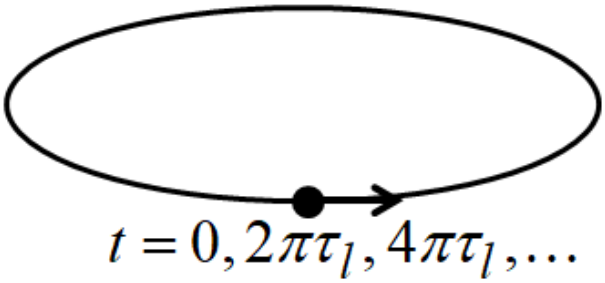

Figure 3.4: The eigenvalues τ_l of $\hat{T}^{\mathcal{PT}}_{S^1}$ describes the periodicity in time of a free particle moving in a ring with a constant velocity.

locity of the particle, the particle does not have a fixed momentum: Otherwise, the wave function of the particle is a momentum eigenstate ψ_l and expectation value with this state leads to contradictions (see Sec.(3.2.1)). On the other hand, one can verify that the expectation value of Eq. (3.26) with biorthogonal quantum mechanics implies $\langle\chi_l|[\hat{H}, \hat{T}^{\mathcal{PT}}_{S^1}]|\phi_l\rangle = -\hbar\,\langle\chi_l|\hat{W}|\phi_l\rangle = 0$. Therefore, our interpretation of time eigenvalues does not disagree with the commutation relation. More generally, a wave function in the domain of $\hat{T}^{\mathcal{PT}}_{S^1}$ has the form $|\Psi\rangle = \sum_l a_l\,|\phi_l\rangle$ and the corresponding wave function in the domain of $(\hat{T}^{\mathcal{PT}}_{S^1})^\dagger$ has the form $\langle\bar{\Psi}| = \sum_l a_l^*\,\langle\chi_l|$. Arrival time with these states is

$$\langle\bar{\Psi}|\hat{T}^{\mathcal{PT}}_{S^1}|\Psi\rangle = \sum_l |a_l|^2\tau_l. \tag{3.31}$$

Commutation relations do not have a vanishing expectation value in this case.

In the infinite-radius limit I obtain

$$\hat{T}^{\mathcal{PT}}_{S^1} \to i\hat{T}^{\mathrm{NH}}_{\mathbb{R}} = i\hat{T}_{\mathbb{R}} - \frac{m\hbar}{2}\hat{p}^{-2}. \tag{3.32}$$

As discussed after Eq. (3.21), the last additional term describes a "matter wave clock" which reflects the wave nature of a particle. The non-Hermitian time operator $\hat{T}^{\mathrm{NH}}_{\mathbb{R}}$ (which is not $\mathcal{PT}$-symmetric) was studied in ref. [73]. This operator satisfies the canonical commutation relation

$$[\hat{H}, \hat{T}^{\mathrm{NH}}_{\mathbb{R}}] = [\hat{H}, (\hat{T}^{\mathrm{NH}}_{\mathbb{R}})^\dagger] = [\hat{H}, \hat{T}_{\mathbb{R}}] = i\hbar. \tag{3.33}$$

Note that $\hat{T}_{S^1}^{\mathcal{PT}}$ is also non-Hermitian for finite radius. Also note that other $\mathcal{PT}$-symmetric time operators can be defined naturally. $i\hat{T}_{\mathbb{R}}^{\mathrm{NH}}$ is a non-Hermitian $\mathcal{PT}$-symmetric operator, but $\hat{T}_{\mathbb{R}}^{\mathrm{NH}}$ is not $\mathcal{PT}$-symmetric. In addition, $i\hat{T}_{\mathbb{R}}$ is $\mathcal{PT}$-symmetric as well, but $i\hat{T}_{\mathbb{R}}$ and $\hat{T}_{\mathbb{R}}$ give the same time-energy uncertainty relation as shown in the next section.

3.5 TIME-ENERGY UNCERTAINTY RELATIONS

In the previous sections we saw that generalized commutation relations are necessary to define position operators and time operators in ring systems. Similarly, position-momentum uncertainty relations and time-energy uncertainty relations are modified in ring systems. This modification is motivated by the following example. Consider the uncertainty relations $\Delta\hat{\theta}\Delta\hat{\pi}_\theta \geq \hbar/2$ and $\Delta\hat{H}\Delta\hat{T} \geq \hbar/2$. For a momentum eigenstate we obtain $\Delta\hat{\pi}_\theta = 0$ and $\Delta\hat{H} = 0$ which imply the contradiction $0 \geq \hbar/2$. This contradiction is avoided with generalized uncertainty relations.

The Robertson uncertainty relation [82] between two Hermitian operators is given by

$$(\Delta\hat{A})_\psi(\Delta\hat{B})_\psi \geq \frac{1}{2}\left|\langle[\hat{A},\hat{B}]\rangle_\psi\right|, \tag{3.34}$$

where $\langle\hat{A}\rangle_\psi = \langle\psi|\hat{A}|\psi\rangle$ and $(\Delta\hat{A})_\psi = \sqrt{\langle\hat{A}^2\rangle_\psi - \langle\hat{A}\rangle_\psi^2}$ is the norm of the state $[\hat{A} - \langle\hat{A}\rangle_\psi]\,|\psi\rangle$. For non-Hermitian operators we have a natural extension $(\Delta\hat{A})_\psi = \sqrt{\langle\hat{A}^\dagger\hat{A}\rangle_\psi - |\,\langle\hat{A}\rangle_\psi\,|^2}$. In general, $(\Delta\hat{A})_\psi \neq (\Delta\hat{A}^\dagger)_\psi$ if $\hat{A}$ and $\hat{A}^\dagger$ do not commute. Now, the Robertson uncertainty relation for non-Hermitian operators has a few different generalizations. First, Dou and Du [83] proposed the following generalization (which is written here with a different notation)

$$(\Delta\hat{A})_\psi^0(\Delta\hat{B})_\psi^0 \geq \frac{1}{2}\left|\langle[\hat{A},\hat{B}]\rangle_\psi\right|, \tag{3.35}$$

with $(\Delta\hat{A})_\psi^0 = [(\Delta\hat{A})_\psi + (\Delta\hat{A}^\dagger)_\psi]/2$ and so on. Therefore, we have, for instance, the

following uncertainty relations

$$(\Delta\hat{\pi}_\theta)_\psi(\Delta\hat{W})_\psi \geq \frac{\hbar}{2}\left|\langle\hat{W}\rangle_\psi\right|, \tag{3.36}$$

$$(\Delta\hat{H})_\psi(\Delta\hat{T}^{\mathcal{PT}}_{S^1})^0_\psi \geq \frac{\hbar}{2}\left|\langle\hat{W}\rangle_\psi\right|. \tag{3.37}$$

Second, Tanimura [31] proposed the following uncertainty relation

$$\begin{aligned}(\Delta\hat{\pi}_\theta)_\psi(\Delta\hat{W})_\psi &\geq \left|\langle[\hat{\pi}_\theta - \langle\hat{\pi}_\theta\rangle_\psi][\hat{W} - \langle\hat{W}\rangle_\psi]\rangle_\psi\right| \\ &= \left|\langle\hat{\pi}_\theta\hat{W}\rangle_\psi - \langle\hat{\pi}_\theta\rangle_\psi\langle\hat{W}\rangle_\psi\right|,\end{aligned} \tag{3.38}$$

which is a direct consequence of the Cauchy-Schwarz inequality. Similarly, we can obtain

$$(\Delta\hat{\pi}_\theta)_\psi(\Delta\hat{W})_\psi \geq \left|\langle\hat{\pi}_\theta\rangle_\psi\langle\hat{W}\rangle_\psi - \langle\hat{W}\hat{\pi}_\theta\rangle_\psi\right|, \tag{3.39}$$

$$(\Delta\hat{H})_\psi(\Delta\hat{T}^{\mathcal{PT}}_{S^1})_\psi \geq |\,\langle\hat{H}\hat{T}^{\mathcal{PT}}_{S^1}\rangle_\psi - \langle\hat{H}\rangle_\psi\langle\hat{T}^{\mathcal{PT}}_{S^1}\rangle_\psi\,|, \tag{3.40}$$

$$(\Delta\hat{H})_\psi(\Delta\hat{T}^{\mathcal{PT}\dagger}_{S^1})_\psi \geq |\,\langle\hat{H}\rangle_\psi\langle\hat{T}^{\mathcal{PT}}_{S^1}\rangle_\psi - \langle\hat{T}^{\mathcal{PT}}_{S^1}\hat{H}\rangle_\psi\,|. \tag{3.41}$$

We note that the uncertainty relations (3.38), (3.40) and (3.41) are not unique but other equivalent or inequivalent forms can be defined if necessary. Adding (3.40) to (3.41) and using the triangle inequality we obtain (3.37). Moreover, by adding (3.38) to the inequivalent form (3.39) we obtain (3.36). For the self-adjoint operators we have the usual Robertson uncertainty relations (3.34). $\mathcal{PT}$-symmetric time operators satisfy Eq. (3.35). In all cases, we see that the angle-angular momentum uncertainty relations and the energy-time uncertainty relations have the same lower bound. This fact is a consequence of the similarities between the commutation relations (3.26), (3.21), (3.19), (3.23) and (3.9), (3.13), (3.14), (3.15) respectively.

The significance of these uncertainty relations is as follows. First, uncertainties depend on wave functions and, on the contrary, the state of a ring system can be inferred from uncertainties. Under some circumstances, such as for non-localized momentum eigenstates, we have $|\,\langle[\hat{H},\hat{T}]\rangle\,| = 0$, i.e. uncertainty vanishes. We have $|\,\langle[\hat{H},\hat{T}]\rangle\,| = \hbar$ for a well-

localized state or for time operators introduced by Galapon and collaborators [65, 67], which reduces to the case of canonical commutation relation. In fact, the same situation appears for angle-angular momentum commutation relations as studied in ref. [31]. Second, we cannot completely distinguish from the canonical commutation relation and from the time-energy uncertainty relation if a time operator is self-adjoint or $\mathcal{PT}$ symmetric: For instance, the Aharonov-Bohm time operator defined in Eq. (3.2) and the non-Hermitian time operator in Eq. (3.32) give the same commutation relation (Eq. (3.33)) and the same time-energy uncertainty relation (as it can be seen from Eq. (3.33), Eq. (3.34), Eq. (3.35)). Therefore, if time operators are defined based on uncertainty relations, then it is possible that such time operators are not self-adjoint (not even Hermitian) but $\mathcal{PT}$-symmetric.

3.6 DISCUSSION AND CONCLUSION

First, I summarize the main results in this chapter. I defined a self-adjoint time operator $\hat{T}$ and a $\mathcal{PT}$-symmetric time operator $\hat{T}_{S^1}^{\mathcal{PT}}$ for a free particle on a ring system. $\hat{T}$ is a generalized time operator which satisfies Eq. (3.3). I studied three cases of $\hat{T}$, namely 1) $\hat{T}_{S^1}$ which can be used for the QTC model in Chapter 2, 2) $\hat{T}_{S^1}^{\mathrm{Re}}$ which can be interpreted as a "matter wave clock", and $\hat{T}_{\Theta}$ which is interpreted as a time-of-arrival operator and contains Galapon's time operator for periodic systems as a special case. The eigenstates and eigenvalues of $\hat{T}_{S^1}^{\mathcal{PT}}$ are calculated using biorthogonal quantum mechanics [80]. A summary of various time operators is given in Fig. 3.5[2].

Second, Galapon *et al.* also defined a self-adjoint time operator for a confined system with and without periodic boundary conditions which satisfy Eq. (3.1) [65, 66]. However, as we saw in this chapter, time operators should be defined based on the real-space topology of a quantum system. So, Eq. (3.1) may not be the only possibility to define self-adjoint time operators. The current work may also be generalized to relativistic particles,

[2]Note that $\mathcal{PT}$-symmetric operators are not necessarily non-Hermitian: For instance, $\hat{p}^2$ is a $\mathcal{PT}$-symmetric Hermitian operator.

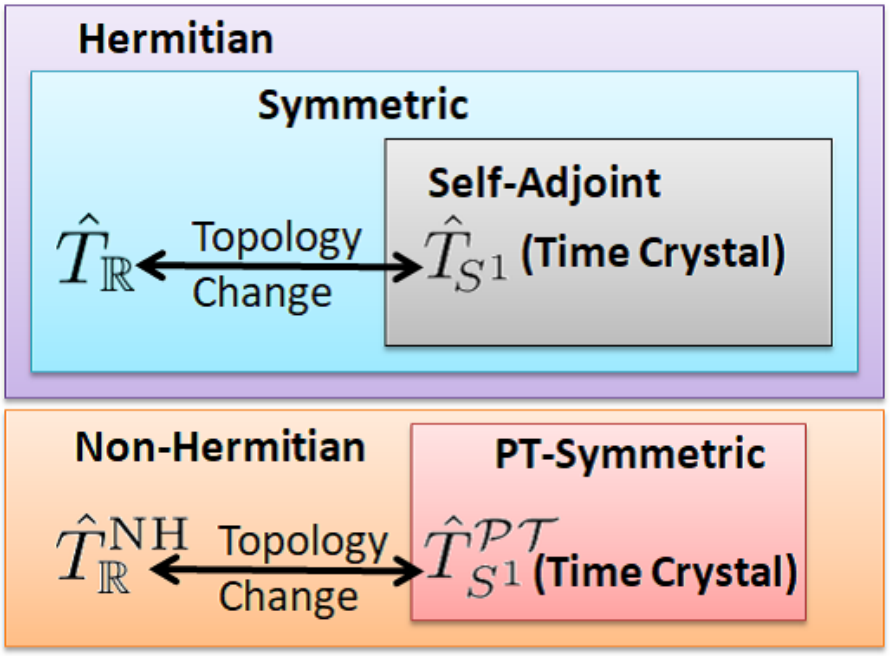

Figure 3.5: $\hat{T}_{S^1}$ [Eq. (3.18)] is a self-adjoint time operator and $\hat{T}^{\mathcal{PT}}_{S^1}$ [Eq. (3.27)] is a $\mathcal{PT}$-symmetric time operator. In the large radius limit (from S^1 to $\mathbb{R}$), $\hat{T}_{S^1}$ reduces to the Aharonov-Bohm time operator $\hat{T}_{\mathbb{R}}$ [Eq. (3.2)] and $\hat{T}^{\mathcal{PT}}_{S^1}$ reduces to the non-Hermitian operator $\hat{T}^{\mathrm{NH}}_{\mathbb{R}}$ [Eq. (3.32)].

but quantization of constrained systems is still a subject of current active research and the commutation relations Eq. (3.9) are not guaranteed to hold for relativistic systems as well.

Third, in order to obtain additional insights into QTC and understand how to create many other models of QTC, a universal origin of this periodicity is required. I surmise that this origin can be understood from Eq. (3.3) and $\hat{K}'(t)$ characterizes the time-periodic evolution of a QTC: For the QTC model in Chapter 2, this quantity is the charge density expectation value; and for Floquet time crystals [21–28] this quantity is typically the single-ion magnetization. In addition, we note that most of the early models of QTC [1, 15, 53, 54] are ring systems but the infinite-radius approximation was used. Therefore, these models should be reconsidered in light of the GWWR.

Fourth, an operator being $\mathcal{PT}$-symmetric does not necessarily mean that it has real eigenvalues, but it can have real eigenvalues. Typically, in $\mathcal{PT}$-symmetric systems with gain and loss (denoted by $\pm\gamma$) all eigenvalues are real below a critical value $\gamma < \gamma_c$ and

$\mathcal{PT}$-symmetry is spontaneously broken otherwise. Therefore, it would be an interesting problem to consider the spontaneous breaking of $\mathcal{PT}$-symmetry (of the Hamiltonian or the time operator) in periodically driven Floquet time crystals [21–28].

Moreover, possible applications of this work are as follows. 1) As I discussed in Sec. 3.3, the GWWR may give a universal explanation of time crystals [1, 15, 21–28, 53, 54, 79], where the commutation factor $\hat{K}(t) = \hat{T}(t) - \hat{T}$ gives the temporal structure of the system. 2) Time operators in this chapter are defined based on the position operator $\hat{f}$. Therefore, this work may be used to reexamine the relationship between space and time in the level of operators and understand space-time structures in quantum mechanics. 3) More radically, analysis of the cosmic microwave background anisotropy suggests that our universe is finite with a periodic structure [84]. If this is correct, then this work may be applied in quantum cosmology to understand, for instance, the space-time structure of the early (topologically non-trivial) universe.

Finally, we discuss the results in this chapter in the context of experiments. Suppose we want to study experimentally the arrival time or other events of a ring system. According to Sec. 3.2.1, time operators with the CCR can be studied only for special kinds of states and experiments must be well-designed. On the other hand, the new time operators can be used to study time of arrival or other events for general states in ring systems. For example, the order parameter of a charge density wave (CDW) or a superconductor is a complex scalar $\Delta = |\Delta| e^{i\theta}$. Suppose that we can quantize the phase θ by $\hat{W} = \widehat{e^{i\theta}}$. Then the commutation relations Eq. (3.19), Eq. (3.21), and Eq. (3.26) give the Heisenberg equations of motion of the time operators $\frac{d}{dt}\hat{T}_{S^1}(t) = -\hat{C}(t)$, $\frac{d}{dt}\hat{T}^{\mathrm{Re}}_{S^1}(t) = \hat{S}(t)$, and $\frac{d}{dt}\hat{T}^{\mathcal{PT}}_{S^1}(t) = -i\hat{W}(t)$, respectively. $\hat{C}(t)$ can describe charge density fluctuation and $\hat{S}(t)$ can describe time-dependent Josephson current. Therefore, time operators in periodic systems can be tested in various CDW [40, 79] or superconducting systems (including topological crystals such as ring crystals and Möbius strip crystals [40, 85]), in superfluid systems with spatial periodicity [15, 54] and in ring-shaped ion traps [53, 56]. $\hat{T}_\Theta$ can

be tested by measuring the time-of-arrival of a sine-Gordon-soliton in annular Josephson junctions [55], or we can use any other systems which can be described by a single particle in S^1. Experimental uncertainties will be given by the results in Sec. 3.5. Uncertainties depend on wave functions and, on the contrary, the state of a ring system can be inferred from uncertainties. Furthermore, uncertainties themselves can be used to test the new formulation in experiments. Experimental scheme to study uncertainty relations is possible and has been used in a different context (such as [86]). I believe similar experiments are possible for ring systems.

Chapter 4

MULTIVALLEY FREE ENERGY LANDSCAPE AND THE ORIGIN OF STRIPE AND QUASI-STRIPE CDW STRUCTURES IN MONOLAYER MX_2 COMPOUNDS

4.1 INTRODUCTION

Usually, anisotropic structures such as stripe phases can be explained by anisotropic interaction between the constituent atoms, electrons, or liquid crystal polymers [87]. Anisotropic structures in charge density waves (CDWs) have been explained likewise [88–91]. CDWs are periodic modulations of electric charge density in low-dimensional conductors [38, 92–95]. The stability of CDWs containing domain stripe walls [96, 97] and quasi-stripe (triclinic) domain walls [97–99] have been explained by Coulomb interaction between domain walls [88, 89] and three-dimensional stacking [90]. However, are these interactions indispensable? It would be ideal to have rich structures with the least amount of interactions.

In this chapter, I report that stripe and quasi-stripe CDW domain walls can appear without anisotropic interactions and explain the origin of these phases by topological defect theory (line defects in a two-dimensional plane) and interference between harmonics of macroscopic CDW wave functions. I consider the transition metal dichalcogenide (MX2) compounds $1T$-TaS_2 and $2H$-$TaSe_2$ (Figure 1 (a),(b)) which have recently experienced a resurgence of interest due to rich physical content and potential applicability to nanoscale electromechanics [34, 100–111]. These materials exhibit various CDW phases which are characterized by domain walls (topological defects). Interesting CDW phases are the stripe phase with stripe domain walls and the triclinic (T) phase with quasi-stripe domain walls, which surprisingly show up only on heating from the C phase (Figure 1 (d) and (e)).

I use a Ginzburg-Landau model for a CDW with general wave vectors $\mathbf{Q}^{(i)}$ $(i = 1, 2, 3)$ satisfying the triple-Q condition $\mathbf{Q}^{(1)} + \mathbf{Q}^{(2)} + \mathbf{Q}^{(3)} = 0$ (see Methods for detail). Historically, this model was first introduced by McMillan [88, 112, 113] to explain the IC-C phase transition. Nakanishi and Shiba analyzed this model more carefully by including higher order harmonics in the order parameter configuration, finding the presence of the NC phase in $1T$-TaS_2 [114] (Figure 4.1 (d)). Experimentally, the C, IC, NC, stripe and T phases all satisfy the triple-Q condition. It is noteworthy that the free energy only involves the terms compatible with the crystal symmetry and the only input from experimental data is the incommensurate wave vectors $\mathbf{Q}_{\mathrm{IC}}^{(i)}$ for the IC phase. Nevertheless, as shown by Nakanishi and Shiba, the free energy has an unexpected minimum corresponding to the NC phase. Thus, it is natural to ask whether there are more hidden minima if one makes a thorough search which was not feasible at the time.

I revisit the McMillan-Nakanishi-Shiba models [88, 114, 115] for monolayer $1T$-TaS_2 and $2H$-$TaSe_2$ without interaction between domain walls. First, I show that CDWs with $\mathbf{Q}^{(i)}$s that are separated by 120° (i.e. the three-fold rotation symmetry of the underlying

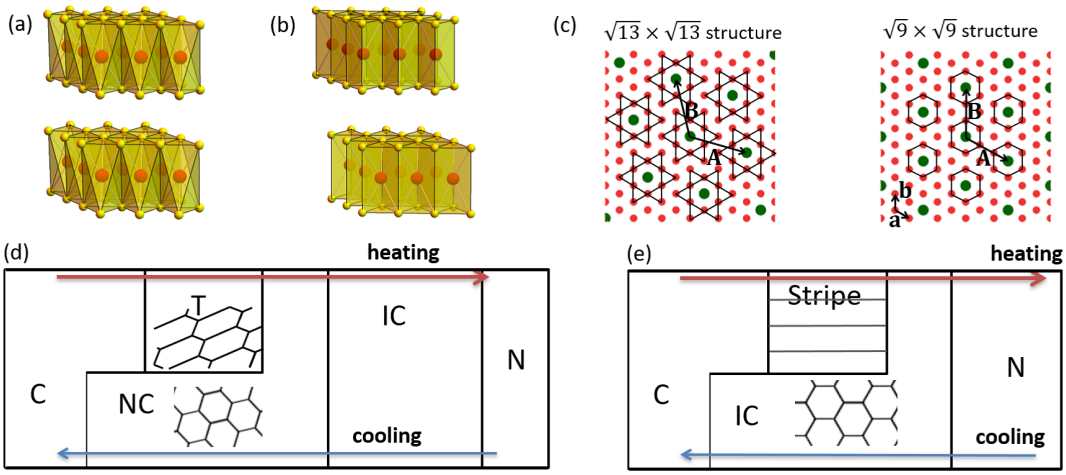

Figure 4.1: CDW structures in $1T$-TaS_2 and $2H$-$TaSe_2$. (a): Structure of $1T$-TaS_2. (b): Structure of $2H$-$TaSe_2$. These materials have two-dimensional layered crystalline structures like graphene. Here, the red spheres represent Ta atoms and the yellow spheres represent S or Se atoms. (c):The Ta atoms form a triangular lattice with superlattice vectors a and b. $\mathbf{A}$ and $\mathbf{B}$ are commensurate (C) CDW superlattice vectors ($\mathbf{A} = (\mu + \nu)\mathbf{a} + \nu\mathbf{b}, \mathbf{B} = -\nu\mathbf{a} + \mu\mathbf{b}, |\mathbf{A}| = |\mathbf{B}| = \sqrt{\mu^2 + \mu\nu + \nu^2}|\mathbf{a}|$). The CCDW superlattice for $1T$-TaS_2 ($\mu = 3, \nu = 1$) and $2H$-$TaSe_2$ ($\mu = 3, \nu = 0$) are shown with green spheres. (d) Temperature dependence of CDW phases in bulk $1T$-TaS_2. (e) Temperature dependence of CDW phases in bulk $2H$-$TaSe_2$. The blue arrows represent the cooling cycle. The red arrows represent the heating cycle. C: Commensurate phase, T: Triclinic phase with quasi-stripe domain walls (stretched honeycomb lattice). NC: Nearly commensurate phase, IC: Incommensurate phase, N: Normal metal phase. The domain walls of the NC, T, and stripe phases are depicted with solid lines.

lattice) contain a free-energy landscape with many local minima. Then, I remove this constraint and show that free energy local minima corresponding to the stripe and T phases appear. Finally, I explain the origin of stripe and T domain walls and discuss the implication of our results.

