Liquid-crystalline chiral phase formation and structural transitions of dumbbell-shaped colloids

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Abstract

Colloidal liquid crystals (LCs) are an emerging class of soft materials that naturally combine the unique properties of both LC molecules and colloidal particles. Chiral LC blue phases are highly attractive in fast optical displays and electrooptical devices, but the formation of chiral structures from achiral colloidal particles is rare. Herein we demonstrate that achiral dumbbell-shaped colloids can assemble into a rich variety of characteristic LC phases, including nematic phases with lock structures, smectic phases, and particularly double-twisted chiral columnar phases. Phase diagrams from experiments and simulations show that the existence and stable regions of different LC phases are strongly dependent on the geometry of colloids. Furthermore, the LC phases can be dynamically tuned by external magnetic fields. This work paves a new route to the development of stimuli-responsive functional LC mesostructures for LC display and laser applications.
Introduction

Liquid crystals (LCs) are a state of matter that bears liquid-like fluidity and crystal-like ordering, which are ubiquitous in the natural world and our everyday life. Chiral LCs possess helical structures arising from the spontaneous twist of LC mesogens. They are intriguing for applications in areas, such as display devices and photonic materials. Interestingly enough, chiral LCs can be formed from the chiral organization of achiral objects (e.g., molecules and colloids) that do not have a chiral center. This phenomenon is of paramount importance from both theoretical and practical perspectives because the synthesis of chiral objects is often difficult and costly. The formation of chiral LCs from achiral building blocks has been predicted by numerous computational researches, however, experimental demonstrations have been rare. Experimental examples of chiral LCs from achiral mesogens have been limited to cholesteric and chiral smectic phases formed by banana-shaped molecules and their colloidal analogues (i.e., bent-core colloids), chiral smectic phase from odd-membered mesogenic dimers, and twist grain boundary phase assembled from graphene. Compared with molecular LCs, colloid-based LC materials are thermally stable, inexpensive, and possess enhanced susceptibility to external fields, including shear fields, electric fields, and magnetic fields, which makes them attractive for a wide range of applications such as shearing microlithography, photonics, and LC displays. Moreover, because of their unique dimensions (from submicrometer to micrometer scale), colloidal particles are ideal model systems for studying LC phase behavior of their molecular counterparts which are difficult to be directly observed and quantitatively studied. Therefore, there is an urgent need to develop new levels of understanding of the chiral formation from achiral colloidal mesogens with novel shapes.

Dumbbell-shaped colloids (DBCs) are featured with distinctive concave geometry and unique packing behavior, making them attractive as building blocks for functional optical materials. Nanosized dumbbells with charge patches can pack into zippers, cross-stacks, and open-lattice crystals. Dumbbell-like nano-arrows can assemble into net-like, zipper-like, and weave-like two-dimensional lattices, as well as non-close-packed three-dimensional supercrystals, depending on the aspect ratio of the colloids. Moreover, dumbbell-shaped molecules exhibit geometry dependent assembly behavior to produce helical structures, suggesting the concave geometry can twist the packing of dumbbell-shaped building blocks. Nevertheless, to date, the manner in which the concave geometry of DBCs determines their LC phase behavior has not been explored experimentally, largely due to current challenges in the large-scale synthesis of these colloids with precisely controlled geometries.
This work presents a systematic study of the LC phase behavior of DBCs, which resemble dumbbell-shaped LC mesogens, with precisely tunable geometry$^{32,33}$. We observe that these DBCs assemble into a rich variety of characteristic LC phases, including double-twisted chiral columnar phase (N*) with core geometry similar to that of blue phases$^{8,9}$, nematic phases with one-lock (N1) and two-lock (N2) structures, and smectic A (SmA) phase (Fig. 1). In particular, the chirality formation in N* phases from achiral building blocks is of significant interest and has been rarely reported in colloidal systems$^{3,5,34}$. The equilibrium phases are found to be dictated by the two critical geometric parameters of DBCs, namely, the ratio of diameters ($R_D$) of end blocks ($D_e$) to central blocks ($D_c$), and the ratio of lengths ($R_L$) of end blocks ($L_e$) to central blocks ($L_c$). Brownian dynamic simulations confirm the geometry dependent formation of mesophases and show good agreement with experimental results. The experimental and computational results are summarized in phase diagrams of stable phases and phase transitions in the $R_D$ and $R_L$ space. Moreover, we also demonstrate that the colloidal LC phases can be dynamically manipulated using magnetic fields. This study provides new insights into the phase behavior of dumbbell-shaped objects, and the fabrication of stimuli-responsive functional colloidal materials with desired electronic and photonic properties for LC display and LC laser applications.

