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Multiple- Q instability under fourth-order inplane single-ion anisotropy

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Abstract

Multiple- Q instability under tetragonal single-ion anisotropy is theoretically investigated. By performing the simulated annealing for a spin model with the momentum-resolved interaction and fourth-order inplane single-ion anisotropy on a square lattice, we find that the interplay between the competing exchange interactions in momentum space and single-ion anisotropy leads to various field-induced double- Q states in the ground state. In particular, we show that a double- Q vortex crystal emerges when the interactions at high-harmonic wave vectors are comparable to those at wave vectors that give the dominant contributions. Our results underscore the importance of magnetic anisotropy, which brings about the multiple- Q instability in centrosymmetric magnets.

Keywords: multiple- Q magnetic state, single-ion anisotropy, square lattice, noncoplanar spin texture, magnetic frustration

1. Introduction

Multiple- Q magnetic states as a consequence of a superposition of multiple spin density waves have long been studied in condensed matter physics [1, 2, 3, 4, 5, 6, 7]. Depending on its constituent waves, a plethora of noncollinear and noncoplanar spin textures, such as a skyrmion, hedgehog, and vortex, emerge in magnetic systems [8], which lead to unconventional phenomena like the topological Hall and Nernst effects [9, 10, 11, 12, 13, 14, 15, 16]. In addition, multiple- Q states often give rise to further intriguing physical phenomena, such as the electric polarization [17, 18, 19, 20, 21], charge density waves [22, 23, 24], antisymmetric spin polarization [25], nonreciprocal transport [26], and ferroaxial nature [27], owing to the symmetry lowering under noncollinear and noncoplanar spin textures.

Despite its complicated spin textures, multiple- Q states have been ubiquitously found in magnetic materials irrespective of the lattice structures and

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chemical elements. For example, magnetic skyrmion crystal (SkX) and hedgehog crystal have been observed in materials with different lattice structures [28]; triangular SkX in noncentrosymmetric d -electron compounds, MnSi [29, 30, 11], $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ [31, 32], FeGe [33, 34], and Cu_2OSeO_3 [35, 36, 37], and the f -electron compound, EuPtSi [38, 39, 40, 41, 42], and the centrosymmetric d -electron compound, MnSc_2S_4 [43, 44, 45, 46], and f -electron compounds, Gd_2PdSi_3 [15, 47, 48, 49] and $\text{Gd}_3\text{Ru}_4\text{Al}_{12}$ [50, 51, 52, 53], square SkX in centrosymmetric f -electron compounds, GdRu_2Si_2 [54, 55, 56] and EuAl_4 [57, 58, 59, 60, 61], and hedgehog crystals in the noncentrosymmetric d -electron compound $\text{MnSi}_{1-x}\text{Ge}_x$ [62, 63, 64, 65, 66, 67] and the centrosymmetric d -electron compound SrFeO_3 [68, 69, 70]. Simultaneously, various stabilization mechanisms to realize multiple- Q states have been clarified, such as the multiple-spin interaction [71, 72, 73], frustrated competing interaction [74, 75, 76], long-range interaction [77, 78, 79, 80], and effective interaction arising from the Fermi surface instability [81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91].

Magnetic anisotropy is another source to stabilize the multiple- Q states. One of the most familiar examples is the Dzyaloshinskii-Moriya (DM) interaction [92, 93, 1, 2, 3, 5, 94, 95, 96], which originates from relativistic spin-orbit coupling in noncentrosymmetric lattice systems. The competition between the ferromagnetic exchange interaction and DM interaction leads to instability toward the spiral (single- Q) state, which results in the emergence of the SkXs under an external magnetic field. Recently, it was shown that a staggered-type DM interaction induces the SkXs even in centrosymmetric systems [97, 98, 99]. Another example is the symmetric anisotropic exchange interaction, which also arises from spin-orbit coupling but does not require the inversion symmetry breaking. By combining the competing exchange interactions, such a symmetric anisotropic exchange interaction can become a source of SkXs [100, 101, 102, 103, 104, 105, 106] and vortex crystals [107, 108, 109] in centrosymmetric hosts.

In the present study, we focus on another type of magnetic anisotropy, a single-ion anisotropy, which also tends to stabilize the SkX and other multiple- Q states. For example, the uniaxial easy-axis anisotropy leads to the instability toward the SkX and other multiple- Q states under an external magnetic field in both localized spin systems [110, 111, 112, 113] and itinerant electron systems [114, 90]. Furthermore, the effect of the higher-order single-ion anisotropy has been recently investigated in hexagonal- and trigonal-lattice systems in a systematic way, where the emergence of not only the SkX but also other multiple- Q states has been identified [115]. We extend such an analysis incorporating the higher-order single-ion anisotropy to tetragonal-lattice cases, where the fourth-order inplane single-ion anisotropy is inherent owing to the fourfold rotation symmetry of tetragonal systems. By performing the simulated annealing for the effective spin model with momentum-resolved interaction on a two-dimensional square lattice, we find that only the single- Q spiral state appears under the fourth-order inplane single-ion anisotropy in contrast to the conventional (second-order) single-ion anisotropy, the latter of which tends to favor the SkX. On the other hand, we show that various double- Q instabilities

occur by taking into account the competing exchange interaction in momentum space [116]. Our result indicates a possibility of the multiple- Q formation in magnetic systems with higher-order single-ion anisotropy.

The rest of this paper is organized as follows. In Sec. 2, we introduce an effective spin model consisting of the momentum-resolved interaction, fourth-order inplane single-ion anisotropy, and external magnetic field. We also describe a numerical method based on simulated annealing. In Sec. 3, we show the result in the presence of the fourth-order inplane single-ion anisotropy, where only the single- Q spiral instability is found. Then, we discuss a possible double- Q instability by additionally considering the effect of competing interactions in momentum space in Sec. 4. Finally, Sec. 5 is devoted to a summary of this paper.

