THE JOURNAL OF PHYSICAL CHEMISTRY B

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Heat-Driven Rigid-Body Rotation of a Mixture of Cholesteric Liquid Crystal Droplets and Colloids

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Cite This: J. Phys. Chem. B 2020, 124, 6170–6174 Read Online

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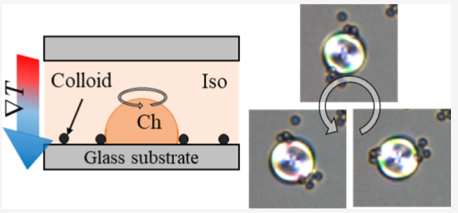
ABSTRACT: We show that cholesteric (Ch) liquid crystal droplets with cylindrically symmetric orientation dispersing in an isotropic (Iso) phase exhibited unidirectional rotation under a heat flux along the symmetry axis. By introducing colloidal particle adhesive to the Ch droplet surface, we traced the translational motion of the colloids and found that the colloids rotated unidirectionally around the center of each Ch droplet. The director configuration of the droplets was not distorted either spatially or temporally, while the colloids rotated constantly. The results suggest that the Ch droplets under the heat flux should rotate as a rigid body. Using this heat-driven rotation of the Ch droplets, we designed new geometries of various composites of Ch droplets and colloids and succeeded in driving intriguing complex dynamics.



I. INTRODUCTION

A chiral structure subjected to a uniform flux often shows nontrivial and nonequilibrium dynamics.[1−4] For example, in a cholesteric (Ch)−isotropic (Iso) coexisting phase, Ch droplets dispersing in an Iso phase rotate unidirectionally under a heat flux.[5−7] The rotation is considered to be driven by the cross-correlation between the heat flux and torque due to the mirror symmetry breaking.[8] Recently, the heat-driven rotation of Ch droplets has been studied not only for academic interests but also as a soft active matter[9−14] because they exhibit a variety of motions under a heat flux depending on the director configuration, which can be controlled by changing the droplet size, chirality, and boundary condition.

In a Ch droplet, the simplest director configuration is a single-twisted (ST) structure, in which the director is twisted with a constant pitch along one helical axis (h). When an ST droplet is applied with a heat flux perpendicular to h, the texture of the droplet rotates unidirectionally under polarized optical microscopy (POM). Two interpretations to explain the textural rotation of Ch droplets have been proposed. One is rigid-body rotation,[6,15] where the center of mass of each constituent liquid crystal (LC) molecule rotates around the droplet center, and the other is director rotation,[16,17] in which only the direction of the molecular long axis rotates, while the center of mass of each LC molecule is fixed. To clarify which interpretation is more appropriate for the heat-driven rotation of Ch droplets, in a previous paper, we introduced micron-size particles adhering to ST droplets and confirmed the rotation of the particles together with the helical texture of the droplets.[15] This result suggests the rigid-body rotation of ST Ch droplets. However, another interpretation was also proposed on the particle rotation; the anchoring between the particles and the droplets converts the director rotation to the transport of the particles,[17] which is based on the assumption that the anchoring energy should depend on the position of the droplets and should be minimal at a certain point. To confirm the propriety of the two interpretations, it should be revealed whether the particles select the position with a special director configuration. If the particles can adhere to the droplet surface at any position with various director orientations, then we should examine how the position of the particles on the droplets affects the rotational behavior.

To investigate the rotational mechanism of Ch droplets, we used droplets with a double-twisted (DT) structure. As shown in Figure 1, the DT droplet has cylindrical symmetry, where the director along the center axis of the droplets is aligned parallel to the symmetry axis and twists radially toward the side.[18] When the droplet diameter is smaller than the helical pitch, the DT structure is stable[19] and is not easily distorted. When a heat flux is applied to the DT droplet along the cylindrical axis, the torque caused by the thermomechanical coupling should be exerted on the director. However, due to the cylindrical symmetry, the director cannot rotate without distorting the DT structure, which prohibits the pure director rotation. Therefore, if the DT droplets rotate under a heat flux along the cylindrical axis without director distortion, the rotation must be rigid-body rotation. So far, we have not observed the rotational motion of an isolated DT droplet subjected to a heat flux along the symmetry axis.[20] This result suggests the possibility that when the symmetry axis of the DT