**Results**

**LC phases assembled from DBCs with various geometries.** DBCs with controlled $R_D$ (1.06–1.77), $R_L$ (0.08–1.81), and $L/D_e$ (1.55–9.93, where $L$ is the overall length of the DBC, i.e., $L=2L_e+L_c$) were synthesized via a wet-chemical method$^{32}$. The DBCs were dispersed in dimethyl sulfoxide (DMSO) in capillary tubes and left to slowly settle to the bottom (Supplementary Fig. 1)$^{13,14}$. The LC phases were formed after ~30 days of sedimentation with a volume fraction of ~50%. Notably, similar to molecular LCs, the colloidal LC phases exhibited fluidic behavior and could flow upon tilting the capillary tube (Supplementary Fig. 2).Depending on $R_D$ and $R_L$, the DBCs assembled into a variety of entropy driven LC mesophases, including N*, N2, N1, and SmA phases. The long-range ordering and detailed organization of individual DBCs within all the formed phases were characterized by polarized optical microscopy (POM) and scanning electron microscopy (SEM), respectively.

For DBCs with two very short e-blocks (e.g., $R_L = 0.10$), the colloids organized into the N* phase with a racemic mixture of double-twisted chiral columns, although the constituent DBCs are achiral (Fig. 2a–c). Within each double-twisted chiral column, the DBCs were rotated in a helical fashion along the long-axis ($t_1$) of each column and also formed a barrel-like chiral twist with respect to its layer normal ($t_2$) (Fig. 1). Statistically, the population of
right- and left-handed chiral columns is approximately equal at a 100 μm length scale. Thus, the N* phase shows locally defined handedness and helical pitch (P) that varies slightly from domain to domain. For example, the helical structure in Fig. 2b displays a left-handed column and a right-handed column with a half P (1/2P) of ~5 μm. The helical twist can be interrupted when columns with different handedness merge. Within the barrel twist, the central DBC does not tilt with \( t_2 \), and the tilt angle of DBCs increases from the inner to the outer regime (Fig. 1). The formation of the N* phase was further confirmed by the characteristic texture in the POM image of periodic lines with a spacing of ~5 μm, corresponding to 1/2P (Fig. 2c)\(^1.5\).

The dumbbell shape endows DBCs with interlocking ability. DBCs with two short e-blocks (e.g., \( R_L = 0.56 \)) assembled into an N2 phase (Fig. 2d–f). In the nematic phase, the degree of orientational order can be described by the order parameter \( S = \frac{1}{2}(3 \cos^2 \theta - 1) \), where \( \theta \) is the angle between the director and the long axis of each DBC, and the brackets denote an average over all the measured DBCs\(^3.5\). \( S \) varies from 0 (isotropic) to 1 (crystal). As shown in SEM images (Fig. 2d,e), the DBCs show a high degree of long-range orientational order with \( S \approx 0.7 \). Moreover, neighboring DBCs are packed in a slipped mode to form two-lock structures; that is, the two thick e-blocks of two end-to-end adjacent DBCs are embedded within the groove (i.e., thin c-block) of a third DBC. The localized POM image (Fig. 2f) shows the typical schlieren texture of the N2 LC phase. DBCs with two intermediate e-blocks (e.g., \( R_L = 0.99 \)) organized into the N1 phase (Fig. 2g–i). The detailed packing of DBCs is shown in Fig. 2g,h. The concavity of one DBC does not have sufficient room to accommodate two e-blocks of neighboring DBCs to form the two-lock structure. Instead, the groove of each DBC fits one e-block of an adjacent DBC to form the one-lock structure with \( S \approx 0.8 \). The N1 phase also exhibits a schlieren texture (Fig. 2i). DBCs with two long e-blocks (e.g., \( R_L = 1.29 \)) exhibited a SmA phase with the average rod alignment perpendicular to the smectic layer (Fig. 2j–l). The individual DBCs are located within the smectic layer (as shown in Fig. 2j,k). The formation of the SmA phase was also confirmed by the characteristic striped texture in the POM image (Fig. 2l).