4.2 RESULTS

4.2.1 $1T$-TaS_2 CDW states with 120° constraint

First, I consider the special case that the CDW wave vectors $\mathbf{Q}^{(i)}$ ($i = 1, 2, 3$) are separated by 120°. In this case, the triple-Q condition $\mathbf{Q}^{(1)} + \mathbf{Q}^{(2)} + \mathbf{Q}^{(3)} = 0$ implies $|\mathbf{Q}^{(1)}| = |\mathbf{Q}^{(2)}| = |\mathbf{Q}^{(3)}|$. Numerical results of CDW free energy $F[\{\mathbf{Q}^{(i)}\}; T]$ at tempera-

ture T are shown in Figure 2. Each $\mathbf{Q}^{(i)}$s ($i = 1, 2, 3$) define a two-dimensional reciprocal space. To characterize the set $\{\mathbf{Q}^{(i)}\}$, I use two components of $\mathbf{Q}^{(1)}$ since they uniquely determine $\mathbf{Q}^{(2)}$ and $\mathbf{Q}^{(3)}$ by the triple-Q condition and the 120° constraint. The CDW free energy can be visualized by varying these components.

Figure 2 (a) reproduces the results by Nakanishi-Shiba which uses the special wave vector path $\mathbf{Q}^{(1)} = \mathbf{Q}(x)$ (shown in Figure 4.2 (b) with black lines). I have used the free-energy parameters in ref. [114]. Important wave vectors are the commensurate wave vectors $\mathbf{Q}_{\mathrm{C}}^{(i)}$ and the incommensurate wave vectors $\mathbf{Q}_{\mathrm{IC}}^{(i)}$ which have the norms $|\mathbf{Q}_{\mathrm{C}}^{(i)}|/|\mathbf{G}_i| = 1/\sqrt{13} \approx 0.277$ and $|\mathbf{Q}_{\mathrm{IC}}^{(i)}|/|\mathbf{G}_i| = 0.283$, respectively, and are tilted from the primitive reciprocal lattice vectors $\mathbf{G}_i$ by angles of 13.9° and 0°, respectively. $N = 0$ represents the charge density modulation by the fundamental wave $\mathbf{Q}^{(i)}$. The important idea by Nakanishi-Shiba is to include higher order harmonics with $N = 1, 2, 3, \ldots$ (see Figure 4.6). In fact, the NC phase appears for $N > 0$.

In this extended analysis, I have used two types of free energies: the first one (type-1) uses phenomenological parameters which reproduce a ring-like diffuse scattering obtained by an electron diffraction experiment [116], while the second one (type-2) uses McMillan's original free energy [88] analyzed with the method of Nakanishi-Shiba (see the Method section for more detail). The type-2 free energy is important because ring-like diffuse scattering were not observed for ultra-thin sheet of $1T$-TaS_2 including monolayer [34]. Figure 4.2 (b), (c), (d) show the free energy in the $\mathbf{Q}^{(1)}$ space. Note that the same free energy is obtained for $\mathbf{Q}^{(2)}$ and $\mathbf{Q}^{(3)}$ because of the three-fold rotational symmetry.

Surprisingly, the free energy has a "multivalley landscape": there are many local minima besides those corresponding to the well-known IC, NC, and C CDWs. Each of these local minima correspond to new CDW states with three-fold rotational symmetry. Despite of different global texture, type-1 and type-2 free energies show almost identical local minima. If the 120° constraint is removed, then these local minima are expected to

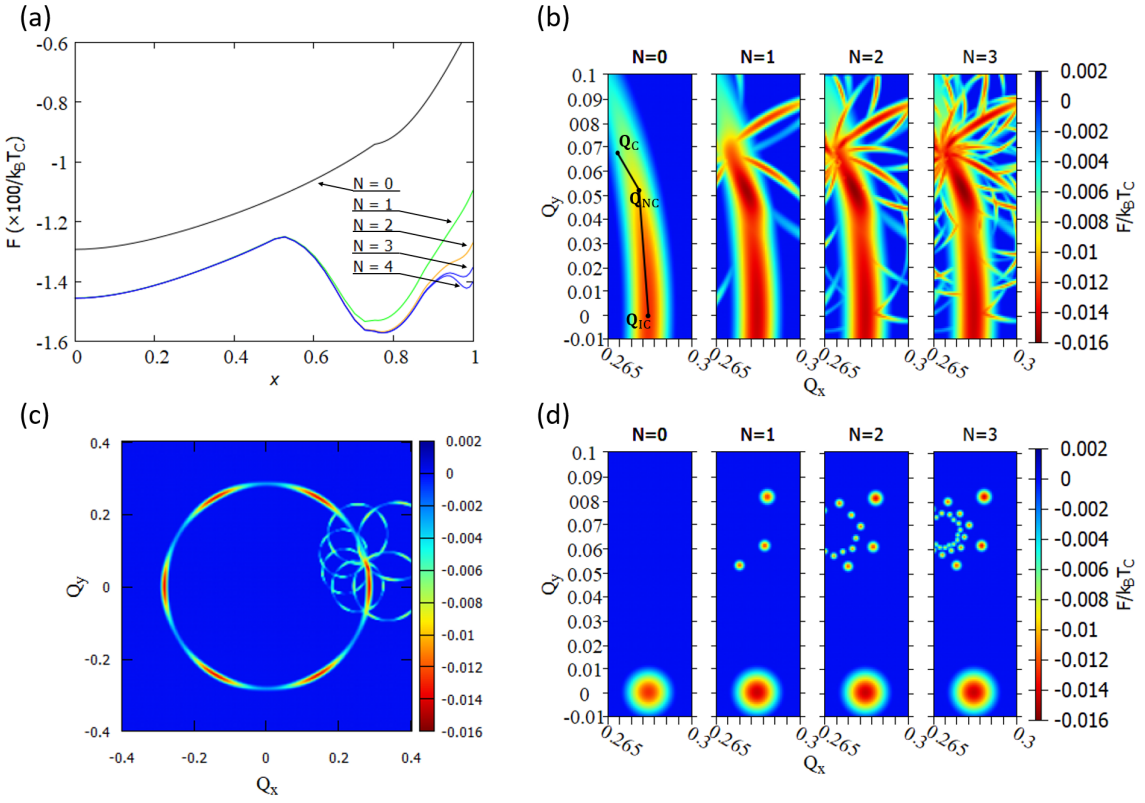

Figure 4.2: Numerical results of $1T$-TaS_2 free energy in the $\mathbf{Q}^{(1)} = (Q_x, Q_y)$ space (in units with $|\mathbf{G}_i| = 1$). (a) Reproduction of Nakanishi-Shiba's result [114]. x parameterizes the wave vector $\mathbf{Q}(x)$ with $\mathbf{Q}(0) = \mathbf{Q}_{\rm IC}$, $\mathbf{Q}(x^*) = \mathbf{Q}_{\rm NC}$ (nearly commensurate), and $\mathbf{Q}(1) = \mathbf{Q}_{\rm C}$. $\mathbf{Q}(x)$ is shown with solid black lines in (b). The "multivalley landscape" of the type-1 free energy with $N = 0$, $N = 1$, $N = 2$, and $N = 3$ are given in (b). (c) show the type-1 free energy for larger Q_x, Q_y values. Note that the number of branches and local minima increase as N increases. New local minima appear near $\mathbf{Q}_{\rm C}$. (d) The type-2 free energy with $N = 0$, $N = 1$, $N = 2$, and $N = 3$ show almost identical local minima, but it is easier to see how the CCDW states is obtained as $N \to \infty$.

move to new minima corresponding to the T phase.

4.2.2 $2H$-$TaSe_2$ CDW states with 120° constraint

The multivalley free energy structure also exists in $2H$-$TaSe_2$ as shown in Figure 4.3. The commensurate wave vectors $\mathbf{Q}_{\rm C}^{(i)}$ and the incommensurate wave vectors $\mathbf{Q}_{\rm IC}^{(i)}$ have the norms $|\mathbf{Q}_{\rm C}^{(i)}|/|\mathbf{G}_i| = 1/\sqrt{9} \approx 0.333$ and $|\mathbf{Q}_{\rm IC}^{(i)}|/|\mathbf{G}_i| = 0.325$, respectively, and they are parallel to the primitive reciprocal lattice vectors $\mathbf{G}_i$. Figure 4.3 (a) reproduces the free energy along the special line $Q_x = 0$ obtained by Nakanishi-Shiba [115] which explains the IC-C phase transition. I extend their analysis to the two-dimensional plane by varying

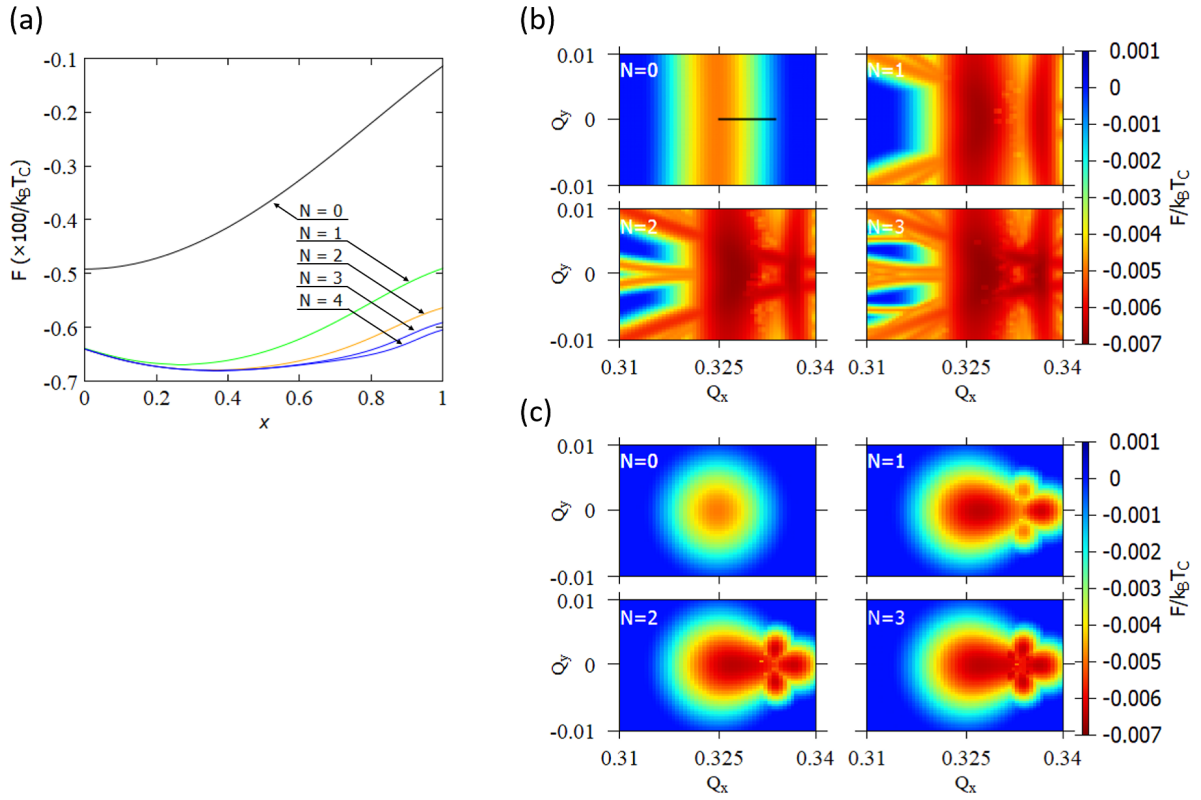

Figure 4.3: Numerical results of $2H$-$TaSe_2$ free energy in the $\mathbf{Q}^{(1)} = (Q_x, Q_y)$ space (in units with $|\mathbf{G}_i| = 1$). (a): Reproduction of the result by Nakanishi-Shiba [115]. x parameterizes the wave vector $\mathbf{Q}(x)$ with $\mathbf{Q}(0) = \mathbf{Q}_{\rm IC}$ and $\mathbf{Q}(1) = \mathbf{Q}_{\rm C}$. $\mathbf{Q}(x)$ is shown with a black line in (b) The "multivalley landscape" of the free energy with $N = 0$, $N = 1$, $N = 2$ and $N = 3$ are given for the type-1 (b) and type-2 (c) free energies

Q_x and find new local free energy minima like those of $1T$-TaS_2. Here, I have used the free-energy parameters in ref. [115]. These local minima correspond to CDW states with three-fold rotational symmetry. Despite different global texture, type-1 and type-2 free energies show almost identical local minima. If the 120° constraint is removed, then these local minima are expected to move to new minima corresponding to the stripe phase.

4.2.3 Anisotropic CDW Phases

Here, I consider CDW states without the 120° constraint. Let $R_\pm$ denote the matrices which rotate a wave vector $\mathbf{Q} = (Q_x, Q_y)$ by $\pm 120°$. If we set the 120° constraint, then the vectors $\{\mathbf{Q}^{(1)}, R_-\mathbf{Q}^{(2)}, R_+\mathbf{Q}^{(3)}\}$ are identical. If the 120° constraint is removed, then the triple-Q condition implies that the vertices of $\{\mathbf{Q}^{(1)}, R_-\mathbf{Q}^{(2)}, R_+\mathbf{Q}^{(3)}\}$ form a regular tri-

angle (see Figure 4.6). Consequently, stable or quasi-stable CDW states with anisotropic domain walls are obtained if local minima from the free energy shown with $\mathbf{Q}^{(1)}, \mathbf{Q}^{(2)}, \mathbf{Q}^{(3)}$ form a regular triangle.

First, I consider the T phase. From the triple-Q condition there are four independent degrees of freedom. I fix two degrees of freedom, namely, the angles between domain walls, to visualize the free energy. The angles between domain walls are known from experiments [99]]. Here, I consider the angles $\delta\phi_1 = 360^\circ - \delta\phi_2 - \delta\phi_3$, $\delta\phi_2 = 180^\circ - \phi_{\rm C}, \delta\phi_3 = 150^\circ$, where $\phi_{\rm C} \approx 13.9^\circ$ is the angle between $\mathbf{Q}_{\rm C}^{(i)}$ and $\mathbf{Q}_{\rm IC}^{(i)}$. Figure 4.4 (a) shows the free energy with $N = 1$. I have used the free energy parameters from Nakanishi-Shiba [114]. Then, I find a triplet of local minima which forms a regular triangle (black). This result is surprising because the T phase was previously explained by inter-layer interaction [90], but our calculation is done for a monolayer crystal. Experimental result of the T phase in bulk $1T$-TaS_2 (green triangle) [99] is in good agreement with the numerical calculation. The domain size in our calculation is smaller than bulk crystal but may become closer to experiment if we further consider Coulomb interaction between domain walls or inter-layer interaction. Aside from this quantitative detail, I conclude that anisotropic domain walls for the T phase can be formed without inter-layer interaction and, hence, in a monolayer $1T$-TaS_2. Next, I consider the stripe phase. According to experimental results [96], the stripe phase is obtained by minimizing the free energy with the constraint $\mathbf{Q}^{(1)} = \mathbf{Q}_{\rm C}^{(1)}$. Figure 4.4 (b) is the free energy ($N = 1$) visualized with $\mathbf{Q}^{(2)}$ and $\mathbf{Q}^{(3)}$. Clearly, these local minima form a regular triangle with $\mathbf{Q}_{\rm C}^{(1)}$. These domain walls correspond to the stripe phase. Again, the domain size is smaller than the stripe domain of bulk $2H$-$TaSe_2$ (green triangle) [96]. The domain size may become closer to experiment if we further consider Coulomb interaction between domain walls. Therefore, we conclude that anisotropic domain walls for the stripe phase can be formed naturally even in a monolayer $2H$-$TaSe_2$ with the least amount of interaction.

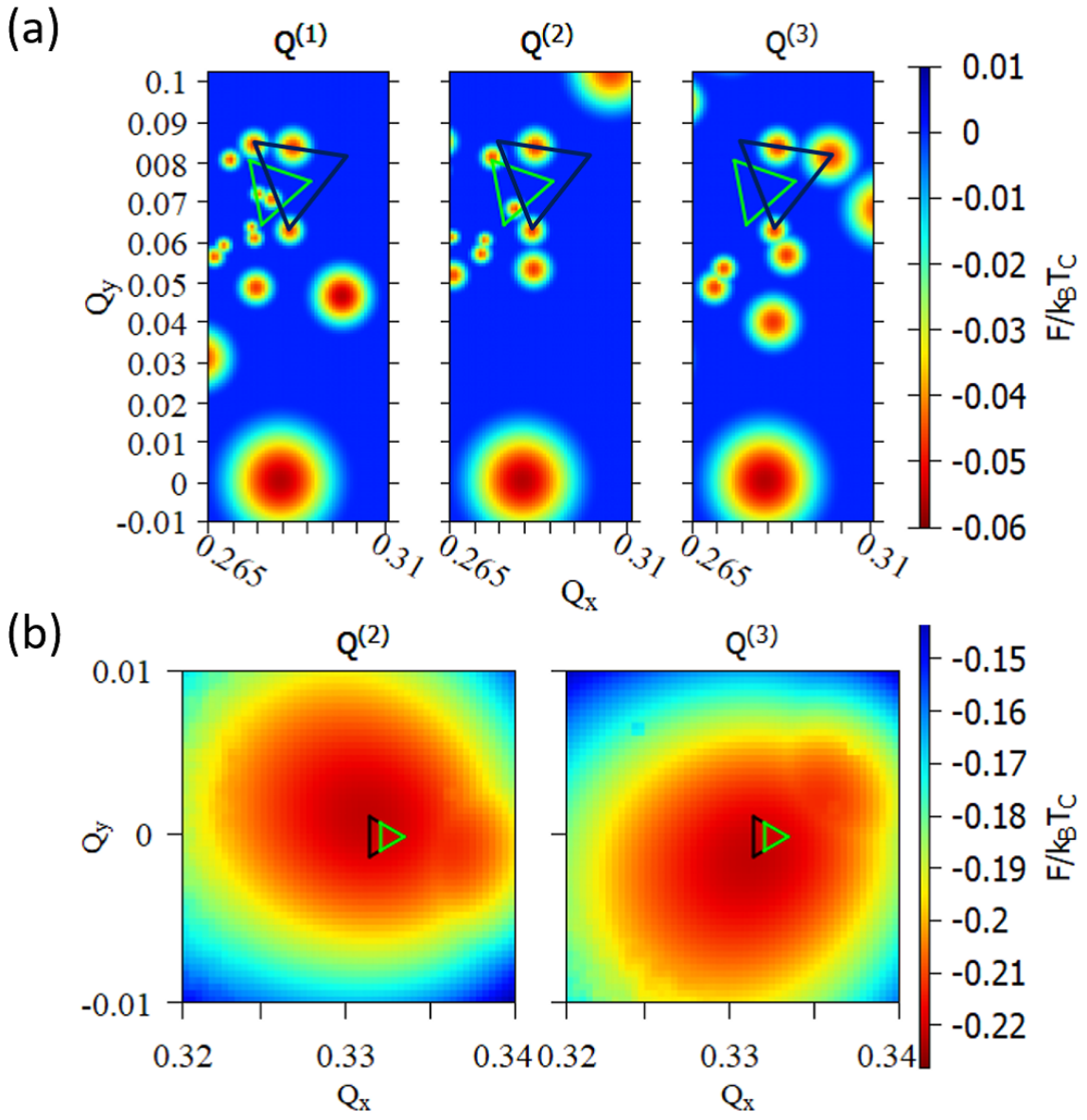

Figure 4.4: Free energy of T and stripe phases. (a) Free energy of $1T$-TaS_2 without the 120° constraint ($T = 225$K). Local minima for $\mathbf{Q}^{(1)}$, $\mathbf{Q}^{(2)}$ and $\mathbf{Q}^{(3)}$ form a regular triangle. These local minima correspond to the T phase with quasi-stripe domain walls. (b) Free energy of $2H$-$TaSe_2$ without the 120° constraint ($T = 100$K). The point $\mathbf{Q}^{(1)} = \mathbf{Q}_{\mathrm{C}}^{(1)}$ and local minima for $\mathbf{Q}^{(2)}$ and $\mathbf{Q}^{(3)}$ form a regular triangle. Therefore, these local minima correspond to the stripe phase.

4.3 DISCUSSION AND CONCLUSION

First, I summarize the results presented in the previous sections. I revisited the McMillan-Nakanishi-Shiba models for $1T$-TaS_2 and $2H$-$TaSe_2$ with general CDW wave vectors $\mathbf{Q}^{(i)}$ $(i = 1, 2, 3)$ separated by 120°. A "bird's-eye view" of CDW free energy landscape with multivalley structure reveal the presence of multiple local minima which correspond to different CDW states. Then, I removed the 120° constraint and observed local minima for the stripe and T phases. These results are surprising because we resorted to neither Coulomb interaction between domain walls nor inter-layer interaction, which had been considered as the origins of the stripe and phases.

To explain these results, I consider the following mechanism. CDW phases between the IC and C phases are accompanied with domain walls, where each domain contains a CCDW with a different phase: such domain walls minimize the commensurability energy between the CDW and the underlying lattice. The average domain size is inversely proportional to $|\mathbf{Q}^{(i)} - \mathbf{Q}_{\mathrm{C}}^{(i)}|$. As temperature decreases, $\mathbf{Q}^{(i)}$ approaches $\mathbf{Q}_{\mathrm{C}}^{(i)}$ and the domain size diverges: The ground state is the C phase without domain walls. As temperature increases from the ground state, energy increase due to thermal fluctuation induces domain walls. Since the formation of domain walls requires energy, these domain walls are created one by one. These domain walls are expected to be anisotropic rather than hexagonal, since the formation of hexagonal domain walls require a global arrangement. This model is analogous to the formation of the Abrikosov vortex lattice in a type-2 superconductor, where vortices are formed one by one and finally form a lattice structure as the number of vortices increase.

Moreover, for $1T$-TaS_2, the C phase is closer (in the reciprocal lattice space) to the T phase than the NC phase, i.e. the T phase has larger domains which is energetically favored. Therefore, as temperature increases, $1T$-TaS_2 will first make a phase transition from the C phase to the T phase; phase transition to the NC phase occurs when the NC domain size and the T domain size become comparable (Figure 4.5) [99].

Note that this induced anisotropy model has experimental support. The low temperature CDW phase of a free-standing monolayer $1T$-TaS_2 with multiple T domain walls has been observed with scanning transmission electron microscopy [34]. In addition, $1T$-TaS_2 with a single domain wall has been observed using scanning tunneling microscopy [117].

Next, I explain what condition distinguishes stripe and T domain walls. While $2H$-$TaSe_2$ has a parity-conserving commensurate structure (Figure 4.1(c)), $1T$-TaS_2 breaks parity in the first place, and hence they cannot have stripe domain walls. So, from the results in this chapter, I predict that MX_2 compounds with triple-CDW can have domain walls with an angle of $180° - \phi_{\mathrm{C}}$, where ϕ_{C} is the angle between $\mathbf{Q}_{\mathrm{C}}^{(i)}$ and $\mathbf{Q}_{\mathrm{IC}}^{(i)}$. From the commensurability condition conditions $\mu\mathbf{Q}_{\mathrm{C}}^{(i)} - \nu\mathbf{Q}_{\mathrm{C}}^{(i+1)} = \mathbf{G}_i$ where $\mathbf{G}_i$ are reciprocal lattices of the crystal, one can obtain obtain $\phi_{\mathrm{C}} = \cos^{-1}(\mu + \nu/2)/\sqrt{\mu^2 + \mu\nu + \nu^2}$. Then, we see that $\mu \neq 0$ and $\nu \neq 0$ gives T domain walls (such as $1T$-TaS_2 with $\mu = 3$ and $\nu = 1$), while $\mu \neq 0$ and $\nu = 0$ gives stripe domain walls (such as $2H$-$TaSe_2$ with $\mu = 3$ and $\nu = 0$). In particular, I predict that the ultrathin $TaSe_2$ with a new $\sqrt{7} \times \sqrt{7}$ commensurate structure (that is, $\mu = 2$ and $\nu = 1$) [118] can also have T domain walls.

