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ANCHORING OF TWISTED CHIRAL NEMATIC LIQUID CRYSTALS ON A THIN POLYMERIC FILM

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A generalized form of anisotropic surface energy for the description of twisted chiral nematic samples with side-chain polymeric layers is proposed, and the anchoring properties of such samples are analyzed, using the equivalent anchoring energy method proposed by Alexe-Ionescu *et al.* Our results give a more general expression for the equivalent anchoring energy, the new variables included in the expression are determined by the anchoring of the side chains and by the sterical interaction of the side chains with the nematic liquid crystals (NLCs). In addition, the coupling of the polymer side chains with the surface, reduces the equivalent anchoring energy as well as the threshold field.

Keywords: Twisted chiral nematic liquid crystal cell; weak anchoring; side-chain polymer; threshold field; surface anchoring.

1. Introduction

In liquid crystals (LCs), surface effects have been studied mainly for nematic phases.[1] The translational symmetry and often also the rotational symmetry of the nematic phase are broken when it encounters an interface.[2] In the absence of an external electric or magnetic field (field free condition), the alignment of nematic liquid crystals (NLCs) is entirely dictated by the surface forces, which depend on the presence of surface layers.[3] In the special case where the surface layer is an ordered medium particular effects are expected, because in addition to the physicochemical interactions, the steric interaction also has to be taken into account.[4–6]

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If the surface layer is polymeric film, the LC molecules are aligned by the sterical interactions between the polymer molecules and the LC molecules.[7]

For a NLC sample with weak anchoring surfaces, the phenomenological expression for the surface energy density proposed by Rapini–Papoular (RP),[8] which describes the anisotropic interaction between the nematic director and the substrate, is expressed by:

$$f_s = -\frac{1}{2}\alpha(\mathbf{n} \cdot \mathbf{e})^2 \,, \tag{1}$$

where $\mathbf{e}$ is the easy direction, $\mathbf{n}$ is the NLC director at the surface, and the anchoring strength parameter α measures the ability of the director to deviate from the easy direction. The RP formula only includes the lowest terms of the lowest order, while it can describe many surface effects on the phenomenological level successfully. With Eq. (1) Sugimura *et al.* have discussed the threshold and the saturation fields for twisted chiral nematic (TCN) layers with weak boundary coupling.[9,10]

Recently, Alexe-Ionescu *et al.*[11] have investigated the anchoring of a side-chain polymer for a homogeneous NLC sample. These nanostructured polymeric layers seem very promising for application in display technology because they allow a continuous control of the pretilt angle of NLCs.[12] They assumed an expression of the anisotropic surface energy for this special case where the aligning layer is a side-chain polymer and performed analysis in the presence and in the absence of external electric field. By comparing the results of standard analysis with the special case, they found that the equivalent anchoring energy depends on the coupling of the nematic with the polymer side groups as well as on the coupling of the polymer side groups with the surface. However, the anisotropic surface energy they proposed is only suitable for untwisted NLC samples.

In this paper, we extend the anisotropic surface energy proposed in Ref. 11 to a more general form, for the description of a special TCN sample with side-chain polymeric layers, as seen in Appendix A, and then we investigate the anchoring properties of a side-chain polymer for a TCN sample phenomenologically, using the equivalent anchoring energy method proposed by Alexe-Ionescu *et al.*[11]

2. Theoretical Basis

In the surface layer, there exist an anisotropic surface field characterized by azimuthal anchoring and polar anchoring. Experimentally determined values of azimuthal anchoring energy ($\sim 10^{-5}$ J/m^2) is usually an order of magnitude lower than polar anchoring energy ($\sim 10^{-4}$ J/m^2),[13–15] so we assume that the coupling of the nematic with the polymer side groups is symmetric, while the coupling of the polymer side groups with the surface is anisotropic. In this assumption, Appendix A gives the anisotropic surface energy of TCN samples with side-chain polymer layers as:

$$f_s = -\frac{1}{2}\alpha(\mathbf{n} \cdot \mathbf{w})^2 + \frac{1}{2}\beta_a(\mathbf{w} \cdot \mathbf{e}_y)^2 + \frac{1}{2}\beta_p(\mathbf{w} \cdot \mathbf{e}_z)^2 \,, \tag{2}$$

where $\mathbf{w}$ is the direction of the polymer side groups. We suppose that in the absence of any imposed deformation the polymer side group is parallel to the x-axis. The parameters β_a, β_p are azimuthal and polar anchoring strength cofficients connected with the restoring torque of elastic origin acting on the lateral group, respectively. Assuming mesogenic side groups, the parameter α takes into account the tendency of the nematic molecules to be parallel, for sterical reasons, to the side-chain groups. Here we do not take into account the presence of aliphatic chains, imposing homeotropic alignment, as mentioned in Ref. 11.