Received: May 10, 2020
Revised: June 18, 2020
Published: June 19, 2020

https://dx.doi.org/10.1021/acs.jpcb.0c04186
J. Phys. Chem. B 2020, 124, 6170–6174

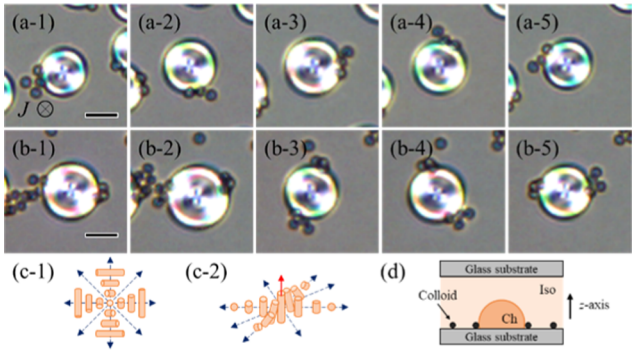


Figure 1. POM images of DT droplets, to the surface of which colloids adhere at (a) one point and (b) two points. The droplets were subjected to a downward heat flux (~1 mK/μm). The diameters of the DT droplets in (a) and (b) are 16.0 ± 1.4 and 17.8 ± 1.1 μm, respectively. The snapshots were taken every 30 min (a) and 20 min (b), respectively. The scale bars indicate 10 μm. See also Supporting Movies ESI 1a and ESI 1b. (c-1) Schematic top view and (c-2) perspective view of the DT structure. (d) Schematic side view of a DT droplet. We defined the z-axis as the direction parallel to the center axis of the DT droplet (perpendicular to the substrates).

structure corresponds with the rotational center axis, even if the DT droplet rotation is rigid-body, the rotation cannot be detected under POM.

In this paper, to visualize the rigid-body rotation of DT droplets, we added colloidal particles to the LC samples and traced their translational motion. In the Ch−Iso coexisting phase, since the Ch and the phases are composed of identical material, the interfacial energy is intrinsically low. In this study, we introduced colloids with high affinity for the Ch−Iso interface and showed that the Ch droplets rotate as a rigid body. If the heat flux drives the unidirectional vortex flow that can be converted into the translational motion of the colloids, the system may be applied to soft motors. We also investigated some novel dynamics of the composite system of the Ch droplets and the colloids.

II. METHODS

We prepared two kinds of LC samples for DT-LC and ST-LC. As DT (ST)-LC, we used the mixture of 0.4 wt % (1.1 wt %) (*S*)-4-{[1-(methylheptyl) oxy]carbonyl} phenyl-4(hexyloxy) benzoate (DIC), 13.4 wt % (37.3 wt %) 5CB, 85.9 wt % (60.8 wt %) No. 270032 (LCC), and 0.3 wt % (0.7 wt %) azobenzene polymer, poly(ethylene oxide-*b*-11-[4-(4-butylphenylazo) phenoxy]-undecyl methacrylate) (Polymer Source) (hereinafter PEO-PAzo). We added 4-μm-sized silica beads (SEKISUI CHEMICAL) to the Ch samples and obtained the mixture of Ch LCs and colloids.

The mixture was sandwiched by two 1.4-mm-thick glass plates, and the sample thickness was adjusted to be 30 μm by inserting bead spacers. The temperature of the top of the upper glass was set to be ~1 K higher than that of the bottom of the lower glass, which provided ~1 mK/μm temperature gradient to the LC sample in the perpendicular direction to the substrate. To observe the position of the colloids and the director configuration inside the droplets simultaneously, we conducted POM observation with a white light source, in which a polarizer and an analyzer were set to form a 45° angle with each other. In a previous work, we confirmed that the amphiphilic PEO-PAzo considerably decreased the friction between glass substrates and Ch droplets,[12,21] so that the Ch droplets in this experiment are expected to be lubricated.