The appearance of the stripe and T phases with the least amount of interaction can also be explained in the context of entropy. The Kosterlitz-Thouless transition [119, 120], for instance, is stabilized by the production of vortices. Creation of vortices requires energy, but vortices also increase entropy and minimize the free energy of the system. Similarly, stabilization of the stripe and T phases is related to entropy release/increase which is asymmetric with heating/cooling. The stripe phase and the T phase break the 3-fold rotational symmetry of the CCDW state. The T phase further breaks parity. Lower symmetry due to production of symmetry-breaking domain walls implies larger entropy, i.e. lower free energy. From these discussions, we conclude that the T and stripe phases can appear naturally even in monolayer MX_2: Coulomb interaction between domain walls or inter-layer interaction may be secondary factors for the appearance of these phases.

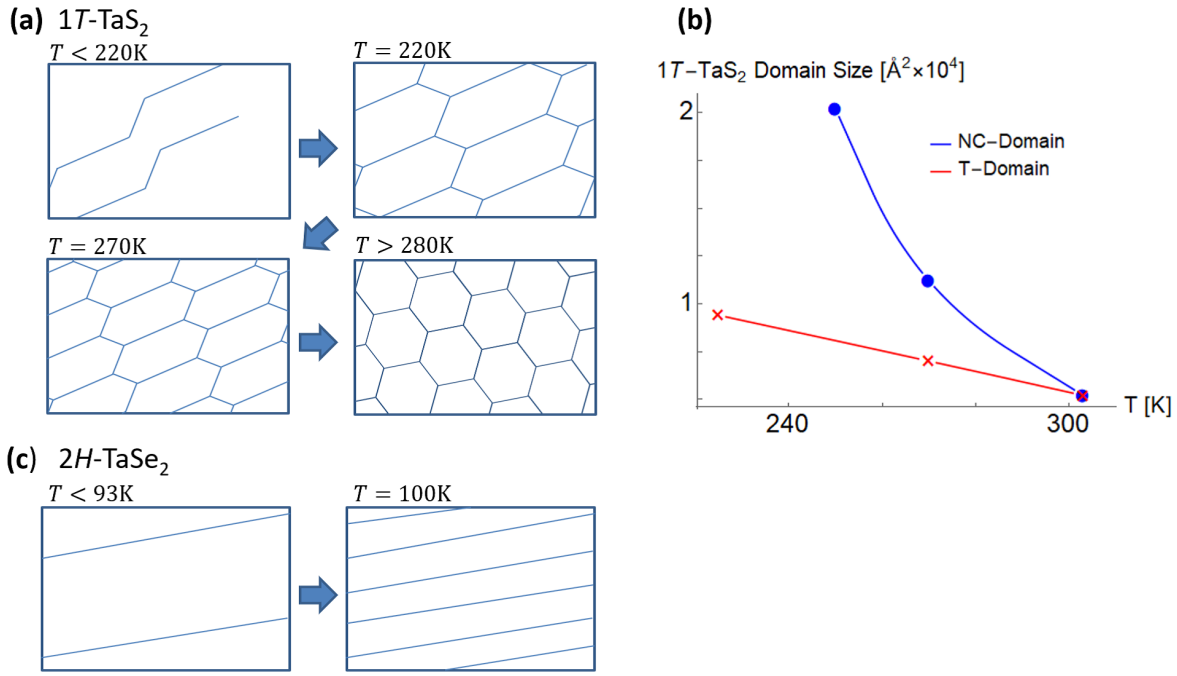

Figure 4.5: A model for the formation of anisotropic domain walls. (a): The ground state of $1T$-TaS_2 is the C phase without domain walls. As temperature increases, domain walls are formed one by one. As the number of domain walls increase, we obtain the T phase. The domain size decreases as temperature increase further. Then, T-NC phase transition occur when the NC domain size and the T domain size become comparable (280K for bulk $1T$-TaS_2). The temperature dependence is shown here for bulk $1T$-TaS_2 [99]. (b) Bulk data of $1T$-TaS_2 domain size. The T domain size (heating) are from ref. [99]. The NC domain size (cooling) are from ref. [125]. We note that the same NC domain sizes were verified but not reported in ref. [99]. (c) Similarly, the ground state of $2H$-$TaSe_2$ is the CCDW state without domain walls. As temperature increases, we speculate that domain walls are formed one by one. The temperature dependence is shown here for bulk $2H$-$TaSe_2$ [96].

Note that these results does not contradict with Nakanishi-Shiba's model of stripe and T phases based on inter-layer stacking [90]: their work still apply for MX_2 with higher number of layers. These results also explains my collaborated experimental result, namely the appearance of the T phase in monolayer $1T$-TaS_2. As future applications of this work, the appearance of various local minima may explain mysterious CDW states such as the "hidden CDW state" [108, 121–123] and these minima also predict new CDW phases, hence our model may become the basis to study CDW further. The analysis in this chapter may be extended to study the interplay between supersolid symmetry and lattice symmetry, and application to other van der Waals structures [124].

4.4 METHODS

McMillan's phenomenological free energy for triple CDW is [88, 112, 113]

$$F = \int d^2r \left\{ a(\mathbf{r})\alpha^2(\mathbf{r}) - b(\mathbf{r})\alpha^3(\mathbf{r}) + c(\mathbf{r})\alpha^4(\mathbf{r}) + \sum_{i=1}^{3} \psi_i^*(\mathbf{r}) e(-i\nabla)\psi_i(\mathbf{r}) \right. \tag{4.1}$$

$$\left. + d(\mathbf{r})[|\psi_1(\mathbf{r})\psi_2(\mathbf{r})|^2 + |\psi_2(\mathbf{r})\psi_3(\mathbf{r})|^2 + |\psi_3(\mathbf{r})\psi_1(\mathbf{r})|^2] \right\} \tag{4.2}$$

Here the complex order parameters $\psi_i(\mathbf{r})$ at position $\mathbf{r}$ are related to the electron charge density $\rho(\mathbf{r}) = \rho_0(\mathbf{r})1 + \alpha(\mathbf{r})$, $\alpha(\mathbf{r}) = \sum_{i=1}^{3} \mathrm{Re}[\psi_i(\mathbf{r})]$. $\rho_0(\mathbf{r})$ is the charge density in the normal state. The index i $(= 1, 2, 3)$ specifies the three components of the triple CDW. $a(\mathbf{r})$, $b(\mathbf{r})$, $c(\mathbf{r})$, $d(\mathbf{r})$ are position-dependent coefficients which have the periodicity of the crystal lattice and can be written in the form $a_0 + a_1 \sum_{i=1}^{6} e^{i\mathbf{G}_i\cdot\mathbf{r}}$, etc., where $\mathbf{G}_i$ the six shortest reciprocal lattice vectors. The commensurate wave vectors $\mathbf{Q}_{\mathrm{C}}^{(i)} = |\mathbf{Q}_{\mathrm{C}}^{(i)}|(\cos\phi_{\mathrm{C}i}, \sin\phi_{\mathrm{C}i})$ are defined by the conditions $\mu\mathbf{Q}_{\mathrm{C}}^{(i)} - \nu\mathbf{Q}_{\mathrm{C}}^{(i+1)} = \mathbf{G}_i$ $(i = 1, 2, 3)$, $|\mathbf{Q}_{\mathrm{C}}^{(i)}| = |\mathbf{G}_i|/\sqrt{\mu^2 + \mu\nu + \nu^2}$. Type-1 and Type-2 free energies explained in the Result section correspond to different forms of $e(-i\nabla)$. In their study of $1T$ and $2H$-polytypes, Nakanishi and Shiba considered $(\mu, \nu) = (3, 1)$ and $(\mu, \nu) = (3, 0)$, respectively, but their method can be generalized and applied to other CDW materials with different (μ, ν) values. The complex order parameters of a CCDW state is $\psi_i(\mathbf{r}) = \Delta_{\mathrm{C}} e^{i\mathbf{Q}_{\mathrm{C}}^{(i)}\cdot\mathbf{r} + i\theta_{(}\mathrm{C}i)}$ where $\mathbf{Q}_{\mathrm{C}}^{(i)}$ are the wave vectors of the CCDW state and $\theta_{\mathrm{C}i}$ are constant phases which minimize the free energy. For other CDW states, $\psi_i(\mathbf{r})$ can be expanded as a function of the deviation vectors $\mathbf{q}^{(i)} = \mathbf{Q}^{(i)} - \mathbf{Q}_{\mathrm{C}}^{(i)}$. Then we may expand $\psi_i(\mathbf{r})$ as $\psi_i(\mathbf{r}) = \sum_{l,m,n\geq 0, l\cdot m\cdot n=0} \Delta_{lmn}^{(i)} e^{i\mathbf{Q}_{lmn}^{(i)}\cdot\mathbf{r}}$ where l, m, n are integers and $\mathbf{Q}_{lmn}^{(i)} = l\mathbf{k}^{(i)} + m\mathbf{k}^{(i+1)} + n\mathbf{k}^{(i+2)} + \mathbf{Q}^{(i)}$, $\mathbf{k}^{(i)} = \mu\mathbf{q}^{(i)} - \nu\mathbf{q}^{(}(i+1))$ and $\mathbf{q}^{(i)} = \mathbf{Q}^{(i)} - \mathbf{Q}_{\mathrm{C}}^{(i)}$, and we have introduced the cyclic notations such as $\mathbf{q}^{(4)} = \mathbf{q}^{(1)}$ and $\mathbf{q}^{(5)} = \mathbf{q}^{(2)}$. The free-energy is obtained by varying general $\mathbf{Q}^{(i)}$s. In this case, we assume that each $\Delta_{lmn}^{(i)}$ are real. These wave vectors are depicted in Figure 4.6. $\Delta_{lmn}^{(i)}$ are determined by numerically calculating the set of coupled non-linear differential equations $\frac{\partial F}{\partial \Delta_{lmn}^{(i)}} = 0$.

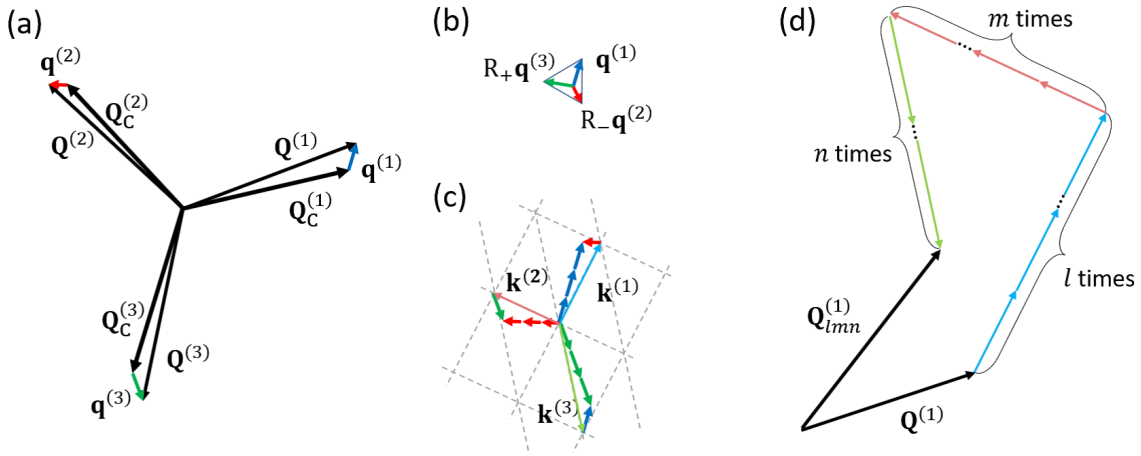

Figure 4.6: The elements of the McMillan-Nakanishi-Shiba model. (a) CCDW wave vectors $\mathbf{Q}_{\mathrm{C}}^{(i)}$ define the CCDW charge density $\rho(\mathbf{r}) = \rho_0[1 + \Delta_{\mathrm{C}} \sum_{i=1}^{3} \cos(\mathbf{Q}_{\mathrm{C}}^{(i)} \cdot \mathbf{r} + \theta_{\mathrm{C}i})]$. The free energy is calculated as a function of a general wave vector $\mathbf{Q}^{(i)}$ with the triple-Q condition $\mathbf{Q}^{(1)} + \mathbf{Q}^{(2)} + \mathbf{Q}^{(3)} = 0$. (b) The deviation vectors $\mathbf{q}^{(i)} = \mathbf{Q}^{(i)} - \mathbf{Q}_{\mathrm{C}}^{(i)}$ projected on the $\mathbf{Q}^{(1)}$ space form a regular triangle. Here, $R_{\pm}$ are rotation matrix of $\pm 120°$. If $\mathbf{Q}^{(i)}$ are separated by 120°, then this triangle reduces to a point. (c) The harmonics form a triangular lattice. $\mathbf{k}^{(i)}$ are wave vectors responsible for the formation of domain walls (the image shows an example for $(\mu, \nu) = (3, 1)$). (d) A general CDW state is given by $\rho(\mathbf{r}) = \rho_0[1 + \sum_{i=1}^{3} \sum_{l,m,n \geq 0, l \cdot m \cdot n = 0} \Delta_{lmn}^{(i)} \cos \mathbf{Q}_{lmn}^{(i)} \cdot \mathbf{r}]$ with $3N(N+1)$ higher harmonics.

Chapter 5

CONCLUSION OF THESIS

I studied crystalline orders in terms of macroscopic wave functions. In particular, I studied quantum time crystals and quantum space crystals as new orders in CDW systems. Like superconductors, the order parameter that characterize different CDW phases is a macroscopic wave function. However, unlike superconductors, CDWs are electronic crystals which break spatial translation symmetry. Therefore, CDWs were ideal to directly observe the effect of quantum mechanics on crystal phases. The main results are summarized as follows.

In chapter 2, I showed that time translation symmetry of a ring system with a macroscopic wave function is broken by decoherence. An important subtlety was that, because of strong quantum fluctuation, the ground state of an isolated finite-size quantum crystal is a superposition of lattices with different vertices. We had the same difficulty for a quantum time crystal in finite-size system, namely a superposition of time-periodic oscillation which preserves the symmetry of a Hamiltonian. So, I considered symmetry breaking by decoherence. In particular, I used a ring-shaped incommensurate charge density wave (ICDW ring) threaded by a fluctuating magnetic flux: the Caldeira-Leggett model was used to model the fluctuating flux as a bath of harmonic oscillators. The charge density expectation value of a quantized ICDW ring coupled to its environment oscillates periodically. This periodicity is a consequence of interference of macroscopic CDW wave

Table 5.1: Summary of time crystals and time operators

	Time crystal possible?	Topology	Macroscopic wave function?	Decoherence?	Time operator
Wilczek	Yes	Thermodynamic limit	Superconductor wave function	×	△
Bruno/ Watanabe-Oshikawa	No	Thermodynamic limit	Anything	×	×
Volovik	Yes (finite lifetime)	$\mathbb{R}^3$	Superfluid ^{3}He-A	×	△
This model	Yes (finite lifetime)	S^1	Ring CDW	◯	◯
Floquet time crystal	Excited state (finite lifetime)	Anything	Many-body interacting state	×	×

function in ring systems. Therefore, this model forms a metastable quantum time crystal with a finite length in space and in time.

In chapter 3, I investigated time operators in the context of quantum time crystals in ring systems. A generalized commutation relation called the *generalized weak Weyl relation* was used to derive a class of self-adjoint time operators for ring systems. I also revealed the relationship between these time operators and a $\mathcal{PT}$-symmetric time operator. Then, I discussed the connection between time operators, time crystals and real-space topology. In particular, the new time operators can be interpreted as order parameters which describe the "temporal structure" of a time crystal.

A summary of time crystals and time operators is given Table 5.1

In chapter 4, I use the McMillan-Nakanishi-Shiba free energy of study the effect of macroscopic wave function in two-dimensional CDW systems. I made a thorough numerical research of CDW free energy structure and discovered a multivalley free-energy landscape with many local minima. In addition, I discovered local minima which correspond to the triclinic phase and to the stripe phase. These results can be explained by topological defect theory (line defects in a two-dimensional plane) and interference between harmonics of macroscopic CDW wave functions, hence indicate the appearance of anisotropic CDW phases without inter-layer interaction (hence also in monolayer). Moreover, I discussed the temperature dependence of successive phase transition, generalized

the McMillan-Nakanishi-Shiba free energy theory for two-dimensional CDW phase transition, and predict the appearance of new CDW phases in $1T$-$TaSe_2$.

As a general conclusion of from these results, I discovered that the interference of macroscopic wave functions in topological systems give rise to new crystal orders in space and time.

Appendix A

POSITIVE-OPERATOR VALUED MEASUREMENT OF ICDW RING

A.1 PREPARATION

The operators, wave functions and density matrices for the ICDW ring-Probe system are summarized in Table A.1. Suppose we want to measure an ICDW ring from time $t = 0$ to $t = T$. The Hamiltonian of the entire system is

$$\hat{H}(t) = \hat{H}_s + \hat{V}(t) + \hat{H}_d \tag{A.1}$$

$$\hat{H}_s = \frac{(\hat{\pi}_\theta - \alpha\hbar)^2}{2I} \tag{A.2}$$

$$\hat{V}(t) = \delta(t - T)\hat{V}_0 \tag{A.3}$$

$$\hat{H}_d = \frac{\hat{P}^2}{2M}. \tag{A.4}$$

Here, we assume that the probe is heavy and $\hat{H}_d$ can be ignored. $\hat{V}_0$ is an interaction potential which depends on the operators $\hat{W}, \hat{\pi}_\theta, \hat{X}, \hat{P}$. Moreover, assume the form $\hat{V}_0 =$

	ICDW ring	Probe	Composite System
Commutation relation	$\hat{\pi}_\theta$, $[\hat{\pi}_\theta, \hat{W}] = \hbar\hat{W}$	$[\hat{X}, \hat{P}] = i\hbar$	
Wave function	$\psi(\theta) = \sum_l c_l e^{il\theta}$	$\Phi(X)$	$\Psi(\theta, X) = \psi(\theta)\Phi(X)$

Table A.1: Summary of operators and wave functions for the ICDW ring-Probe system

$g\hat{A}\otimes\hat{P}$ where g is the interaction strength $\hat{A}$ is a function of $\hat{W}$. From the Hamiltonians (A.1) we define the time evolution operators

$$U_s(t) \equiv \exp\left\{-\frac{i}{\hbar}\int_0^t \hat{H}_s dt'\right\} = \exp\left\{-\frac{it}{2I\hbar}(\hat{\pi}_\theta - \alpha\hbar)^2\right\} \tag{A.5}$$

$$U_d(t) \equiv \exp\left\{-\frac{i}{\hbar}\int_0^t \hat{V}(t')dt'\right\} = \exp\left\{-\frac{i}{\hbar}h(T-t)\hat{V}_0\right\} \tag{A.6}$$

$$U(t) \equiv \exp\left\{-\frac{i}{\hbar}\int_0^t \hat{H}(t')dt'\right\} = \exp\left\{-\frac{it}{2I\hbar}(\hat{\pi}_\theta - \alpha\hbar)^2 - \frac{i}{\hbar}h(T-t)\hat{V}_0\right\}. \tag{A.7}$$

Here, $h(T-t) = \int_0^t \delta(t-T)dt$ is the Heaviside step function. In general, we don't have $U(t) = U_s(t)U_d(t)$, so we compute the time evolution via the interaction picture. For the state $\Psi = \psi\otimes\Phi$ we define $\Psi_I(t) = U_s^\dagger(t)\Psi(t)$. Then, the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\Psi(t) = \hat{H}(t)\Psi(t) \tag{A.8}$$

is equivalent to

$$i\hbar\frac{\partial}{\partial t}\Psi_I(t) = \hat{H}_I(t)\Psi_I(t), \tag{A.9}$$

where

$$\hat{H}_I(t) = U_s^\dagger(t)\hat{V}(t)U_s(t) = \delta(t-T)U_s^\dagger(t)\hat{V}_0U_s(t) \tag{A.10}$$

and the time evolution of $|\Psi_I(t)\rangle$ is given by

$$\Psi_I(t) = U_I(t)\Psi_I, \tag{A.11}$$

$$U_I(t) = \mathcal{T}\exp\left\{-\frac{i}{\hbar}\int_0^t dt'\hat{H}_I(t')\right\} \tag{A.12}$$

$$= \mathcal{T}\exp\left\{-\frac{i}{\hbar}\int_0^t dt'\delta(t'-T)U_s^\dagger(t)\hat{V}_0U_s(t)\right\} \tag{A.13}$$

$$= \mathcal{T}\exp\left\{-\frac{i}{\hbar}U_s^\dagger(T)\hat{V}_0U_s(T)\right\} \tag{A.14}$$

$$= U_s^\dagger(T)U_d(T)U_s(T) \tag{A.15}$$

where $\mathcal{T}$ denotes time ordering.

A.2 WAVE FUNCTION AND DENSITY MATRIX AFTER MEASUREMENT

Now, the time evolution of the state $\Psi = \psi \otimes \Phi$ for time $t > T$ can be written as

$$\Psi(t) = U_s(t)\Psi_I(t) \tag{A.16}$$
$$= U_s(t)U_I(t)\Psi \tag{A.17}$$
$$= U_s(t)U_s^\dagger(T)U_d(T)U_s(T)\Psi \tag{A.18}$$
$$= U_s(t-T)U_d(T)U_s(T)\Psi \tag{A.19}$$

If we choose $\hat{V}_0 = g\hat{A}\otimes\hat{P}$ then $U_d(t)$ can be regarded as a translation operator $U_d(T)\hat{X}U_d(T) = \hat{X} + g\hat{A}$. Then, using $U_s^\dagger(t)\hat{X}U_s(t) = \hat{X}\ \forall t$ and $U_s^\dagger(T)(\hat{X}+\hat{A})U_s(T) = \hat{A}(t)$, the expectation value of $\hat{X}$ can be calculated directly.

$$\langle\Psi(t), \hat{X}\Psi(t)\rangle = \langle U_s(t-T)U_d(T)U_s(T)\Psi, \hat{X}U_s(t-T)U_d(T)U_s(T)\Psi\rangle \tag{A.20}$$
$$= \langle\Psi, U_s^\dagger(T)U_d^\dagger(T)U_s^\dagger(t-T)\hat{X}U_s(t-T)U_d(T)U_s(T)\Psi\rangle \tag{A.21}$$
$$= \langle\Psi, U_s^\dagger(T)U_d^\dagger(T)\hat{X}U_d(T)U_s(T)\Psi\rangle \tag{A.22}$$
$$= \langle\Psi, U_s^\dagger(T)(\hat{X}+\hat{A})U_s(T)\Psi\rangle \tag{A.23}$$
$$= \langle\Psi, (\hat{X} + g\hat{A}(T))\Psi\rangle \tag{A.24}$$
$$= \langle\Psi, \hat{X}\Psi\rangle + \langle\Psi_I, g\hat{A}(T)\Psi\rangle \tag{A.25}$$

Finally, assume that $\Phi(X) = \Phi(-X)$ and we obtain $\langle\Psi_I, \hat{X}\Psi_I\rangle = 0$. Therefore,

$$\langle\Psi(t), \hat{X}\Psi(t)\rangle = g\langle\Psi, \hat{A}(T)\Psi\rangle . \tag{A.26}$$

This result can be understood intuitively. Suppose that a charge particle (probe) with position $\hat{X}$ arrives nearby the CDW and interacts with the CDW with strength g. Then, for $\hat{A} = \hat{n}$ (charge density operator) the probe trajectory is deviated by the charge density expectation value.

