

2-9-2011

Comment on "Raman Scattering Study of Phase Biaxiality in a Thermotropic Bent-Core Nematic Liquid Crystal" Reply

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Recommended Citation

Acharya, Bharat R.; Choi, Hyunchul; Srinivasarao, Mohan; and Kumar, Satyendra (2011). Comment on "Raman Scattering Study of Phase Biaxiality in a Thermotropic Bent-Core Nematic Liquid Crystal" Reply. *Physical Review Letters* 107(10). doi: 10.1103/PhysRevLett.107.109802 Retrieved from <https://digitalcommons.kent.edu/phyypubs/20>

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Comment on “Raman Scattering Study of Phase Biaxiality in a Thermotropic Bent-Core Nematic Liquid Crystal”

Recently, Park *et al.* [1] reported their use of polarized Raman scattering to measure biaxial order parameters [2,3] in the nematic phases of a bent-core liquid crystal. Their work closely follows our approach [2] that deduced uniaxial and biaxial order parameters for the oxydiazole compound C5-Ph-ODBP-Ph-OC12. However, they consider a different material (A131) and their analysis does not take molecular bend into account. Park *et al.* report a second-order uniaxial-biaxial transition via analysis of the order parameters and unusually low values of the uniaxial order which they attribute to fluctuations relating to the biaxial phase.

That Park *et al.* did not consider molecular bend, which is how biaxiality is included in the Raman analysis for bent-core systems [2], has significant implications for their data. Their neglect of molecular bend offers a straightforward explanation for their low P_{200} and P_{400} order parameter values. Figure 1 compares P_{200} and P_{400} for C5-Ph-ODBP-Ph-OC12 [2] by using two different data analysis models. Systematically lower values result from the model that takes no account of molecular bend, the approach followed by Park *et al.* We recently reported P_{200} and P_{400} for six oxydiazole compounds [4], taking account of molecular bend. Three of the materials in Ref. [4] have a reported biaxial nematic phase [2,5,6]—none show unusual behavior of P_{200} and P_{400} ; indeed, the parameters reported are in excellent agreement with theory.

Park *et al.* are mistaken in stating that our biaxial order parameters [2] do not agree with theory. The relevant theoretical limits are $|P_{220}| \leq 0.25$, $|P_{420}| \leq 0.0536$, and $|P_{440}| \leq 0.0625$. Our measurements have two sources of uncertainty, discussed in detail in Ref. [2]: those due to the measurement (± 0.03) and, importantly, an *additional* uncertainty of ± 0.02 due to uncertainty in the molecular bend angle. In considering the combined uncertainty, only one of our biaxial order parameter data points ($\sim 5\%$ of our data) falls outside 2 standard deviations of the limit in the biaxial regime and 90% are within 1 standard deviation. It is clear that our data are in excellent agreement with the theoretical limits.

We reported [3] that while one of the biaxial parameters, specifically P_{440} , may take nonzero values in the uniaxial phase, both P_{220} and P_{420} *must* be strictly zero. However, more than half the values for P_{220} in Ref. [1] are nonzero in the uniaxial phase (outside 2 standard deviations). No physical explanation is offered for the origin or meaning of these nonzero P_{220} values in the uniaxial regime. It is difficult to attribute such significant nonzero values of P_{220} to a second-order uniaxial-biaxial phase transition since it is reasonable to expect P_{220} to be zero in the uniaxial phase, growing uniformly in the biaxial phase, as seen in our work [2].

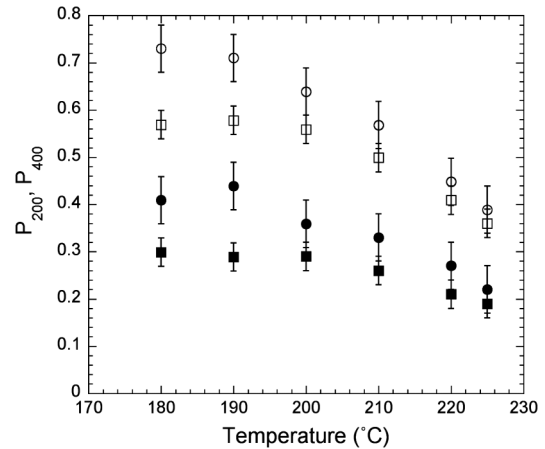


FIG. 1. P_{200} (open symbols) and P_{400} (filled symbols) for C5-Ph-ODBP-Ph-OC12. The circles result from a model that includes a 140° molecular bend angle (two scatterers) as in Ref. [2]; the squares are the systematically lower values obtained when the bend angle is not included.

Polarized Raman scattering is clearly an important methodology in studying biaxial order in liquid crystals. However, any approach that requires an appropriate model and employs fitting parameters with interdependencies requires extreme care in interpreting unusual behavior in the resulting data. Park *et al.*'s nonzero P_{220} values in the uniaxial regime are unphysical, and anomalously low P_{200} and P_{400} values are not observed in analogous systems. We believe their data should be reanalyzed with rigorous consideration of the influence of the bend angle before their interpretation of low uniaxial order parameters due to fluctuations can be realistically suggested.

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Received 28 July 2010; published 2 September 2011

DOI: 10.1103/PhysRevLett.107.109801

PACS numbers: 61.30.-v

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