**Phase diagram and geometric effects based on experimental observations.** The assembled phases of DBCs were mapped out in a phase diagram in the space of \( R_D \) and \( R_L \) (Fig. 3a). The achiral DBCs with two very short e-blocks (\( R_D > 1.1 \) and \( R_L < 0.45 \)) underwent spontaneous chiral symmetry breaking, leading to the N* phase with chiral superstructures (Supplementary Fig. 3). Due to the DBCs’ strong tendency to align with each other and the adjacent DBCs not being confined to a planar surface, the steric hindrance between adjacent DBCs induces twists,
which eventually generates chiral assembled structures\textsuperscript{36}. DBCs can twist in two equivalent ways (Fig. 1), thus leading to the formation of racemic mixtures with equal probability of right- and left-handed chiral domains. It is found that $P$ is strongly dependent on the value of $L_c/D_e$ (Fig. 3b). $P$ increases from 7 to 17 μm with $L_c/D_e$ increasing from 3.0 to 8.5. To the best of our knowledge, this is the first experimental demonstration of the formation of a double-twisted LC phase from achiral DBCs. This phenomenon is in agreement with previous simulation studies on polymer-tethered nanorods\textsuperscript{7}, bent-core, and linear rigid particles\textsuperscript{8,9}.

As $R_l$ increases, DBCs with two short e-blocks ($1.3 < R_D < 1.6$ and $R_l \sim 0.50$) form the N2 phase, as DBCs with two-lock arrangements can pack more efficiently than the simple nematic phase (Supplementary Fig. 4). With the further increase of $R_l$, $L_e$ becomes comparable to $L_c$. The shape complementarity in the morphology leads to the N1 phase with zipper structures (Supplementary Fig. 5)\textsuperscript{26} for DBCs with two intermediate e-blocks ($1.1 < R_D < 1.6$ and $R_l \sim 1.00$). Notably, the dashed lines at $R_l$ of 0.50 and 1.00 locate around the centers of N2 and N1 phases, respectively. For DBCs with long e-blocks ($R_l > 1.35$) or DBCs with small diameter differences ($R_D < 1.1$), SmA phases are formed due to the mismatch between $L_e$ and $L_c$ of DBCs and the nearly rod-like shape of DBCs (Supplementary Fig. 6)\textsuperscript{37}. For DBCs with large diameter difference ($R_D > 1.6$), isotropic phases (i.e., without orientational and positional order) were found (Supplementary Fig. 7). The formation of the isotropic phase can be attributed to the large energy barriers for the assembly of DBCs. In summary, these results indicate that subtle variation in the geometry of DBCs has a significant impact on their phase behavior.

**Simulations based on a coarse-grained Brownian dynamics model.** We simulated the LC phase formation of DBCs with different geometric parameters using a coarse-grained Brownian dynamics model (see Methods for simulation details)\textsuperscript{38}. Briefly, the DBCs were constructed by freezing linearly arranged rigid spherical beads into a dumbbell rod geometry in accordance with the size and shape of real DBCs. The e-blocks with a large diameter $D_e$ were located at both ends of the central c-block with a small diameter $D_c$. The interactions between DBCs were set to be purely repulsive Weeks-Chandler-Andersen (WCA) potential\textsuperscript{39}. All simulations were carried out in a cubic cell with periodic boundary conditions. To simulate the phase condition with high densities, the length of box size was iteratively increased and decreased upon changing the temperature.