A.3 MEASUREMENT OPERATOR

Consider a measurement of CDW at time $t = 0^+$. Then, from the interaction picture we obtain

$$\Psi \to U_I(0^+)\Psi = U_s^\dagger(0^+)U_d(0^+)U_s(0^+)\,|\psi\rangle\,|\Phi\rangle \approx U_d(0)\Psi. \tag{A.27}$$

Now, define $\hat{M}(X) = [U_d(0^+)\Phi](X)$, then the reduced density matrix of the CDW becomes

$$\hat{\rho}_s^{red} \equiv= \mathrm{tr}_d[\rho] \int dX \hat{M}(X)\hat{\rho}_s\hat{M}^\dagger(X). \tag{A.28}$$

From the trace identities $\mathrm{tr}[\hat{A}\hat{B}] = \mathrm{tr}[\hat{B}\hat{A}]$ and $\mathrm{tr}[\rho] = 1$ we obtain

$$\begin{aligned} 1 &= \mathrm{tr}_s[\hat{\rho}_s^{red}] && \text{(A.29)}\\ &= \int dX \mathrm{tr}_s[\hat{M}(X)\hat{\rho}_s\hat{M}^\dagger(X)] && \text{(A.30)}\\ &= \int dX \mathrm{tr}_s[\hat{M}^\dagger(X)\hat{M}(X)\hat{\rho}_s] && \text{(A.31)} \end{aligned}$$

which implies the normalization condition

$$\int dX \hat{M}^\dagger(X)\hat{M}(X) = 1. \tag{A.32}$$

If we define $\hat{E}(X) = \hat{M}^\dagger(X)\hat{M}(X)$ then the normalization condition is equivalent to

$$\int dX \hat{E}(X) = 1. \tag{A.33}$$

Moreover, by the definition of $\hat{M}(x)$ and $\hat{E}(x)$ one can show that

$$\langle\psi|\hat{E}(X)|\psi\rangle > 0 \tag{A.34}$$

i.e. $\hat{E}(X)$ is a positive operator. From this result and Eq. (A.33) we can define the set $\{\hat{E}(X)\}$ as a positive-operator valued measure.

Now, consider a projection measurement of ICDW ring. The charge density in the true ground state $\psi = \psi_0$ (zero-momentum state) is uniform

$$\langle\psi_0|\hat{\rho}(x,t)|\psi_0\rangle = \rho_0, \tag{A.35}$$

but the true ground state is projected to an effective state as the ICDW system is measured. The effective ground state is then expected to relax to the true ground state within a finite time. Define an effective ground state

$$|\xi_a\rangle = \sum_{n\in\mathbb{Z}} \int_{-\infty}^{\infty} A_a e^{-\frac{(\xi'-\xi+2\pi n)^2}{2a^2}} |\xi'\rangle \, d\xi' \tag{A.36}$$

where A_a is a normalization constant. This wave function can be obtained formally by a POVM

$$\hat{M}_a \equiv \sum_{n\in\mathbb{Z}} \int_{-\infty}^{\infty} A_a e^{-\frac{(\xi'-\xi+2\pi n)^2}{2a^2}} |\xi'\rangle \langle\xi'| \, d\xi'. \tag{A.37}$$

For $a < \pi$, this states can be approximated by a Gaussian state

$$|\xi_a\rangle \approx \int_{-\infty}^{\infty} \left(\frac{1}{2\pi a^2}\right)^{1/4} e^{-\frac{(\xi'-\xi)^2}{2a^2}} |\xi'\rangle \, d\xi'. \tag{A.38}$$

Then, using $\langle\xi_a, \hat{W}\xi_a\rangle = e^{i\xi}e^{-a^2/4}$, the charge density becomes

$$\langle\xi_a|\hat{\rho}(x,0)|\xi_a\rangle = \rho_0 + e^{-a^2/4}\rho_1 \cos(2k_{\mathrm{F}}x + \xi). \tag{A.39}$$

In the $a \to \infty$ limit (or, more practically, for $a \gg \pi$), the electron charge density is uniform as in equation (A.35). This is the true ground state of the ICDW ring system. On the other hand, in the $a \to 0$ limit, the charge density is equivalent to the classical charge density Eq. (2.1). The energy difference between the measured state and the true ground state is

$$\Delta E \approx \frac{\hbar^2}{4Ia^2}, \tag{A.40}$$

where I is the moment of inertial of the ICDW, and the measured state will relax to the

true ground state within

$$\tau_E \sim \frac{\hbar}{\Delta E} \approx \frac{4IRa^2}{\hbar}. \tag{A.41}$$

Appendix B

QUANTUM BROWNIAN MOTION IN S^1

B.1 REDUCED DENSITY MATRIX ON S^1

Consider an ICDW ring A coupled to its environment B. Let ρ_A and ρ_B denote the density operators of A and B, respectively. Let θ and ϕ be angular coordinates on the ring system A and let $\mathbf{p} = \{p_k : k = 1, \mathcal{N}\}$ and $\mathbf{s} = \{s_k : k = 1, \mathcal{N}\}$ be momentum coordinates of the bath B. Then, the density matrix of the coupled system can be written

$$\begin{aligned}\rho(\theta, \mathbf{p}, \phi, \mathbf{s}) &= \langle \theta, \mathbf{p} | \rho_{AB}(t) | \phi, \mathbf{s} \rangle \\ &= \int_0^{2\pi} d\theta' d\phi' \int_{-\infty}^{\infty} d\mathbf{p}' d\mathbf{s}' K(\theta, \mathbf{p}, t; \theta', \mathbf{p}', 0) K^*(\phi, \mathbf{s}, t; \phi', \mathbf{s}', 0) \\ &\quad \times \langle \theta', \mathbf{p}' | \rho_{AB}(0) | \phi', \mathbf{s}' \rangle\end{aligned}$$

where

$$K(\theta, \mathbf{p}, t; \theta', \mathbf{p}', 0) = \langle \theta, \mathbf{p} | e^{-\frac{i}{\hbar}\hat{H}t} | \theta', \mathbf{p}' \rangle \tag{B.1}$$

and

$$K^*(\phi, \mathbf{s}, t; \phi', \mathbf{s}', 0) = \langle \phi', \mathbf{s}' | e^{\frac{i}{\hbar}\hat{H}t} | \phi, \mathbf{s} \rangle \tag{B.2}$$

are recognized as Feynman propagators if we notice that $|\mathbf{p}\rangle$ and $|\mathbf{s}\rangle$ are actually position states after canonical transformation. The propagators (B.1) and (B.2) can be written using path integrals by dividing the time t into N time steps of length $\epsilon = t/(N+1)$, $\mathbf{p} = \mathbf{p}_N$, $\mathbf{p}' = \mathbf{p}_0$, $\theta = \theta_N$ and $\theta' = \theta_0$. For $N \to \infty$ the propagator becomes

$$\begin{aligned} K(\theta, \mathbf{p}, t; \theta', \mathbf{p}', 0) &= \left\langle \theta, \mathbf{p} \left| \lim_{\epsilon \to 0} \left(\exp -\frac{i\epsilon}{\hbar} \hat{H} \right)^N \right| \theta', \mathbf{p}' \right\rangle \\ &= \lim_{\epsilon \to 0} \int_0^{2\pi} \left(\prod_{n=1}^{N-1} d\theta_n \right) \int_{-\infty}^{\infty} \left(\prod_{n=1}^{N-1} d\mathbf{p}_n \right) \prod_{n=1}^{N} K_n(\theta_n, \mathbf{p}_n, \epsilon; \theta_{n-1}, \mathbf{p}_{n-1}, 0). \end{aligned}$$

The Hamiltonian operator $\hat{H}$ can be decomposed into the kinetic part $\hat{\mathcal{K}}$ and the potential part $\hat{\mathcal{V}}$, i.e. $\hat{H} = \hat{\mathcal{K}} + \hat{\mathcal{V}}$, which satisfy the eigenvalue equations

$$\hat{\mathcal{K}} \left| \psi_l, \mathbf{q} \right\rangle = \mathcal{K}(l, \mathbf{P}) \left| \psi_l, \mathbf{q} \right\rangle, \quad \hat{\mathcal{V}} \left| \theta, \mathbf{p} \right\rangle = \mathcal{V}(\theta, \mathbf{R}(\theta)) \left| \theta, \mathbf{p} \right\rangle. \tag{B.3}$$

Then we obtain

$$\begin{aligned} &K(\theta_n, \mathbf{p}_n, \epsilon; \theta_{n-1}, \mathbf{p}_{n-1}, 0) \\ &= \sum_{l_n} \int_{-\infty}^{\infty} d\mathbf{q}_n \left\langle \theta_n, \mathbf{p}_n \left| \exp\left(-\frac{i\epsilon}{\hbar} \hat{\mathcal{K}} \right) \right| \psi_{l_n}, \mathbf{q}_n \right\rangle \left\langle \psi_{l_n}, \mathbf{q}_n \left| \exp\left(-\frac{i\epsilon}{\hbar} \hat{\mathcal{V}} \right) \right| \theta_{n-1}, \mathbf{p}_{n-1} \right\rangle \\ &= \sum_{l_n=-\infty}^{\infty} \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{d\mathbf{q}_n}{(2\pi\hbar)^{\mathcal{N}}} \exp\left\{ -\frac{i\epsilon}{\hbar} \mathcal{K}(l_n, \mathbf{P}_n) - \frac{i\epsilon}{\hbar} \mathcal{V}(\theta_{n-1}, \mathbf{R}_{n-1}(\theta_{n-1})) \right\} \\ &\quad \times \exp\left\{ i l_n (\theta_n - \theta_{n-1}) - \frac{i}{\hbar} \mathbf{q}_n \cdot (\mathbf{p}_n - \mathbf{p}_{n-1})) \right\}. \end{aligned}$$

Next, define $A_n = \sum_j c q_{j,n} = \sum_j \frac{C_j}{m\omega_j^2} P_{j,n}$ and obtain

$$\begin{aligned} &K(\theta_n, \mathbf{p}_n, \epsilon; \theta_{n-1}, \mathbf{p}_{n-1}, 0) \\ &= \sum_{l_n=-\infty}^{\infty} \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{d\mathbf{q}_n}{(2\pi\hbar)^{\mathcal{N}}} \exp\left\{ -\frac{i\epsilon}{\hbar} \mathcal{K}(l_n, \mathbf{P}_n) - \frac{i\epsilon}{\hbar} \mathcal{V}(\theta_{n-1}, \mathbf{R}_{n-1}(\theta_{n-1})) \right\} \\ &\quad \times \exp\left\{ i(l_n - A_n/\hbar)(\theta_n - \theta_{n-1}) + \frac{i}{\hbar} \mathbf{P}_n \cdot (\mathbf{R}_n(\theta_n) - \mathbf{R}_{n-1}(\theta_{n-1})) \right\}. \end{aligned}$$

The sum over l_n can be replaced by a sum of integrals using the Poisson resummation formula

$$\sum_{l\in\mathbb{Z}} f(l) = \sum_{l\in\mathbb{Z}} \int_{-\infty}^{\infty} f(\zeta)\exp(2\pi l\zeta)d\zeta. \tag{B.4}$$

Then,

$$\begin{aligned}
&K(\theta_n, \mathbf{p}_n, \epsilon; \theta_{n-1}, \mathbf{p}_{n-1}, 0) \\
&\quad= \sum_{l_n=-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{d\zeta_n}{2\pi} \int_{-\infty}^{\infty} \frac{d\mathbf{q}_n}{(2\pi\hbar)^{\mathcal{N}}} \exp\left\{-\frac{i\epsilon}{\hbar}\mathcal{K}(\zeta_n, \mathbf{P}_n) - \frac{i\epsilon}{\hbar}\mathcal{V}(\theta_{n-1}, \mathbf{R}_{n-1}(\theta_{n-1}))\right\} \\
&\qquad\times \exp\left\{i(\zeta_n - A_n/\hbar)(\theta_n - \theta_{n-1} + 2\pi l_n) + \frac{i}{\hbar}\mathbf{P}_n\cdot(\mathbf{R}_n(\theta_n + 2\pi l_n) - \mathbf{R}_{n-1}(\theta_{n-1}))\right\}.
\end{aligned}$$

For our Hamiltonian of an ICDW ring threaded by a fluctuating magnetic flux we have

$$\begin{aligned}
\mathcal{K}(\zeta_n, \mathbf{P}_n) &= \frac{(\zeta_n\hbar - A_n)^2}{2I} + \sum_j \frac{1}{2m}P_{j,n}^2 \\
\mathcal{V}(\theta_{n-1}, \mathbf{R}_{n-1}(\theta_{n-1})) &= \sum_j \frac{1}{2}m\omega_j^2\left(R_{j,n-1}(\theta_{n-1}) - \frac{C_j\theta_{n-1}}{m\omega_j^2}\right)^2.
\end{aligned}$$

We note that that the potential $\mathcal{V}(\theta, \mathbf{R}(\theta))$ is rotationally invariant. That is, for an arbitrary rotation $\theta \to \theta + \delta$, the potential is $\mathcal{V}(\theta+\delta, \mathbf{R}(\theta+\delta)) = \mathcal{V}(\theta, \mathbf{R}(\theta))$. Let $\tilde{\zeta}_n = \zeta_n - A_n/\hbar$. We use the fact that the phase space volume is conserved under canonical transformation. Then,

$$\begin{aligned}
&K(\theta, \mathbf{p}, t; \theta', \mathbf{p}', 0) \\
&\quad= \left(\prod_{j=1}^{\mathcal{N}} \frac{1}{m\omega_j}\right) \lim_{\epsilon\to 0}\left(\prod_{n=1}^{N-1}\int_0^{2\pi} d\theta_n \int_{-\infty}^{\infty} d\mathbf{R}_n(\theta_n)\right)\left(\prod_{n=1}^{N}\sum_{l_n=-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{d\mathbf{P}_n}{(2\pi\hbar)^{\mathcal{N}}}\frac{d\tilde{\zeta}_n}{2\pi}\right) \\
&\qquad\times \exp\sum_{n=1}^{N}\left\{-\frac{i\epsilon}{\hbar}\left(\frac{\hbar^2}{2I}\tilde{\zeta}_n^2 + \sum_j \frac{1}{2m}P_{j,n}^2\right) - \frac{i\epsilon}{\hbar}\mathcal{V}(\theta_{n-1}, \mathbf{R}_{n-1}(\theta_{n-1}))\right\} \\
&\qquad\times \exp\sum_{n=1}^{N}\left\{i\tilde{\zeta}_n(\theta_n - \theta_{n-1} + 2\pi l_n) + \frac{i}{\hbar}\mathbf{P}_n\cdot(\mathbf{R}_n(\theta_n + 2\pi l_n) - \mathbf{R}_{n-1}(\theta_{n-1}))\right\}
\end{aligned}$$

Solve the $\tilde{\zeta}_n$ and $P_{j,n}$ integrals to obtain

$$
\begin{aligned}
&K(\theta, \mathbf{p}, t; \theta', \mathbf{p}', 0) \\
&= \left(\prod_{j=1}^{\mathcal{N}} \frac{1}{m\omega_j} \right) \lim_{\epsilon \to 0} \left(\prod_{n=1}^{N-1} \int_0^{2\pi} d\theta_n \int_{-\infty}^{\infty} d\mathbf{R}_n(\theta_n) \right) \left(\prod_{n=1}^{N} \sum_{l_n=-\infty}^{\infty} \sqrt{\frac{I}{2\pi i\epsilon\hbar}} \sqrt{\frac{m}{2\pi i\epsilon\hbar}}^{\mathcal{N}} \right) \\
&\times \exp \sum_{n=1}^{N} \left\{ \frac{i}{\hbar} \frac{I}{2\epsilon} (\theta_n - \theta_{n-1} + 2\pi l_n)^2 + \frac{i}{\hbar} \frac{m}{2\epsilon} (\mathbf{R}_n(\theta_n + 2\pi l_n) - \mathbf{R}_{n-1}(\theta_{n-1}))^2 \right\} \\
&\times \exp \sum_{n=1}^{N} \left\{ -\frac{i\epsilon}{\hbar} \mathcal{V}(\theta_{n-1}, \mathbf{R}_{n-1}(\theta_{n-1})) \right\}.
\end{aligned}
$$

The integral including θ_{N-1} is

$$
\begin{aligned}
I_{N-1} = &\sum_{l_N=-\infty}^{\infty} \sum_{l_{N-1}=-\infty}^{\infty} \sqrt{\frac{I}{2\pi i\epsilon\hbar}} \sqrt{\frac{m}{2\pi i\epsilon\hbar}}^{\mathcal{N}} \int_0^{2\pi} d\theta_{N-1} \int_{-\infty}^{\infty} d\mathbf{R}_{N-1}(\theta_{N-1}) \\
&\times \exp \left\{ \frac{i}{\hbar} \frac{I}{2\epsilon} (\theta_N - \theta_{N-1} + 2\pi l_N)^2 + \frac{i}{\hbar} \frac{I}{2\epsilon} (\theta_{N-1} - \theta_{N-2} + 2\pi l_{N-1})^2 \right\} \\
&\times \exp \left\{ \frac{i}{\hbar} \frac{m}{2\epsilon} [\mathbf{R}_N(\theta_N + 2\pi l_N) - \mathbf{R}_{N-1}(\theta_{N-1})]^2 \right\} \\
&\times \exp \left\{ \frac{i}{\hbar} \frac{m}{2\epsilon} [\mathbf{R}_{N-1}(\theta_{N-1} + 2\pi l_{N-1}) - \mathbf{R}_{N-2}(\theta_{N-2})]^2 \right\} \\
&\times \exp \left\{ -\frac{i\epsilon}{\hbar} \mathcal{V}(\theta_{N-1}, \mathbf{R}_{N-1}(\theta_{N-1})) \right\}.
\end{aligned}
$$

The sum over l_{N-1} can be absorbed into the θ_{N-1} integral by changing the domain of θ_{N-1} integration from $[0, 2\pi)$ to $(-\infty, \infty)$. This is done by the following procedure:

1. transform l_N to $\tilde{l}_N = l_N + l_{N-1}$ and write $\tilde{\theta}_{N-1} = \theta_{N-1} + 2\pi l_{N-1}$ to obtain

$$
\begin{aligned}
I_{N-1} = &\sum_{l_N=-\infty}^{\infty} \sum_{l_{N-1}=-\infty}^{\infty} \sqrt{\frac{I}{2\pi i\epsilon\hbar}} \sqrt{\frac{m}{2\pi i\epsilon\hbar}}^{\mathcal{N}} \int_0^{2\pi} d\theta_{N-1} \int_{-\infty}^{\infty} d\mathbf{R}_{N-1}(\theta_{N-1}) \\
&\times \exp \left\{ \frac{i}{\hbar} \frac{I}{2\epsilon} (\tilde{\theta}_N - \theta_{N-1} + 2\pi \tilde{l}_N)^2 + \frac{i}{\hbar} \frac{I}{2\epsilon} (\tilde{\theta}_{N-1} - \theta_{N-2})^2 \right\} \\
&\times \exp \left\{ \frac{i}{\hbar} \frac{m}{2\epsilon} [\mathbf{R}_N(\theta_N + 2\pi \tilde{l}_N) - \mathbf{R}_{N-1}(\tilde{\theta}_{N-1})]^2 \right\} \\
&\times \exp \left\{ \frac{i}{\hbar} \frac{m}{2\epsilon} [\mathbf{R}_{N-1}(\tilde{\theta}_{N-1}) - \mathbf{R}_{N-2}(\theta_{N-2})]^2 \right\} \\
&\times \exp \left\{ -\frac{i\epsilon}{\hbar} \mathcal{V}(\tilde{\theta}_{N-1}, \mathbf{R}_{N-1}(\tilde{\theta}_{N-1})) \right\}.
\end{aligned}
$$

2. Change the variable of integration $\int_0^{2\pi} d\theta_n \to \int_{2\pi n}^{2\pi(n+1)} d\tilde{\theta}_n$ and change the domain of θ_{N-1} integration from $[0, 2\pi)$ to $(-\infty, \infty)$ and use the periodicity of $\mathcal{V}(\theta_{N-1}, \mathbf{R}_{N-1}(\theta_{N-1}))$ to obtain

$$
\begin{aligned}
I_{N-1} = \sum_{l_N=-\infty}^{\infty} \sqrt{\frac{I}{2\pi i\epsilon\hbar}}\sqrt{\frac{m}{2\pi i\epsilon\hbar}}^{\mathcal{N}} \int_{-\infty}^{\infty} d\theta_{N-1} \int_{-\infty}^{\infty} d\mathbf{R}_{N-1}(\theta_{N-1}) \\
\times \exp\left\{\frac{i}{\hbar}\frac{I}{2\epsilon}(\theta_N - \theta_{N-1} + 2\pi l_N)^2 + \frac{i}{\hbar}\frac{I}{2\epsilon}(\theta_{N-1} - \theta_{N-2})^2\right\} \\
\times \exp\left\{\frac{i}{\hbar}\frac{m}{2\epsilon}[\mathbf{R}_N(\theta_N + 2\pi l_N) - \mathbf{R}_{N-1}(\theta_{N-1})]^2\right\} \\
\times \exp\left\{\frac{i}{\hbar}\frac{m}{2\epsilon}[\mathbf{R}_{N-1}(\theta_{N-1}) - \mathbf{R}_{N-2}(\theta_{N-2})]^2\right\} \\
\times \exp\left\{-\frac{i\epsilon}{\hbar}\mathcal{V}(\theta_{N-1}, \mathbf{R}_{N-1}(\theta_{N-1}))\right\}.
\end{aligned}
$$

Repeat this procedures for the integrals involving θ_n from $n = N-2$ to $n = 1$, and obtain

$$
\begin{aligned}
&K(\theta, \mathbf{q}, t; \theta', \mathbf{q}', 0) \\
&= \sum_{l=-\infty}^{\infty} \lim_{\epsilon\to 0} \int_{-\infty}^{\infty} \left(\prod_{n=1}^{N-1} d\theta_n\right) \int_{-\infty}^{\infty} \left(\prod_{n=1}^{N-1} d\mathbf{R}_{n-1}(\theta_{n-1})\right) \prod_{n=1}^{N} \sqrt{\frac{I}{2\pi i\epsilon\hbar}}\sqrt{\frac{m}{2\pi i\epsilon\hbar}}^{\mathcal{N}} \\
&\times \exp\left\{\frac{i}{\hbar}\frac{I}{2\epsilon}(\theta_n - \theta_{n-1} + 2\pi l\delta_{n,N})^2\right\} \\
&\times \exp\left\{\frac{i}{\hbar}\frac{m}{2\epsilon}[\mathbf{R}_n(\theta_n + 2\pi l\delta_{n,N}) - \mathbf{R}_{n-1}(\theta_{n-1})]^2 - \frac{i\epsilon}{\hbar}\mathcal{V}(\theta_{n-1}, \mathbf{R}_{n-1}(\theta_{n-1}))\right\}.
\end{aligned}
$$

Define

$$
\mathcal{A}_A = \sqrt{\frac{2\pi i\epsilon\hbar}{I}}, \qquad \mathcal{A}_B = \sqrt{\frac{2\pi i\epsilon\hbar}{m}}^{\mathcal{N}}, \tag{B.5}
$$

$$
\int \mathcal{D}\theta = \frac{1}{\mathcal{A}_A}\int_{-\infty}^{\infty}\left(\prod_{n=1}^{N-1}\frac{d\theta_n}{\mathcal{A}_A}\right), \qquad \int \mathcal{D}\mathbf{R}(\theta) = \frac{1}{\mathcal{A}_B}\int_{-\infty}^{\infty}\left(\prod_{n=1}^{N-1}\frac{d\mathbf{R}_n(\theta_n)}{\mathcal{A}_B}\right), \tag{B.6}
$$

$$L_n(\theta_n, \mathbf{R}_n(\theta_n)) = \frac{I}{2}\left(\frac{\theta_n - \theta_{n-1} + 2\pi l\delta_{n,N}}{\epsilon}\right)^2 + \frac{m}{2}\left(\frac{\mathbf{R}_n(\theta_n + 2\pi l\delta_{n,N}) - \mathbf{R}_{n-1}(\theta_{n-1})}{\epsilon}\right)^2 - \mathcal{V}(\theta_{n-1}, \mathbf{R}_{n-1}(\theta_{n-1})) \quad \text{(B.7)}$$

where

$$L[\theta, \mathbf{R}(\theta)] = \lim_{\epsilon\to 0} L_n[\theta_n, \mathbf{R}_n(\theta_n)] = L_0[\theta] + L_B[\theta, \mathbf{R}(\theta)] \quad \text{(B.8)}$$

$$L_0[\theta] = \frac{I}{2}\dot{\theta}^2 \quad \text{(B.9)}$$

$$L_B[\theta, \mathbf{R}(\theta)] = \sum_{j=1}^{\mathcal{N}} \frac{m}{2}\dot{R}_j(\theta)^2 - \sum_{j=1}^{\mathcal{N}} \frac{1}{2}m\omega_j^2\left(R_j(\theta) - \frac{C_j\theta}{m\omega_j^2}\right)^2 \quad \text{(B.10)}$$

is the Lagrangian of the coupled system. The action $S[\theta, \mathbf{R}(\theta)]$ is given by

$$S[\theta, \mathbf{R}(\theta)] = \int_0^t L[\theta, \mathbf{R}(\theta)]ds = S_0[\theta] + S_B[\theta, \mathbf{R}(\theta)] \quad \text{(B.11)}$$

$$S_0[\theta] = \int_0^t d\tau L[\theta] \quad \text{(B.12)}$$

$$S_B[\theta, \mathbf{R}(\theta)] = \int_0^t d\tau L_B[\theta, \mathbf{R}(\theta)]. \quad \text{(B.13)}$$

