

Stability of cholesteric blue phases in the presence of a guest component

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We investigate theoretically, with the aid of numerical calculations based on a Landau-de Gennes theory, how two cubic cholesteric blue phases of a chiral liquid crystal, BP I and BP II, are stabilized when a guest component, such as a polymer network, is introduced and replace energetically costly defect regions. We show that the temperature range of stable BP I is significantly widened by the guest component, while the stability of BP II is only modestly enhanced.

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I. INTRODUCTION

Liquid crystals [1] have intrigued physicists as well as material scientists because they exhibit various phases with spontaneous orientational and/or positional molecular order. Cholesteric blue phases (BPs) of a highly chiral liquid crystal are good examples of such intriguing liquid crystalline phases that have challenged scientists for a long time. BPs have unique optical properties including the presence of Bragg reflections in the visible wavelength range implying periodic structures whose periodicity is a few hundred nanometers, and the absence of birefringence in spite of local orientational order, in contrast to usual nematic liquid crystals. Extensive experimental and theoretical studies in the 1980s and 1990s, summarized in a number of review articles [2–8], revealed that two thermodynamically stable BPs possess cubic symmetry; one phase referred to as BP I has the symmetry of O^8 [$I4_132$], while the symmetry of the other phase, BP II, is O^2 [$P4_232$] (a third BP, BP III, is believed to have an amorphous structure [9]). They comprise a regular array of so-called double-twist cylinders and a network of line disclinations of winding number $-1/2$. At the axis of a double-twist cylinder, the local orientation profile, or the director \mathbf{n} , exhibits twist distortions along all the directions perpendicular to the cylinder axis. Though the local free energy density of a liquid crystal at the axes of double-twist cylinders is lower than that of a single twist of a well-known chiral nematic (or cholesteric) phase (N^*), double-twist configurations cannot fill the whole space without introducing line disclinations as singularities in \mathbf{n} . BPs have thus attracted interest of physicists as a fascinating example of frustrated order. Recently, attention is being paid to BPs from other fields of physics because of the similarities between BPs and chiral ferromagnets [10], and also because double-twist configurations in a liquid crystal can be regarded as an example of Skyrmion excitations [11,12] whose crucial role has been discussed in various condensed matter systems [13] including two-dimensional electron gases [14], Bose-Einstein condensates [15], and thin films of chiral ferromagnets [16], as well as elementary particle systems where Skyrmion excitations were originally proposed [13,17].

The temperature range of stable BPs is a few Kelvin or less in usual liquid crystal materials, possibly because of

the delicate balance between the energetic gain of double-twist configurations and the loss of disclination lines. This narrow temperature range hindered the use of BPs in practical applications, though their characteristic optical properties easily tuned by an applied electric field was highly appealing. However, BPs with wider stable temperature range were recently reported by several groups [18–20]. Among them, Kikuchi *et al.* [18] demonstrated that introducing photo-cross-linked polymers as a guest component greatly stabilizes BPs over 60 K. We note that doping nanoparticles have been shown to stabilize BPs [21–25] though its effect is not as dramatic as that of photo-cross-linked polymers. These BPs with greater stability drew considerable attention to possible applications of BPs, including mirrorless lasing [26,27] and fast-switching displays [28–30].

Although there have been several theoretical studies concerning the stability of BPs of pure liquid crystals [4,6,31–35], the stabilization of BPs by introducing other components has only been given little attention theoretically [36], and it is not yet clear whether polymer-doped cholesteric blue phases are stabilized thermodynamically or kinetically. Recently, we discussed in a quantitative manner how a guest component can stabilize BP I [37]. We focused on its thermodynamic stability, and showed that the replacement of energetically costly disclination core regions by the guest component and the resulting reduction of the total free energy of BP I can give a good account for the wide temperature range of stable BP I observed experimentally [18]. In our previous study [37], we restricted our attention to moderate strength of chirality which yields BP I as the only stable BP between the isotropic phase (Iso) and N^* . When the chirality of the liquid crystal is strong enough, BP II appears as a thermodynamically stable phase between Iso and BP I (here we will not discuss BP III that could appear between Iso and BP II). It was shown experimentally [38] that, in contrast to the above-mentioned cases of BP I, the stabilization of BP II is highly difficult and requires a very careful choice of experimental conditions. Similar difficulties in stabilizing BP II were reported [23] when metal nanoparticles were doped as a guest component; the authors of Ref. [23] showed that longer sputter time (and hence more doped nanoparticles) yielded more stable BP I and that doped nanoparticles did not enhance the stability of BP II.