2. Model and method

We start by considering a localized spin model on a two-dimensional square lattice belonging to the point group D_{4h} , whose Hamiltonian is given by

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - B \sum_i \{[(S_i^x)^2 - (S_i^y)^2]^2 - 4(S_i^x)^2(S_i^y)^2\} - H \sum_i S_i^z, \quad (1)$$

where $\mathbf{S}_i = (S_i^x, S_i^y, S_i^z)$ is the classical localized spin at site i ; the amplitude of $\mathbf{S}_i$ is set to be one. The first term represents the exchange interaction between i th and j th spins. We tune the interaction parameters so that the Fourier transform of J_{ij} , i.e., $J(\mathbf{q})$, shows the minima at $\mathbf{Q}_1 = (\pi/4, 0)$ and $\mathbf{Q}_2 = (0, \pi/4)$, where we set the lattice constant of the square lattice as unity. For example, such peak structures at $\mathbf{Q}_1$ and $\mathbf{Q}_2$ are constructed by choosing $(J_1, J_2, J_3) = (-1, 2/5, \sqrt{2}/20)$, $(J_1, J_2, J_3) = (-1, 3/10, \sqrt{2}/10)$, and so on, where J_1 , J_2 , and J_3 are the nearest-neighbor, second-nearest-neighbor, and third-nearest-neighbor exchange interactions, respectively. By focusing on the ground-state spin configuration of the model, where the internal energy is almost determined by the contribution at the dominant ordering vectors, we simplify the interaction term as follows:

$$\sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \sim 2J \sum_{\nu} \mathbf{S}_{\mathbf{Q}_{\nu}} \cdot \mathbf{S}_{-\mathbf{Q}_{\nu}}, \quad (2)$$

where ν is the label of the dominant ordering vectors; we take $\nu = 1-2$ in Sec. 3 and $\nu = 1-6$ in Sec. 4. We set $\mathbf{Q}_1 = (\pi/4, 0)$ and $\mathbf{Q}_2 = (0, \pi/4)$ in the following calculations. The prefactor 2 of the right side in Eq. (2) is due to the contributions at $-\mathbf{Q}_{\nu}$. We neglect the contributions from other wave vectors; we consider the contributions from high-harmonic wave vectors in Sec. 4. We set $J = 1$ as the energy unit of the model in Eq. (1).

The second term in Eq. (1) represents the fourth-order single-ion anisotropy that arises from the spin-orbit coupling under the tetragonal symmetry. As its angle dependence is given by $\cos 4\theta$ for $S^x = \cos \theta$ and $S^y = \sin \theta$, the energy

tends to become lower when the spins align the $\pm x$ and $\pm y$ directions. The sign of B is irrelevant in the following results, as its reversal just gives the rotation of the spin structure by $\pi/4$; we set $B > 0$. The third term in Eq. (1) represents the Zeeman coupling to an external magnetic field along the out-of-plane direction, $H > 0$.

The ground-state spin patterns while changing B and H are calculated by performing the simulated annealing, where the local updates for localized spins are done based on the Metropolis algorithm. In the simulations, an initial spin configuration is randomly given at high temperatures $T_0 = 1-10$. Then, the temperature is gradually reduced from T_0 to the final low temperature $T_f = 0.01$ by the annealing rate $\alpha = 0.99999-0.999999$ and $T_{n+1} = \alpha T_n$ in each Monte Carlo step. After equilibration at T_f , 10^5-10^6 Monte Carlo sweeps are used for measurements. The above process is independently done in each set of model parameters. The system size is set as $N = 32^2$ to be commensurate with $\mathbf{Q}_\nu$.

We calculate the magnetic moment at wave vector $\mathbf{q}$ and spin component $\mu = x, y, z$, $m_\mathbf{q}^\mu$, in the resultant spin configurations, which is given by

$$m_\mathbf{q}^\mu = \sqrt{\frac{S_s^{\mu\mu}(\mathbf{q})}{N}}, \quad (3)$$

$$S_s^{\mu\mu}(\mathbf{q}) = \frac{1}{N} \sum_{i,j} \langle S_i^\mu S_j^\mu \rangle e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}, \quad (4)$$

where $\langle \dots \rangle$ represents the average over the Monte Carlo samplings. The net magnetization along the z direction is given by $M^z = (1/N) \sum_i S_i^z$.

3. $xy(x^2 - y^2)$ -type tetragonal single-ion anisotropy

Let us discuss the optimal spin configurations against H for several B . Figures 1(a) and 1(b) show the H dependence of the magnetization M^z and the magnetic moments at $\mathbf{Q}_1 = (\pi/4, 0)$ and $\mathbf{Q}_2 = (0, \pi/4)$ and $\mu = x, y, z$, $(m_\mathbf{q}^\mu)^2$, respectively, at $B = 0.1$. At $H = 0$, the single- Q vertical spiral state is stabilized, whose spin configuration in real space is shown in Fig. 2(a); the spin rotates in the yz plane. Owing to the presence of inplane anisotropy B , $(m_{\mathbf{Q}_1}^y)^2 > (m_{\mathbf{Q}_1}^z)^2$, as shown in Fig. 1(b). It is noted that another spiral state, whose spiral plane lies on the xz plane, is energetically degenerate. In other words, the cycloidal spiral state whose spiral plane lies on the xz plane is not distinguished from the proper-screw spiral state whose spiral plane lies on the yz plane when the ordering vector is along $\mathbf{Q}_1$. This is why we refer to this state as the single- Q vertical spiral state [110, 111].

