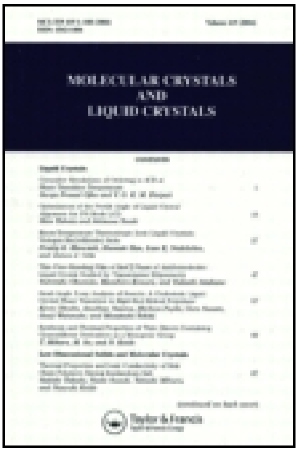

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Azimuthal Anchoring Energy at a SiO_x-Nematic Interface

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AZIMUTHAL ANCHORING ENERGY AT A SiO_x-NEMATIC INTERFACE.

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Abstract The azimuthal anchoring energy at the interface between the nematic liquid crystal pentylcyanobiphenyl (5CB) and an obliquely evaporated SiO_x substrate is measured by using a reflectometric method. The time response of the director at the surface is investigated and two very different regimes are evidenced. The azimuthal anchoring energy coefficient exhibits an almost-critical behaviour near the clearing temperature and is almost independent on the thickness d of the SiO_x layer but it exhibits a sharp decrease for $d \le d_c \approx 25$ Å. The anchoring function is found to depend on the azimuthal director angle φ as $W(\varphi) = W_a/2 \sin^2(\varphi)$.

Keywords: nematic interfaces, anchoring, time response

INTRODUCTION

Let's consider the interface between an NLC and a solid substrate lying on the x-y plane where the director $\vec{n}$, makes the polar angle θ with the z-axis and the azimuthal angle φ with the x-axis. In the absence of external torques the anchoring energy function[1-5] $W(\theta,\varphi)$ is minimised if the director is aligned along a well defined "*easy axis*" defined by the two angles θ_0 and φ_0. By using external torques (magnetic, electric...) one can rotate the director from the easy axis. In most of the experiments the anchoring is strong and thus, one can only explore the anchoring energy function near the equilibrium point $(\theta \approx \theta_0, \varphi \approx \varphi_0)$:

$$W(\theta,\varphi) \approx \frac{W_a}{2}(\varphi - \varphi_0)^2 + \frac{W_p}{2}(\theta - \theta_0)^2 \tag{1}$$

where W_a and W_p are the azimuthal and the polar anchoring energy coefficients, respectively. This difficulty can be overcome if the anchoring is weak[6] , if very high magnetic fields ($\approx$ 100 KG) are available[7] or if an electric field is applied to a NLC with a large dielectric anisotropy[8,9].

Some years ago[10] one of us investigated the behaviour of the azimuthal anchoring coefficient at the SiO_x-nematic interface by using a torsion pendulum technique. In this paper we study the azimuthal anchoring energy at the SiO_x-nematic interface by using a more sensitive reflectometric method. The dependence of the anchoring energy coefficient on the temperature and on the thickness of the SiO_x layer are investigated. The anchoring energy function is found to depend on the azimuthal angle as $W= W_a/2 \sin^2\varphi$.

EXPERIMENTAL METHOD

The reflectometric technique used to measure the azimuthal anchoring energy has been already described in detail in ref.[11].The NLC sample is sandwiched between two SiO_x-treated glass plates separated at the ends by two mylar spacers having different thickness (130 μm and 260 μm) in such a way to make a wedge. The glass plates too are wedge shaped with a wedge angle 0.4°. The NLC lies between the two polar expansions of a electromagnet giving magnetic fields up to 23.5 KG and its temperature is stabilized within 0.02°C. A 5 mW He-Ne laser beam is polarized by a polarizer and impinges at almost normal incidence (θ_i< 1°) on the NLC layer. The laser beam reflected from the first SiO_x-NLC interface passes through a crossed analyser and is collected by a photodetector (due to the wedge this beam can be easily separated from beams reflected by other interfaces). The ratio between the signal of this photodetector and that of a reference photodetector is measured to eliminate fluctuations of the laser intensity.