In this paper, we employ our previous theoretical argument [37] to discuss the stability of BP I and BP II when the chirality of the liquid crystals is strong enough. We show that indeed

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BP II is not significantly stabilized compared with BP I and give a discussion on its reason by comparing the profiles of free energy densities of BP I and BP II in a quantitative manner. After presenting our theoretical argument in Sec. II we present and discuss our results in Sec. III. Section IV concludes this paper.

II. THEORETICAL ARGUMENT

Our theoretical argument here is the same as that presented in our previous paper [37], where we discussed only the stability of BP I by choosing the parameters in the numerical calculations so that BP II did not appear in the phase sequence. In the present study we consider a situation where, in the absence of the guest component, BP II appears as a thermodynamically stable phase between an isotropic phase and BP I. Here we describe only the essential features of our theoretical argument and do not repeat the details already presented in [37].

The orientational order of cholesteric blue phases is discussed on the basis of a Landau–de Gennes theory which employs a second-rank symmetric and traceless tensor as an order parameter. After appropriate rescaling of the free energy density, the order parameter, and the length, the free energy density φ in terms of the rescaled order parameter $\chi_{\alpha\beta}$ becomes [6,37]

$$\varphi = \tau \text{Tr}\chi^2 - \sqrt{6} \text{Tr}\chi^3 + (\text{Tr}\chi^2)^2 + \kappa^2 \{[(\nabla \times \chi)_{\alpha\beta} + \chi_{\alpha\beta}]^2 + \eta[(\nabla \cdot \chi)_{\alpha}]^2\}, \quad (1)$$

where Tr represents the trace of a tensor. The rescaled temperature τ satisfies $\tau \simeq 1 \times (T - T^*)$, where T is the absolute temperature in Kelvin and T^* is the temperature at which the isotropic phase becomes unstable. The parameter κ , inversely proportional to the cholesteric pitch, measures the strength of chirality, and η concerns the anisotropy of liquid crystal elasticity. We choose $\kappa = 0.7$, which corresponds to the cholesteric pitch of approximately 160 nm (in our previous study [37], we chose $\kappa = 0.4$). We also set $\eta = 1$ (the so-called one-constant approximation). See Ref. [37] for the correspondence between rescaled variables and (dimensional) material parameters.

The profile of the orientational order $\chi_{\alpha\beta}$ without the guest component is calculated by solving the Euler-Lagrange equation $\delta F / \delta \chi_{\alpha\beta}(\mathbf{r}) - \lambda \delta_{ij} = 0$, where λ is a Lagrange multiplier ensuring $\text{Tr}\chi_{\alpha\beta} = 0$, and $F = \int_{\Omega_{\text{tot}}} d\mathbf{r} \varphi$ is the total free energy. The volume of integration Ω_{tot} is set on a $N^3 = 64^3$ cubic lattice with periodic boundary conditions. We use the numerical scheme developed in Ref. [39], in which not only the order parameter χ but also the lattice spacings are relaxed in the course of calculation. With the choice of $\kappa = 0.7$ and $\eta = 1$, we find that the ranges of stable BP I and BP II are $-3.44 \lesssim \tau \lesssim -0.277$ and $-0.277 \lesssim \tau \lesssim 0.224$, respectively and at lower and higher temperatures the stable phases are N^* and Iso, respectively.