The formation of all the phases observed in the experiments was confirmed by the simulations. Figure 4 shows four characteristic snapshots of the assembled DBCs with different values for $R_D$ and $R_l$. The simulation results indicate that the assembled phases of DBCs are strongly dependent on $R_l$, which is consistent with the
experimental results. When $R_D$ was fixed as 1.40, the DBCs with $R_L = 0.15$ accommodated in an $N^\ast$ phase. Figure 4a,e show the configuration of a single column extracted from the simulation box. The snapshots clearly show that, rather than packing parallel to each other, the DBCs form a regular chiral twist both within and about the column. In this case, the e-blocks prevent regular close packing of the DBCs, which would otherwise lead to the formation of the smectic phase. As a result, the neighboring DBCs are closely packed with a tilted angle to form the twisted structure. It should be noted that these assembled structures exhibit a discontinuity in the twist of the column because of the convergence of columns with different handedness. DBCs with $R_L = 0.50$ form an $N_2$ phase (Fig. 4b). Typically, two e-blocks of two end-to-end adjacent DBCs are trapped in one concave groove of an adjacent DBC. In the local structure of the $N_2$ phase, a total of six DBCs (three green e-blocks above and three blue e-blocks below) packed around the central red c-block. In other words, two layers of e-blocks from surrounding rods are trapped by the central c-block along its long axis. In contrast, DBCs with $R_L = 1.00$ exhibit an $N_1$ phase (Fig. 4c). One e-block of each DBC is preferentially embedded in the concave groove of an adjacent DBC. This configuration allows DBCs to pack closely with neighboring ones, resulting in a zipper structure. As $R_L$ increases, DBCs with $R_L = 1.80$ become stable in the SmA phase (Fig. 4d). In this case, since the e-block is longer than the c-block, the e-block cannot be fit into the concave groove of a DBC. Therefore, the effect of the diameter difference of blocks becomes negligible, making them behave like conventional colloidal rods. Finally, it was found that only DBCs with $1.1 \leq R_D < 1.6$ could exhibit assembly behavior that are distinct from rod-like colloids. DBCs with even larger $R_D$ ($\geq 1.6$) tend to form an isotropic phase (i.e., without positional and orientational order) due to the high energy required for the assembly of DBCs (Supplementary Fig. 8).

**Phase diagram and geometric effects based on simulations.** We evaluated the formation and transitions of mesophases by systematically varying $R_D$ and $R_L$ in simulations. The results are summarized in the phase diagram shown in Fig. 5. The phase regions are in good agreement with the experimental phase diagram. The formation of mesophases is determined by both $R_D$ and $R_L$. The $N^\ast$ phase is stable only when $R_L$ is small, i.e., the DBCs possess two very short e-blocks (Supplementary Fig. 9). Chirality of the structure arises from the twist arrangement of achiral DBCs. When the e-block becomes longer the formation of the $N^\ast$ phase is suppressed, while the formation of $N_2$ and $N_1$ phases is strongly promoted. The $N_2$ phase is formed when $R_L$ is around 0.5 (i.e., the length of c-block is approximately twice of that of e-block). In this case, the concave groove of a DBC can accommodate two e-blocks in the long axis direction, which is driven by excluded volume interactions (Supplementary Fig. 10). The $N_1$ phase
was observed with further increasing $R_L$ to about 1.0. The comparable length of c-block to e-block enables the trapping of one e-block in each concave groove of DBCs in the long axis direction. When the e-block is longer than the c-block, the DBCs form the SmA phase.

**LC Phase manipulation with magnetic fields.** Thanks to the positive diamagnetic anisotropy of DBCs, the colloidal LCs of DBCs are responsive to external magnetic fields, and the LC phases can be dynamically tuned by controlling the DBC orientation. Upon applying a magnetic field of 5 T, DBCs were rotated and aligned their long axis along the field direction, resulting in their visual texture changing from N* to regular nematic phase as shown in the POM images (Supplementary Fig. 11). The strip pattern of nematic phase is well-aligned to the magnetic field. In addition, fewer defects were observed after the application of the magnetic field, indicating the efficient alignment of DBCs by the magnetic field. This result suggests the potential of such DBC-based colloidal phases for application in stimuli-responsive and magnetically switchable systems.

**Discussion**

In conclusion, we have systematically studied the phase behavior of the colloidal analogues of dumbbell-shaped molecules, so-called dumbbell-shaped colloids (DBCs). We demonstrated, using both experimental and simulation data, the critical role of colloidal geometry in the formation of mesoscopically ordered phases. Our experimental results showed that DBCs with $R_D$ ranging from 1.0 to 1.8 and $R_L$ ranging from 0.1 to 1.8 assembled into a variety of LC phases including N*, N2, N1, and SmA phases. The phases and onset of phase transitions were found to be strongly dependent on the geometric parameters (i.e., $R_D$ and $R_L$) of the DBCs. It is worth noting that double-twisted chiral superstructures with racemic mixtures can be produced from rigid achiral colloids. Brownian dynamics simulation was used to further establish the correlation between the geometry and phase formation of DBCs. The simulation results are in good agreement with the experimental data, providing a theoretical basis for the future design of complex mesogenic systems. In addition, we show that the colloidal LC phases can be dynamically tuned by controlling the orientation of DBCs with magnetic fields. This work provides new insight into the rules required for the design of particles of different shapes to achieve superstructures with increased complexity and desired stimuli-responsive electronic and photonic properties.
Methods