We consider a TCN cell of thickness d located between the two planes $z = \pm d/2$ and with mirror symmetry with respect to the middle plane $z = 0$, for which, $\alpha = \alpha^- = \alpha^+$, $\beta_a = \beta_a^- = \beta_a^+$, $\beta_p = \beta_p^- = \beta_p^+$, and then for any ϕ_t describing the total twist angle of the TCN, we have $\phi(-z) = \phi_t - \phi(z)$ and $\theta(-z) = \theta(z)$. The surface tilt angles are taken to be the same at both surfaces, $\theta_0 = \theta(-d/2) = \theta(d/2)$. The direction $\mathbf{w}$ of the polymer side groups at $z = -d/2$ and the director $\mathbf{n}$ at the z layer can be expressed as:

$$\mathbf{w} = \left(\cos\theta_w \cos\phi_w, \cos\theta_w \sin\phi_w, \sin\theta_w \right), \tag{3}$$

$$\mathbf{n} = \left(\cos\theta \cos\phi, \cos\theta \sin\phi, \sin\theta \right), \tag{4}$$

where θ_w, ϕ_w are the tilt angle and azimuthal angle of the polymer side groups $\mathbf{w}$, and $\theta = \theta(z)$, $\phi = \phi(z)$ are the tilt angle and azimuthal angle of the director $\mathbf{n}$.

According to Eq. (2), the surface energy density at $z = -d/2$ is given by:

$$f_s^- = -\frac{1}{2}\alpha[\cos\theta_0 \cos\phi_0 \cos\theta_w \cos\phi_w + \cos\theta_0 \sin\phi_0 \cos\theta_w \sin\phi_w + \sin\theta_0 \sin\theta_w]^2$$

$$+ \frac{1}{2}\beta_a \cos^2\theta_w \sin^2\phi_w + \frac{1}{2}\beta_p \sin^2\theta_w , \tag{5}$$
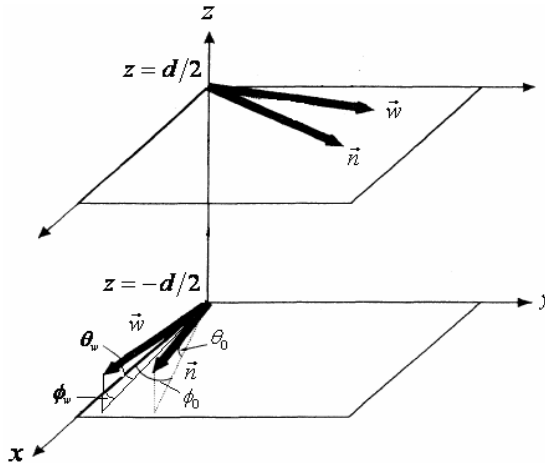

Fig. 1.   The geometry of the twisted chiral nematic cell located between the two paltes $z = \pm d/2$.

where $\phi_0 = \phi(-d/2)$. We assume that the dielectric anisotropy of the NLC is positive ($\varepsilon_a > 0$), and the applied electric field is parallel to the z-axis, $\mathbf{E} = (0, 0, E)$, then the free energy density in the bulk up to the second order derivative of the director is given by:

$$f_b = \frac{1}{2} f(\theta)(d\theta/dz)^2 + \frac{1}{2} h(\theta)(d\phi/dz)^2 + k_2 \cos^2 \theta (d\phi/dz)$$

$$+ \frac{k_2^2}{2K_{22}} - \frac{1}{2} \varepsilon_a E^2 \sin^2 \theta \,, \tag{6}$$

where

$$f(\theta) = K_{11} \cos^2 \theta + K_{33} \sin^2 \theta \,,$$
$$f(\theta) = \cos^2 \theta (K_{22} \cos^2 \theta + K_{33} \sin^2 \theta) \,, \tag{7}$$
$$k_2 = -2\pi K_{22}/p_0 \,,$$

and K_{11}, K_{22}, K_{33} are the splay, twist and bend elastic constants of the NLC, respectively, p_0 denotes the pitch of the material induced by a chiral dopant. The surface-like term $K_s \equiv K_{22} + K_{24}$ is omitted here as it does not contribute to the free energy when $\mathbf{n}$ depends just on a single Cartesian coordinate.