When the free energy density profile $\varphi(\mathbf{r})$ or φ_i ($i = 1, 2, \dots, N^3$ labels the numerical lattice points) is calculated, we can determine how the liquid crystal accommodates the guest component. In our previous study [37], we assumed strong segregation with sharp interfaces between the liquid

crystal and the guest component, and weak anchoring at the interfaces without disturbing the liquid crystal alignment, and gave a detailed argument on the validity of these assumptions. We adopt the same assumptions here, and then the guest component just replaces the regions of liquid crystal with larger free energy density. As in [37], we sort the set $\{\varphi_i\}$ so that f_i becomes a monotonically decreasing function of i . When the volume fraction of the guest component is ϕ , the liquid crystal with higher φ must be replaced by the guest component, and the index i of the lattice point to be replaced satisfies $i/N^3 < \phi$. For the convenience of the following discussion, we define $\bar{\varphi}(\phi) \equiv \varphi_{i=\phi N^3}$. From its construction mentioned above, $\bar{\varphi}(\phi)$ is a monotonically decreasing function of ϕ . As we will show below, $\bar{\varphi}(\phi)$ can give a simple and clear insight into how large the defect regions with higher φ are, and how the regions with lower φ are distributed.

When we denote the region replaced by the guest component by Ω_c , the total free energy of the system of cholesteric blue phases with the guest component is given by

$$F_{\text{BP I, II}} = \int_{\Omega_{\text{tot}} - \Omega_c} d\mathbf{r} \varphi(\mathbf{r}) + \Omega_{\text{tot}} \phi \varphi_g, \quad (2)$$

where φ_g is the free energy density of the guest component assumed to be independent of position. From the definition of $\bar{\varphi}(\phi)$, Eq. (2) can be also written as

$$F_{\text{BP I, II}} = \Omega_{\text{tot}} \left(\int_{\phi}^1 d\phi' \bar{\varphi}(\phi') + \phi \varphi_g \right). \quad (3)$$

The free energy of N^* to be compared with $F_{\text{BP I, II}}$ is

$$F_{N^*} = \Omega_{\text{tot}} \{(1 - \phi) \varphi_{N^*} + \phi \varphi_g\}, \quad (4)$$

where φ_{N^*} is the free energy density of the N^* phase that is again position independent. We note that mere replacement of a part of liquid crystal volume by a guest component of equal volume in the above argument does not change the volume or the dimension of the unit cell. Although the interaction between the liquid crystal and the guest component leading to surface anchoring, which we assume to be negligible as

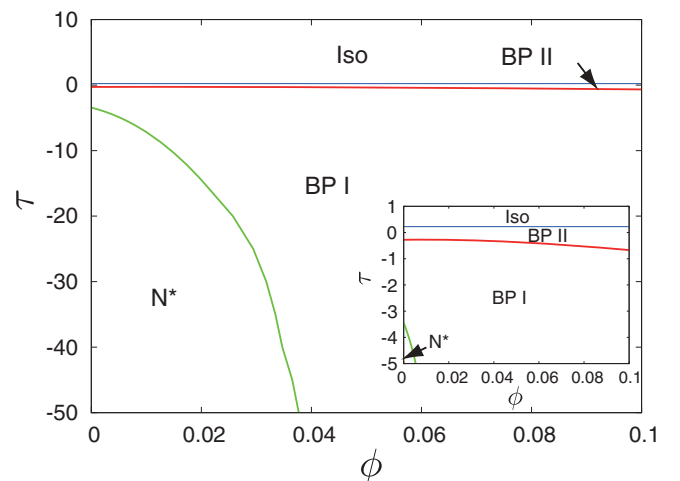


FIG. 1. (Color online) Calculated phase diagram in the presence of a guest component of volume fraction ϕ . For reference, the boundary between BP II and Iso ($\tau \simeq 0.224$) is also shown as a dotted line. Inset is a magnified one around $\tau = 0$.

mentioned above, could change the dimension of the unit cell, this effect is beyond the scope of the present work.