III. RESULTS AND DISCUSSION

When the DT-LC sample was cooled from the Iso phase temperature to the Ch−Iso coexisting temperature (~82.8 °C), a number of Ch droplets randomly appeared in the Iso phase. When the diameter of the Ch droplets is smaller than the helical pitch of DT-LC (~25 μm, the chiral dopant concentration being 0.4 wt %), the director configuration in the Ch droplets should be the DT structure. Under POM with the polarizer and the analyzer forming an angle of 45° with each other, we observed the typical DT texture, as shown in Figure 1a,b. Here, the DT droplets have the cylindrical symmetry axis perpendicular to the glass substrates through the center of the droplets (Figure 1c). One can notice that some colloids are sticking to the Ch−Iso interface, while the director configuration of the DT structure is unchanged, even in the very vicinity of the colloids within the experimental resolution of the POM. Due to gravity, the silica colloids tend to be located near the bottom of the cell and thereby adhere to the hemispherical droplet along the circumference of the base, as schematically shown in Figure 1d. In this geometry, the director should be planar to the Ch−Iso interface at any point for the colloids to touch.

When we applied a heat flux to the mixture of the DT droplets and the colloids, the colloids rotated in the counterclockwise (CCW) direction around the center of the droplets, as shown in Figure 1a and Supporting Movie ESI 1a. The rotational direction of the colloids agreed with that of previously reported ST Ch droplets with the same chirality.[6] Under the temperature gradient of $\nabla T \sim 1$ mK/μm, the angular velocity of the colloids was constant as $\sim 8 \times 10^{-4}$ rad/s within our observation time (150 min). The rotation of the colloids around the droplet center axis indicates the in-plane rotational flow along the interface of Ch droplets. On the other hand, in the z-direction parallel to the center axis (Figure 1d), the colloids never showed any translational motion, indicating that there is no flow parallel to the heat flux. In contrast to the in-plane rotation of the colloids, the texture of the DT droplets exhibited no change. This is reasonable because if the DT droplet rotates as a rigid body without distorting the director, no change should be observed in the POM images due to the cylindrical symmetry, and only the rotation of the colloids attached to the droplet can be noticed. Next, we applied the heat flux to the DT droplet, to which the two aggregates of the colloids adhered at the two different points, as shown in Figure 1b and Supporting Movie ESI 1b. Both the aggregates of the colloids rotated in the CCW direction at the same speed, the snapshots of which are given in Figure 1b. It should be noted that the relative position of the aggregates of the colloids was maintained within the observation time. The rotation of the colloids reveals that the DT droplets with cylindrical symmetry exhibit unidirectional rigid-body rotation under a heat flux along the cylindrical axis.

We next investigated the rotational behavior of ST droplets without cylindrical symmetry. Figure 2a−c shows the POM images of the ST droplets with the colloids. The striped texture corresponds to the ST structure, in which the helical axis of the director is parallel to the glass substrate and perpendicular to the stripe. The stripe width corresponds to half the helical pitch of ST-LC (~10 μm, the chiral dopant concentration being 1.1 wt %). In contrast to the DT droplets, since the

https://dx.doi.org/10.1021/acs.jpcb.0c04186
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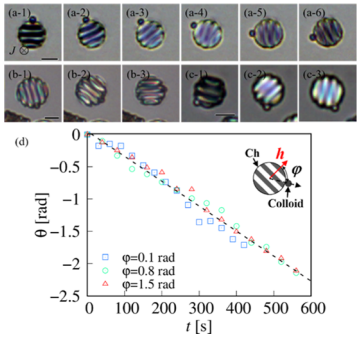

Figure 2. (a) POM images of the ST droplets dispersing in the Iso phase attached by the colloid positioning at φ = 0.1 rad. The diameter of the droplet is 18.1 ± 0.3 μm. The snapshots were taken every 90 s. (b) POM images of the ST droplets attached by the colloid at φ =1.5 rad (the diameter is 22.2 ± 1.5 μm) and (c) at φ =0.8 rad (the diameter is 16.4 ± 0.2 μm). The samples are subjected to a downward heat flux (~1 mK/μm). The snapshots were taken every 180 s. The scale bars indicate 10 μm. See also Supporting Movies ESI 2a, ESI 2b, and ESI 2c. (d) The time evolution of the rotational angle θ of the colloids. The inset shows the schematics of an ST droplet. We defined the angle between the helical axis h and the vector from the center of the droplets to a colloid as φ.

cylindrical symmetry is broken in the ST droplets, the director at the Ch−Iso interface is not always planar but inhomogeneous, depending on the position. As is clearly shown in Figure 2, the colloids, being located at the bottom due to gravity, adhere to the Ch−Iso interface independent of the angle φ, defined as the angle between the helical axis h and the vector from the center of the droplets to each colloid (see the inset of Figure 2d). This suggests that the anchoring of the colloids is so weak that they do not adhere to the position with specific director configuration (planar or homeotropic). Figure 2a−c shows the ST droplets with φ = 0.1, 1.5, and 0.8 rad, respectively. As with the DT droplets, no director distortion of the ST droplets was observed in the vicinity of the colloids, which also proves the weak anchoring of the colloids.