The total free energy per unit area of the cell is given by:

$$F = \int_{-d/2}^{d/2} f_b dz + 2f_s^- - \Delta E^2 \sin^2 \theta_w \,, \tag{8}$$

where $\Delta = \varepsilon_{ap}\ell$, ε_{ap} being the dielectric anisotropy of the polymer side groups, and ℓ the length of the chain connecting the lateral group to the surface. $\Delta E^2 \sin^2 \theta_w$ represents the interaction energy between the polymer side group and the electric field. Minimization of the free energy in the bulk yields the stable director configuration for any given field. The variational calculation[16] of Eq. (6) yields

$$f(\theta)(d^2\theta/dz^2) + \frac{1}{2} f_\theta (d\theta/dz)^2 - \frac{1}{2} h_\theta (d\phi/dz)^2 + 2k_2 \sin \theta \cos \theta (d\phi/dz)$$

$$+ \varepsilon_a E^2 \sin \theta \cos \theta = 0 \,, \tag{9}$$

$$h(\theta)(d\phi/dz) + k_2 \cos^2 \theta = C \,, \tag{10}$$

where C is a constant of integration, $f_\theta = df/d\theta$, $h_\theta = dh/d\theta$.

3. Fréedericksz Transition

Threshold electric field E_c means the value on which the director of LCs starts to generate a deformation. We suppose that $\theta_w = 0$ for $E < E_c$. In the vicinity of the threshold field, the deformations of nematic and side groups are very small, that is for $E \sim E_c$, $\theta \sim 0$ and $\theta_w \sim 0$. Considering this limit and neglecting the higher

ranks, Eq. (7) gives $f(\theta) \sim K_{11} + (K_{33} - K_{11})\theta^2$, $h(\theta) \sim K_{22} + (K_{33} - 2K_{22})\theta^2$, $f_\theta \sim 2(K_{33} - K_{11})\theta$ and $h_\theta \sim 2(K_{33} - 2K_{22})\theta$, and Eqs. (8)–(10) change to

$$F = \int_{-d/2}^{d/2} \left[\frac{1}{2} K_{11}(d\theta/dz)^2 + \frac{1}{2}[K_{22} + (K_{33} - 2K_{22})\theta^2](d\phi/dz)^2 \right.$$

$$\left. + k_2(d\phi/dz) + \frac{k_2^2}{2K_{22}} - \frac{1}{2}\varepsilon_a E^2\theta \right] dz$$

$$+ \beta_a - \beta_a \cos^2 \phi_w + [\beta_a \cos^2 \phi_w - \beta_a + \beta_p - \Delta E^2 + \alpha \cos^2(\phi_0 - \phi_w)]\theta_w^2$$

$$+ \alpha\theta_0^2 \cos^2(\phi_0 - \phi_w) - 2\alpha\theta_0\theta_w \cos(\phi_0 - \phi_w) - \alpha \cos^2(\phi_0 - \phi_w) , \tag{11}$$

$$K_{11}(d^2\theta/dz^2) + (2K_{22} - K_{33})\theta(d\phi/dz)^2 + 2k_2\theta(d\phi/dz) + \varepsilon_a E^2\theta = 0 , \tag{12}$$

$$K_{22}(d\phi/dz) + k_2 = C . \tag{13}$$

According to Eq. (37) in Ref. 10, we have

$$C - k_2 = K_{22}(\phi_t - 2\phi_0)/d . \tag{14}$$

Equations (13) and (14) lead to the expression of $d\phi/dz$, namely

$$d\phi/dz = (\phi_t - 2\phi_0)/d . \tag{15}$$

Substituting Eq. (15) into Eq. (12) and defining the electric coherence length

$$\xi = \left[\frac{K_{11}}{(2K_{22} - K_{33})[(\phi_t - 2\phi_0)/d]^2 + 2k_2(\phi_t - 2\phi_0)/d + \varepsilon_a E^2} \right]^{1/2} , \tag{16}$$