In our previous study [37], the effect of the energy of the interface between the liquid crystal and the guest component was discussed. It was shown that, with a reasonable value of the interfacial energy $\sigma \simeq 1 \times 10^{-5} \text{ J m}^{-2}$, the stability of blue phases was only slightly influenced by the interfacial energy. Moreover, we assumed that the guest components macroscopically phase separate in the N^* phase of the liquid crystal, which leads to the absence of the contribution of σ in the thermodynamic limit. This assumption is in fact unreasonable in the case of photopolymerized polymer network as the guest component. Then the contribution of σ is nonzero in F_{N^*} , which cancels out that in $F_{\text{BP I, II}}$. Therefore, in the present discussion, we do not consider the contribution of the interfacial energy.

The phase boundaries between the two of BP I, BP II, and N^* in the (ϕ, τ) phase diagram are determined by solving $F_{\text{BP I, II}} - F_{N^*} = 0$ and $F_{\text{BP I}} - F_{\text{BP II}} = 0$ (note that $F_{\text{BP I, II}}$ and F_{N^*} are now regarded as a function of ϕ and τ). In the experiments of Kikuchi *et al.* [18], the thermodynamic stability

of BP I over Iso at higher temperatures is not significantly affected by the guest component. It is possibly due to the fact that at the BP I–Iso transition the local nematic order loses its thermodynamic stability, and the guest component does not influence the local nematic order. Therefore we do not discuss here the effect of the guest component on the stability of BP I over Iso as in the previous study [37].

III. RESULTS AND DISCUSSIONS

In Fig. 1, we show the calculated (ϕ, τ) phase diagram of a chiral liquid crystal in the presence of a guest component. As already shown in our previous work [37], the temperature range of stable BP I becomes remarkably wider with the increase of ϕ . From the relation $\tau \simeq 1 \times (T - T^*)$, BP I can be stable over the temperature range of larger than 60 K by introducing a guest component of volume fraction less than 10%, despite the fact that it is stable only in the range of less than 4 K in the absence of a guest component. This is in good agreement with experiments of Kikuchi *et al.* [18] using a photo-cross-linked polymer as a guest component. On