When the magnetic field is increased, the single- Q vertical spiral state turns into the single- Q conical spiral state with a jump of M^z , as shown in Fig. 1(a). Accordingly, the spiral plane changes from the xz (or yz) plane to the xy plane, as found in nonzero $(m_{\mathbf{Q}_1}^x)^2, (m_{\mathbf{Q}_1}^y)^2$ but $(m_{\mathbf{Q}_1}^z)^2 = 0$ in Fig. 1(b); the amplitudes of x and y components are the same, i.e., $(m_{\mathbf{Q}_1}^x)^2 = (m_{\mathbf{Q}_1}^y)^2$. The real-space spin configuration of the single- Q conical spiral state is shown in Fig. 2(b). It

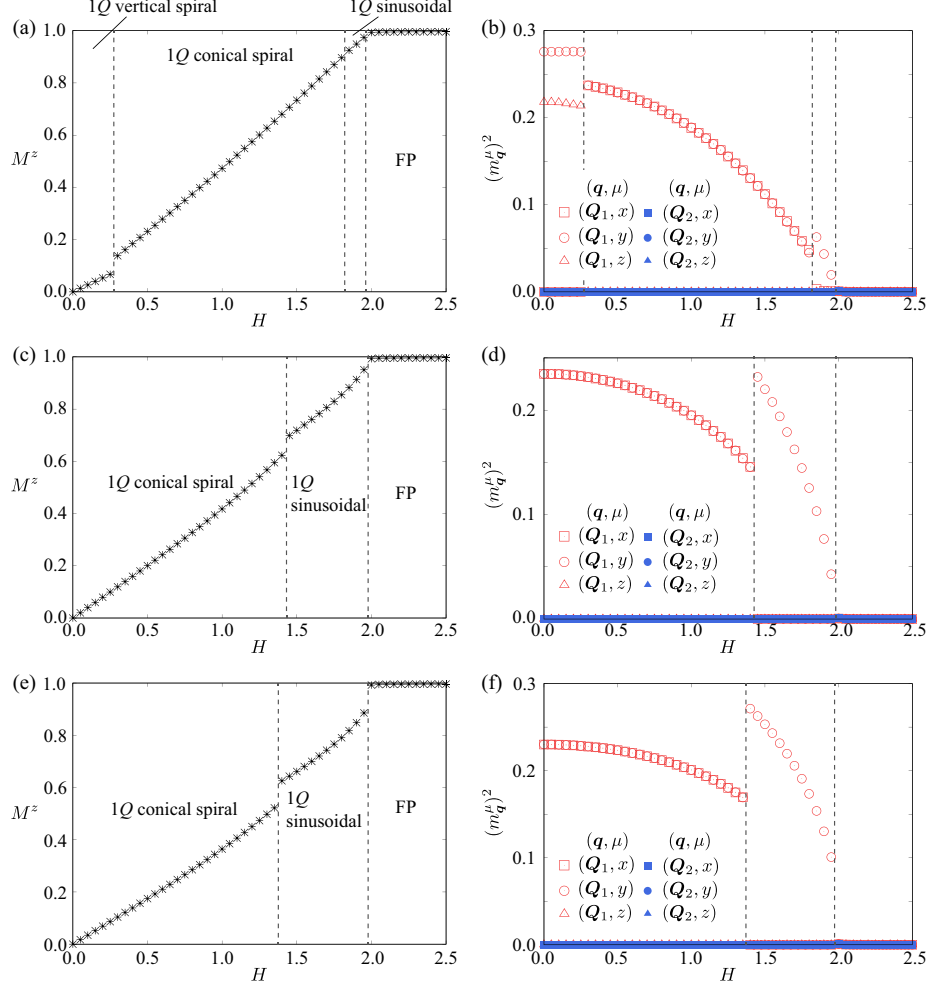


Figure 1: H dependence of (a,c,e) the magnetization, M^z , and (b,d,f) the magnetic moments for $\mathbf{q} = \mathbf{Q}_1, \mathbf{Q}_2$ and $\mu = x, y, z$, $(m_q^\mu)^2$, at (a,b) $B = 0.1$, (c,d) $B = 0.2$, and (e,f) $B = 0.3$. The dashed vertical lines represent the phase boundaries between different magnetic phases. FP represents the fully-polarized state. 1Q represents the single-Q state.

is noted that the spiral plane is not circular owing to the presence of inplane anisotropy B , which tends to align the spins in the x or y direction. Indeed, the spin directions seem to be fixed almost in the x or y direction in the obtained spin configuration in Fig. 2(b).

With a further increase of B , the single-Q conical spiral state is replaced by the single-Q sinusoidal state. Although the change of M is small in the transition shown in Fig. 1(a), the magnetic moments behaves discontinuously; $(m_{Q_1}^x)^2$ becomes almost zero in Fig. 1(b). In other words, the spin spatially

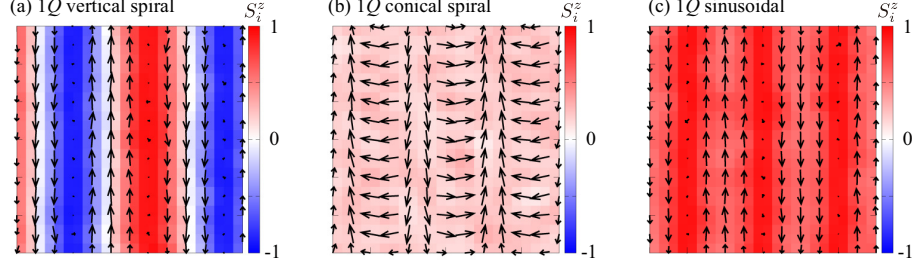


Figure 2: Real-space spin configurations obtained by simulated annealing in (a) the 1 Q vertical spiral state at $B = 0.1$ and $H = 0.2$, (b) the 1 Q conical spiral state at $B = 0.2$ and $H = 0.5$, and (c) the 1 Q sinusoidal state at $B = 0.5$ and $H = 1.5$. The arrows represent the xy components of spins, (S_i^x, S_i^y) , and the color stands for the z component, S_i^z .

oscillates only in the y direction, as shown in the real-space spin configuration in Fig. 2(c). This state changes into a fully-polarized state when the magnetic field reaches around 2.

When the magnitude of B is increased, the single- Q vertical spiral state is no longer stabilized, as shown in the case of $B = 0.2$ in Fig. 1(c) and $B = 0.3$ in Fig. 1(e). Both regions of the single- Q conical spiral and sinusoidal states tend to be extended to a lower-field region. The behavior of magnetic moments at $\mathbf{Q}_1$ in both phases are qualitatively similar to that at $B = 0.1$, as shown in Figs. 1(b), 1(d), and 1(f). In this way, one finds that the introduction of B is not enough to induce the multiple- Q instability. This feature is in contrast to the result in the system with the hexagonal-type inplane single-ion anisotropy on a triangular lattice, where several triple- Q states emerge depending on the magnitude of anisotropy and magnetic field [115].