Synthesis of DBCs. DBCs were synthesized by a recently developed emulsion-templated wet-chemical approach. In a typical experiment, 500 mg polyvinylpyrrolidone ($M_w = 40,000$, Sigma-Aldrich) was dissolved in 5.0 ml 1-pentanol (99%, Sigma-Aldrich) in a 10 ml glass vial under sonication. 140 μl deionized water (18.2 MΩ), 50 μl 0.18 M sodium citrate dihydrate (99%, Sigma-Aldrich) aqueous solution, 475 μl anhydrous ethanol (Pharmco-Aaper), and 100 μl ammonium hydroxide solution (28 wt%, Sigma-Aldrich) were added to the glass vial and vortexed for 1 min. After the mixture was left for 5 min to release gas bubbles, 50 μl tetraethylorthosilicate (TEOS, 98%, Sigma-Aldrich) was added and the solution was then gently shaken for 30 s. The $L_e$ (160−900 nm) and $L_c$ (170−3360 nm) of DBCs can be individually controlled by tuning the reaction time during the growth of each block. The $D_e$ of DBCs can be tuned from 190 to 460 nm by adjusting the amount of ammonium hydroxide. The $D_c$ of DBCs can be adjusted in the range of 140−340 nm by varying the strength of perturbation in the reaction temperature (25−70 °C) during the DBC growth. A higher temperature during the perturbation period produces a smaller $D_c$. For example, for DBCs with $L_e = 160$ nm, $L_c = 1660$ nm, $D_e = 315$ nm, and $D_c = 240$ nm, the reaction mixture was incubated at 25 °C for 30 min, 60 °C for 20 min, and 25 °C for another 4 h.

Purification. The as-synthesized DBCs were purified by centrifugation at 6000 rpm for 10 min. The precipitated DBCs were then washed three times with ethanol under centrifugation at 3000 rpm. Finally, the DBCs were suspended in ethanol and centrifuged at 1000 rpm for 10 min to remove small particles and other lightweight impurities.

Assembly of DBCs into LCs. The DBC dispersions in DMSO (99.9%, Fisher Chemical) with a volume fraction of 20% were filled into one-end-sealed glass capillaries (Stuart) with an outside diameter of 1.9 mm, an inside diameter of 1.3 mm, and a length of 100 mm, and rectangular capillaries (VitroCom) with 2.0 mm in width, 0.2 mm in inner-thickness, 0.14 mm in wall thickness, and 50 mm in length. The other ends of the capillaries were then sealed by melting. Subsequently, the capillaries were left in a vertical position for the sedimentation of DBCs. After one month, the capillaries were opened, and the sediment was taken out and slowly dried in air at room temperature for SEM characterizations. The samples in rectangular capillaries were directly used for POM characterization.

Phase switching with magnetic fields. The switching of colloidal LC phases was conducted under a magnetic field generated by 9 T Room Temperature Bore Magnet System (Cryomagnetics.Inc). The samples in rectangular
capillaries upon sedimentation were allowed to sit undisturbed for 1 hour under a magnetic field of 5 T and then examined using POM.

**Characterizations.** The SEM images of DBCs and their assemblies were taken using a Hitachi SU-70 Schottky field emission gun SEM at an operation voltage of 10 kV. POM images were obtained by an Olympus BX50F4 Microscope with crossed polarizing filters.

**Computer simulations.** Computer simulations on the formation mechanism of different structures of DBCs were approached as follow: the e-block and c-block with the diameters of $D_e$ and $D_c$ were constructed by freezing linearly arranged spherical beads, respectively. Two e-blocks with a diameter of $D_e$ were located at both ends of a DBC. Each DBC was treated as a rigid body in the simulation. Non-bonding interactions between DBCs were described by the WCA potential. The WCA potential is the 12-6 Lennard-Jones potential truncated at the energy minimum and shifted vertically by $\varepsilon/k_B T$; it is a purely repulsive potential featuring the volume occupied by DBCs. The Brownian dynamics of rigid bodies in the canonical ensemble was adopted for the simulations. The details of the simulation method can be found in Ref. 38. The length and the energy units of the simulation system are $\sigma$ and $\varepsilon$, respectively. The time step is in a unit of $\sigma\sqrt{m/\varepsilon}$, while the unit of temperature is $\varepsilon/k_B$. A leapfrog algorithm of rigid bodies with a time step of $\Delta t = 0.01$ was adopted to integrate the equations of motion