Now, suppose that we initially have the total density operator given by

$$\hat{\rho}_{AB}(0) = \hat{\rho}_A(0)\hat{\rho}_B(0). \quad \text{(B.14)}$$

Then, the reduced density matrix is

$$\begin{aligned}\rho(\theta, \phi, t) &= \int_{-\infty}^{\infty} d\mathbf{R}(\theta)d\mathbf{Q}(\theta)\delta(\mathbf{R}(\theta) - \mathbf{Q}(\theta))\rho(\theta, \mathbf{p}, \phi, \mathbf{s}, t) \\ &= \int_0^{2\pi} d\theta' d\phi' J(\theta, \phi, t; \theta', \phi', 0)\rho_A(\theta', \phi', 0)\end{aligned} \quad \text{(B.15)}$$

where

$$J(\theta, \phi, t; \theta', \phi', 0) = \sum_{l=-\infty}^{\infty}\sum_{l'=-\infty}^{\infty}\int_{\theta'}^{\theta+2\pi l}\mathcal{D}\theta\int_{\phi'}^{\phi+2\pi l'}\mathcal{D}\phi^* \exp\frac{i}{\hbar}\left(S_0[\theta] - S_0[\phi]\right)\mathcal{F}[\theta, \phi] \quad \text{(B.16)}$$

is the propagator of the density matrix,

$$\mathcal{F}[\theta,\phi] = \int_{-\infty}^{\infty} d\mathbf{R}(\theta)d\mathbf{Q}(\phi)d\mathbf{R}'(\theta')d\mathbf{Q}'(\phi')\delta(\mathbf{R}(\theta)-\mathbf{Q}(\phi))\rho_B(\mathbf{p}',\mathbf{s}',0)$$
$$\times \int_{\mathbf{R}'(\theta')}^{\mathbf{R}(\theta)} \mathcal{D}\mathbf{R}(\theta) \int_{\mathbf{Q}'(\phi')}^{\mathbf{Q}(\phi)} \mathcal{D}\mathbf{Q}(\phi) \exp\frac{i}{\hbar}\left(S_B[\theta,\mathbf{R}(\theta)] - S_B[\phi,\mathbf{Q}(\phi)]\right)$$

is the influence functional and $Q(\phi) = -(s_j - c\phi)/m\omega_j$. Supposed that the density operator $\hat{\rho}_B$ can be written as a canonical ensemble at $t = 0$. That is,

$$\rho_B = \frac{\exp(-\beta\hat{H}_B)}{\mathrm{tr}_B[\exp(-\beta\hat{H}_B)]} \tag{B.17}$$

Introduce the imaginary time $\tau = -i\hbar\beta$, then the density matrix of the environment at $t = 0$ is

$$\rho_B(\mathbf{p}',\mathbf{s}',0) = \frac{\left\langle \mathbf{p}' \left| \exp\left(-\frac{i\tau}{\hbar}\hat{H}_B\right) \right| \mathbf{s}' \right\rangle}{\mathrm{tr}_B\left[\exp\left(-\frac{i\tau}{\hbar}\hat{H}_B\right)\right]} = \frac{K_B(\mathbf{p}',\tau;\mathbf{s}',0)}{\int d\mathbf{q} K_B(\mathbf{q},\tau;\mathbf{q},0)}.$$

B.2 MOMENTUM REPRESENTATION OF THE DENSITY MATRIX OF A HARMONIC OSCILLATOR

The propagator of a harmonic oscillator with the Lagrangian

$$L = \frac{m}{2}\dot{x}^2 - \frac{1}{2}m\omega x^2 \tag{B.18}$$

has a well known solution

$$K(x,t;x',0) = \int \mathcal{D}x \exp\frac{i}{\hbar}\int_0^t L ds \tag{B.19}$$
$$= \sqrt{\frac{m\omega}{2\pi i\hbar\sin\omega t}} \exp\left\{\frac{i}{\hbar}\frac{m\omega}{2\sin\omega t}[(x^2+x'^2)\cos\omega t - 2xx']\right\} \tag{B.20}$$

$K(p,t;p',0)$ is obtained by a Fourier transformation

$$K(p,t:p',0) = \int_{-\infty}^{\infty} \frac{dxdx'}{2\pi\hbar} K(x,t;x',0) \exp \frac{i}{\hbar} \left(px - p'x'\right). \tag{B.21}$$

Let $A = \frac{m\omega \cos \omega t}{2 \sin \omega t}$ and $B = \frac{m\omega}{2 \sin \omega t}$, then

$$K(p,t;p',0) = \frac{F(t)}{\sqrt{4B^2 - 4A^2}} \exp \frac{i}{\hbar} \frac{Ap^2 + Ap'^2 - 2Bpp'}{4B^2 - 4A^2}$$

where

$$4B^2 - 4A^2 = \frac{m^2\omega^2(1 - \cos^2 \omega t)}{\sin^2 \omega t} = m^2\omega^2, \tag{B.22}$$

so,

$$K(p,t;p',0) = \frac{F(t)}{m\omega} \exp \frac{i}{\hbar} S[p(t)/m\omega]. \tag{B.23}$$

Therefore, using $\tau = -i\hbar\beta$, $\beta = 1/k_BT$, $\nu_j = \hbar\omega_j\beta$ and $i \sinh x = \sin ix$ we have

$$\langle \mathbf{p}' | \exp(-i\tau \hat{H}_B/\hbar) | \mathbf{s}' \rangle = \prod_{j=1}^{\mathcal{N}} \frac{1}{m\omega_j} \sqrt{\frac{m\omega_j}{2\pi\hbar \sinh \nu_j}} \exp - \frac{m\omega_j}{2\hbar \sinh \nu_j} \left[\frac{p'^2_j + s'^2_j}{m^2\omega_j^2} \cosh \nu - 2 \frac{p'_j s'_j}{m^2\omega_j^2} \right], \tag{B.24}$$

and

$$\mathrm{tr}_B[\exp(-i\tau \hat{H}_B/\hbar)] = \int_{-\infty}^{\infty} d\mathbf{q} \, \langle \mathbf{q} | \exp(-i\tau \hat{H}_B/\hbar) | \mathbf{q} \rangle = \prod_{j=1}^{\mathcal{N}} \frac{1}{2 \sinh \frac{\nu_j}{2}}.$$

We are assuming that the environment is not coupled to the system at $t = 0$, so $R'_j = p'_j/m\omega_j$ and $Q'_j = s'_j/m\omega_j$. Therefore,

$$\rho_B(\mathbf{p}', \mathbf{s}', 0) = \left(\prod_{j=1}^{\mathcal{N}} \frac{1}{m\omega_j} \right) \rho_B(\mathbf{R}', \mathbf{Q}', 0) \tag{B.25}$$

where

$$\rho_B(\mathbf{R}',\mathbf{Q}',0)=\prod_{j=1}^{\mathcal{N}}2\sinh\frac{\nu_j}{2}\sqrt{\frac{m\omega_j}{2\pi\hbar\sinh\nu_j}}\exp-\frac{m\omega_j}{2\hbar\sinh\nu_j}\left[({R'}_j^2+{Q'}_j^2)\cosh\nu_j-2{R'}_j{Q'}_j\right]. \tag{B.26}$$

Let us rewrite this density matrix into a simpler form. Define $x_{0j}=R'_j+Q'_j$ and $x'_{0j}=R'_j-Q'_j$. Then, using

$$({R'_j}^2+{Q'_j}^2)\cosh\nu_j-2{R'}_j{Q'}_j=\frac{1}{2}(\cosh\nu_j-1)x_{0j}^2+\frac{1}{2}(\cosh\nu_j+1){x'_{0j}}^2 \tag{B.27}$$

and the identity

$$\frac{\cosh\nu_j-1}{\sinh\nu_j}=\frac{\sinh\nu_j}{\cosh\nu_j+1}=\tanh\frac{\nu_j}{2}, \tag{B.28}$$

we obtain

$$\rho_B(\mathbf{R}',\mathbf{Q}',0)=\prod_{j=1}^{\mathcal{N}}\sqrt{\frac{m\omega_j}{\pi\hbar\mu_j}}\exp-\frac{m\omega_j}{4\hbar}\left[\frac{x_{0j}^2}{\mu_j}+{x'_{0j}}^2\mu_j\right]. \tag{B.29}$$

$$\mu_j=\coth\frac{\nu_j}{2}. \tag{B.30}$$

B.3 THE INFLUENCE FUNCTIONAL $\mathcal{F}[\theta,\phi]$

Now, we calculate the density functional $\mathcal{F}[\theta,\phi]$ explicitly. We will write $\mathbf{R}$ and $\mathbf{Q}$ instead of $\mathbf{R}[\theta]$ and $\mathbf{Q}[\phi]$ for simplicity, but keep in mind that they depend on θ and ϕ, respectively. Then, the influence functional can be written as

$$\begin{aligned}\mathcal{F}[\theta,\phi]=&\int_{-\infty}^{\infty}d\mathbf{x}_t d\mathbf{x}'_t d\mathbf{x}_0 d\mathbf{x}'_0(1/2)^{\mathcal{N}}\delta(\mathbf{x}'_t)\rho_B(\mathbf{R}',\mathbf{Q}',0)\int_{\mathbf{R}'}^{\mathbf{R}}\mathcal{D}\mathbf{R}\int_{\mathbf{Q}'}^{\mathbf{Q}}\mathcal{D}\mathbf{Q}^*\\&\times\exp\left\{\frac{i}{\hbar}\int_0^t ds\sum_j\left[\frac{m}{2}(\dot{R}_j^2-\dot{Q}_j^2)-\frac{1}{2}m\omega_j^2(R_j^2-Q_j^2)+C_j(\theta R_j-\phi Q_j)-\frac{C_j^2}{2m\omega_j^2}(\theta^2-\phi^2)\right]\right\}.\end{aligned} \tag{B.31}$$

To evaluate this we introduce the variables

$$\begin{aligned} \varphi &= \theta + \phi, \qquad \varphi' = \theta - \phi, \\ x_j &= R_j + Q_j, \qquad x'_j = R_j - Q_j. \end{aligned} \tag{B.32}$$

Then, the influence functional becomes

$$\begin{aligned} \mathcal{F}[\theta,\phi] &= \int_{-\infty}^{\infty} d\mathbf{x}_t d\mathbf{x}'_t d\mathbf{x}_0 d\mathbf{x}'_0 (1/2)^{\mathcal{N}} \delta(\mathbf{x}'_t) \rho_B(\mathbf{R}',\mathbf{Q}',0) \int \mathcal{D}\mathbf{x}\mathcal{D}\mathbf{x}' \\ &\times \exp\left\{ \frac{i}{\hbar} \int_0^t ds \sum_j \left[\frac{m}{2}\dot{x}_j\dot{x}'_j - \frac{1}{2}m\omega_j^2 x_j x'_j + \frac{C_j}{2}(\varphi x'_j + \varphi' x_j) - \frac{C_j^2}{2m\omega_j^2}\varphi'\varphi \right] \right\} \\ &= \int_{-\infty}^{\infty} d\mathbf{x}_t d\mathbf{x}'_t d\mathbf{x}_0 d\mathbf{x}'_0 (1/2)^{\mathcal{N}} \delta(\mathbf{x}'_t) \rho_B(\mathbf{R}',\mathbf{Q}',0) \\ &\times \exp\left\{ \frac{im}{2\hbar} \sum_j (x_{tj}\dot{x}'_{tj} - x_{0j}\dot{x}'_{0j}) - \frac{i}{\hbar}\int_0^t ds \sum_j \frac{C_j^2}{2m\omega_j^2}\varphi'\varphi \right\} \\ &\times \int \mathcal{D}\mathbf{x}\mathcal{D}\mathbf{x}' \exp\left\{ \frac{i}{\hbar}\int_0^t ds \sum_j \left[-g(x'_j)x_j + \frac{C_j}{2}\varphi x'_j \right] \right\} \end{aligned}$$

where

$$g(x'_j) = \frac{m}{2}\ddot{x}'_j + \frac{1}{2}m\omega_j^2 x'_j - \frac{C_j}{2}\varphi'. \tag{B.33}$$

We note that the classical solution $x'_{j,cl}$ satisfies the Euler-Lagrange equation

$$g(x'_{j,cl}) = 0, \quad g(x'_j(0)) = 0, \quad g(x'_j(t)) = 0. \tag{B.34}$$

The path integral over $\mathbf{x}$ can be done first. Calling this integral $I_{\mathbf{x},\mathbf{x}'}$ we obtain

$$I_{\mathbf{x},\mathbf{x}'} = \int \mathcal{D}\mathbf{x}\mathcal{D}\mathbf{x}' \exp\left\{ \frac{i}{\hbar}\int_0^t ds \sum_j \left[-g(x'_j)x_j + \frac{C_j}{2}\varphi x'_j \right] \right\} \tag{B.35}$$

$$= \frac{1}{|\mathcal{A}_B|^2} \int_{-\infty}^{\infty} \left(\prod_{n=1}^{N-1} \frac{d\mathbf{x}d\mathbf{x}'}{|\mathcal{A}_B|^2} \right) \prod_{n=1}^{N} \prod_j \exp \frac{i\epsilon}{\hbar} \left[-g(x'_{n,j})x_{n,j} + \frac{C_j}{2}\varphi x'_j \right] \tag{B.36}$$

$$= \frac{1}{|\mathcal{A}_B|^2} \int_{-\infty}^{\infty} \left(\prod_{n=1}^{N-1} \frac{d\mathbf{x}'}{|\mathcal{A}_B|^2} \right) \prod_j \left(\prod_{n=1}^{N-1} \delta\left[\frac{\epsilon}{2\pi\hbar} g(x'_{n,j}) \right] \right) \prod_{n=1}^{N} \exp \frac{i\epsilon}{\hbar}\frac{C_j}{2}\varphi x'_{n,j}. \tag{B.37}$$

$|\mathcal{A}|^{-2}$ is absorbed into the delta functions. The interpretation of the delta function is to insert the classical solution $x'_{j,cl}$ into x'_j which satisfy $g(x'_{j,cl}) = 0$. The outcome is,

$$I_{\mathbf{x},\mathbf{x}'} = \frac{1}{|\mathcal{A}_B|^2} \exp\left(\frac{i}{\hbar}\int_0^t ds \sum_j \frac{C_j}{2}\varphi x'_{cl}\right). \tag{B.38}$$

Therefore, the influence functional is

$$\begin{aligned}\mathcal{F}[\theta,\phi] = \int_{-\infty}^{\infty} d\mathbf{x}_t d\mathbf{x}'_t d\mathbf{x}_0 d\mathbf{x}'_0 (1/2)^{\mathcal{N}} \delta(\mathbf{x}'_t)\rho_B(\mathbf{R}',\mathbf{Q}',0) \exp\frac{i}{\hbar}\frac{m}{2}\sum_j (x_{tj}\dot{x}'_{tj} - x_{0j}\dot{x}'_{0j}) \\ \times \frac{1}{|\mathcal{A}_B|^2}\exp\left(\frac{i}{\hbar}\int_0^t ds \sum_j \left[-\frac{C_j^2}{2m\omega_j^2}\varphi'\varphi + \frac{C_j}{2}\varphi x'_{cl}\right]\right).\end{aligned}$$

The $\mathbf{x}_t$ integral gives a delta function of $\dot{\mathbf{x}}'_t$

$$\begin{aligned}\mathcal{F}[\theta,\phi] = \int_{-\infty}^{\infty} d\mathbf{x}'_t d\mathbf{x}_0 d\mathbf{x}'_0 (1/2)^{\mathcal{N}} \delta(\mathbf{x}'_t)\delta(\dot{\mathbf{x}}'_t)\rho_B(\mathbf{R}',\mathbf{Q}',0) \\ \times \exp\left(\frac{i}{\hbar}\int_0^t ds \sum_j \left[-\frac{C_j^2}{2m\omega_j^2}\varphi'\varphi + \frac{C_j}{2}\varphi x'_{cl}\right] + \frac{i}{\hbar}\frac{m}{2}\sum_j x_{0j}\dot{x}'_{0j}\right).\end{aligned}$$

Insert equation (B.29) for the density matrix and do the $\mathbf{x}_0$ integral, then we obtain

$$\begin{aligned}\mathcal{F}[\theta,\phi] = \int_{-\infty}^{\infty} d\mathbf{x}'_t d\mathbf{x}'_0 \delta(\mathbf{x}'_t)\delta(\dot{\mathbf{x}}'_t) \\ \times \prod_j \exp\left(\frac{i}{\hbar}\int_0^t ds \left[-\frac{C_j^2}{2m\omega_j^2}\varphi'\varphi + \frac{C_j}{2}\varphi x'_{cl}\right] - \frac{m\mu_j\omega_j}{4\hbar}\left(\frac{\dot{x}'^{\,2}_{0j}}{\omega_j^2} + x'^{\,2}_{0j}\right)\right).\end{aligned}$$

The two delta functions $\delta(\mathbf{x}'_t)$ and $\delta(\dot{\mathbf{x}}'_t)$ can be interpreted as boundary conditions on the classical solution of $g(\mathbf{x}'(s)) = 0$. The result of doing the remaining integrals would be to substitute the classical solution of x'_{cl}, $\dot{x}'_{0j}$ and x'_{0j} which satisfy these boundary conditions. The solution to the classical solution of $g(x) = 0$, that is

$$\ddot{x}'_j + \omega_j^2 x'_j - \frac{C_j}{m}\varphi' = 0. \tag{B.39}$$

is

$$x_j(\tau) = -\int_\tau^t \frac{C_j^2 \varphi'(s)}{m\omega_j} \sin\omega_j(\tau - s)ds + x'_{tj}\cos\omega_j(t-\tau) - \frac{\dot{x}'_{tj}}{\omega_j}\sin\omega_j(t-\tau). \tag{B.40}$$

For our boundary condition this solution reduces to

$$x'_j(\tau) = -\int_\tau^t \frac{C_j \varphi'(s)}{m\omega_j} \sin\omega_j(\tau - s)ds \tag{B.41}$$

$$\dot{x}'_j(\tau) = -\int_\tau^t \frac{C_j \varphi'(s)}{m} \cos\omega_j(\tau - s)ds \tag{B.42}$$

Therefore, the result of integration is

$$\mathcal{F}[\theta,\phi] = \exp\left(-\sum_j \frac{i}{\hbar}\frac{C_j^2}{2m\omega_j}\int_0^t\int_\tau^t \varphi(\tau)\left(\frac{2}{\omega_j}\delta(\tau - s) - \sin\omega_j(\tau - s)\right)\varphi'(s)d\tau ds\right)$$
$$\times \exp\left(-\sum_j \frac{\mu_j}{4\hbar}\int_0^t\int_0^t \frac{C_j^2}{m\omega_j}\varphi'(\tau)\cos\omega_j(\tau - s)\varphi'(s)dsd\tau\right).$$

$$\int_0^t\int_0^t E(\tau,s)dsd\tau = 2\int_0^t\int_0^\tau E(\tau,s)dsd\tau. \tag{B.43}$$

The first integral can be written as follows (see figure B.1 b))

$$\int_0^t\int_\tau^t E(\tau,s)dsd\tau = \int_0^t\int_0^\tau E(s,\tau)dsd\tau. \tag{B.44}$$

Then, the influence functional becomes

$$\mathcal{F}[\theta,\phi] = \exp i\Phi[\theta,\phi] \tag{B.45}$$

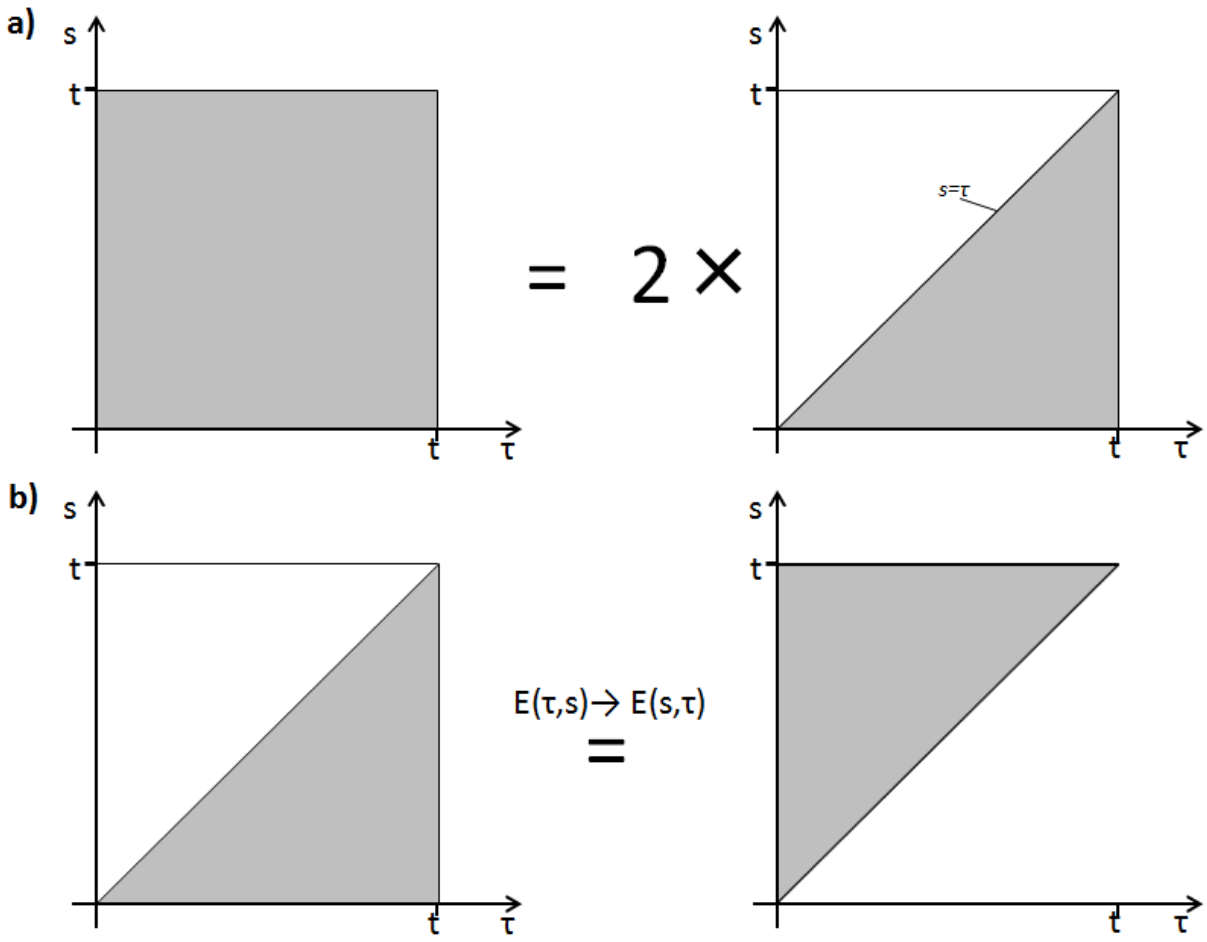

Figure B.1: The domain of integration is shown. If the integrand is symmetric, then the domain of integration can be changed according to a). The domain of integration can be changed according to b) if the variables τ and s in the integrand $E(\tau, s)$ are exchanged.