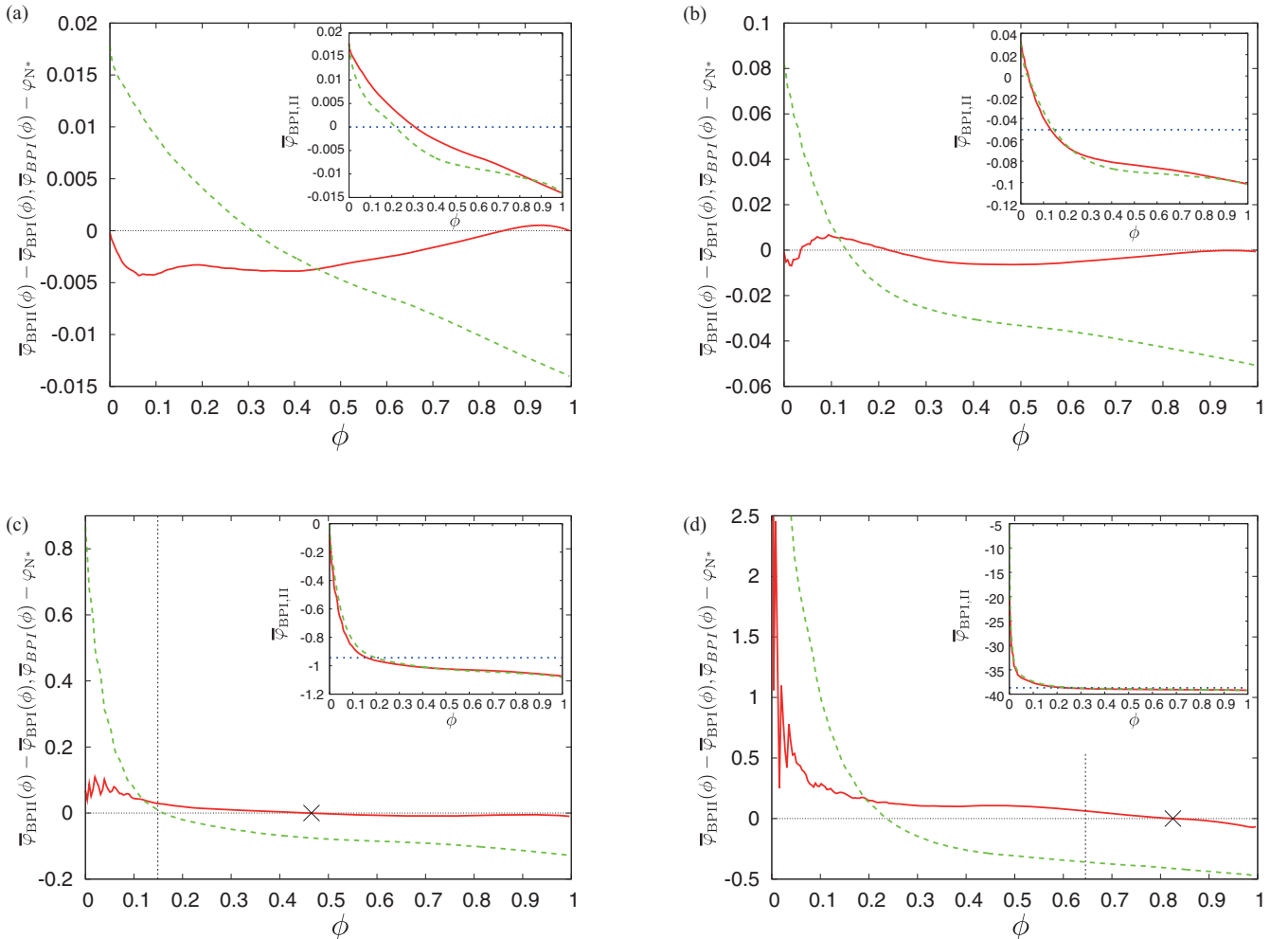


FIG. 2. (Color online) Plots of $\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi)$ (red solid line) and $\bar{\varphi}_{\text{BP I}}(\phi) - \varphi_{N^*}$ (green dashed line) for $\tau =$ (a) 0.2, (b) 0, (c) -1 , and (d) -10 . In (c) and (d), the values of ϕ satisfying $\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi) = 0$ and $F_{\text{BP I}} = F_{\text{BP II}}$ are highlighted by a cross (x) and a vertical dashed line, respectively. Irregular oscillations of $\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi)$ at small ϕ are a numerical artifact. Insets are the plots of $\bar{\varphi}_{\text{BP II}}(\phi)$ (green dashed line), $\bar{\varphi}_{\text{BP I}}(\phi)$ (red solid line), and, for reference, φ_{N^*} (blue dotted horizontal line).

the other hand, the temperature range of stable BP II is not significantly altered by the increasing ϕ , again in agreement with experimental findings [38].