4. Effect of competing interactions in momentum space

To enhance the multiple- Q instability, we additionally take into account the contributions of the interaction from other wave vectors in Eq. (2). In particular, we consider the contribution of the interaction at high-harmonic wave vectors, such as $\mathbf{Q}_1 + \mathbf{Q}_2$. This is because such an interaction assists the formation of the multiple- Q state rather than the single- Q state [113, 116]. Specifically, we consider two types of interactions, which are represented by

$$\mathcal{H}' = J' \sum_{\nu=3,4} \mathbf{S}_{\mathbf{Q}_\nu} \cdot \mathbf{S}_{-\mathbf{Q}_\nu}, \quad (5)$$

$$\mathcal{H}'' = J'' \sum_{\nu=5,6} \mathbf{S}_{\mathbf{Q}_\nu} \cdot \mathbf{S}_{-\mathbf{Q}_\nu}, \quad (6)$$

where $\mathbf{Q}_3 = \mathbf{Q}_1 + \mathbf{Q}_2$, $\mathbf{Q}_4 = \mathbf{Q}_1 - \mathbf{Q}_2$, $\mathbf{Q}_5 = (\mathbf{Q}_1 + \mathbf{Q}_2)/2$, and $\mathbf{Q}_6 = (\mathbf{Q}_1 - \mathbf{Q}_2)/2$, as shown in Figs. 3(a) and 3(b). Thus, $\mathbf{Q}_3$ and $\mathbf{Q}_4$ are high-harmonic wave

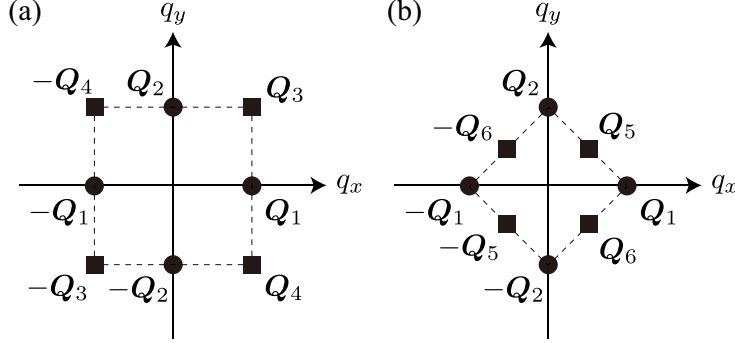


Figure 3: Interaction channels in momentum space. The interactions at $\pm\mathbf{Q}_1 = \pm(Q, 0)$ and $\pm\mathbf{Q}_2 = \pm(0, Q)$ are set to be dominant, while those at $\pm\mathbf{Q}_3 = \pm(\mathbf{Q}_1 + \mathbf{Q}_2)$ and $\pm\mathbf{Q}_4 = \pm(\mathbf{Q}_1 - \mathbf{Q}_2)$ [$\pm\mathbf{Q}_5 = \pm(\mathbf{Q}_1 + \mathbf{Q}_2)/2$ and $\pm\mathbf{Q}_6 = \pm(\mathbf{Q}_1 - \mathbf{Q}_2)/2$] are set to be subdominant in (a) [(b)].

vectors of $\mathbf{Q}_1$ and $\mathbf{Q}_2$ [Fig. 3(a)], while $\mathbf{Q}_1$ and $\mathbf{Q}_2$ are high-harmonic wave
vectors of $\mathbf{Q}_5$ and $\mathbf{Q}_6$ [Fig. 3(b)]. We set $J' = \kappa_1 J$ and $J'' = \kappa_2 J$, where κ_1
and κ_2 represent the ratio of the interaction. The values of κ_1 and κ_2 can be
close to one depending on the real-space exchange interaction. For example
in the case of the J_1 - J_3 Heisenberg model, $\kappa_1 \simeq 0.976$ and $\kappa_2 \simeq 0.995$ for
 $(J_1, J_2, J_3) = (-1, 3/10, \sqrt{2}/10)$. Such a situation often happens when the shape
of the energy landscape of $J(\mathbf{q})$ [$J(\mathbf{q})$ is the Fourier transform of the real-space
exchange interactions J_{ij}] is relatively close to circular; the energy for the spiral
waves at $\mathbf{Q}_1$ and $\mathbf{Q}_2$ along the $\langle 100 \rangle$ directions is close to those at $\mathbf{Q}_3$ - $\mathbf{Q}_6$ along
the $\langle 110 \rangle$ directions [104]. In the following, we discuss the result for $\mathcal{H} + \mathcal{H}'$ in
Sec. 4.1 and that for $\mathcal{H} + \mathcal{H}''$ in Sec. 4.2.

4.1. Interaction at $\mathbf{Q}_1 \pm \mathbf{Q}_2$

Figure 4 shows the phase diagram while changing κ_1 and H for $B = 0.2$. For
 $\kappa_1 = 0$, only the single- Q conical spiral and single- Q sinusoidal states appear, as
discussed in Sec. 3. When increasing κ_1 , one finds that the double- Q sinusoidal
state appears in the high-field region of the single- Q sinusoidal state for $\kappa_1 \gtrsim 0.3$.
The transition from the single- Q sinusoidal state to the double- Q sinusoidal state
is of first order with a slight jump of M^z in Fig. 5(a). As shown in Fig. 6(a),
this state is characterized by $(m_{\mathbf{Q}_1}^y)^2 = (m_{\mathbf{Q}_2}^y)^2$. It is noted that this state also
has nonzero magnetic moments at $\mathbf{Q}_3$ and $\mathbf{Q}_4$, as shown in Fig. 6(b). Owing to
nonzero $m_{\mathbf{Q}_3}^z = m_{\mathbf{Q}_4}^z$, the interactions at $\mathbf{Q}_3$ and $\mathbf{Q}_4$ contributes to the energy
gain, which results in stabilizing the double- Q state instead of the single- Q state.
The real-space spin configuration is shown in Fig. 7(a), where the regions with
 $S_i^y \sim +1$ and $S_i^y \sim -1$ form two interpenetrating square lattice with the length
of 8. Meanwhile, the z -spin configuration is modulated to form another square
lattice with the length of $4\sqrt{2}$, which corresponds to nonzero $m_{\mathbf{Q}_3}^z$ and $m_{\mathbf{Q}_4}^z$.
This state breaks fourfold rotational symmetry.