The thin SiO_x-layer is evaporated at the incidence angle of 60° on the glass plates of the cell to induce a planar homogeneous alignment of the director[12]. Most of the SiO_x layers are evaporated by using a molybdenum source having an aperture area of 4 x 10 mm (Balzers BD482048) at a distance of 40 cm from the glass plate. In some cases an evaporation source of smaller aperture (1 x 1 mm) was used. The residual pressure in the evaporation bell-jar was lower than 10^{-6} Torr and the evaporation rate was about 0.2 A/sec. The thickness of the SiO_x-layer was measured by a control quartz plate put close to the glass plate in the bell-jar. The NLC used for the experiments was pentylcyanobiphenil (5CB) purchased from BDH chemicals and used without further purification(T_c=35.3 °C). The homogeneous director alignment of the samples was controlled by conoscopy.

For a uniformly aligned nematic sample (monocrystal) the light intensity after the crossed analyser is given by:

$$I=I_0 \sin^2[2(\delta-\varphi)] \tag{2}$$

where δ is the angle between the polariser axis and an x-axis in the SiO_x plane and φ is the azimuthal angle between the director and the x-axis at the interface . I_0 can be measured by rotating the polarizer axis and measuring the maximum reflected intensity. This parameter is proportional to the square power of the anisotropy of refractive indices of the NLC and thus, it is almost proportional to the square power of the order parameter of the NLC. In effect I_0 measure a sort of average value $\overline{S_s}$ of the order parameter in a thin interfacial layer of thickness $\lambda/2\pi$ (λ = 6328 Å is the wavelength of the laser beam). Above the clearing temperature, I_0 abruptly reduces to about 1/200 of its value for T = T_c. This indicates that $\overline{S_s}$ in the isotropic phase becomes smaller than 0.02 in a good agreement with results obtained by using transmitted light methods[13,15] and in a complete disagreement with experimental results obtained by Mada et al.[16]. Mada et al. measured the reflectivity coefficients for the ordinary and for the extraordinary polarization of the laser beam. A large difference between these two coefficients was found also in the isotropic phase of the NLC corresponding to a residual order parameter at the surface $\overline{S_s} = .13$. We point out that our method is more direct than the Mada one, since the I_0 intensity directly measures the anisotropy of surface refractive indices.

In the presence of a magnetic field H, widely exceeding the Freedericks threshold value H_c ($H_c \sim 150$ Gauss for our samples), the director tends to align parallel to the magnetic field everywhere in the NLC but in a thin twisted layer close to the SiO_x-nematic interfaces. The balance of the anchoring and elastic surface torques for $H \gg H_c$ gives:

$$\frac{\partial W(\varphi)}{\partial \varphi} = \sqrt{K_{22}\chi_\alpha}\, H \sin(\beta - \varphi) \tag{3}$$

where β is the angle which the magnetic field makes with the easy axis x , K_{22} is the twist elastic constant and χ_α is the diamagnetic anisotropy. Therefore the anchoring torque $\partial W / \partial \varphi$ and thus, the azimuthal anchoring energy W (φ), can be obtained by measuring the surface director angle φ for different H values. The surface director angle φ is obtained by measuring the intensity of the reflected beam (see eq. 2). Corrections to eq.(2) due to the twist of the director near the interface produced by the magnetic field are negligible in our experimental conditions and their effect can be further reduced by using the experimental procedure discussed in ref.(11). Let's assume that the x-axis coincides with the easy axis at the SiO_x-nematic interface ($\varphi_o = 0$). For $\varphi \ll 1$, the anchoring energy function is given by eq.(1) and thus, eq.(3) becomes:

$$W_a = \frac{\sqrt{K_{22}\chi_\alpha}\; H \sin\beta}{\varphi} \tag{4}$$

We point out that <u>the measurement of the surface director orientation by using the reflectometric method does not require the knowledge of the optical refractive indices and of the magnetic and elastic constants of the nematic LC</u>. The very slow sensitivity of the reflectometric method to bulk effects makes this method very attractive for measurements of azimuthal anchoring energies with respect to transmitted light methods[17,18].

EXPERIMENTAL RESULTS

Fig.1a shows the typical time-variation of the intensity of the reflected beam between crossed polarizers (eq.2) for $\delta = 22.5^o$ when a magnetic field H = 7.5 KG is switched on at the time t=0 and switched off at the time $t = 4 \times 10^3$ sec. By using eq.(2) we obtains the corresponding time-variation of the azimuthal angle φ shown in fig 1b. One can distinguish two very different regimes: a fast relaxation characterized by the short time τ_f after which the director reaches an almost stationary value φ_1 and a slow relaxation characterized by the long time τ_l ($\approx$ 60 min) during which the director reaches the stationary value φ_2 ($\varphi_2 \approx 3/2\ \varphi_1$).