To clarify why BP II is not significantly stabilized by a guest component, we discuss the behavior of free energy densities $\bar{\varphi}(\phi)$ for BP I and BP II [which we will denote by $\bar{\varphi}_{\text{BP I}}(\phi)$ and $\bar{\varphi}_{\text{BP II}}(\phi)$, respectively]. In Fig. 2 we plot $\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi)$ and $\bar{\varphi}_{\text{BP I}}(\phi) - \varphi_{N^*}$ as well as $\bar{\varphi}_{\text{BP I}}(\phi)$ and $\bar{\varphi}_{\text{BP II}}(\phi)$ at various temperatures. When $\tau = 0.2$, close to the transition between BP II and Iso, $\bar{\varphi}_{\text{BP II}}(\phi)$ is smaller than $\bar{\varphi}_{\text{BP I}}(\phi)$ except at ϕ close to 1 as seen in Fig. 2(a). This clearly indicates that BP II is more favorable energetically than BP I at this temperature. However, Figs. 2(b)–2(d) indicate that, as the temperature is lowered, $\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi)$ becomes positive at smaller values of ϕ (except at those very close to zero for $\tau = 0$). Moreover, in the range of ϕ where $\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi) < 0$, the energetic gain of BP II over BP I, i.e., $|\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi)|$, becomes smaller for lower τ . We also note that the value of ϕ at which $\bar{\varphi}_{\text{BP II}}(\phi) = \bar{\varphi}_{\text{BP I}}(\phi)$ becomes larger. These properties of $\bar{\varphi}_{\text{BP II}}(\phi)$ are responsible for the fact that BP II becomes less and less favorable energetically than BP I when the temperature is lowered.

The function $\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi)$ characterizes the energetic advantage of BP II over BP I, and $\bar{\varphi}_{\text{BP I}}(\phi) - \varphi_{N^*}$ characterizes that of BP I over N^* . It is well known [6] that double-twist distortions in BP I and BP II are locally more favorable energetically than single-twist ones in N^* and compensate for the energetic loss in the defect regions. This is clearly seen in the behavior of $\bar{\varphi}_{\text{BP I}}(\phi) - \varphi_{N^*}$, and once the energetically costly region with small ϕ is removed by the guest component, BP I can be energetically more favorable than N^* . However, for BP II to become the most stable phase, its free energy must be lower than that of BP I as well as that of N^* . The energetic gain of BP II over BP I, $|\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi)|$, at ϕ close to 1 is much smaller than that of BP I over N^* , $\bar{\varphi}_{\text{BP I}}(\phi) - \varphi_{N^*}$. This is because in BP II, as in BP I, energetically favorable regions involve double-twist distortions and their free energy densities φ cannot be much smaller than that in BP I, except at higher temperatures [Fig. 2(a)]. Moreover, when the temperature τ is lowered, the range of ϕ with $\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi) < 0$ shrinks [see Figs. 2(b)–2(d)]. Therefore, a large amount of volume at which $\bar{\varphi}_{\text{BP II}}(\phi) - \bar{\varphi}_{\text{BP I}}(\phi) > 0$ must be removed by a guest component for the free energy of BP II to be lower than that of BP I. This qualitatively explains why BP II is not easy to stabilize by introducing a guest component as observed experimentally [23,38].

Here we comment on a previous study by Ravnik *et al.* [21] who discussed the effect of spherical colloidal particles on the stability of cholesteric blue phases. By numerical calculations based on a Landau–de Gennes theory, they showed that in the case of small cholesteric pitch BP II can be successfully stabilized by doping colloidal particles. They also visualized the colloidal arrangement that minimizes the free energy and colloidal particles are at the disclination lines (we also note that a recent experiment [40] clearly demonstrated the trapping of colloidal particles at a disclination line of a nematic liquid crystal). In their case, where colloidal particles reside in a unit cell of BP II depends sensitively on surface anchoring strength, particle radius, number density (or volume fraction) of particles, and so on (the effect of particle size is more