where the influence phase $\Phi[\theta,\phi]$ can be written as

$$i\Phi[\theta,\phi] = -\frac{i}{\hbar}\int_0^t\int_0^\tau d\tau ds\varphi(s)\alpha_I(\tau-s)\varphi'(\tau) - \frac{1}{\hbar}\int_0^t\int_0^\tau dsd\tau\varphi'(\tau)\alpha_R(\tau-s)\varphi'(s) \tag{B.46}$$

$$\alpha_R(\tau-s) = \sum_j \frac{C_j^2}{2m\omega_j}\coth\frac{\hbar\omega_j}{2k_BT}\cos\omega_j(\tau-s) \tag{B.47}$$

$$\alpha_I(\tau-s) = \sum_j \frac{C_j^2}{2m\omega_j}\left(\frac{2}{\omega_j}\delta(\tau-s)+\sin\omega_j(\tau-s)\right). \tag{B.48}$$

Therefore, we obtain the density matrix propagator $J(\theta,\phi,t;\theta',\phi',0)$

$$J(\theta_\mathrm{f},\phi_\mathrm{f},t;\theta\mathrm{i},\phi_\mathrm{i},0) \tag{B.49}$$

$$= \int_{\theta_\mathrm{i}}^{\theta_\mathrm{f}} D\theta\int_{\phi_\mathrm{i}}^{\phi_\mathrm{f}} D\phi^*\exp\left\{\frac{i}{\hbar}S[\varphi^+,\varphi^-]-\Gamma[\varphi^-]\right\}\Big|_{\substack{\varphi^+=(\theta+\phi)/2\\ \varphi^-=\phi-\theta}}, \tag{B.50}$$

the classical action $S[\varphi^+,\varphi^-]$ is

$$\begin{aligned} S[\varphi,\varphi'] = &-\int_0^t d\tau I\dot\varphi^+(\tau)\dot\varphi^-(\tau) \\ &+2\int_0^t d\tau\int_0^s ds\varphi^-(\tau)\alpha_\mathrm{I}(\tau-s)\varphi^+(s) \end{aligned}$$

and the $\Gamma[\varphi^-]$ is given by

$$\Gamma[\varphi^-] = \frac{1}{2\hbar}\int_0^t d\tau\int_0^t ds\varphi^-(\tau)\alpha_\mathrm{R}(\tau-s)\varphi^-(s).$$

The path integral in (B.50) can be done exactly and we obtain

$$J(\theta_\mathrm{f},\phi_\mathrm{f},t;\theta_\mathrm{i},\phi_\mathrm{i},0) = F^2(t)\exp\left(\frac{i}{\hbar}S[\varphi_\mathrm{cl}^+,\varphi_\mathrm{cl}^-]-\Gamma[\varphi_\mathrm{cl}^-]\right) \tag{B.51}$$

$\varphi_{\rm cl}^{\pm}$ are the classical coordinates obtained from the Euler-Lagrange equation

$$I\ddot{\varphi}_{\rm cl}^{-}(u) + 2\int_{u}^{t} d\tau \varphi_{\rm cl}^{-}(\tau)\alpha_{\rm I}(\tau - u) = 0 \tag{B.52}$$

$$I\ddot{\varphi}_{\rm cl}^{+}(u) + 2\int_{0}^{u} d\tau \varphi_{\rm cl}^{+}(\tau)\alpha_{\rm I}(u - \tau) = 0 \tag{B.53}$$

whose solution are given in terms of boundary conditions $\varphi_{\rm i}^{\pm} = \varphi^{\pm}(0), \varphi_{\rm f}^{\pm} = \varphi^{\pm}(t)$:

$$\varphi_{\rm cl}^{+}(u) = \kappa_i(u;t)\varphi_{\rm i}^{+} + \kappa_f(u;t)\varphi_{\rm f}^{+} \tag{B.54}$$

$$\varphi_{\rm cl}^{-}(u) = \kappa_i(t-u;t)\varphi_{\rm f}^{-} + \kappa_f(t-u;t)\varphi_{\rm i}^{-} \tag{B.55}$$

$$\kappa_i(u;t) = \dot{G}(u) - \frac{\dot{G}(t)}{G(t)}G(u), \qquad \kappa_f(u;t) = \frac{G(u)}{G(t)}. \tag{B.56}$$

Then, the classical action reduces to

$$S[\varphi_{\rm cl}^{+}, \varphi_{\rm cl}^{-}] = -I[\dot{\varphi}_{\rm cl}^{+}(t)\varphi_{\rm f}^{-} - \dot{\varphi}_{\rm cl}^{+}(0)\varphi_{\rm i}^{-}]. \tag{B.57}$$

Appendix C

CALCULATION OF CHARGE DENSITY EXPECTATION VALUE

Here, I calculate the time evolution of charge density expectation value for an ICDW ring via the Caldeira-Leggett model.

C.1 SUMMARY OF KNOWN RESULTS

First, I summarize known results from previous sections. The charge density expectation value is

$$\langle \hat{W}(t) \rangle = \frac{1}{2} \int_{-\pi}^{\pi} d\theta_{\rm f} \int_{-\pi}^{\pi} d\phi_{\rm f} \left(e^{i\theta_{\rm f}} \rho(\theta_{\rm f}, \phi_{\rm f}, t) + e^{i\phi_{\rm f}} \rho(\phi_{\rm f}, \theta_{\rm f}, t) \right) \delta(\theta_{\rm f} - \phi_{\rm f}) \tag{C.1}$$

$$= \frac{1}{2} \int_{-\pi}^{\pi} d\theta_{\rm f} \int_{-\pi}^{\pi} d\phi_{\rm f} \left(e^{i\theta_{\rm f}} \rho(\theta_{\rm f}, \phi_{\rm f}, t) + e^{i\phi_{\rm f}} \rho(\theta_{\rm f}, \phi_{\rm f}, t)^* \right) \delta(\theta_{\rm f} - \phi_{\rm f}). \tag{C.2}$$

The delta function $\delta(\theta_{\rm f} - \phi_{\rm f})$ was included for later convenience. The reduced density matrix is

$$\rho(\theta_{\rm f}, \phi_{\rm f}, t) = \int_0^{2\pi} d\theta_{\rm i} d\phi_{\rm i} \sum_{l_1, l_2 = -\infty}^{\infty} J(\theta_{\rm f} + 2\pi l_1, \phi_{\rm f} + 2\pi l_2, t; \theta_{\rm i}, \phi_{\rm i}, 0) \rho(\theta_{\rm i}, \phi_{\rm i}, 0) \tag{C.3}$$

whose time evolution is given by the propagator

$$J(\theta,\phi,t;\theta',\phi',0)=\int_{\theta'}^{\theta}D\theta\int_{\phi'}^{\phi}D\phi^{*}\exp\left\{\frac{i}{\hbar}S_0[\varphi^{+},\varphi^{-}]-\frac{1}{\hbar}\Xi[\varphi,\varphi']\right\}, \tag{C.4}$$

where $\varphi^{+}=(\theta+\phi)/2$ and $\varphi^{-}=\phi-\theta$ are center-of mass coordinate and relative coordinate, respectively. The action of ICDW ring without environment is

$$S_0[\varphi^{+},\varphi^{-}]=-I\int_0^t d\tau\dot{\varphi}^{+}(\tau)\dot{\varphi}^{-}(\tau), \tag{C.5}$$

where I is the moment of inertia of the ICDW ring. Effect of environment is given by the influence phase

$$\Xi[\varphi^{+},\varphi^{-}]=\Xi^{(N)}[\varphi^{-}]+i\Xi^{(F)}[\varphi^{+},\varphi^{-}] \tag{C.6}$$

$$\Xi^{(N)}[\varphi^{-}]=\int_0^t\int_0^{\tau}dsd\tau\varphi^{-}(\tau)\alpha_R(\tau-s)\varphi^{-}(s) \tag{C.7}$$

$$\Xi^{(F)}[\varphi^{+},\varphi^{-}]=-2\int_0^t\int_0^{\tau}dsd\tau\varphi^{-}(\tau)\alpha_I(\tau-s)\varphi^{+}(s) \tag{C.8}$$

$$\alpha_R(\tau-s)=\sum_j\frac{C_j^2}{2m\omega_j}\coth\frac{\hbar\omega_j}{2k_{\mathrm{B}}T}\cos\omega_j(\tau-s) \tag{C.9}$$

$$\alpha_I(\tau-s)=I\gamma(0)\delta(\tau-s)+\frac{I}{2}\frac{d}{d(\tau-s)}\gamma(\tau-s) \tag{C.10}$$

$$\gamma(t)=\frac{1}{I}\sum_j\frac{C_j^2}{m\omega_j^2}\cos\omega_j t \tag{C.11}$$

The influence action $\Xi^{(F)}[\varphi^{+},\varphi^{-}]$ introduces friction into the system. On the other hand, the noise action $\Xi^{(N)}[\varphi^{-}]$ may pump energy back in a random way.

C.2 EQUATION OF MOTION

Next, I calculate the Euler-Lagrange equation from the action $S[\varphi^+, \varphi^-] = S_0[\varphi^+, \varphi^-] - \Xi^{(F)}[\varphi^+, \varphi^-]$. The variation of $S_0[\varphi^+, \varphi^-]$ and $\Xi^{(F)}[\varphi^+, \varphi^-]$ are given by

$$\delta S_0[\varphi^+, \varphi^-] = -I \int_0^t d\tau [\delta\dot{\varphi}^+(\tau)\dot{\varphi}^-(\tau) + \dot{\varphi}^+(\tau)\delta\dot{\varphi}^-(\tau)] = I \int_0^t d\tau [\delta\varphi^+(\tau)\ddot{\varphi}^-(\tau) + \ddot{\varphi}^+(\tau)\delta\varphi^-(\tau)],$$
$$\delta\Xi^{(F)}[\varphi^+, \varphi-] = -2 \int_0^t \int_0^\tau ds d\tau \alpha_I(\tau - s)[\delta\varphi^-(\tau)\varphi^+(s) + \varphi^-(\tau)\delta\varphi^+(s)]$$

so, using $\frac{\delta\varphi^+(\tau)}{\delta\varphi^+(u)} = \frac{\delta\varphi^-(\tau)}{\delta\varphi^-(u)} = \delta(\tau - u)$ and $\frac{\delta\varphi^-(\tau)}{\delta\varphi^+(u)} = \frac{\delta\varphi^+(\tau)}{\delta\varphi^-(u)} = 0$, I obtain

$$\frac{\delta S_0[\varphi^+, \varphi^-]}{\delta\varphi^+(u)} = I \int_0^t d\tau \ddot{\varphi}^-(\tau)\delta(\tau - u) = \frac{I}{2}\ddot{\varphi}^-(u),$$
$$\frac{\delta S_0[\varphi^+, \varphi^-]}{\delta\varphi^-(u)} = I \int_0^t d\tau \ddot{\varphi}^+(\tau)\delta(\tau - u) = \frac{I}{2}\ddot{\varphi}^+(u),$$
$$\frac{\delta\Xi^{(F)}[\varphi^+, \varphi^-]}{\delta\varphi^+(u)} = -2 \int_0^t d\tau \varphi^-(\tau) \int_0^\tau ds \alpha_I(\tau - s)\delta(s - u) = -\int_u^t d\tau \alpha_I(\tau - u)\varphi'(\tau),$$
$$\frac{\delta\Xi^{(F)}[\varphi^+, \varphi^-]}{\delta\varphi^-(u)} = -2 \int_0^t d\tau \delta(\tau - u) \int_0^\tau ds \alpha_I(\tau - s)\varphi^+(s) = -\int_0^u d\tau \varphi^+(\tau)\alpha_I(u - \tau),$$

hence the Euler-Lagrange equations for φ^+ and φ^- are, respectively,

$$\frac{\delta S[\varphi^+, \varphi^-]}{\delta\varphi^+(u)} = 0 \qquad \Longrightarrow \qquad I\ddot{\varphi}^-(u) + 2\int_u^t d\tau \alpha_I(\tau - u)\varphi^-(\tau) = 0, \tag{C.12}$$
$$\frac{\delta S[\varphi^+, \varphi^-]}{\delta\varphi^-(u)} = 0 \qquad \Longrightarrow \qquad I\ddot{\varphi}^+(u) + 2\int_0^u d\tau \alpha_I(u - \tau)\varphi^+(\tau) = 0 \tag{C.13}$$

C.3 GENERAL SOLUTION

Finally, I calculate the solutions of $\varphi^+(u), \varphi^-(u)$ with arbitrary initial position and arbitrary initial velocity. Eq. (C.12) and Eq. (C.13) are related by the transformation $\varphi^-(u) = \varphi^+(t - u)$ [126]. Insert Eq. (C.10) into Eq. (C.13), then the generalized Langevin equation is given by

$$\ddot{\varphi}^+(u) + \int_0^u \dot{\varphi}^+(\tau)\gamma(u - \tau) + \gamma(u)\varphi^+(0) = 0. \tag{C.14}$$

Laplace transform each terms of (C.14)

$$\hat{\varphi}^+(z) = \mathcal{L}[\varphi^+(u)] = \int_0^\infty du\varphi^+(u)e^{-zt}, \tag{C.15}$$

$$\hat{\gamma}(\omega) = \mathcal{L}[\gamma(u)] = \int_0^\infty du\gamma(u)e^{-zt}, \tag{C.16}$$

$$\mathcal{L}[\dot{\varphi}^+(u)] = \int_0^\infty du\dot{\varphi}^+(u)e^{-zu} = -\varphi^+(0) + z\hat{\varphi}^+(z), \tag{C.17}$$

$$\mathcal{L}[\ddot{\varphi}^+(u)] = \int_0^\infty du\ddot{\varphi}^+(u)e^{-zu} = -\dot{\varphi}^+(0) - z\varphi^+(0) + z^2\hat{\varphi}^+(z), \tag{C.18}$$

$$\mathcal{L}[(\dot{\varphi}^+ * \gamma)(u)] = \mathcal{L}[\dot{\varphi}^+(u)]\mathcal{L}[\gamma(u)] = [-\varphi^+(0) + z\hat{\varphi}^+(z)]\hat{\gamma}(z), \tag{C.19}$$

and I obtain

$$[z^2 + z\hat{\gamma}(z)]\hat{\varphi}^+(z) = \dot{\varphi}^+(0) + z\varphi^+(0). \tag{C.20}$$

Assuming $z^2 + z\hat{\gamma}(z) \neq 0$ one can define the fundamental solution

$$G(u) = \mathcal{L}^{-1}\left[\frac{z}{z^2 + z\hat{\gamma}(z)}\right](u) = \frac{1}{2\pi i}\int_{-i\infty}^{i\infty} \frac{e^i zu}{z^2 + z\hat{\gamma}(z)}dz \tag{C.21}$$

and the solution of $\varphi^+(u)$ is

$$\varphi^+(u) = G(u)\dot{\varphi}^+(0) + \dot{G}(u)\varphi^+(0). \tag{C.22}$$

From the initial conditions $\varphi_i^+ = \varphi^+(0)$ and $\varphi_f^+ = \varphi^+(t)$ I obtain

$$\dot{\varphi}^+(0) = \frac{\varphi_f^+ - \dot{G}(t)\varphi_i^+}{G(t)}. \tag{C.23}$$

Therefore, the classical solutions are

$$\varphi^+(u) = \kappa_i(u;t)\varphi_{\rm i}^+ + \kappa_f(u;t)\varphi_{\rm f}^+ \tag{C.24}$$

$$\varphi_{\rm cl}^-(t-u) = \kappa_i(u;t)\varphi_{\rm f}^- + \kappa_f(u;t)\varphi_{\rm i}^- \tag{C.25}$$

$$\varphi_{\rm cl}^-(u) = \kappa_i(t-u;t)\varphi_{\rm f}^- + \kappa_f(t-u;t)\varphi_{\rm i}^- \tag{C.26}$$

$$\kappa_i(u;t) = \dot{G}(u) - \frac{\dot{G}(t)}{G(t)}G(u), \qquad \kappa_f(u;t) = \frac{G(u)}{G(t)} \tag{C.27}$$

Our next task is to compute the density matrix propagator J. I expand the action $S[\varphi,\varphi'] = S_0[\varphi,\varphi'] - \Xi^{(F)}[\varphi,\varphi']$ around classical trajectories as follows:

$$\theta = \theta_{\rm cl} + \tilde{\theta}, \qquad \phi = \phi_{\rm cl} + \tilde{\phi}, \qquad \varphi^+ = \varphi_{\rm cl}^+ + \tilde{\varphi}^+, \qquad \varphi^- = \varphi_{\rm cl}^- + \tilde{\varphi}^-. \tag{C.28}$$

Expand S and $\Xi^{(N)}$ in terms of classical path and quantum fluctuation

$$S[\varphi_{\rm cl}^+ + \tilde{\varphi}^+, \varphi_{\rm cl}^- + \tilde{\varphi}^-] = S[\varphi_{\rm cl}^+, \varphi_{\rm cl}^-] + S[\tilde{\varphi}^+, \tilde{\varphi}^-] + S_\times[\varphi_{\rm cl}^+, \varphi_{\rm cl}^-, \tilde{\varphi}^+, \tilde{\varphi}^-] \tag{C.29}$$

$$\Xi^{(N)}[\varphi_{\rm cl}^- + \tilde{\varphi}^-] = \Xi^{(N)}[\varphi_{\rm cl}^-] + \Xi^{(N)}[\tilde{\varphi}^-] + 2\Xi_\times^{(N)}[\varphi_{\rm cl}^-, \tilde{\varphi}^-] \tag{C.30}$$

where

$$S_\times[\varphi_{\rm cl}^+, \varphi_{\rm cl}^-, \tilde{\varphi}^+, \tilde{\varphi}^-] = -\int_0^t d\tau I[\dot{\varphi}_{\rm cl}^+(\tau)\dot{\tilde{\varphi}}^-(\tau) + \dot{\tilde{\varphi}}^+(\tau)\dot{\varphi}_{\rm cl}^-(\tau)] \tag{C.31}$$

$$+ 2\int_0^t \int_0^\tau ds d\tau \alpha_I(\tau - s)[\varphi_{cl}^-(\tau)\tilde{\varphi}^+(s) + \tilde{\varphi}^-(\tau)\varphi_{\rm cl}^+(s)] \tag{C.32}$$

$$\Xi_\times^{(N)}[\varphi_{\rm cl}^-, \tilde{\varphi}^-] = \frac{1}{2}\int_0^t \int_0^t ds d\tau \varphi_{\rm cl}^-(\tau)\tilde{\varphi}^-(s)\alpha_{\rm R}(\tau - s) \tag{C.33}$$

The classical action is calculation using integration by parts and the Euler-Lagrange equation (C.13), then I obtain

$$S[\varphi_{\rm cl}^+, \varphi_{\rm cl}^-] = -I[\dot{\varphi}_{\rm f}^+\varphi_{\rm f}^- - \dot{\varphi}_{\rm i}^+\varphi_{\rm i}^-]. \tag{C.34}$$

Similarly, one can show that $S_\times[\varphi_{\rm cl},\varphi'_{\rm cl},\tilde{\varphi},\tilde{\varphi}']=0$. Then, the propagator J becomes

$$J(\theta_{\rm f},\phi_{\rm f},t;\theta_{\rm i},\phi_{\rm i},0) = e^{\frac{i}{\hbar}S[\varphi^+_{\rm cl},\varphi^-_{\rm cl}]-\frac{1}{\hbar}\Xi^{(N)}[\varphi^-_{\rm cl}]}\mathcal{G}(\varphi^+_{\rm f},\varphi^-_{\rm f},t;\varphi^+_{\rm i},\varphi^-_{\rm i},0) \tag{C.35}$$

$$= e^{\frac{i}{\hbar}S(\varphi^+_{\rm f},\varphi^-_{\rm f},t;\varphi^+_{\rm i},\varphi^-_{\rm i},0)-\frac{1}{\hbar}\Xi^{(N)}(\varphi^-_{\rm f},t;\varphi^-_{\rm i},0)}\mathcal{G}(\varphi^+_{\rm f},\varphi^-_{\rm f},t;\varphi^+_{\rm i},\varphi^-_{\rm i},0) \tag{C.36}$$

$$\mathcal{G}(\varphi^+_{\rm f},\varphi^-_{\rm f},t;\varphi^+_{\rm i},\varphi^-_{\rm i},0) = \int_0^0 D\tilde{\theta}\int_0^0 D\tilde{\phi}^*\exp\left\{\frac{i}{\hbar}S[\tilde{\varphi}^+,\tilde{\varphi}^-]-\frac{1}{\hbar}\Xi^{(N)}[\tilde{\varphi}^-]-\frac{2}{\hbar}\Xi^{(N)}_\times[\varphi^-_{cl},\tilde{\varphi}^-]\right\} \tag{C.37}$$

Here, I assume that $\tilde{\theta}$ and $\tilde{\phi}$ are weak perturbation which do no change winding numbers l_1 and l_2 in Eq. (C.4). Then, $\mathcal{G}(\varphi^+_{\rm f},\varphi^-_{\rm f},t;\varphi^+_{\rm i},\varphi^-_{\rm i},0)$ is formally equivalent to the one calculated by Caldeira and Leggett [42] and it is as function of time, independent of ICDW coordinates:

$$\mathcal{G}(\varphi^+_{\rm f},\varphi^-_{\rm f},t;\varphi^+_{\rm i},\varphi^-_{\rm i},0) = F^2(t). \tag{C.38}$$

$F^2(t)$ is calculated from the normalization condition

$$\begin{aligned}\langle 1\rangle &= \int_{-\pi}^{\pi}d\theta_{\rm f}\int_{-\pi}^{\pi}d\phi_{\rm f}\int_{-\pi}^{\pi}d\theta_{\rm i}\int_{-\pi}^{\pi}d\phi_{\rm i}\delta(\theta_{\rm f}-\phi_{\rm f})\\ &\quad\times\sum_{l_1,l_2=-\infty}^{\infty}J(\theta_{\rm f}+2\pi l_1,\phi_{\rm f}+2\pi l_2,t,\theta_{\rm i},\phi_{\rm i},0)\rho(\theta_{\rm i},\phi_{\rm i},0)\\ &= 1\end{aligned}$$

Here, the delta function is periodic by definition:

$$\delta(\theta)\equiv\sum_{n=-\infty}^{\infty}\frac{1}{2\pi}e^{in\theta}=\sum_{n=-\infty}^{\infty}\frac{1}{2\pi}e^{in(\theta+2\pi n)}=\delta(\theta+2\pi n), \tag{C.39}$$

and $\delta(\theta_{\rm f}-\phi_{\rm f}-)=\delta(\theta_{\rm f}+2\pi l_1-\phi_{\rm f}-2\pi l_2)$. So, because one can write $\sum_{l=-\infty}^{\infty}\int_{-\pi}^{\pi}f(\theta+2\pi l)d\theta=\int_{-\infty}^{\infty}f(\theta)d\theta$ for any function $f(\theta)$, I obtain

$$\begin{aligned}\langle 1\rangle &= \int_{-\infty}^{\infty}d\theta_{\rm f}\int_{-\infty}^{\infty}d\phi_{\rm f}\int_{-\pi}^{\pi}d\theta_{\rm i}\int_{-\pi}^{\pi}d\phi_{\rm i}\delta(\theta_{\rm f}-\phi_{\rm f})J(\theta_{\rm f},\phi_{\rm f},t,\theta_{\rm i},\phi_{\rm i},0)\rho(\theta_{\rm i},\phi_{\rm i},0)\\ &= \int_{-\infty}^{\infty}d\theta_f\int_{-\pi}^{\pi}d\theta_i\int_{-\pi}^{\pi}d\phi_i J(\theta_{\rm f},\theta_{\rm f},t,\theta_{\rm i},\phi_{\rm i},0)\rho(\theta_{\rm i},\phi_{\rm i},0)\end{aligned}$$

which implies $\varphi_{\rm f}^+ = \theta$ and $\varphi_{\rm f}^- = 0$, $\Xi^{(N)}[\varphi_{\rm cl}^-]$ is a function of $\varphi_{\rm i}^-$ only, and

$$S[\varphi_{\rm cl}^+] = I\frac{\theta_{\rm f}\varphi_{\rm i}^- - \varphi_{\rm i}^-\varphi_{\rm i}^+\dot{G}(t)}{G(t)}.$$