Eq. (12) can be rewritten as:

$$\xi^2(d^2\theta/dz^2) + \theta = 0 . \tag{17}$$

Taking into account the symmetry condition $\theta(-z) = \theta(z)$, from Eq. (17) we obtain

$$\theta = A \cos(z/\xi) . \tag{18}$$

Substituting Eqs. (15) and (18) into F given by Eq. (11), we get

$$F = -\frac{1}{2}\frac{K_{11}}{\xi} \sin(d/\xi)A^2 + \frac{1}{2}K_{22}(\phi_t - 2\phi_0)^2/d + k_2(\phi_t - 2\phi_0) + \frac{k_2^2}{2K_{22}}d$$

$$+ \beta_a - \beta_a \cos^2 \phi_w + [\beta_a \cos^2 \phi_w - \beta_a + \beta_p - \Delta E^2 + \alpha \cos^2(\phi_0 - \phi_w)]\theta_w^2$$

$$+ \alpha A^2 \cos^2(d/2\xi) \cos^2(\phi_0 - \phi_w) - 2\alpha A \cos(d/2\xi)\theta_w \cos(\phi_0 - \phi_w)$$

$$- \alpha \cos^2(\phi_0 - \phi_w) . \tag{19}$$

The stable state demands

$$\frac{\partial F}{\partial A} = \frac{\partial F}{\partial \theta_w} = \frac{\partial F}{\partial \phi_0} = \frac{\partial F}{\partial \phi_w} = 0 . \tag{20}$$

From Eq. (19), the relations $\partial F/\partial A = \partial F/\partial \theta_w = 0$ give

$$\left[-\frac{K_{11}}{\xi} \sin(d/\xi) + 2\alpha \cos^2(d/2\xi) \cos^2(\phi_0 - \phi_w) \right] A$$
$$- 2\alpha \cos(d/2\xi) \cos(\phi_0 - \phi_w)\theta_w = 0 \,, \tag{21}$$

$$[\beta_a \cos^2 \phi_w - \beta_a + \beta_p - \Delta E^2 + \alpha \cos^2(\phi_0 - \phi_w)]\theta_w$$
$$- \alpha \cos(d/2\xi) \cos(\phi_0 - \phi_w)A = 0 \,. \tag{22}$$

Then the relations $\partial F/\partial \phi_0 = \partial F/\partial \phi_w = 0$ give

$$-\frac{K_{22}}{d}(\phi_t - 2\phi_0) - k_2 + \alpha \cos(\phi_0 - \phi_w) \sin(\phi_0 - \phi_w) = 0 \,, \tag{23}$$

$$\beta_a \cos \phi_w \sin \phi_w - \alpha \cos(\phi_0 - \phi_w) \sin(\phi_0 - \phi_w) = 0 \,. \tag{24}$$

Equations (23) and (24) show that, for certain values of ϕ_t, the solutions of ϕ_0 and ϕ_w can be determined completely by α and β_a, and the details is given in Appendix B. Equations (21) and (22) form a homogeneous system. The solution $\theta_w = A = 0$, corresponding to the undistorted state, is stable if

$$\frac{\partial^2 F}{\partial A^2} > 0, \quad \frac{\partial^2 F}{\partial A^2}\frac{\partial^2 F}{\partial \theta_w^2} - \frac{\partial^2 F}{\partial A \partial \theta_w} > 0 \,,$$

which gives

$$-\frac{K_{11}}{\xi} \sin(d/\xi) + 2\alpha \cos^2(d/2\xi) \cos^2(\phi_0 - \phi_w) > 0 \,, \tag{25}$$

$$\beta_a \cos^2 \phi_w \alpha \cos^2(\phi_0 - \phi_w) - [\beta_a \cos^2 \phi_w - \beta_a + \beta_p - \Delta E^2 + \alpha \cos^2(\phi_0 - \phi_w)]$$
$$\times \frac{K_{11}}{\xi} \tan(d/2\xi) - \alpha^2 \cos^2(\phi_0 - \phi_w) \sin^2(\phi_0 - \phi_w) > 0 \,. \tag{26}$$

Since ℓ is a molecular dimension $\Delta E^2 \ll \beta_\alpha$ and $(\phi_0 - \phi_w)$ is very small, Eqs. (25) and (26) can be rewritten as:

$$\tan(d/2\xi) < \xi\alpha \cos^2(\phi_0 - \phi_w)/K_{11} \,, \tag{27}$$

$$\tan(d/2\xi) < \xi w_e/K_{11} \,, \tag{28}$$

respectively, where

$$w_e = \frac{\alpha \cos^2(\phi_0 - \phi_w)\beta_a \cos^2 \phi_w}{\alpha \cos^2(\phi_0 - \phi_w) + \beta_a \cos^2 \phi_w + \beta_p - \beta_a} \,. \tag{29}$$

As has been pointed out above, the values of ϕ_0 and ϕ_w in Eq. (29) can be determined from Eqs. (23) and (24). The more restrictive inequality between (27) and (28) is the second one. Consequently the threshold we are seeking is given by:

$$\tan(d/2\xi_c) = \xi_c w_e/K_{11} \,. \tag{30}$$

According to Eq. (16), the threshold electric field E_c is given by:

$$E_c = \left[\frac{K_{11}d^2/\xi_c^2 - (2K_{22} - K_{33})(\phi_t - 2\phi_0)^2 - 2k_2(\phi_t - 2\phi_0)d}{\varepsilon_a d^2} \right]^{1/2} . \tag{31}$$

where ξ_c are the solutions of the transcendental equation (30).