When we applied a heat flux (~1 mK/μm) to the sample, both the texture of the ST droplets and the colloids sticking to the Ch−Iso interface rotated unidirectionally in the CCW direction, as shown in Figure 2a−c and Supporting Movies ESI 2a, ESI 2b, and ESI 2c. In the same way as the DT droplets, no translational motion of the colloids was observed in the z-direction. This result indicates that in-plane rotational flow about the droplet center axis should exist along the Ch−Iso interface. Within our observation time (~600 s), φ was constant and the angular velocity of the textural rotation of the ST droplets was the same as that of the rotation of the colloids.

We checked the relationship between the rotational velocity of the colloids and the director orientation in the vicinity of the colloids. Figure 2d shows the time evolution of the rotation angle θ of the colloids with φ = 0.1, 0.8, and 1.5 rad, in which the clockwise direction is taken to be positive. As shown in the figure, θ decreased linearly with time, indicating that the angular velocity of the colloids, ω, was constant. More importantly, ω resulted in approximately the same value of ~4 mrad/s independent of φ, yet with a slight variation due to the difference of the droplets size and thermal fluctuations. The result shows that the director configuration in the vicinity of the colloid does not affect the rotational behavior of the colloids.

Using the rigid-body rotation of the Ch droplets and the adhering colloids, it is possible to drive different dynamics in another geometry. We first fabricated the composite of the ST droplets and the colloids, as shown in Figure 3a. Here, the two

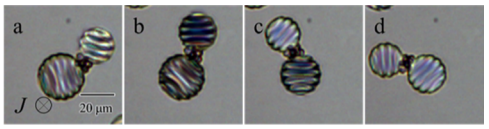

Figure 3. Heat-driven dynamics of the composite of ST droplets and colloids. The sample was subjected to a downward heat flux (~1 mK/μm). The snapshots were taken every 5 min. See also Supporting Movie ESI 3.

ST droplets are connected by the aggregate of the colloids, resulting in the formation of the dumbbell-like shape. When we applied a downward heat flux of ~1 mK/μm to the sample, the dumbbell-like composite rotated in the CCW direction (Supporting Movie ESI 3), as shown in Figure 3a−d. The rotational direction agreed with that of the heat-driven rotations of the ST and DT droplets.[12,13] The heat-driven torque on the two ST droplets should be transferred to each other via the colloids, which results in the complex dynamics of the dumbbell.

We next examined the dynamics of the DT droplet when the adhering colloid was confined to the substrate. We fabricated a DT droplet whose cylindrical symmetry axis was parallel to the glass substrate (called "rugby ball-type" in our previous work[19,20]) and attached colloids to the Ch−Iso interface. When one of the colloids was confined on the glass substrate, under a heat flux, not the colloid but the DT droplet rotated around the center of the colloid in the CCW direction, as shown in Figure 4 and Supporting Movie ESI 4. The DT droplet rotated at a constant speed of ~0.17 rad/min, which lasted for more than 90 min, within our observation time.

As mentioned above, all the experimental results consistently suggest that both the DT and the ST droplets should rotate as a rigid body under a heat flux. From the fact that the colloids can adhere to the surface of the ST droplets at any point with various director configurations and that the director orientation is not influenced by the attached colloids, the anchoring force between the colloids and the director should be weak. In such a system, the weak anchoring can hardly transfer the pure director rotation to the rotation of the colloids at the same speed as the texture.