Then, the integral can be evaluated easily (using $\mu = I/\hbar$)

$$\begin{aligned}
\langle 1\rangle &= F^2(t)\int_{-\infty}^{\infty}d\theta_f\int_{-\pi}^{\pi}d\theta_i\int_{-\pi}^{\pi}d\phi_i\exp\left\{i\mu\frac{\theta_{\rm f}\varphi_{\rm i}^- - \varphi_{\rm i}^-\varphi_{\rm i}^+\dot{G}(t)}{G(t)} - \frac{1}{\hbar}\Xi^{(N)}[\varphi^-]\right\}\rho(\theta_{\rm i},\phi_{\rm i},0)\\
&= F^2(t)\int_{-\pi}^{\pi}d\theta_i\int_{-\pi}^{\pi}d\phi_i\exp\left\{-i\mu\frac{\varphi_{\rm i}^-\varphi_{\rm i}^+\dot{G}(t)}{G(t)} - \frac{1}{\hbar}\Xi^{(N)}[\varphi^-]\right\}\delta\left(\frac{\mu\varphi_{\rm i}^-}{G(t)}\right)\rho(\theta_{\rm i},\phi_{\rm i},0)\\
&= F^2(t)\int_{-\pi}^{\pi}d\theta_i\left|\frac{G(t)}{\mu}\right|\rho(\theta_{\rm i},\theta_{\rm i},0)\\
&= F^2(t)\left|\frac{G(t)\hbar}{I}\right|
\end{aligned}$$

which implies

$$F^2(t) = \left|\frac{\mu}{G(t)}\right|$$

Calculation of $\langle\hat{W}(t)\rangle$ is almost equivalent and I obtain

$$\begin{aligned}
\langle\hat{W}(t)\rangle &= \frac{F^2(t)}{2}\int_{-\infty}^{\infty}d\theta_f\int_{-\pi}^{\pi}d\theta_i\int_{-\pi}^{\pi}d\phi_i e^{i\theta}\exp\left\{i\mu\frac{\theta_{\rm f}\varphi_{\rm i}^- - \varphi_{\rm i}^-\varphi_{\rm i}^+\dot{G}(t)}{G(t)} - \frac{1}{\hbar}\Xi^{(N)}[\varphi^-]\right\}\rho(\theta_{\rm i},\phi_{\rm i},0)\\
&+ \frac{F^2(t)}{2}\int_{-\infty}^{\infty}d\theta_f\int_{-\pi}^{\pi}d\theta_i\int_{-\pi}^{\pi}d\phi_i e^{i\theta}\exp\left\{-i\mu\frac{\theta_{\rm f}\varphi_{\rm i}^- - \varphi_{\rm i}^-\varphi_{\rm i}^+\dot{G}(t)}{G(t)} - \frac{1}{\hbar}\Xi^{(N)}[\varphi^-]\right\}\rho(\theta_{\rm i},\phi_{\rm i},0)^*\\
&= \frac{F^2(t)}{2}\int_{-\pi}^{\pi}d\theta_i\int_{-\pi}^{\pi}d\phi_i\exp\left\{-i\mu\varphi_{\rm i}^-\varphi_{\rm i}^+\frac{\dot{G}(t)}{G(t)} - \frac{1}{\hbar}\Xi^{(N)}[\varphi^-]\right\}\delta\left(1+\mu\varphi_{\rm i}^-\frac{1}{G(t)}\right)\rho(\theta_{\rm i},\phi_{\rm i},0)\\
&+ \frac{F^2(t)}{2}\int_{-\pi}^{\pi}d\theta_i\int_{-\pi}^{\pi}d\phi_i\exp\left\{i\mu\varphi_{\rm i}^-\varphi_{\rm i}^+\frac{\dot{G}(t)}{G(t)} - \frac{1}{\hbar}\Xi^{(N)}[\varphi^-]\right\}\delta\left(1-\mu\varphi_{\rm i}^-\frac{1}{G(t)}\right)\rho(\theta_{\rm i},\phi_{\rm i},0)^*\\
&= \frac{1}{2}\int_{-\pi}^{\pi}d\theta_i\exp\left\{i\dot{G}(t)\theta_i - i\frac{G(t)\dot{G}(t)}{2\mu} - \frac{1}{\hbar}\Xi^{(N)}[\varphi^-\big|_{\varphi_i^-=-G(t)/\mu}]\right\}\rho(\theta_{\rm i},\theta_{\rm i}-G(t)/2\mu,0)\\
&+ \frac{1}{2}\int_{-\pi}^{\pi}d\theta_i\exp\left\{i\dot{G}(t)\theta_i + i\frac{G(t)\dot{G}(t)}{2\mu} - \frac{1}{\hbar}\Xi^{(N)}[\varphi^-\big|_{\varphi_i^-=-G(t)/\mu}]\right\}\rho(\theta_{\rm i},\theta_{\rm i}+G(t)/2\mu,0)^*.
\end{aligned}$$

Now, the noise action $\Xi^{(N)}$ is a function of t only, so I write

$$\Gamma(t) = \frac{1}{\hbar}\Xi^{(N)}[\varphi^-\big|_{\varphi_i^-=-G(t)/\mu}]. \tag{C.40}$$

For the ground state $\psi_{\rm GS}(\theta) = 1/\sqrt{2\pi}$ I obtain

$$\langle\psi_{\rm GS}, \hat{W}(t)\psi_{\rm GS}\rangle = \mathrm{sinc}\left(\pi\dot{G}(t)\right)\cos\left(\frac{G(t)\dot{G}(t)}{2\mu}\right)e^{-\Gamma(t)} \tag{C.41}$$

and the charge density expectation value is

$$\langle\psi_{\rm GS}, \hat{n}(x,t)\psi_{\rm GS}\rangle = \mathrm{sinc}\left(\pi\dot{G}(t)\right)\cos\left(\frac{G(t)\dot{G}(t)}{2\mu}\right)\cos(2k_{\rm F}x)e^{-\Gamma(t)}. \tag{C.42}$$

Eq. (C.41) and Eq. (C.42) are the main results of this appendix.

Appendix D

SUPPLEMENTARY INFORMATION TO THE NAKANISHI-SHIBA MODEL

D.1 $\mathbf{Q}(x)$ FOR $1T$-TaS_2

(a)Let $\mathbf{Q}(x)$ be the wave vector path parametrized by $x \in [0,1]$. The endpoints of this path are $\mathbf{Q}(0) = \mathbf{Q}_{\mathrm{IC}}, \mathbf{Q}(1) = \mathbf{Q}_{\mathrm{C}}$, and the NCCDW state is given by $\mathbf{Q}(x^*) = \mathbf{Q}_{\mathrm{NC}}$ where

$$x^* = \frac{d(\mathbf{Q}_{\mathrm{IC}}, \mathbf{Q}_{\mathrm{NC}})}{d(\mathbf{Q}_{\mathrm{IC}}, \mathbf{Q}_{\mathrm{NC}}) + d(\mathbf{Q}_{\mathrm{NC}}, \mathbf{Q}_{\mathrm{C}})} < 1.$$

$d(\mathbf{A}, \mathbf{B}) = \sqrt{(\mathbf{A}-\mathbf{B}) \cdot (\mathbf{A}-\mathbf{B})}$ is the distance between vectors $\mathbf{A}, \mathbf{B}$. Then, $\mathbf{Q}(x)$ can be written as

$$\mathbf{Q}(x) = \begin{cases} \mathbf{Q}_{\mathrm{IC}} + \frac{x}{x^*}(\mathbf{Q}_{\mathrm{NC}} - \mathbf{Q}_{\mathrm{IC}}) & \text{if } 0 \leq x \leq x^* \\ \mathbf{Q}_{\mathrm{NC}} + \frac{x-x^*}{1-x^*}(\mathbf{Q}_{\mathrm{C}} - \mathbf{Q}_{\mathrm{NC}}) & \text{if } x* \leq x \leq 1 \end{cases}.$$

D.2 METHOD

D.2.1 NAKANISHI-SHIBA-MCMILLAN FREE ENERGY

McMillan's phenomenological free energy of triple CDW is [88, 112, 113, 127]

$$F = \int d^2r \Big\{ a(\mathbf{r})\alpha^2(\mathbf{r}) - b(\mathbf{r})\alpha^3(\mathbf{r}) + c(\mathbf{r})\alpha^4(\mathbf{r}) + d(\mathbf{r})\big[|\psi_1\psi_2|^2 + |\psi_2\psi_3|^2 + |\psi_3\psi_1|^2\big] + \sum_{i=1}^{3} \psi_i^* e_i(-i\nabla)\psi_i \Big\} \tag{D.1}$$

where the complex order parameters $\psi_i(\mathbf{r})$ are related to the electron charge density

$$\rho(\mathbf{r}) = \rho_0(r)[1 + \alpha(\mathbf{r})] \tag{D.2}$$

and

$$\alpha(\mathbf{r}) = \mathrm{Re}\left[\psi_1(\mathbf{r}) + \psi_2(\mathbf{r}) + \psi_3(\mathbf{r})\right]. \tag{D.3}$$

$\rho_0(\mathbf{r})$ is the charge density in the normal state. The indices 1, 2, 3 specify the three components of the triple CDW. Each coefficient has lattice periodicity and can be written as

$$b(\mathbf{r}) = b_0 + b_1 \sum_{i=1}^{3} e^{i\mathbf{G}_i\cdot\mathbf{r}}, \text{ etc.}, \tag{D.4}$$

where $\mathbf{G}_i$ are reciprocal lattice vectors. The function $e_i(-i\nabla)$ represents a gradient operator. First, we expand the α^2 term in Eq. (D.1). Using $a(\mathbf{r}) = a_0 + a_1 \sum_{i=1}^{3} e^{i\mathbf{G}_i\cdot\mathbf{r}}$, $\alpha(\mathbf{r}) = \mathrm{Re}\sum_{i=1}^{3}\psi_i(\mathbf{r})$ and $\psi_i(\mathbf{r}) = e^{i\mathbf{Q}_{\mathrm{C}}^{(i)}\cdot\mathbf{r}}\phi_i(\mathbf{r})$ we have

$$\int d^2r\ a(\mathbf{r})\alpha^2(\mathbf{r}) = a_0 \int d^2r \left[\mathrm{Re}\sum_{i=1}^{3} e^{i\mathbf{Q}_{\mathrm{C}}^{(i)}\cdot\mathbf{r}}\phi_i(\mathbf{r})\right]^2 + a_1 \int d^2r \sum_{i=1}^{3} e^{i\mathbf{G}_i\cdot\mathbf{r}} \left[\mathrm{Re}\sum_{j=1}^{3} e^{i\mathbf{Q}_{\mathrm{C}}^{(j)}\cdot\mathbf{r}}\phi_j(\mathbf{r})\right]^2.$$

Oscillating terms vanish after integration, so the non-zero terms are those with absolute values $|\phi_i|^2$:

$$\int d^2r\ a(\mathbf{r})\alpha^2(\mathbf{r}) = \frac{a_0}{2}\sum_{i=1}^{3}\int d^2r\ |\phi_i|^2.$$

Similarly,

$$\int d^2r\ b(\mathbf{r})\alpha^3(\mathbf{r}) =$$
$$= -\int d^2r\left\{\frac{3b_0}{4}(\phi_1\phi_2\phi_3 + \text{c.c.}) + \frac{3b_1}{8}\sum_{i=1}^{3}(\phi_i^2\phi_{i+1}^* + \text{c.c.})\delta_{q,2}\delta_{p,1} + \frac{b_1}{8}\sum_{i=1}^{3}(\phi_i^3 + \text{c.c.})\delta_{q,3}\delta_{p,0}\right\}$$

and

$$\int d^2r\ c(\mathbf{r})\alpha^4(\mathbf{r}) = \int d^2r\sum_{i=1}^{3}\left\{\frac{c_0}{8}(3|\phi_i|^4 + 12|\phi_i\phi_{i+1}|^2) + \frac{c_1}{4}(\phi_i^3\phi_{i+1}^* + \text{c.c.})\delta_{q,3}\delta_{p,1}\right.$$
$$\left. + \frac{3c_1}{4}(\phi_i^2\phi_{i+1}^*\phi_{i+2}^* + \text{c.c.})\delta_{q,3}\delta_{p,0} + \frac{3c_1}{4}(\phi_i\phi_{i+1}^{*2}\phi_{i+2}^* + \text{c.c.})\delta_{q,2}\delta_{p,1} + \frac{c_1}{4}(\phi_i^3\phi_{i+2} + \text{c.c.})\delta_{q,2}\delta_{p,1}\right\}.$$

The term proportional to b_0 arises from the triple-Q condition $\mathbf{Q}_{\text{C}}^{(1)} + \mathbf{Q}_{\text{C}}^{(2)} + \mathbf{Q}_{\text{C}}^{(3)} = \mathbf{0}$. The terms proportional to a_1, b_1 and c_1 arise from the commensurate structure $q\mathbf{Q}_{\text{C}}^{(i)} - p\mathbf{Q}_{\text{C}}^{(i+1)} = \mathbf{G}_i$, or equivalently,

$$\mathbf{Q}_{\text{C}}^{(i)} = \frac{q^2\mathbf{G}_i + qp\mathbf{G}_{i+1} + p^2\mathbf{G}_{i+2}}{q^3 - p^3}. \tag{D.5}$$

After some rearrangement we obtain

$$F = F_A + F_B + F_C + F_D + F_{(q,p)}, \tag{D.6}$$
$$F_A = \int d^2r\sum_{i=1}^{3}\phi_i^* A_i(\mathbf{Q}_{\text{C}}^{(i)} - i\nabla)\phi_i, \tag{D.6a}$$
$$F_B = \int d^2r B\sum_{i=1}^{3}|\phi_i|^4, \tag{D.6b}$$
$$F_C = \int d^2r C\sum_{i=1}^{3}|\phi_i\phi_{i+1}|^2, \tag{D.6c}$$
$$F_D = \int d^2r\frac{D}{2}(\phi_1\phi_2\phi_3 + \text{c.c.}), \tag{D.6d}$$

and the form of commensurate free energies $F_{(q,p)}$ depend on the (q,p) values:

$$F_{(2,1)} = \int d^2r \sum_{i=1}^{3} \left\{ \frac{U}{2}(\phi_i^2 \phi_{i+1}^* + \text{c.c.}) + \frac{V}{2}(\phi_i \phi_{i+1}^{*2} \phi_{i+2}^* + \text{c.c.}) + \frac{Z}{2}(\phi_i^3 \phi_{i+2} + \text{c.c.}) \right\}, \tag{D.6e}$$

$$F_{(3,0)} = \int d^2r \sum_{i=1}^{3} \left\{ \frac{Y}{2}(\phi_i^3 + \text{c.c}) + \frac{W}{2}(\phi_i^2 \phi_{i+1}^* \phi_{i+2}^* + \text{c.c.}) \right\}, \tag{D.6f}$$

$$F_{(3,1)} = \int d^2r \sum_{i=1}^{3} \frac{E}{2}(\phi_i^3 \phi_{i+1}^* + \text{c.c}). \tag{D.6g}$$

Here, we have $A_i(\mathbf{Q},T) = \frac{1}{2}a_0 + e_i(\mathbf{Q})$, $B = \frac{3}{8}c_0$, $C = \frac{3}{2}c_0 + d_0$, $D = -\frac{3}{2}b_0$, $E = \frac{1}{2}c_1$, $Y = -\frac{1}{4}b_1$, $W = \frac{3}{2}c_1$, $U = -\frac{3}{4}b_1$, $V = \frac{3}{2}c_1$, $Z = \frac{1}{2}c_1$.

D.2.2 TYPE 1 AND TYPE 2 FREE ENERGIES

Consider two types of free energies which use different forms of $A_i(\mathbf{Q},T)$. The first one (type 1) is the free energy considered by Nakanishi and Shiba [114] which uses phenomenological parameters which reproduce a ring-like diffuse scattering obtained in and electron diffraction experiment:

$$A_i(\mathbf{Q},T) = T - T_C + (1-\xi_i)u + v\xi_i(1-\cos(6\phi_i)). \qquad \text{(Type 1)} \tag{D.7}$$

where

$$\begin{aligned} \xi_i &= 1 - s(|\mathbf{Q}| - |\mathbf{Q}_{\text{IC}}^{(i)}|)^2/|\mathbf{G}_i|^2 \quad \text{for } (|\mathbf{Q}| - |\mathbf{Q}_{\text{IC}}^{(i)}|)^2/|\mathbf{G}_i|^2 < s^{-1} \\ &= 0 \qquad\qquad\qquad\qquad\qquad\qquad\quad \text{otherwise,} \end{aligned}$$

u and v are parameters; ϕ_i is the angle between $\mathbf{Q}$ and $\mathbf{G}_i$, $\mathbf{Q}_{\text{IC}}^{(i)}$ is the fundamental wave vector of the ICCDW phase. This $A_i(\mathbf{Q},T)$ has a circular ditch with the radius $\mathbf{Q}_{\text{IC}}^{(i)}$ around $\mathbf{Q} = 0$. The second one (type 2) uses McMillan's free energy [88] which has the

gradient term

$$\int d^2r \sum_{i=1}^{3} \psi_i^* e_i(-i\nabla) = \int d^2r \sum_{i=1}^{3} \left[e|(\mathbf{Q}_{\mathrm{IC}}^{(i)} \cdot \nabla - i|\mathbf{Q}_{\mathrm{CI}}^{(i)}|^2)\psi_i|^2 + f|\mathbf{Q}_{\mathrm{IC}}^{(i)} \times \nabla\psi_i|^2 \right]$$
$$= \int d^2r \sum_{i=1}^{3} \psi_i^* \left[e(\mathbf{Q}_{\mathrm{IC}}^{(i)} \cdot (-i\nabla) - |\mathbf{Q}_{\mathrm{IC}}^{(i)}|^2)^2 + f|\mathbf{Q}_{\mathbf{IC}}^{(i)} \times (-i\nabla)|^2 \right] \psi_i.$$

where e and f are constants and we have done integration by parts in the second equality assuming that ψ_i are non-decaying periodically oscillating functions. Then, we have

$$\begin{aligned} e_i(\mathbf{Q}) &= e(\mathbf{Q}_{\mathrm{IC}}^{(i)} \cdot \mathbf{Q} - |\mathbf{Q}_{\mathrm{IC}}^{(i)}|^2)^2 + f|\mathbf{Q}_{\mathrm{IC}}^{(i)} \times \mathbf{Q}|^2 \\ &= e(|\mathbf{Q}_{\mathrm{IC}}^{(i)}||\mathbf{Q}|\cos\phi_i - |\mathbf{Q}_{\mathrm{IC}}^{(i)}|^2)^2 + f|\mathbf{Q}_{\mathrm{IC}}^{(i)}|^2|\mathbf{Q}|^2\sin^2\phi_i \\ &= e|\mathbf{Q}_{\mathrm{IC}}^{(i)}|^2(|\mathbf{Q}|^2 - 2|\mathbf{Q}_{\mathrm{IC}}^{(i)}||\mathbf{Q}|\cos\phi_i + |\mathbf{Q}_{\mathrm{IC}}^{(i)}|^2) + (f-e)|\mathbf{Q}_{\mathrm{IC}}^{(i)}|^2|\mathbf{Q}|^2\sin^2\phi_i \\ &= e|\mathbf{Q}_{\mathrm{IC}}^{(i)}|^2|\mathbf{Q} - \mathbf{Q}_{\mathrm{IC}}^{(i)}|^2 + (f-e)|\mathbf{Q}_{\mathrm{IC}}^{(i)} \times \mathbf{Q}|^2. \end{aligned}$$

Now, assuming $e = f = s/(|\mathbf{Q}_{\mathrm{IC}}^{(i)}|^2|\mathbf{G}_i|^2)$ we obtain the type 2 free energy with

$$A_i(\mathbf{Q}, T) = T - T_C + s|\mathbf{Q} - \mathbf{Q}_{\mathrm{IC}}^{(i)}|^2/|\mathbf{G}_i|^2. \qquad \text{(Type 2)} \tag{D.8}$$

This $A_i(\mathbf{Q})$ has a circular (or elliptical if $e \neq f$) bulge around $\mathbf{Q}_{\mathrm{IC}}^{(i)}$. In their study of $2H$-$TaSe_2$, Nakanishi and Shiba obtained this form as an approximate form of Eq. (D.7) for $\mathbf{Q} \approx \mathbf{Q}_{\mathrm{IC}}^{(i)}$ [115], but it is actually also a special case of McMillan's original definition of free energy. Therefore, assuming $e = f$, the type 2 free energy can be used for general values of $\mathbf{Q}$.

D.2.3 CCDW STATE

For the C-CDW state we have $\phi_i(\mathbf{r}) = \Delta_{\mathrm{C}} e^{i\theta_{\mathrm{C}i}}$, where $\theta_{\mathrm{C}i}$ are constant phases which minimize the free energy F. Substituting this to Eq. (D.6), we obtain

$$F_{\mathrm{C}} = \mathrm{Area} \cdot f_{\mathrm{C}} \tag{D.9}$$

where Area $= \int d^2r$ and

$$f_{\rm C} = \sum_{i=1}^{3} A_i(\mathbf{Q}_{\rm C})\Delta_{\rm C}^2 + 3(B+C)\Delta_{\rm C}^4 + D\Delta_{\rm C}^3\cos(\theta_{\rm C1}+\theta_{\rm C2}+\theta_{\rm C3})$$
$$+\sum_{i=1}^{3}\begin{cases} U\Delta_{\rm C}^3\cos(2\theta_{{\rm C}i}-\theta_{{\rm C}i+1}) + V\Delta_{\rm C}^4\cos(\theta_{{\rm C}i}-2\theta_{{\rm C}i+1}-\theta_{{\rm C}i+2}) & \\ \qquad +Z\Delta_{\rm C}^4\cos(3\theta_{{\rm C}i}+\theta_{{\rm C}i+2}) & ,\ \text{if } (q,p)=(2,1) \\ Y\Delta_{\rm C}^4\cos(3\theta_{{\rm C}i}) + W\Delta_{\rm C}^4\cos(2\theta_{{\rm C}i}-\theta_{{\rm C}i+1}-\theta_{{\rm C}i+2}) & ,\ \text{if } (q,p)=(3,0) \\ E\Delta_{\rm C}^4\cos(3\theta_{{\rm C}i}-\theta_{{\rm C}i+1}) & ,\ \text{if } (q,p)=(3,1) \end{cases}$$

We assume that F_D and $F_{(q,p)}$ stabilize the triple CDW co-operatively, i.e. $D, E, Y, W, U, V, Z < 0$. In this case the free energy is minimized if θ_i satisfy the following conditions:

$$\theta_{\rm C1} = \frac{2\pi}{q^3-p^3}(q^2l+qpm+p^2n),$$
$$\theta_{\rm C2} = \frac{2\pi}{q^3-p^3}(q^2m+qpn+p^2l),$$
$$\theta_{\rm C3} = \frac{2\pi}{q^3-p^3}(q^2n+qpl+p^2m),$$
$$l+m+n = (q-p)\nu.$$

Note that these phases have the same structure as Eq. (D.5). Then, the commensurate free energy density becomes

$$f_{\rm C} = \sum_{i=1}^{3} A_i(\mathbf{Q}_{\rm C})\Delta_{\rm C}^2 + 3(B+C)\Delta_{\rm C}^4 + D\Delta_{\rm C}^3$$
$$+\begin{cases} 3U\Delta_{\rm C}^3 + 3V\Delta_{\rm C}^4 + 3Z\Delta_{\rm C}^4 & ,\ \text{if } (q,p)=(2,1) \\ 3Y\Delta_{\rm C}^4 + 3W\Delta_{\rm C}^4 & ,\ \text{if } (q,p)=(3,0) \\ 3E\Delta_{\rm C}^4 & ,\ \text{if } (q,p)=(3,1) \end{cases}$$

The magnitude of $\Delta_{\rm C}$ is obtained by $\partial F_{\rm C}/\partial\Delta_{\rm C} = 0$.

	u	v	s	B	C	D	E	Y	W	U	V	Z
$(3,1)$ values [114]	1	0.05	1200	1	2	-1	-1.5					
$(3,0)$ values [115]	1	0	1000	1	2	-0.2		-0.3	-1			
$(2,1)$ values	1	0.05	1000	1	2	-0.2				-0.3	-1.0	-1.5

D.2.4 ICCDW STATE

For other CDW states, $\phi_i(\mathbf{r})$ can be expanded as

$$\phi_i(\mathbf{r}) = \sum_{\substack{l,m,n\geq 0 \\ l\cdot m\cdot n=0}} \Delta^{(i)}_{lmn} \exp[i\mathbf{q}^{(i)}_{lmn}\cdot\mathbf{r}], \tag{D.10}$$

where l,m,n are integers and $\mathbf{q}^{(i)}_{lmn} = l\mathbf{k}^{(i)} + m\mathbf{k}^{(i+1)} + n\mathbf{k}^{(i+2)} + \mathbf{q}^{(i)}$, $\mathbf{k}^{(i)} = q\mathbf{q}^{(i)} - p\mathbf{q}^{(i+1)}$ and $\mathbf{q}^{(i)} = \mathbf{Q}^{(i)} - \mathbf{Q}^{(i)}_c$, and we have defined $\mathbf{q}^{(4)} = \mathbf{q}^{(1)}$, $\mathbf{q}^{(5)} = \mathbf{q}^{(2)}$. After substituting Eq. (D.10) into Eq. (D.6), the free energy is minimized to find the Δ_{lmn} values. The order parameters are determined by the non-linear coupled equation $\partial F/\partial\Delta^{(i)}_{lmn} = 0$.