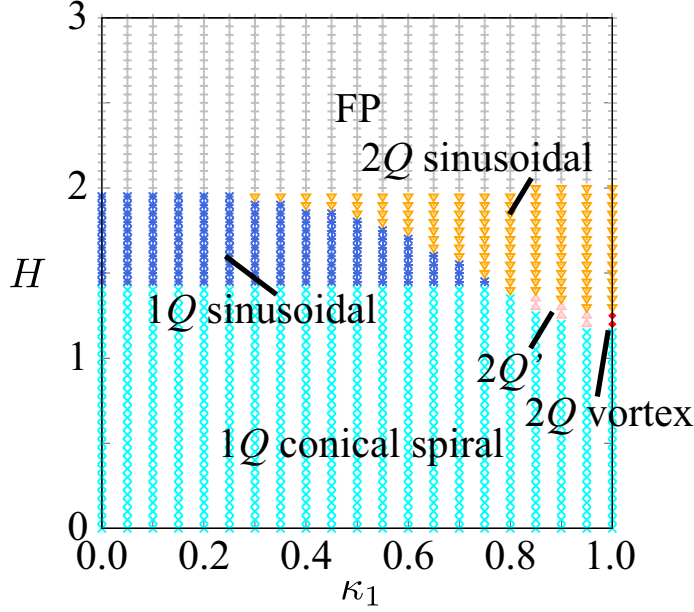


Figure 4: Magnetic phase diagram in the plane of κ_1 and H at $B = 0.2$ ($\kappa_2 = 0$). FP represents the fully-polarized state. 1Q and 2Q represent the single-Q and double-Q states, respectively.

When κ_1 is comparable to 1, i.e., $J' \simeq J$, two additional states, the double-Q' and double-Q vortex states, appear between the single-Q conical spiral and double-Q sinusoidal states, as shown in Fig. 4. The double-Q' state appears for $0.85 \lesssim \kappa_1 \lesssim 0.95$ in the phase diagram. The magnetization in the double-Q' state is smoothly connected to that in the single-Q conical state, as shown in Fig. 5(b). In addition, the spin texture in this state is similar to that in the single-Q conical state, but the additional magnetic moment, $(m_{\mathbf{Q}_2}^z)^2$, becomes nonzero, as shown in Fig. 6(c). The amplitude of $(m_{\mathbf{Q}_2}^z)^2$ is much smaller than those of $(m_{\mathbf{Q}_1}^x)^2$ and $(m_{\mathbf{Q}_1}^y)^2$; Q' means the different amplitudes at $\mathbf{Q}_1$ and $\mathbf{Q}_2$. Reflecting the double-Q structure, the magnitudes of $(m_{\mathbf{Q}_3}^x)^2$ and $(m_{\mathbf{Q}_4}^x)^2$ take nonzero values in Fig. 6(d). The real-space spin texture in Fig. 7(b) is similar to that in the single-Q conical state except for the slight modulation of the z component along the $\mathbf{Q}_2$ direction. This state breaks both fourfold rotational symmetry and spatial inversion symmetry.

For $\kappa_1 \simeq 1$, the double-Q vortex state, which breaks fourfold rotational symmetry, appears in Fig. 4. This state is distinguished from the single-Q conical state and the double-Q sinusoidal state by the uniform magnetization; there is a clear jump of M_z , as shown in Fig. 5(c). This state is characterized by the dominant contributions from $(m_{\mathbf{Q}_1}^y)^2$ and $(m_{\mathbf{Q}_2}^y)^2$ with equal amplitude in Fig. 6(e) and the subdominant contributions from $(m_{\mathbf{Q}_3}^x)^2 = (m_{\mathbf{Q}_4}^x)^2$ and

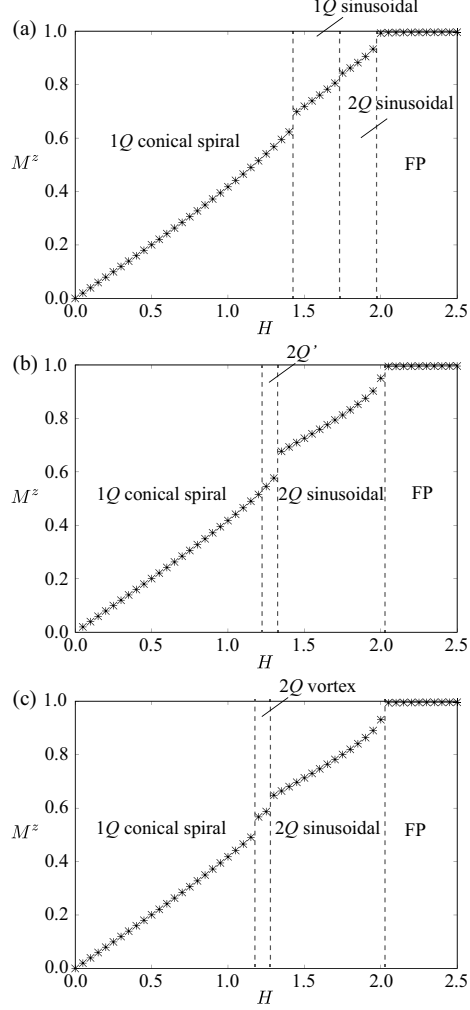


Figure 5: H dependence of M^z at (a) $\kappa_1 = 0.6$, (b) $\kappa_1 = 0.9$, and (c) $\kappa_1 = 1$. The other parameters are set as $B = 0.2$ and $\kappa_2 = 0$. The dashed vertical lines represent the phase boundaries between different magnetic phases.