In the simulations, DBCs in the initial configuration were regularly generated in the simulation box with a volume fraction of 0.48. The regular initial configuration is helpful for testing whether the smectic structure is stable under certain simulation conditions and getting the physical insight of the formation mechanism of different structures. Then the temperature of the system was iteratively increased and decreased between 0.5$k_B T$ and $1.0k_B T$ with an interval of $\Delta T = 0.05k_B T$ every $2 \times 10^5$ timesteps. The box size in z-direction was also iteratively increased and decreased with the variance of temperature. The volume fraction was varied between 0.46 and 0.48. Finally, the system was further equilibrated for $1 \times 10^7$ timesteps at the 0.48 volume fraction. In the present work, the parameter $L/D_e$ was fixed at around 5 except for some special cases. This value corresponds to a moderate rod length according to our previous work. The values of $L/D_e$ in several systems with extremely small $R_L$ (i.e., 0.08, 0.12 and 0.15) were set as 11.5, 7.0 and 8.1, respectively.
Data availability. The authors declare that the data supporting the findings of this study are available within the article and its Supplementary Information files.

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Author contributions

Y.Y., Z.L., K.L., and Z.N. designed research. Y.Y. and G.C. performed experiments. H.P. performed simulations. Y.Y., G.C., H.P., X.Z., W.S., Z.L., K.L., and Z.N. analyzed data. M.L., C.F., and B.Y. provided valuable suggestions for the discussion of the results. Y.Y., Z.L., K.L., and Z.N. prepared the manuscript. All the authors read and commented on the manuscript.

Additional information

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Figure legends

**Figure 1** | Schematic illustration of four typical LC phases assembled from DBCs. Schematic illustrations of the geometric parameters of DBCs and the corresponding assembled N*, N2, N1, and SmA phases.

**Figure 2** | LC phases of DBCs with different geometries. Representative low (a,d,g,j) and high (b,e,h,k) magnification SEM images, and (c,f,i,l) POM images of LC phases assembled from different DBCs: (a,b,c) N* phase assembled from DBCs with $L_e = 160$ nm, $L_c = 1660$ nm, $D_e = 315$ nm, and $D_c = 240$ nm, (d,e,f) N2 phase assembled from DBCs with $L_e = 400$ nm, $L_c = 715$ nm, $D_e = 280$ nm, and $D_c = 190$ nm, (g,h,i) N1 phase assembled from DBCs with $L_e = 515$ nm, $L_c = 520$ nm, $D_e = 235$ nm, and $D_c = 155$ nm, (j,k,l) SmA phase assembled from DBCs with $L_e = 900$ nm, $L_c = 700$ nm, $D_e = 245$ nm, and $D_c = 225$ nm.

**Figure 3** | Correlation between geometry and LC phases of DBCs based on experimental results. (a) Experimental phase diagram of DBCs as a function of $R_D$ and $R_L$. The solid symbols represent the data points in Fig. 2. The black dashed lines indicate superficial boundaries between phases. The red lines correspond to $R_L = 0.5$ and $R_L = 1.0$, respectively. They locate approximately at the center of the corresponding phases, namely N2 and N1 phases. (b) Helical pitch $P$ as a function of $L_c/D_e$.

**Figure 4** | LC phases observed in Brownian dynamics simulations. Simulation results of LC phases assembled from DBCs with (a) $R_D = 1.40$ and $R_L = 0.15$ in N* phase (right: a double-twisted column extracted from left), (b) $R_D = 1.40$ and $R_L = 0.50$ in N2 phase (right: side and bottom view of the typical local structure), (c) $R_D = 1.40$ and $R_L = 1.00$ in N1 phase (right: a cylinder picked out along the z-axis), and (d) $R_D = 1.40$ and $R_L = 1.80$ in the SmA phase (right: a cylinder picked out along the z-axis). (e) Double-twisted structure viewed from different directions.

**Figure 5** | Phase diagram based on simulation results. Computational phase diagram of DBCs as a function of $R_D$ and $R_L$. The boundary dashed lines are taken from the experimental phase diagram.
Figure 1.
Figure 2.
Figure 3.
Figure 4.
Figure 5.