To see the effect of the coupling of the polymer side chains with the surface, we check our generalized side-chain model in the assumption of $\beta_a = \beta_p = \beta$.

Firstly, we discuss our result in comparison with that of Sugimura *et al.*[10] by using numerical calculations. We introduce the similar dimensionless parameter

$$\lambda = \frac{\pi K_{22}}{\alpha d} , \tag{32}$$

and the reduced electric field $u = E_c/E_0$ to those in Ref. 10, where

$$E_0 = \frac{\pi}{d}\sqrt{K_{11}/\varepsilon_a} \tag{33}$$

is the threshold electric field for an untwisted nematic slab ($\phi_t = 0$) with strong anchoring boundary ($\lambda = 0$, i.e., $\alpha \to \infty$). Then we plot the λ dependence of the reduced electric field u for various β/α in Fig. 2, which shows our present result and that reported in Ref. 10 for a twisted nematic cell ($\phi_t = 90°, d/p_0 = 0$) with the same material parameters as those used in Ref. 10, i.e., $K_{33}/K_{11} = 1.5$ and $K_{22}/K_{11} = 0.6$. It is clear that the coupling of the side chains with the surface reduces the threshold field. In addition, as the anchoring parameter β increasing, our result tends to that of Sugimura *et al.* in Ref. 10, which is reasonable.

Secondly, in the assumption of $\beta_a = \beta_p = \beta$, Eq. (29) reduces to

$$w_e = \frac{\alpha \cos^2(\phi_0 - \phi_w)\beta \cos^2 \phi_w}{\alpha \cos^2(\phi_0 - \phi_w) + \beta \cos^2 \phi_w} . \tag{34}$$

It follows that

$$\frac{1}{w_e} = \frac{1}{\alpha \cos^2(\phi_0 - \phi_w)} + \frac{1}{\beta \cos^2 \phi_w} . \tag{35}$$

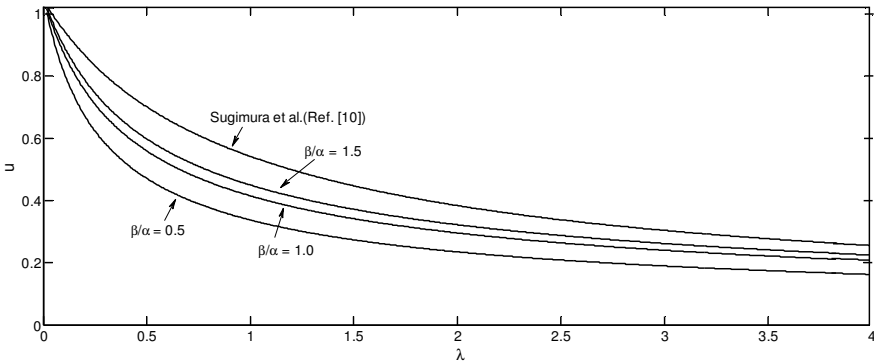Fig. 2. The λ dependence of the reduced electric field u for twisted nematic cells. We show the present theoretical result and that reported in Ref. 10.

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From Eq. (35) we find that if $\beta \to \infty$, i.e., the side groups are fixed and $\phi_w = 0$, $w_e \to \alpha \cos^2 \phi_0$, as expected. Moreover, Eq. (35) also gives that the equivalent anchoring energy w_e is smaller than α and β. Consequently, the coupling of the side chains with the surface reduces the equivalent anchoring energy.

In addition, by comparing Eq. (34) or (35) with the equivalent anchoring energy given by Alexe-Ionescu *et al.*,[11] i.e., $w_e = \alpha\beta/(\alpha + \beta)$, we conclude that, in the linear approximation, we get a more general expression for the equivalent anchoring energy, and the new variables ϕ_w and ϕ_0 included in our generalized expression are determined completely by the anchoring parameters α and β for a certain ϕ_t.

