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DISSERTATION ABSTRACT

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学 位 論 文 題 名

Title of dissertation submitted for the degree

Non-Equilibrium Steady State Response and Fluctuations of Sheared Nematic Liquid Crystals

(せん断を印加したネマチック液晶の非平衡定常応答とゆらぎ)

Liquid crystals (LCs) are intermediate states that exhibit some of the behaviors of a conventional liquid and some of a solid crystal. They possess high fluidity like liquid and anisotropy in their optical, mechanical and electrical properties like solid at the same time. In LCs consisting of rod-like molecules, each molecule tends to point in one direction, resulting in the anisotropy. The molecular orientation and the flow are mutually coupled, and so the change of orientation can give rise to that of flow and vice versa. Utilizing the fact that the orientation can be easily controlled with external electric fields, LCs are applicable to some devices such as electrorheological fluids and micromotors. However, the dynamical properties have not yet been examined in detail, which is necessary to design devices. Therefore, it is worthwhile to investigate the fundamental rheological properties such as the shear stress response to a time-varying electric field under shear flow. On the other hand, the sheared LC is suitable for exploring the physics of non-equilibrium steady states (NESS) as it is taken far away from equilibrium by the application of shear flow, which violates the time-reversal symmetry. In non-equilibrium states, some characteristic features appear different from those in equilibrium states. For example, it is well known that the fluctuation-dissipation theorem, which is the central theorem in equilibrium statistical physics, breaks down.

The main purpose of this dissertation is to investigate the NESS response and fluctuations of sheared LCs both experimentally and theoretically. On the basis of the Ericksen-Leslie (EL) theory the dynamical properties were examined. The shear stress response to an ac electric field was measured in shear flow and dc electric fields, and the results were analyzed in terms of the EL theory.

In Chapter 1, the fundamental properties of liquid crystals were given. Continuum theories used in this dissertation were presented and the reorientation of molecules under shear flow was examined by using it. Earlier studies related to rheology and non-equilibrium were referred. The objectives and outline were given.

In Chapter 2, using the EL theory, phase diagrams in the three-dimensional space of magnetic field at a constant shear rate were obtained for two liquid crystals 5CB and 8CB which have different flow properties. An analytical solution was given for the tumbling state, in which the director rotates about an axis determined as a left eigenvector of the dynamical matrix that governs the motion of the director. It was shown that non-conservative forces caused by shear flow appear when magnetic fields are applied, so that the director points out of the shear plane. The non-conservative forces experimentally observed in the response function of 5CB were thought to originate from a similar

mechanism. Using the Langevin equation, the orientational fluctuations of the director were examined in detail by calculating the time correlation function and the response function, which are related to each other through a modified fluctuation dissipation relation. Critical behaviors such as the divergence of the relaxation time and fluctuations were found at the boundary between the stable and unstable states.

In Chapter 3, the NESS response of an LC to an ac electric field under constant dc electric field and steady shear flow was investigated. The first and second-order shear stress responses were theoretically obtained from the EL theory assuming a mono-domain model. It was found theoretically and experimentally that when a dc electric field is applied both responses remain constant and non-zero even at high frequencies of ac electric field. That is, there is a plateau, which originates from the time derivative of the director in the viscous equation. This plateau is a remarkable feature in the shear stress response brought about by the application of a dc electric field. By performing an optical measurement, the director response was confirmed to vanish at high frequencies, strongly supporting the above-mentioned mechanism for the appearance of the plateau. It was also clarified that the plateau in the second-order response disappears in the absence of a dc electric field, due to the Parodi relation, which is derived from the Onsager reciprocal relation.

In Chapter 4, the field-induced transition from the non-flow-aligning state (NFAS) to the flow-aligning state (FAS) was investigated by measuring the shear stress response to an ac electric field. Totally different frequency dispersions in the two states were observed for both the first- and second-order responses. The slowing down of relaxation time and the increased zero-frequency response were found near the transition point as well as in equilibrium phase transitions. The experimental results in the FAS were in good agreement with the calculated results based on the EL theory, including the plateau observed at high frequencies. As a result, the present measurement method can be expected to be useful for studying non-equilibrium phase transitions under shear flow. As for NFAS, it is necessary to elucidate the director field to calculate the stress response.

In Chapter 5, the general conclusions of the present thesis and future prospects were given.