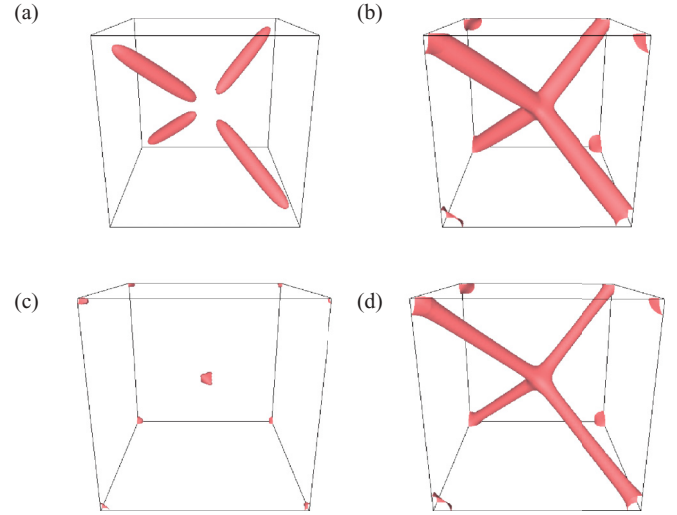


FIG. 3. (Color online) Isosurfaces of free energy density at (a) and (b) $\tau = 0$ and (c) and (d) $\tau = -1$. The value of φ is (a) 0.01, (b) -0.005 , (c) -0.06 , and (d) -0.3 .

clearly manifested in a different work [41]). They discuss the stability of blue phases doped with particles on the basis that the arrangements of particles in BP I and BP II are face-centered and body-centered cubic, respectively, which, they showed, are the optimum arrangements in terms of the free energy. However, the number density of colloidal particles was inevitably fixed, and therefore the effect of its variation cannot be deduced from their argument. Moreover, it is not clear from their text whether the energies of doped BP I and BP II were compared at the same number density of colloidal particles. Therefore, our results cannot be directly compared with those of [21], and further studies will be necessary to clarify the difference between the effect of doping solid particles and that of introducing a polymer network which can adopt an optimum shape. Nevertheless, we believe that the reduction of the free energy by the replacement of defect core is the common mechanism of the stabilization of blue phases, whatever the guest component is.

Finally it is interesting to see how the regions with higher free energy density are distributed in BP II (in the case of BP I, they are straight disclination lines, as demonstrated visually in Refs. [21,37]). In Fig. 3 we show their distributions at different temperatures τ . When $\tau = 0$ (BP II is lower in free energy than BP I), the most energetically costly regions reside in the straight parts of disclination lines (this was already shown in Fig. 1 B of Ref. [21]). On the other hand, they are at the junction point of four disclination lines when $\tau = -1$ (at which BP I is the most stable). Therefore the distribution of free energy density in BP II depends on temperature in a nontrivial manner, and an intuition that the free energy density is the highest at the junction points proves wrong. For BP II to be energetically more favorable than BP I, it might be necessary to reduce the free energy density at the junction points that do not exist in BP I.

IV. CONCLUSION

We discussed, by numerical calculations using a Landau–de Gennes theory, how cubic cholesteric blue phases, BP I and

BP II, are stabilized by introducing a guest component, based on an assumption that the guest component replaces the energetically costly defect regions of the blue phase. In our previous work [37] we showed that BP I can be effectively stabilized by the guest component, in agreement with an experimental demonstration by Kikuchi *et al.* [18]. In this work we considered the cases with larger helical twisting power of a chiral liquid crystal for which both BP I and BP II are stable (without a guest component) in a certain temperature range. We showed that the temperature range of BP II is not significantly widened by the guest component in contrast to that of BP I, which is again in agreement with experimental findings. This result was qualitatively explained by comparing the profiles of the free energy densities of BP I and BP II. BP I after replacing the defects comprises regions with double-twist distortions that are energetically more favorable than single-twist distortions in N^* , which is the main reason for the great enhancement of the stability of BP I over N^* . On the other hand, after the replacement of defects, both BP I and BP II are made up of energetically favorable double-twist regions, and thus the mere replacement of defect regions cannot give a great energetic advantage of BP II over BP I.

Our theoretical argument can be applied to any problem concerning the stability of soft materials with energetically costly topological defects when an immiscible guest component is introduced. One interesting problem could be the stability of another cholesteric blue phase, BP III. It was shown recently [24,25] that BP III could be effectively stabilized by doping nanoparticles. There were considerable debates on the structure of BP III, and an amorphous network of disclination lines was suggested as its probable structure numerically [9]. Our argument, combined with the numerical profiles of BP III, could clarify the nature of the stabilization of BP III which is now entirely unclear.

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