210 $(m_{Q_3}^z)^2 = (m_{Q_4}^z)^2$ in Fig. 6(f). This state exhibits a characteristic feature in
 the real-space spin configuration. As shown in Fig. 7(c), the vortex alignment
 appears in the region for $S_i^z \simeq 1$. There are two vortices: One is the vortex
 with the winding number of $+2$ and the other is that of -2 for the inplane spin
 component. The doubling of the winding number in vortices is attributed to a
 large contribution from the magnetic moments at Q_3 and Q_4 . In other words,
 215 the four- Q -like modulation plays an important role in obtaining the state with
 a high winding number, which was also suggested in the model with different

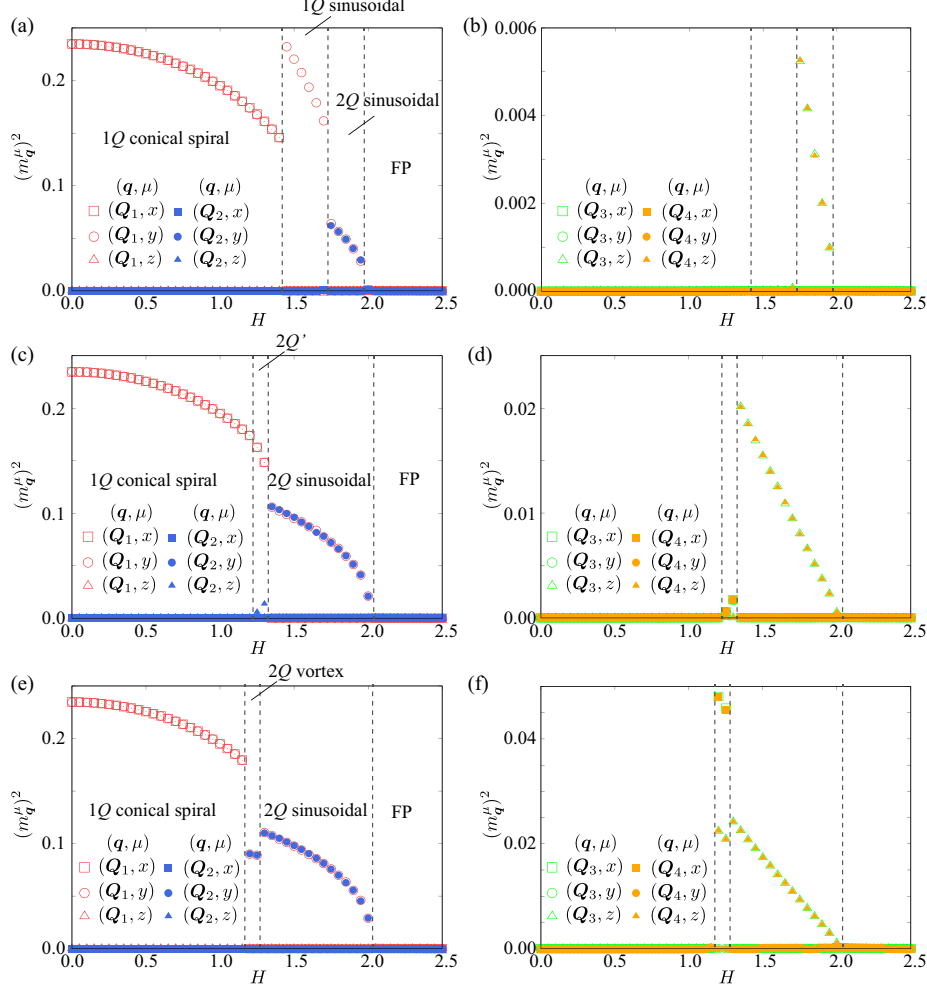


Figure 6: H dependence of (a,c,e) $(m_{Q_1, Q_2}^\mu)^2$ and (b,d,f) $(m_{Q_3, Q_4}^\mu)^2$ for $\mu = x, y, z$ at (a,b) $\kappa_1 = 0.6$, (c,d) $\kappa_1 = 0.9$, and (e,f) $\kappa_1 = 1$. The other parameters are set as $B = 0.2$ and $\kappa_2 = 0$. The dashed vertical lines represent the phase boundaries between different magnetic phases.

types of magnetic anisotropy [104, 116]. Nevertheless, we refer to this state as the double- Q vortex state, since the dominant contributions are expressed as Q_1 and Q_2 .

220

Let us briefly discuss the difference from the result with the easy-axis single-ion anisotropy $-A \sum_i (S_i^z)^2$ [113]. For $A \neq 0$ but $B = 0$, it was shown that the SkX with nonzero integer topological number is stabilized when taking into account κ_1 . Meanwhile, the instability toward the double- Q sinusoidal and double- Q vortex states in Fig. 7 has not been observed. Thus, the multiple- Q

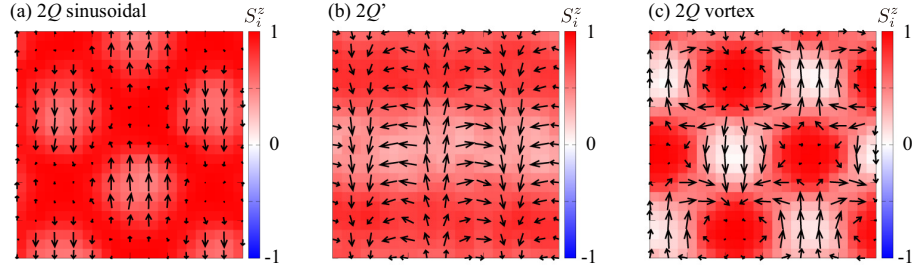


Figure 7: Real-space spin configurations obtained by simulated annealing in (a) the $2Q$ sinusoidal state at $\kappa_1 = 0.6$ and $H = 1.75$, (b) the $2Q'$ state at $\kappa_1 = 0.9$ and $H = 1.3$, and (c) the $2Q$ vortex state at $\kappa_1 = 1$ and $H = 1.2$. The arrows represent the xy components of spins, (S_i^x, S_i^y) , and the color stands for the z component, S_i^z .

