Polar correlations in nematics probed by second harmonic light scattering

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Second harmonic generation (SHG) is a powerful probe of non-centrosymmetric/polar structure in condensed matter. In the conventional (centrosymmetric) nematics, short-range polar correlations can arise due to either static or fluctuating distortions of the director (inducing a flexoelectric polarization) \cite{1-3} or, at least in principle, due to short-range order associated with a polar (vector) or other non-centrosymmetric order parameter. The first of these gives rise only to a weak SH signal, which is incoherent and distributed in a pattern of scattered light that (in contrast to linear scattering from director fluctuations) depends weakly on scattering angle \cite{2,3}. On the other hand, static distortions of the director associated with strong surface anchoring effects may generate substantially higher levels of coherent SHG \cite{1}.

Here we report on experiments using an improved version of the traditional high-sensitivity SHG technique, which we use to characterize short-range polar structure in bulk (thick) nematics with no special surface treatments. Our approach combines a specialized optical telescope and a sensitive, low noise CCD detector to produce a light collection/detection system capable of resolving the pattern of SH light propagating within a large solid angle around the incident direction of the fundamental light. The incident and scattered (SH) polarization can be independently varied from parallel (H) to normal (V) with respect to the plane of the magnetically-aligned director and the incident light.

We compare results from three nematic compounds: a polar, bent-core molecule, a polar calamitic molecule (5CB), and a nonpolar calamitic (used chiefly as a control). On field-aligned bulk samples of the polar molecules, we observe the following features: (1) a symmetric pair of arcs of transmitted SH light with maxima centered along the director at small angles ($\pm \theta$) from the incident light direction; (2) continuously increasing angle $\theta$ with decreasing temperature (in a manner qualitatively similar to the order parameter); (3) significant SH signal for the polarizer/analyzer combinations HV and VV but negligible signal for HH and VH. The signal from the nonpolar nematic compound is barely above background and is polarization insensitive. We describe a model that accounts for the unusual angular pattern of SH light and its temperature and optical polarization dependence in the nematic phase of the polar compounds. We also discuss how our method could prove useful as a tool to characterize and compare polar correlations among a wider range of nematics.

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References:

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