Multimodal nonlinear optical polarizing microscopy of long-range molecular order in liquid crystals

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We demonstrate orientation-sensitive multimodal nonlinear optical polarizing microscopy capable of probing orientational, polar, and biaxial features of mesomorphic ordering in soft matter. This technique achieves simultaneous imaging in broadband coherent anti-Stokes Raman scattering, multiphoton excitation fluorescence, and multiharmonic generation polarizing microscopy modes and is based on the use of a single femtosecond laser and a photonic crystal fiber as sources of the probing light. We show the viability of this technique for mapping of three-dimensional patterns of molecular orientations and show that images obtained in different microscopy modes are consistent with each other.

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Soft matter systems such as liquid crystals (LCs) exhibit polymesomorphism of phase behavior combined with varying degrees of orientational and positional ordering intermediate between that of isotropic fluids and crystalline solids [1]. The long-range orientational order is a salient feature of these systems that results in an unprecedented richness of ground-state structures and textural behavior associated with the uniform alignment of molecules on the scale of nanometers and their slowly varying patterns on the scale of micrometers. In uniaxial nematic LCs, local average molecular orientations are described by the director field $\mathbf{n}(\mathbf{r})$, which is also the optical axis. LC ordering can be polar or nonpolar, uniaxial or biaxial and with varying degrees of positional ordering. There

ferroelectric SmC* (Felix 015-100), SmA (8CB, 4-cyano-4-octylbiphenyl), or cholesteric obtained by mixing 5CB (4-cyano-4-pentylbiphenyl) with a chiral dopant (cholesteryl pelargonate, Sigma-Aldrich). All LCs were obtained from EM Chemicals. For CARS-PM imaging, we chose the CN stretching vibration (2236 cm^{-1}) of 8CB and 5CB molecules parallel to the long molecular axis.

Figure 2 shows three-photon excitation fluorescence (3PF) and SHG forward-detection images of SmC* in an untreated cell obtained using a 1050 nm excitation pulse (∼1

second-order nonlinear processes, whereas CARS-PM, 3PF, and third-harmonic generation are third-order