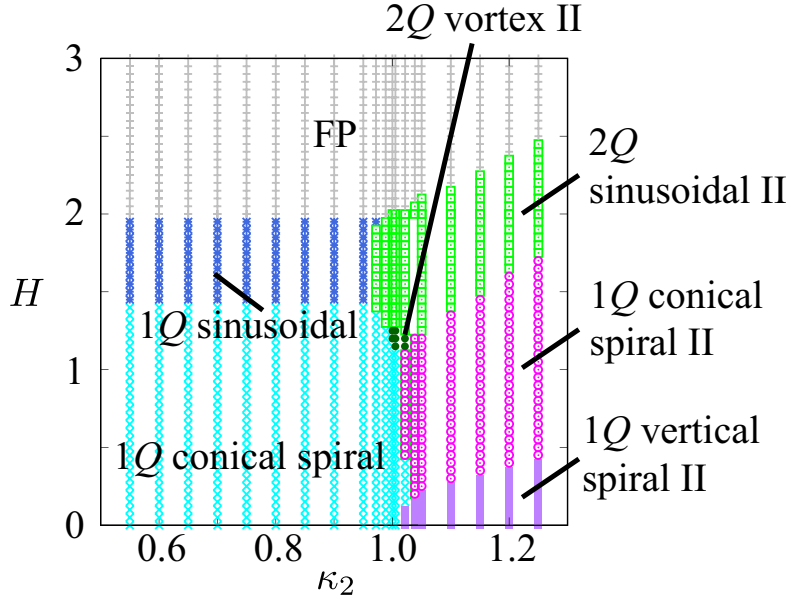


Figure 8: Magnetic phase diagram in the plane of κ_2 and H at $B = 0.2$ ($\kappa_1 = 0$). FP represents the fully-polarized state. $1Q$ and $2Q$ represent the single- Q and double- Q states, respectively.

instability is qualitatively different between the easy-axis single-ion anisotropy and the fourth-order inplane single-ion anisotropy.

4.2. Interaction at $(Q_1 \pm Q_2)/2$

Next, we discuss the case for $\mathcal{H} + \mathcal{H}''$, i.e., $\kappa_1 = 0$ and $\kappa_2 \neq 0$. In this case, Q_1 and Q_2 are regarded as high-harmonic wave vectors of Q_5 and Q_6 . Figure 8

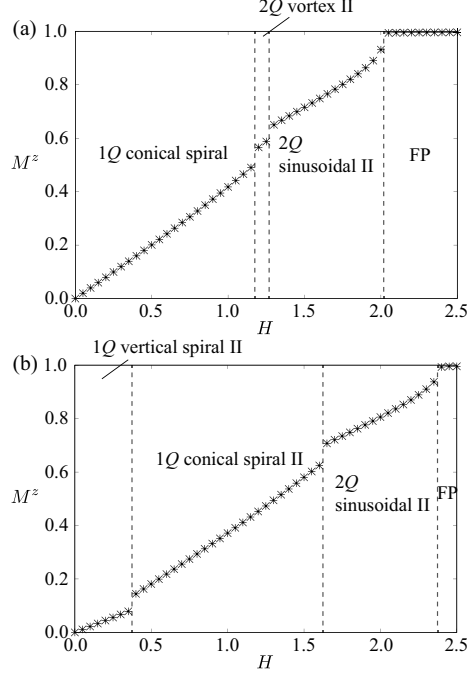


Figure 9: H dependence of M^z for $\mu = x, y, z$ at (a) $\kappa_1 = 1$ and (b) $\kappa_1 = 1.2$. The other parameters are set as $B = 0.2$ and $\kappa_1 = 0$. The dashed vertical lines represent the phase boundaries between different magnetic phases.

230 shows the phase diagram with changing of κ_2 and H at $B = 0.2$ and $\kappa_1 = 0$. For $\kappa_2 \lesssim 0.95$, there is no double- Q instability; only the single- Q conical spiral and single- Q sinusoidal states appear in the phase diagram.

When κ_2 reaches close to one, the double- Q sinusoidal II state appears in the high-field region instead of the single- Q sinusoidal state. Then, the double- Q vortex II state is stabilized in the intermediate-field region between the single- Q conical spiral and double- Q sinusoidal II states. While H is changed, the magnetization M^z shows a jump among the single- Q conical spiral, double- Q vortex II, and the double- Q sinusoidal II states, as shown in Fig. 9(a). The spin configurations of the double- Q vortex II and double- Q sinusoidal II states are qualitatively similar to those of the double- Q vortex and double- Q sinusoidal states in Fig. 4; fourfold rotational symmetry is broken while keeping spatial inversion symmetry. Meanwhile, the dominant peak positions of $\mathbf{m}_{\mathbf{q}}$ are relatively different. In the double- Q sinusoidal II state, the spin configuration consists of nonzero $m_{\mathbf{Q}_5}^y = m_{\mathbf{Q}_6}^y > m_{\mathbf{Q}_1}^z = m_{\mathbf{Q}_2}^z$ in Figs. 10(a) and 10(b). As shown in the real-space spin configuration in Fig. 11(a), the spin texture is similar to that in the double- Q sinusoidal state in Fig. 7(a). In the double- Q vortex II state, the spin configuration is characterized by the dominant contribution $m_{\mathbf{Q}_5}^x = m_{\mathbf{Q}_6}^x$ and the subdominant contributions $m_{\mathbf{Q}_1}^y = m_{\mathbf{Q}_2}^y$ and $m_{\mathbf{Q}_1}^z = m_{\mathbf{Q}_2}^z$.

