Comment on "Rayleigh Scattering from Column Undulations in a Discotic Liquid Crystal"

In an interesting and stimulating Letter [1], the results of Rayleigh scattering on columnar discotics were described and it was found that at long wavelengths as probed by Rayleigh scattering in the visible domain the studied columnar phase reacted like an elastic solid. In Ref. [1] this behavior was attributed to column entanglements and defects in the tubular structure.

Here we point out that this question could not be addressed by the measurements described in Ref. [1], since the columnar phase of the compound studied in Ref. [1] (C8HET) is well known [2] to be of the D_{ho} type. These phases show not only positional order *of* the columns (as is characteristic for columnar phases), but also positional order *within* the columns. This was demonstrated by high-resolution x-ray diffraction experiments [3] (on a similar compound) where it was found that all reflections in the D_{ho} phase were resolution limited; i.e., the D_{ho} phase in fact shows true three-dimensional long-range positional order.

Thus it is clear that D_{ho} has the elasticity of a uniaxial solid [4] and this, indeed, was seen in Ref. [1]. Its elastic energy takes the form [we take z to be the preferred direction parallel to the columns and the displacement vector $\mathbf{u}=(u,v,w)$]

$$
\begin{aligned}
2F_{\mathrm{el}} = \int d\tau \Bigg\{ & c_1 \left[\left(\frac{\partial u}{\partial x} \right)^2 + \left(\frac{\partial v}{\partial y} \right)^2 \right] + c_2 \left(\frac{\partial w}{\partial z} \right)^2 + c_3 \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \frac{\partial w}{\partial z} + c_4 \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right)^2 \\
& + c_5 \left[\left(\frac{\partial u}{\partial z} + \frac{\partial w}{\partial x} \right)^2 + \left(\frac{\partial v}{\partial z} + \frac{\partial w}{\partial y} \right)^2 \right] + K_1 \left[\left(\frac{\partial^2 u}{\partial z \partial x} + \frac{\partial^2 v}{\partial z \partial y} \right)^2 \right] \\
& + K_2 \left[\left(\frac{\partial^2 v}{\partial z \partial x} - \frac{\partial^2 u}{\partial z \partial y} \right)^2 \right] + K_3 \left[\left(\frac{\partial^2 u}{\partial z^2} \right)^2 + \left(\frac{\partial^2 v}{\partial z^2} \right)^2 \right] \Bigg\}, \qquad (1)
\end{aligned}
$$

where we have also listed the curvature terms proportional to K_1, K_2, and K_3, which contain higher-order derivatives of the displacement field [5]. In the case of Rayleigh scattering with visible light one probes the long-wavelength, i.e., small-wave-vector, limit. In this limit the curvature terms are of higher order and the elastic terms dominate in accordance with the observations in Ref. [1]. Since D_{ho} is a comparatively soft solid, however, it is appropriate to keep in (1) the curvature terms such as, e.g., bend, the contribution $\propto K_3$, since those can be expected to be of importance for larger deformations (in which case also nonlinear terms will come into play in general).

When going from the ordered to the disordered columnar discotic phase, D_{hd}, however, the picture changes qualitatively. The x-ray experiments [3] show that there is only liquidlike ordering within the columns and thus the displacement along the axis of the columns is no longer a hydrodynamic variable [6]. Therefore, the contributions proportional to c_2, c_3, and c_5 do not exist and the curvature terms given in Eq. (1) constitute the lowest-order terms describing spatial variations along the axis of the columns. In this phase, D_{hd}, Rayleigh scattering is expected to give qualitatively different results from a 3D crystal, except for the possibility that entanglements and defects in the tubular texture (discussed by Prost in Ref. [10] of Ref. [1]) could be of considerable importance.

Therefore we conclude that the experiments of Ref. [1] carried out in the D_{ho} phase are in full agreement with 3D elasticity as expected from previous high-resolution x-ray diffraction data [3]. The issue of entanglements and defects in the tubular texture, however, could be addressed for the disordered columnar phase D_{hd} for which experiments using Rayleigh scattering therefore seem highly desirable.

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