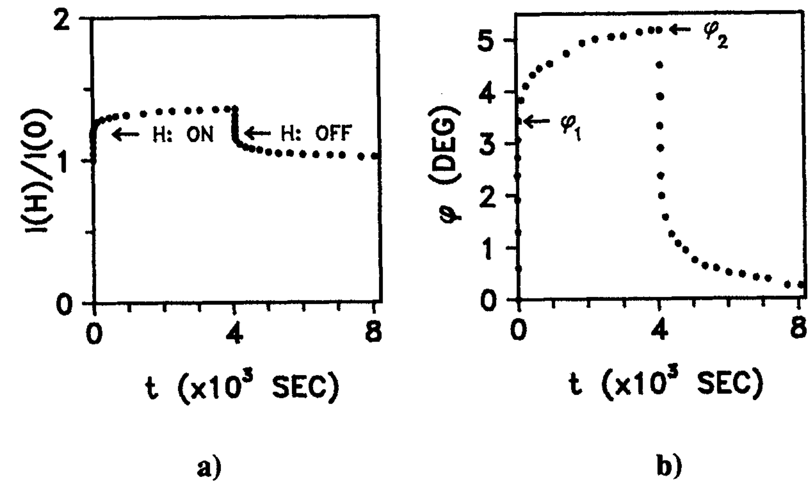

FIGURE 1: a) Time variation of the reflected light intensity between crossed polarizers; b) time variation of the azimuthal angle.

The magnetic field dependence of the short rise time is investigated by reducing the intensity of the magnetic field in such a way that τ_f largely exceeds the magnetic field rise time($\approx$ 2 sec). The rise time is almost proportional to $1/(H^2-H_c^2)$, diverges as H approaches the Freederickz threshold H_c and is of the same order of the bulk reorientation time τ_B ($\tau_B = \gamma/\chi_\alpha(H^2-H_c^2)$, with $\gamma \approx 1$ poise and $\chi_\alpha \approx 10^{-7}$ for 5CB). This indicates that the director surface orientation in the short time regime follows the bulk director reorientation, whilst the long time behaviour seems to be related to a new specific surface phenomenon. The long time tail of the reflected light intensity of fig.1a is not due to electronic or thermal drift effects, although these effects can give contributions up to 3% to the reflected intensity signal at longer times. As a matter of fact opposite long time variations of the reflected light intensity are found at the switching on and off of the magnetic field, respectively (see fig.1a). Furthermore the intensity signal always changes sign if we put the polarizer angle $\delta = -22.5^o$ in place of $\delta = 22.5^o$ according to the predictions of eq.(2) for a rotation of the director. However we point out that the same reflected light variation could be produced by two very different effects: i) a uniform rotation of the director everywhere within the lightned region of the NLC layer(width $\approx$ 1 mm); ii) a reorientation of microscopic domain-like regions near the interface having slightly different director orientations due, for instance, to the granular structure of the SiO_x-NLC interface. In this latter case, the long time variation of the azimuthal director angle of fig.1b could truly represent the time variation of the average director orientation within the lightned area. Measurements performed at different values of the magnetic field indicate that the long time behaviour is poorly affected by the intensity of the applied magnetic field. As a matter of fact no appreciable difference in the response of the director surface angle is found by reducing by a factor 1/4 the intensity of the magnetic field. Both the ratio φ_2/φ_1 and the duration of the long time regime do not change appreciably in this case. However we point out that long time drift effects make very difficult a quantitative investigation of the slow relaxation regime.Therefore the main effect of the reduction of the magnetic field consists on a rescaling by a factor $\approx$ 1/4 of the vertical scale of fig.2.