The observed rigid-body rotation of the DT droplets and the colloids can be explained in the framework of Leslie's theory. According to the simple phenomenological equation given by Leslie, the thermomechanical torque exerted on a chiral LC domain is given as[8]

$$\tau_{\mathrm{Leh}} = \int_b n \times (n \times \nabla T)\mathrm{d}V \tag{1}$$

In our experimental geometry, the local orientation of the director in the DT droplet is denoted as

$$n = (\sin\Theta\cos\Phi, \sin\Theta\sin\Phi, \cos\Theta) \tag{2a}$$

$$\Theta = 2\pi\frac{\sqrt{x^2 + y^2}}{P} \tag{2b}$$

$$\Phi = \tan^{-1}(y/x) + \pi/2 \tag{2c}$$

https://dx.doi.org/10.1021/acs.jpcb.0c04186
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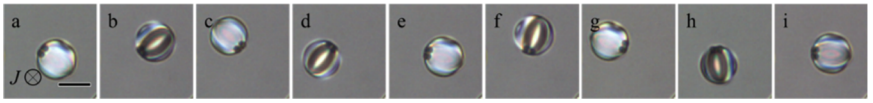

Figure 4. Heat-driven dynamics of the composite of a DT droplet and colloids. The sample was subjected to a downward heat flux (~1 mK/μm). The snapshots were taken every 10 min. The scale bar indicates 20 μm. See also Supporting Movie ESI 4.

where the x- and y-axes are in the plane parallel to the glass substrate, and the z-axis is parallel to the cylindrical symmetry axis.[22] Substituting the director field given by eqs 2 into eq 1, we have $\tau \neq 0$, which can be used to drive the unidirectional rotation of the DT droplets. Although the rigid-body rotation, as well as the director rotation, of an isolated DT droplet without any marker has not been observed because of the cylindrical symmetry, in our previous paper, we reported that the aggregates of the DT cylinders rotate as a rigid body under a heat flux along the symmetry axis,[10,18,19] and from this result, it was predicted that the isolated DT droplets would be able to rotate under a heat flux. Here we gave the evidence for the rigid-body rotation of the DT droplets by showing that the colloids sticking to the DT droplet rotated under a heat flux without distorting the director field.

IV. CONCLUSIONS

We investigated the heat-driven dynamics of a composite of Ch droplets and colloids. By direct microscopy observation, we showed that (1) the colloids adhering to the surface of the DT droplets with the cylindrically symmetric orientation rotated unidirectionally around the droplets' center under a heat flux, while the DT texture was unchanged, (2) the colloids adhered to the surface of the ST droplets at any position independent of the director configuration, and (3) the colloids adhering to the ST droplets rotated at a constant speed independent of the director configuration in the vicinity of the colloids. These results consistently suggest that both ST and DT droplets under heat flux can rotate as a rigid body. In addition, we combined Ch droplets and colloids with various geometries and succeeded in manipulating the translational motion of the center of mass of the droplets and the colloids. The extended variation of the heat-driven rotation of the chiral LCs may be applicable for a heat-driven soft device.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jpcb.0c04186.

Movie corresponding to Figure 1a. A DT Ch droplet attached by one aggregate of colloids, subjected to a heat flux. The recording was performed with 360 times faster speed (Movie ESI 1a). Movie corresponding to Figure 1b. A DT Ch droplet attached by two aggregates of colloids, subjected to a heat flux. The recording was performed with 360 times faster speed (Movie ESI 1b). Movie corresponding to Figure 2a. An ST droplet attached by one colloidal particle at the position of φ = 0.1 rad, subjected to a heat flux. The recording was performed with 60 times faster speed (Movie ESI 2a). Movie corresponding to Figure 2b. An ST droplet attached by one colloidal particle at the position of φ = 1.5 rad, subjected to a heat flux. The recording was performed with 240 times faster speed (Movie ESI 2b). Movie corresponding to Figure 2c. An ST droplet attached by one colloidal particle at the position of φ = 0.8 rad, subjected to a heat flux. The recording was performed with 240 times faster speed (Movie ESI 2c). Movie corresponding to Figure 3. A dumbbell-like composite of two ST droplets connected by aggregating colloids, subjected to a heat flux. The recording was performed with 90 times faster speed (Movie ESI 3). Movie corresponding to Figure 4. A rotating ST droplet partially confined by colloids under a heat flux. The recording was performed with 360 times faster speed (Movie ESI 4) (ZIP).

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Author Contributions

The results and the research plan were discussed by all authors. S.B. and Y.T. wrote the manuscript and performed experiments.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The authors are thankful to Dr. Isa Nishiyama and Dr. Hiroshi Hasebe of DIC Corp. for supplying the LC compounds and the chiral dopants. This work is part of the outcome of research performed under a Waseda University Grant for Special Research Projects (2019C-553).

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