As has mentioned above, in the surface layer, the azimuthal anchoring energy is usually an order of magnitude lower than polar anchoring energy, and thus the condition $\beta_a = \beta_p = \beta$ is seldom fulfilled. The discussion above only gives a qualitative analysis of the effect of the coupling of the polymer side chains with the surface.

4. Conclusions

In this work, extending the work of Alexe-Ionescu *et al.*, we have proposed a generalized form of the anisotropic surface energy [see Eq. (2)] and studied the anchoring properties of a side-chain polymer for filed-controlled TCN samples. Using the treatment method of Alexe-Ionescu *et al.*, we considered the Fréedericksz transition and obtained a more general expression for the equivalent anchoring energy w_e [Eq. (34) or (35)]. We show that the azimuthal angles of side chains and nematic director **n** at the surface, i.e., ϕ_w and ϕ_0, which have important contribution to the equivalent anchoring energy w_e, are determined completely by the anchoring parameters α and β for a certain ϕ_t. In addition, the coupling of the side chains with the surface reduces the equivalent anchoring energy as well as the threshold field.

Acknowledgments

Appendix A. Surface Anchoring Energy of a Side-Chain Polymer Layer

For the case we considered in the text, where the aligning layer is a side-chain polymer, the anisotropic part of the surface anchoring energy depends on the coupling of the nematic with the polymer side groups, as well as on the coupling of the polymer side groups with the surface.

Considering the symmetry of the nematic director **n** with respect to the polymer side group **w**, and assuming that the corresponding anchoring strength coefficient

is α, the finite anchoring energy per unit area for the director orientation can be expressed as:

$$f_{s1} = -\frac{1}{2}\alpha(\mathbf{n} \cdot \mathbf{w})^2 \,. \tag{A.1}$$

Considering the anisotropy of the polymer side group with respect to the surface, we introduce two parameters β_p, β_a describing the polar and azimuthal anchoring strength coefficients respectively, and suppose that in the absence of any imposed deformation, the polymer side group is parallel to the x-axis. Then the finite anchoring energy per unit area describing the elastic restoring torque acting on the polymer side groups is

$$f_{s2} = \frac{1}{2}\beta_a(\mathbf{w} \cdot \mathbf{e}_y)^2 + \frac{1}{2}\beta_p(\mathbf{w} \cdot \mathbf{e}_z)^2 \,. \tag{A.2}$$

Now the total surface energy density of a side-chain polymer layer is

$$f_s = -\frac{1}{2}\alpha(\mathbf{n} \cdot \mathbf{w})^2 + \frac{1}{2}\beta_a(\mathbf{w} \cdot \mathbf{e}_y)^2 + \frac{1}{2}\beta_p(\mathbf{w} \cdot \mathbf{e}_z)^2 \,. \tag{A.3}$$

Appendix B. The Detailed Solutions of ϕ_0 and ϕ_w

According to Eqs. (23) and (24) in the text, we have

$$\frac{K_{22}}{d}(\phi_t - 2\phi_0) + k_2 = \beta_a \cos\phi_w \sin\phi_w \,, \tag{B.1}$$

$$\beta_a \cos\phi_w \sin\phi_w = \alpha \cos(\phi_0 - \phi_w)\sin(\phi_0 - \phi_w) \,. \tag{B.2}$$

Equations (B.1) and (B.2) give

$$\phi_0 = \frac{1}{2}\phi_t + \frac{k_2 d}{2K_{22}} - \frac{\beta_a d \cos\phi_w \sin\phi_w}{2K_{22}} \,, \tag{B.3}$$

$$\phi_0 = \phi_w + \frac{1}{2}\arcsin\left(\frac{2\beta_a \cos\phi_w \sin\phi_w}{\alpha}\right) \,, \tag{B.4}$$

respectively. Elimination of ϕ_0 from Eqs. (B.3) and (B.4) gives

$$\phi_t + \frac{k_2 d}{K_{22}} = 2\phi_w + \arcsin\left(\frac{2\beta_a \cos\phi_w \sin\phi_w}{\alpha}\right) + \frac{\beta_a d \cos\phi_w \sin\phi_w}{K_{22}} \,. \tag{B.5}$$

Equation (B.5) shows that for a certain ϕ_t, the solution of ϕ_w can be determined by the values of α and β_a, and then ϕ_0 is obtained by either of Eq. (B.3) or (B.4). That means that for a certain ϕ_t, using Eqs. (23) and (24) in the text, the solutions of ϕ_w and ϕ_0 can be determined completely by the values of α and β_a.

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