At the present time we are not able to explain the long time behaviour of the surface director azimuthal angle. The very long time associated with this process could be related to a slow reorganization of microscopic dishomogeneities on the surface director orientation and to the presence of somewhat activation energy barrier to be overcome more than that to a real director rotation. More detailed and systematic experiments need in order to explain this new behaviour. If the previous interpretation is correct, only the first transient is related to the director reorientation described by the anchoring energy of eq.(1). Therefore in the following we neglect this long time behaviour and we measure the anchoring energy coefficient by using eq.(4) where φ is substituted by the quasi-equilibrium value φ_1 (it is measured 60 sec after the switching on of the magnetic field). However, since the ratio φ_2/φ_1 is almost independent of the intensity of the applied field, the anchoring energy coefficient obtained by using φ_2 is almost proportional (by a factor 2/3) to the previous one.

Fig.2 shows the azimuthal anchoring energy coefficient W_a versus the temperature for a SiO_x layer having a thickness of 120 Å.

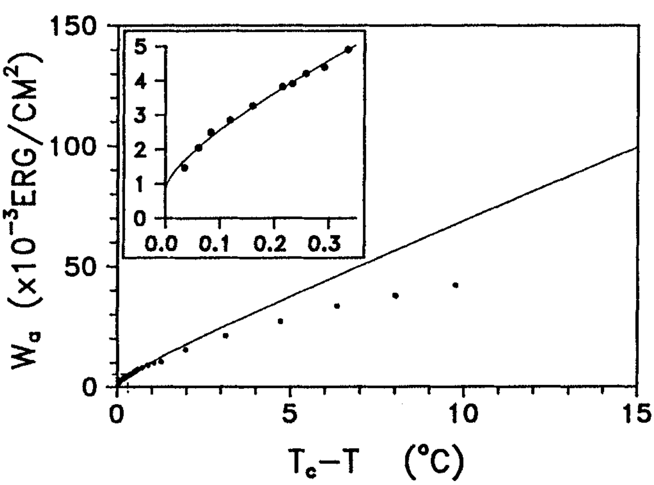

FIGURE 2 : Anchoring energy coefficient versus the temperature.

The full line corresponds to the best fit of the experimental results close to the clearing temperature (insert in fig. 2) to the theoretical expression[10]:

$$W_a = \left[\bar{a} + \bar{b}\,(T_c - T)^{1/2} \right]^2 \qquad (5)$$

where $\bar{a}$ and $\bar{b}$ are two phenomenological coefficients and T_c is the clearing temperature of the NLC. The values obtained by the best fit are 0.0275 $erg^{1/2}/cm$ and 0.073 $erg^{1/2}/cm\,^{o}C^{1/2}$, respectively. Eq.(5) holds very close to the clearing temperature and has been deduced in ref.(10) by making the assumption that the azimuthal anchoring is due to the elastic Berreman mechanism[19], but that the surface order parameter is lower than the bulk one according to the theory of Sluckin et al.[20] and to some experimental results[13,15]. According to Yokoyama et al.[21], also elastic anomalies due to a reduced surface order close to the interface can produce an analogous temperature dependence for both the polar and azimuthal anchoring coefficients.

The almost critical behaviour shown in fig.2 agrees with that already obtained by our group by using the torsion pendulum technique although the present measurements are more accurate due to the better accuracy and sensitivity of the reflectometric method. In particular this higher sensitivity allows us to measure the anchoring energy coefficient in the whole range of existence of the anisotropic phase. Measurements of the temperature dependence of the anchoring energy coefficient have been repeated for different thicknesses d of the SiO_x layer and the same critical behaviour has been observed for all thicknesses of the SiO_x-layer. Unfortunately a quantitative comparison between the present experimental results and those obtained with the torsion pendulum technique[10] cannot be made because the thickness of the SiO_x layer used for the torsion pendulum measurements was not measured. Notice that in ref.(10) a slightly different definition of the anchoring coefficient was used.

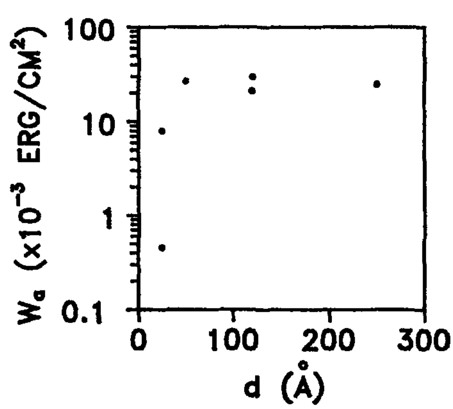

FIGURE 3 : Azimuthal anchoring energy versus the thickness of SiO_x.