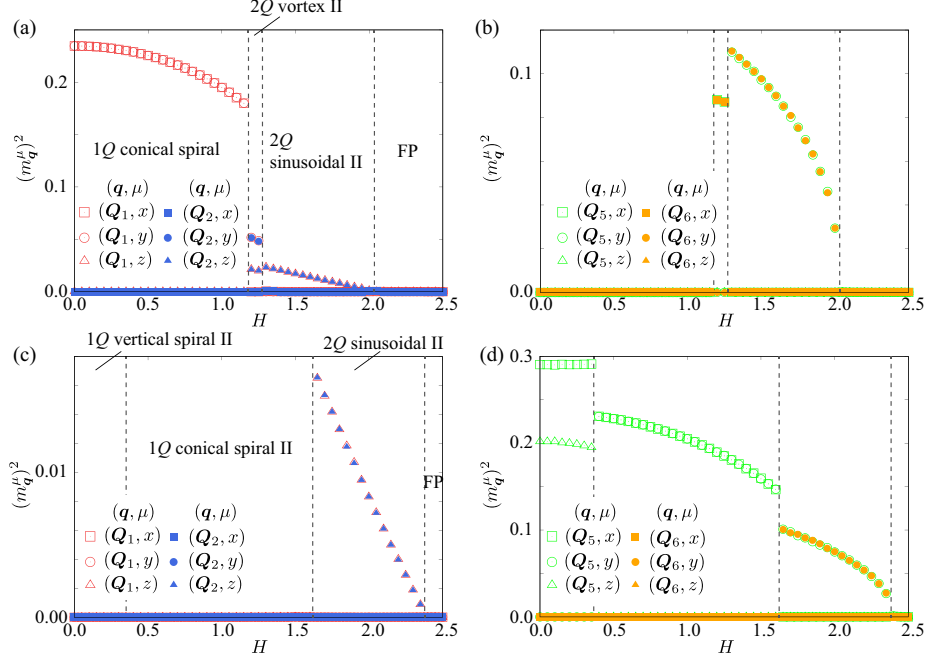


Figure 10: H dependence of (a,b) $(m_{Q_1, Q_2}^\mu)^2$ and (c,d) $(m_{Q_5, Q_6}^\mu)^2$ for $\mu = x, y, z$ at (a,b) $\kappa_1 = 1$ and (c,d) $\kappa_1 = 1.2$. The other parameters are set as $B = 0.2$ and $\kappa_1 = 0$. The dashed vertical lines represent the phase boundaries between different magnetic phases.

in Figs. 10(a) and 10(b), whose real-space spin configuration in Fig. 11(b) is also similar to that in the double- Q vortex state in Fig. 7(c). Thus, the emergence of the double- Q sinusoidal and vortex states tends to be realized when the interactions at high-harmonic wave vectors become important in the presence of fourfold inplane single-ion anisotropy.

When $\kappa_2 > 1$, the single- Q conical spiral state in the low-field region is replaced by the single- Q vertical spiral II and single- Q conical spiral II states, whose spin configurations are shown in Figs. 11(c) and 11(d), respectively. These states are distinguished by different spin configurations; they are characterized by nonzero m_{Q_5} or m_{Q_6} without m_{Q_1} and m_{Q_2} , as shown in Figs. 9(b), 10(c), and 10(d).

5. Summary

To summarize, we have investigated the multiple- Q instability in the tetragonal crystal system by focusing on the role of the fourth-order inplane single-ion anisotropy. Through the simulated annealing for the classical spin model on the two-dimensional square lattice, we showed that no double- Q instability occurs in the presence of the fourth-order inplane single-ion anisotropy, whose tendency

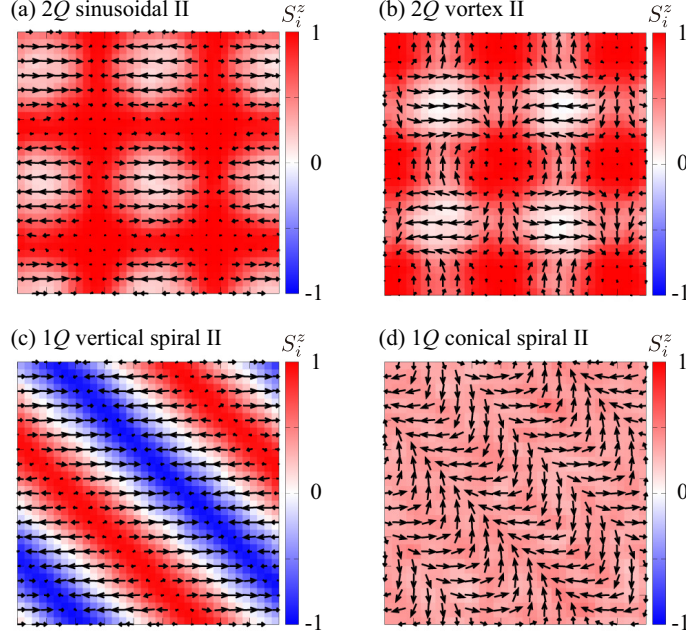


Figure 11: Real-space spin configurations obtained by simulated annealing in (a) the 2Q sinusoidal II state at $\kappa_2 = 1$ and $H = 1.5$, (b) the 2Q vortex II state at $\kappa_2 = 1$ and $H = 1.2$, (c) the 1Q vertical spiral II state at $\kappa_2 = 1.2$ and $H = 0.2$, and (d) the 1Q conical spiral II state at $\kappa_2 = 1.2$ and $H = 1$. The arrows represent the xy components of spins, (S_i^x, S_i^y) , and the color stands for the z component, S_i^z .

is in contrast to the case with the second-order easy-axis single-ion anisotropy. Then, we found that the contribution from the interactions at the high-harmonic wave vectors plays an important role in inducing the double- Q states in the external magnetic field. We obtained different types of the double- Q states, such as the double- Q sinusoidal state and the double- Q vortex state, depending on the model parameters. Our results indicate that the high-order single-ion anisotropy also gives rise to rich multiple- Q states, which will bring about further exotic spin textures. As the recent study suggested the importance of the interplay between magnetic anisotropy and the interactions at the high-harmonic wave vectors in EuAl_4 [60], chemical substitution and external pressure on this material might result in further multiple- Q instabilities as shown in the present result.

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