Next, we expand and integrate F_A. The integral

$$\begin{aligned} F_A &= \int d^2r \sum_{i=1}^{3} \sum_{\substack{l,m,n\geq 0 \\ l\cdot m\cdot n=0}} \sum_{\substack{l',m',n'\geq 0 \\ l'\cdot m'\cdot n'=0}} \Delta^{(i)*}_{lmn}\Delta^{(i)}_{l'm'n'} e^{i\mathbf{q}^{(i)}_{lmn}\cdot\mathbf{r}} A_i(\mathbf{Q}^{(i)}_c - i\nabla) e^{-i\mathbf{q}^{(i)}_{l'm'n'}\cdot\mathbf{r}} \\ &= \int d^2r \sum_{i=1}^{3} \sum_{\substack{l,m,n\geq 0 \\ l\cdot m\cdot n=0}} \sum_{\substack{l',m',n'\geq 0 \\ l'\cdot m'\cdot n'=0}} \Delta^{(i)*}_{lmn}\Delta^{(i)}_{l'm'n'} e^{i(\mathbf{q}^{(i)}_{lmn} - \mathbf{q}^{(i)}_{l'm'n'})\cdot\mathbf{r}} A_i(\mathbf{Q}^{(i)}_c + \mathbf{q}^{(i)}_{l'm'n'}) \end{aligned}$$

is non-zero only if $\mathbf{q}^{(i)}_{lmn} - \mathbf{q}^{(i)}_{l'm'n'} = \mathbf{0}$, i.e.

$$(l-l')\mathbf{k}^{(i)} + (m-m')\mathbf{k}^{(i+2)} + (n-n')\mathbf{k}^{(i+2)} = \mathbf{0}.$$

From the triple-Q condition $\mathbf{Q}^{(1)} + \mathbf{Q}^{(2)} + \mathbf{Q}^{(3)} = \mathbf{0}$ we have $\mathbf{k}^{(i)} + \mathbf{k}^{(i+1)} + \mathbf{k}^{(i+2)} = \mathbf{0}$, so the non-zero terms of F_A are those with the condition $l-l' = m-m' = n-n'$. Moreover, the condition $l\cdot m\cdot n = 0 = l'\cdot m'\cdot n'$ implies that $l = l', m = m', n = n'$. Therefore, we

have

$$F_A = \sum_{i=1}^{3} \sum \Delta_{lmn}^{(i)*} \Delta_{lmn}^{(i)} A_i(\mathbf{Q}_C^{(i)} + q_{lmn}^{(i)}).$$

With similar calculations for F_B, F_C, F_D we obtain the following results:

$$\begin{aligned} F_B &= \int d^2 r B \sum_{i=1}^{3} \sum \Delta_{lmn}^{(i)} \Delta_{l'm'n'}^{(i)*} \Delta_{l''m''n''}^{(i)} \Delta_{l'''m'n'''}^{(i)*} e^{i(\mathbf{q}_{lmn}^{(i)} - \mathbf{q}_{l'm'n'}^{(i)} + \mathbf{q}_{l''m''n''}^{(i)} - \mathbf{q}_{l'''m'''n'''}^{(i)})} \\ &= B \sum_{i=1}^{3} \sum \Delta_{lmn}^{(i)*} \Delta_{l'm'n'}^{(i)} \Delta_{l''m''n''}^{(i)*} \Delta_{l'''m'''n'''}^{(i)} \\ &\quad \times \delta(l - l' + l'' - l''', m - m' + m'' - m''', n - n' + n'' - n'''), \end{aligned}$$

$$\begin{aligned} F_C &= \int d^2 r C \sum_{i=1}^{3} \sum \Delta_{lmn}^{(i)} \Delta_{l'm'n'}^{(i)*} \Delta_{l''m''n''}^{(i+1)} \Delta_{l'''m'''n'''}^{(i+1)*} e^{i(\mathbf{q}_{lmn}^{(i)} - \mathbf{q}_{l'm'n'}^{(i)} + \mathbf{q}_{l''m''n''}^{(i+1)} - \mathbf{q}_{l'''m'''n'''}^{(i+1)}) \cdot \mathbf{r}} \\ &= C \sum_{i=1}^{3} \sum \Delta_{lmn}^{(i)*} \Delta_{l'm'n'}^{(i)} \Delta_{l''m''n''}^{(i+1)*} \Delta_{l'''m'''n'''}^{(i+1)} \\ &\quad \times \delta(l - l' + n'' - n''', m - m' + l'' - l''', n - n' + m'' - m'''), \\ F_D &= \int d^2 r \frac{D}{2} \sum \Delta_{lmn}^{(1)} \Delta_{l'm'n'}^{(2)} \Delta_{l''m''n''}^{(3)} (e^{i(\mathbf{q}_{lmn}^{(1)} + \mathbf{q}_{l'm'n'}^{(2)} + \mathbf{q}_{l''m''n''}^{(3)}) \cdot \mathbf{r}} + \text{c.c.}) \\ &= D \sum \Delta_{lmn}^{(1)} \Delta_{l'm'n'}^{(2)} \Delta_{l''m''n''}^{(3)} \delta(l + n' + m'', m + l' + n'', n + m' + l''), \end{aligned}$$

and the commensurate free energies re

$$
\begin{aligned}
F_{(2,1)} = \int d^2r \sum_{i=1}^{3} \Big\{ & \frac{U}{2} \sum \Delta^{(i)}_{lmn} \Delta^{(i)}_{l'm'n'} \Delta^{(i+1)*}_{l''m''n''} (e^{i(\mathbf{q}^{(i)}_{lmn} + \mathbf{q}^{(i)}_{l'm'n'} - \mathbf{q}^{(i+1)}_{l''m''n''})} + \text{c.c.}) \\
& + \frac{V}{2} \sum \Delta^{(i)}_{lmn} \Delta^{(i+1)*}_{l'm'n'} \Delta^{(i+1)*}_{l''m''n''} \Delta^{(i+2)*}_{l'''m'''n'''} (e^{i(\mathbf{q}^{(i)}_{lmn} - \mathbf{q}^{(i+1)}_{l'm'n'} - \mathbf{q}^{(i+1)}_{l''m''n''} - \mathbf{q}^{(i+2)}_{l'''m'''n'''})} + \text{c.c.}) \\
& + \frac{Z}{2} \sum \Delta^{(i)}_{lmn} \Delta^{(i)}_{l'm'n'} \Delta^{(i)}_{l''m''n''} \Delta^{(i+2)}_{l'''m'''n'''} (e^{i(\mathbf{q}^{(i)}_{lmn} + \mathbf{q}^{(i)}_{l'm'n'} + \mathbf{q}^{(i)}_{l''m''n''} + \mathbf{q}^{i+1}_{l'''m'''n'''})} + \text{c.c.}) \Big\} \\
= U & \sum_{i=1}^{3} \sum \Delta^{(i)}_{lmn} \Delta^{(i)}_{l'm'n'} \Delta^{(i+1)*}_{l''m''n''} \delta(l + l' - n'' + 1, m + m' - l'', n + n' - m'') \\
+ V & \sum_{i=1}^{3} \sum [\Delta^{(i)}_{lmn} \Delta^{(i+1)*}_{l'm'n'} \Delta^{(i+1)*}_{l''m''n''} \Delta^{(i+2)*}_{l'''m'''n'''} \\
& \qquad \times \delta(l - n' - n'' - m''' + 1, m - l' - l'' - n''', n - m' - m'' - l''')] \\
+ Z & \sum_{i=1}^{3} \sum [\Delta^{(i)}_{lmn} \Delta^{(i)}_{l'm'n'} \Delta^{(i)}_{l''m''n''} \Delta^{(i+2)}_{l'''m'''n'''} \\
& \qquad \times \delta(l + l' + l'' + m''' + 1, m + m' + m'' + n''', n + n' + n'' + l''')],
\end{aligned}
$$

$$
\begin{aligned}
F_{(3,0)} = & \frac{Y}{2} \sum_{i=1}^{3} \sum \Delta^{(i)}_{lmn} \Delta^{(i)}_{l'm'n'} \Delta^{(i+1)*}_{l''m''n''} \delta(l + l' - n'' + 1, m + m' - l'', n + n' - m'') \\
& + \frac{W}{2} \sum_{i=1}^{3} \sum [\Delta^{(i)*}_{lmn} \Delta^{(i+1)*}_{l'm'n'} \Delta^{(i+1)*}_{l''m''n''} \Delta^{(i+2)*}_{l'''m'''n'''} \\
& \qquad \times \delta(l - n' - n'' - m''' + 1, m - l' - l'' - n''', n - m' - m'' - l''')],
\end{aligned}
$$

$$
\begin{aligned}
F_{(3,1)} = & \frac{E}{2} \sum_{i=1}^{3} \sum [\Delta^{(i)*}_{lmn} \Delta^{(i)}_{l'm'n'} \Delta^{(i)}_{l''m''n''} \Delta^{(i+1)*}_{l'''m'''n'''} \\
& \qquad \times \delta(l + l' + l'' - n''' + 1, m + m' + m'' - l''', n + n' + n'' - m''')].
\end{aligned}
$$

Here, $\delta(a, b, c)$ is the generalized delta function with $\delta(a, b, c) = 1$ if $a = b = c$ and $\delta(a, b, c) = 0$ otherwise. The sum $\sum$ is taken implicitly over all the integers $l, m, n, l', m', n', \ldots$ with the condition $l \cdot m \cdot n = l' \cdot m' \cdot n' = l'' \cdot m'' \cdot n'' = l''' \cdot m''' \cdot n''' = 0$.

References

[1] F. Wilczek, Phys. Rev. Lett. **109**, 160401 (2012).

[2] P. Holmvall, M. Fogelström, T. Löfwander, and A. B. Vorontsov, Phys. Rev. Research **2**, 013104 (2020).

[3] L.Guo, M. Marthaler, and G. Schön, Phys. Rev. Lett. **111**,205303 (2013).

[4] M. Tsubota, K. Inagaki, T. Matsuura, and S. Tanda, Europhys. Lett. **97**, 57011 (2012).

[5] L.D. Landau, L. P. Pitaevskii, and E. M. Lifshitz, *Statistical Physics, Part 1, 3rd Revised and Enlarged Edition* (Pergamon Press, Oxford, 1980).

[6] P. W. Anderson, *Basic notions of condensed matter physics* (AddisonWesley Publishing company, Reading, Massachusetts,1984).

[7] C. N. Yang, Rev. Mod. Phys. **34**, 694 (1962).

[8] G. Vidal, J. I. Latorre, E. Rico, and A. Kitaev, Phys. Rev. Lett. **90**, 227902(2003).

[9] P. Calabrese and J. Cardy, J. Stat. Mech.: Theory Exp. **2004**, P06002 (2004).

[10] J. van Wezel and J. van den Brink, Am. J. Phys. **75**, 635 (2007).

[11] P. Coleman, Nature **493**, 166 (2013).

[12] P. Bruno, Phys. Rev. Lett. **110**, 118901 (2013).

[13] F. Wilczek, Phys. Rev. Lett. **110**, 118902 (2013).

[14] P. Bruno, Patrick, Phys. Rev. Lett. **111**, 029301 (2013).

[15] P. Nozières, Europhys. Lett. **103**, 57008 (2013).

[16] P. Bruno, Phys. Rev. Lett. **111**, 070402(2013).

[17] H. Watanabe and M. Oshikawa, Phys. Rev. Lett. **114**, 251603 (2015).

[18] N. Yamamoto, Phys. Rev. D **92**, 085011 (2015).

[19] V. K. Kozin and O. Kyriienko, Phys. Rev. Lett. **123**, 210602 (2019).

[20] G. E. Volovik, JETP Letters **98**, 491 (2013).

[21] K. Sacha, Phys. Rev. A **91**, 033617 (2015).

[22] D. V. Else, B. Bauer, and C. Nayak, Chetan, Phys. Rev. Lett. **117**, 09040 (2016).

[23] V. Khemani, A. Lazarides, R. Moessner, and S. L. Sondhi, Phys. Rev. Lett. **116**, 250401 (2016).

[24] C. W. von Keyserlingk and S. L. Sondhi, Phys. Rev. B **93**, 245146 (2016).

[25] N. Y. Yao, A. C. Potter, I. -D. Potirniche, and A. Vishwanath, Phys. Rev. Lett. **118**, 030401 (2017).

[26] D. V. Else, B. Bauer, and C. Nayak, Phys. Rev. X **7**, 011026 (2017).

[27] S. Choi *et. al.*, Nature **543**, 9 (2017).

[28] J. Zhang *et. al.*, Nature **543**, 217 (2017).

[29] A. Arai, *Mathematical Aspects of quantum phenomena*, In Japanese (Asakura-shoten, 2006).

[30] Y. Ohnuki and S. Kitakado, J. Math. Phys. **34**, 2827 (1993).

[31] S. Tanimura, Prog. Theor. Phys. **90**, 271 (1993).

[32] A. Arai and Y. Matsuzawa, Rev. Math. Phys. **20**, 951 (2008).

[33] P. Carruthers and M. M. Nieto, Rev. Mod. Phys. **40**, 411 (1968).

[34] D. Sakabe, Z. Liu, K. Suenaga, K. Nakatsugawa, and S. Tanda, npj Quantum Materials **2**, 22(2017).

[35] A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. P. A. Fisher, A. Garg, and W. Zwerger, Rev. Mod. Phys. **59**, 1 (1987) .

[36] G. Grüner, *Density waves in solids* (Addison-Wesley Pub. Co., Advanced Book Program, 1994).

[37] S. Kagoshima, H. Nagasawa, T. Sambongi, *One-dimensional conductors*, (Springer-Verlag, 1988).

[38] P. Monceau, Adv. Phys. **61**, 325 (2012).

[39] A. Zettl, G. Grüner, and A. H. Thompson, Phys. Rev. B **26**, 5760 (1982).

[40] S. Tanda, T. Tsuneta, Y. Okajima, K. Inagaki, K. Yamaya, and N. Hatakenaka, Nature **417**, 397 (2002).

[41] T. Matsuura, K. Inagaki, and S. Tanda, Phys. Rev. B **79**, 014304 (2009).

[42] A.O. Caldeira and A.J. Leggett, Physica A **121**, 587(1983).

[43] U. Weiss, *Quantum Dissipative Systems* (World Scientific, 1999).

[44] E. N. Bogachek, I. V. Krive, I. O. Kulik, and A. S. Rozhavsky, Phys. Rev. B **42**, 7614 (1990).

[45] J. J. Sakurai, *Modern Quantum Mechanics, rev. ed.* (AddisonWesley, Reading, Massachusetts, 1993).

[46] B. Hall, *Quantum Theory for Mathematicians, Graduate Texts in Mathematics* (Springer, New York, 2013).

[47] P. Hanggi, *Generalized Langevin equations: A useful tool for the perplexed modeller of nonequilibrium fluctuations?* in *Stochastic Dynamics*, Lecture Notes in Physics, Vol. 484 (Springer, Berlin, 1997), pp. 15-22.

[48] H. Grabert and P. Schramm and G.-L. Ingold, Phys. Rep. **168**, 115 (1988).

[49] P. Schramm and H. Grabert, J. Stat. Phys. **49**, 767 (1987).

[50] P. A. Lee, T. M. Rice, and P. W. Anderson, Phys. Rev. Lett. **31**, 462 (1973).

[51] K. Nomura and K. Ichimura, J. Vac. Sci. Technol. A **8**, 504 (1990).

[52] M. Ido, Y. Okajima, H. Wakimoto, and M. Oda, Physica B+C **143**, 54 (1986).

[53] T. Li *et. al.*, Phys. Rev. Lett. **109**, 163001 (2012).

[54] F. Wilczek, Phys. Rev. Lett. **111**, 250402 (2013).

[55] A. V. Ustinov, T. Doderer, R. P. Huebener, N. F. Pedersen, B. Mayer, and V. A. Oboznov, Phys. Rev. Lett. **69**, 1815 (1992).

[56] H.-K. Li *et. al.*, Phys. Rev. Lett. **118**, 053001 (2017).

[57] V. J. Emery and J. D. Axe, Phys. Rev. Lett. **40**, 1507 (1978).

[58] I. U. Heilmann *et. al.*, Phys. Rev. B **20**, 751 (1979).

[59] R. V. Pai and R. Pandit, Phys. Rev. B **71**, 104508 (2005).

[60] R. Blinc and A. P. Levanyuk, *Incommensurate phases in dielectrics* (North-Holland, 1986).

[61] M. Reed and B. Simon, Fourier Analysis, Self-Adjointness (Methods of Modern Mathematical Physics, Vol. 2) (Academic Press, 1975).

[62] J.G. Muga and C.R. Leavens, Phys. Rep. **338**, 353 (2000).

[63] Y. Aharonov and D. Bohm, Phys. Rev. **122**, 1649 (1961).

[64] W. Pauli, in *Handbuch der Physik (Encyclopedia of physics)*, edited by S. Fludge, pp. 1-168.

[65] E. A. Galapon, Proc. Royal Soc. Lond. A: Math. Phys. Eng. Sci. **458**, 451 (2002).

[66] E. A. Galapon, R. F. Caballar, and R. T. Bahague Jr, Phys. Rev. Lett. **93**, 180406 (2004).

[67] E. A. Galapon, Proc. Royal Soc. Lond. A: Math. Phys. Eng. Sci. **458**, 2671 (2002).

[68] A. Arai, Lett. Math. Phys. **87**, 67 (2008).

[69] A. Arai, Rev. Math. Phys. **17**, 1071 (2005).

[70] C. M. Bender, S. Boettcher, and P. N. Meisinger, J. Math. Phys. **40**, 2201 (1999).

[71] C. M. Bender, Rep. Prog. Phys. **70**, 947 (2007).

[72] C. M. Bender, J. Phys. Conf. Ser. **631**, 012002 (2015).

[73] E. Recami, V. S. Olkhovsky, and S. P. Maydanyuk, Int. J. Mod. Phys. A **25**, 1785 (2010).

[74] K. Schmüdgen, J. Funct. Anal. **50**, 8 (1983).

[75] M. Miyamoto, J. Math. Phys. **42**, 1038 (2001).

[76] A. Arai and F. Hiroshima, Ann. Henri Poincaré **18**, 2995 (2017).

[77] L. de Broglie, *Recherches sur la théorie des Quanta*, Ph.D. thesis (University of Paris, 1924).

[78] S.-Y. Lan *et. al.*, Science 339, 554 (2013).

[79] K. Nakatsugawa, T. Fujii, and S. Tanda, Phys. Rev. B **96**, 094308 (2017).

[80] D. C. Brody, J. Phys. A: Math. Theor. **47**, 035305 (2014).

[81] S. Weinberg, *The Quantum Theory of Fields, Volume 1: Foundations* (Cambridge University Press, 2005).

[82] H. P. Robertson, Phys. Rev. **34**, 163 (1929).

[83] Y.-N. Dou and H.-K. Du, J. Math. Phys. **54**, 103508 (2013).

[84] J.-P. Luminet *et. al.*, Nature **425**, 593 (2003).

[85] G. Povie *et. al.*, Science **356**, 172 (2017).

[86] J. Erhart *et. al.*, Nature Phys. **8**, 185 (2012).

[87] E. Fradkin, S. A. Kivelson, and J. M. Tranquada, Rev. Mod. Phys. **87**, 457 (2015).

[88] W. L. McMillan, Phys. Rev. B **14**, 1496 (1976).

[89] P. Bak, D. Mukamel, J. Villain, and K. Wentowska, Phys. Rev. B **19**, 1610 (1979).

[90] K. Nakanishi and H. Shiba, J. Phys. Soc. Jpn. **53**, 1103 (1984).

[91] A. A. Sinchenko, P. D. Grigoriev, P. Lejay, and P. Monceau, Phys. Rev. Lett. **112**, 036601 (2014).

[92] J. A. Wilson, F. J. Di Salvo, and S. Mahajan, Phys. Rev. Lett **32**, 882 (1974).

[93] J. A. Wilson, F. J. Di Salvo, and S. Mahajan, Advances in Physics **24**, 117 (1975).

[94] G. Grüner, Rev. Mod. Phys. **60**, 1129 (1988).

[95] N. P. Ong and P. Monceau, Phys. Rev. B **16**, 3443 (1977).

[96] R. M. Fleming, D. E. Moncton, D. B. McWhan, and F. J. DiSalvo, Phys. Rev. Lett. **45**, 576 (1980).

[97] R. V. Coleman *et. al.*, Adv. Phys. **37**, 559 (1988).

[98] S. Tanda, T. Sambongi, T. Tani, and S. Tanaka, J. Phys. Soc. Jpn. **53**, 476 (1984).

[99] S. Tanda and T. Sambongi, Synth. Met. **11**, 85 (1985).

[100] M. Yoshida *et. al.*, Sci. Rep. **4**, 7302 (2014).

[101] R. Hovden *et. al.*, PNAS **113**,11420 (2016).

[102] Y. Yu *et. al.*, Nat. Nanotechnol. **10**, 270-276 (2015).

[103] A. W. Tsen *et. al.*, PNAS **112**, 5054 (2015).

[104] O. R. Albertin *et. al.*, Phys. Rev. B **93**, 214109 (2016).

[105] R. He *et. al.*, Phys. Rev. B **94**, 201108(R) (2016).

[106] A. Zong *et. al.*, Sci. Adv. **4**, eaau5501 (2018).

[107] G. Lantz *et. al.*, Phys. Rev. B **96**, 224101 (2017).

[108] I. Vaskivskyi *et. al.*, Sci. Adv **1**, e1500168 (2015).

[109] Y. A. Gerasimenko *et. al.*, npj Quantum Materials **4**, 32 (2019).

[110] P. C. Börner *et. al.*, Appl. Phys. Lett.**113**, 173103 (2018).

[111] H. Ryu *et. al.*, Nano Lett. **18**, 689 (2018).

[112] W. L. McMillan, Phys. Rev. B **12**, 1187 (1975).

[113] R. N. Bhatt and W. L. McMillan, Phys. Rev. B **12**, 2042 (1975).

[114] K. Nakanishi and H. Shiba, J. Phys. Soc. Jpn. **43**, 1839 (1977).

[115] K. Nakanishi and H. Shiba, J. Phys. Soc. Jpn. **44**, 1465 (1978).

[116] Y. Yamada, H. Takatera, Solid State Commun. **21**, 41 (1977).

[117] T. Kurosawa and H. Kondo, private communication.

[118] T. Toshima, K. Inagaki, N. Hatakenaka, and S. Tanda, J. Phys. Soc. Jpn **75**, 024706 (2006).

[119] J. M. Kosterlitz and D. J. Thouless, J. Phys. C: Solid State Phys **6**, 1181 (1973).

[120] J. D. Bishop and D. J. Reppy, Phys. Rev. Lett. **40**, 1727 (1978).

[121] L. Stojchevska *et. al.*, Science **344**, 177 (2014).

[122] S. Brazovskii, J. Supercond. Nov. Mag. **28**, 1349 (2015).

[123] J. Ravnik, I. Vaskivskyi, T. Mertelj, and D. Mihailovic, Phys. Rev. B **97**, 075304 (2018).

[124] W. Yao *et. al.*, PNAS **115**, 6928 (2018).

[125] C. B. Scruby, P. M. Williams, and G. S. Parry, Philos. Mag. **31**, 255 (1975).

[126] H.-P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University Press, 2007)

[127] K. Nakanishi, H. Takatera, Y. Yamada, and H. Shiba, J. Phys. Soc. Jpn. **43**, 1509 (1977)