Fig.3 shows the azimuthal anchoring energy coefficient at a given temperature (T_c -T = 3°C) versus the thickness d of the SiO_x layer. We see that the anchoring energy is almost independent of the thickness but abruptly decreases when the thickness becomes lower than 25 Å. Samples having thicknesses of 25 and 23 Å exhibit a not uniform director orientation ; some macroscopic well aligned regions(≈ 20 mm^2 area) are separated by disordered regions. In these samples the anchoring energy coefficients in different aligned regions can differ by a factor greater than two.

The measurements of azimuthal anchoring energy at the SiO_x-nematic interface are complicated by the presence of degradation effects of the interface. In particular the anchoring energy coefficient at a given temperature often tends (but not always) to become weaker with time(variations up to a factor two have been found sometimes after some weeks). By looking at the temperature dependence of the anchoring energy coefficient we find that the $\bar{b}$ coefficient of eq.(5) remains practically unchanged with time whilst the coefficient $\bar{a}$ can decrease appreciably. According to our model of the azimuthal anchoring[10] and that of Yokoyama et al.[21] this behaviour seems to indicate that, in some cases, the surface order parameter at the SiO_x-nematic interface progressively reduces with time. We have also performed some measurements with SiO_x-layers evaporated by using a molybdenum source of smaller aperture (≈ 1 mm^2). In this case, too, the anchoring energy exhibits the almost-critical behaviour of fig.2. Preliminary results indicate that the coefficient $\bar{a}$ of eq.(5) is much larger (≈ ten times) than the corresponding value obtained by using the larger source whilst the coefficients $\bar{b}$ are of the same order of magnitude. According to our theory this could be explained by assuming that the surface order parameter is much greater in these latter samples. Direct measurements of the surface order parameter in these cases should be very important.

As shown in fig.3 the anchoring energy for the samples of 25 and 23 Å is weak. This allows us to investigate the dependence of the azimuthal anchoring energy on the azimuthal angle φ far from the easy angle $\varphi_0=0$. Fig. 4a shows the azimuthal angle of the director at the interface versus the magnetic field H for some values of the angle β between the magnetic field and the easy axis at the interface for a SiO_x-layer of thickness d= 25 Å and at the temperature T =35.16 °C (T_c-T =0.14 °C). The estimated accuracy on the measurement of the angle β is ± 1°. As shown in fig. 4a the director at the surface tends to become parallel to the magnetic field at higher magnetic field values. Theory predicts a sharp transition to a complete reorientation of the director along the magnetic field for β=90° and for a magnetic field greater than a critical saturation value. If the φ-dependence of the azimuthal anchoring energy is assumed to be :

$$W(\varphi) = \frac{W_a}{2}\sin(\varphi)^2 \qquad (6)$$

then the critical saturation magnetic field[20] is $H'_c = W_a/\sqrt{K_{22}\chi_\alpha}$.
Unfortunately when the angle β becomes very close to 90° (within about 1°), a lot of small regions having opposite twists of the director-field occur in the lightened region of the layer. These regions do not relax to a

homogeneous twist and thus, the critical transition for $\beta=90^{\circ}$ cannot be investigated. However a clear pretransitional behaviour can be seen by looking at the experimental points of fig.4a for $\beta=89^{\circ}$(upper curve).

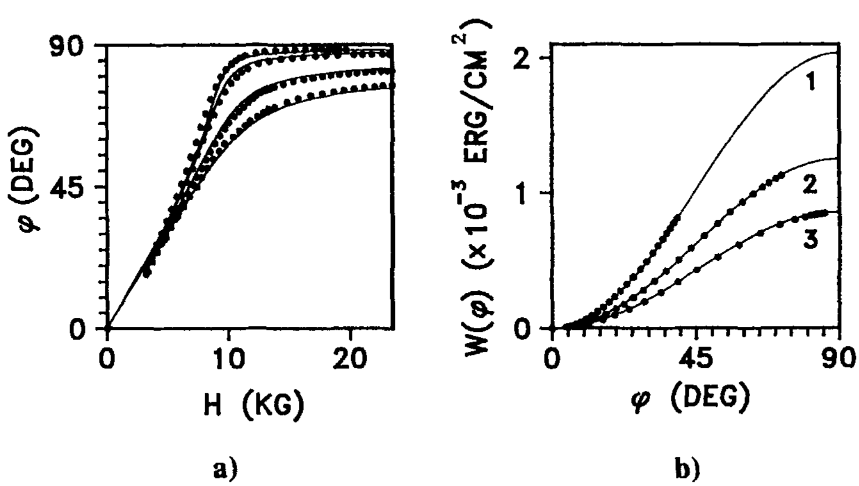

FIGURE 4 : a) Azimuthal angle φ versus the magnetic field H for some values of the angle β between the m.f. and the easy axis at the temperature T=35.16 ^{o}C. In order from bottom to the top : β= 81.2^{o}, β=85.3^{o}, β =88.4^{o}, β =89^{o}. b) Anchoring function versus the azimuthal angle for some temperatures : (1) T = 34.95^{o}C, (2) T = 35.11^{o}C ,(3) T= 35.16^{o}C.

The full lines correspond to the best fit of experimental results to the theoretical predictions for the anchoring function of eq.(6). Small differences between theory and experiment can be explained as due to the uncertainty of the value of the β-angle ($\Delta\beta \approx 1^{o}$). As a matter of fact a much better agreement is found by making the fit with the β angle as a free parameter. Measurements performed with β very close to 90^{o} allow us to investigate the behaviour of the azimuthal anchoring energy function in a large range of φ-values. The experimental procedure consists of measuring the φ-angle as a function of H as shown in fig.4a. Then the corresponding value of the azimuthal anchoring torque is determined by substituting φ , H and β in the r.h.s of eq.(3) and by using the known values of the elastic and magnetic constants of the NLC [22,23]. Then the anchoring function W(φ) is obtained by numerical integration of the anchoring torque $\partial W/\partial\varphi$. Fig. 4b shows the experimental value of the anchoring energy function W(φ) for three different values of the temperature close to the clearing value. Full lines in fig. 4b correspond to the best fits of the experimental results with respect to the anchoring energy function of eq.(6). The experiment clearly shows that, in spite of what is found in the case of polar anchoring at the same kind of interface[8] and at other interfaces[6,7], the azimuthal anchoring is well described by the Rapini potential of eq.(6). Note that, for polar anchoring, Barbero et.al. [24] showed that corrections to the Rapini form found in the experiments could not be related to intrinsic surface phenomena, but to flexoelectricity and ordoelectricity. Both these effects do not affect the azimuthal anchoring measurements.

CONCLUSIVE REMARKS

The almost-critical behaviour of the anchoring energy coefficient which was been previously observed close to the clearing temperature of the NLC by a surface pendulum technique has been confirmed by reflectometric measurements. The higher sensitivity and accuracy of the reflectometric method allows us to measure the temperature dependence of the anchoring energy in the whole temperature range of the NLC and for different values of the thickness d of the SiO_x-layer. Reflectometric measurements evidence for the first time an unusual relaxation behaviour of the azimuthal director angle characterized by two very different times. Finally the shape of the azimuthal anchoring function $W(\varphi)$ is investigated and is found to be well represented by a sine square dependence in spite of what is found for the polar anchoring.

The azimuthal anchoring energy of an NLC at the SiO_x- nematic interface is usually interpreted in terms of the Berreman mechanism[15] for the anchoring at a grooved interface. In fact the experimental observation of the SiO_x surface by using electron microscopy clearly indicates that the SiO_x surface is not flat but consists of a periodic pattern of parallel grooves with an average alignment along a given x-axis. According to the Berreman model, the elastic interactions between NLC molecules near the interface favour the director alignment parallel to the x-axis. By making the simplifying assumptions that the elastic constants of the NLC have the same values ($K_{11}=K_{22}=K_{33}$) and the local tilt angle of the SiO_x surface is small, Berreman obtained just eq.(6) with the azimuthal anchoring coefficient proportional to the twist elastic constant of the nematic LC. The Berreman model has been recently generalized in order to account for the presence of a finite value of the polar anchoring energy and of a reduced surface nematic order near the interface[10,25]. Both these contributions greatly affect the value of the azimuthal anchoring coefficient but not the φ-dependence of the anchoring energy. Therefore our experimental results of fig. 4b agree with the theoretical predictions of the modified Berreman elastic model.

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