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Nonlinear Optics in Chiral Media

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Chiral molecules play an important role in biology, chemistry, and medicine. Conventional techniques to probe molecular chirality, such as circular dichroism and optical rotation, are electric dipole forbidden. They have limited sensitivity and hence difficulty to detect chirality from a monolayer or thin film that is important in a chemical or biological system. Recently, second-harmonic generation (SHG) and sum-frequency generation (SFG) have been considered potentially useful for probing chirality. As second-order nonlinear optical processes, they are allowed under the electric-dipole approximation in media without inversion symmetry, which is the case for all chiral materials. They also have the sensitivity to probe a monolayer or submonolayer. In contrast to linear optical activity, SHG and SFG have the advantage that both chiral and achiral nonlinear polarizability elements are electric dipole allowed. This should make the corresponding nonlinear optical activity easy to detect.

In this paper, we have studied chiral responses in optical sum-frequency generation (Fig. 1) near electronic resonances from surface monolayer and from bulk solution of 1,1′-bi-2-naphthol (BN)[1,2]. As shown in Fig. 2, the molecule is composed of two linked 2-naphthol monomers and has C_2-symmetry. Steric repulsion of the two monomers leads to a twist of 100° between the two, which makes the BN molecule chiral. The polarization dependence of the spectra from the surface monolayer indicates that the molecules are well oriented at the surface with their symmetry axis along the surface normal. The strengths of the resonances, in comparison with the corresponding in bulk solution can be quantitatively understood with the help of a coupled-oscillator model for BN. Orientational ordering effectively enhances the chiral response per BN molecule.

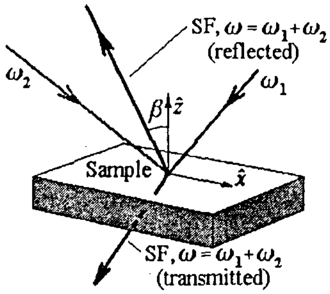

Fig. 1. Experimental arrangement.

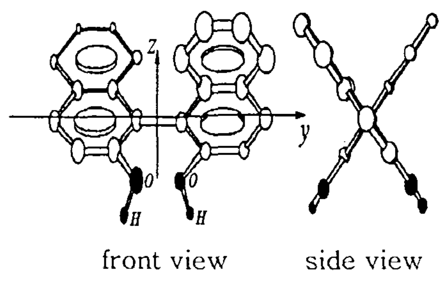

Fig. 2. Structure of the BN molecule.

And also we have measured optically active second-harmonic generation from a chiral liquid crystal (LC)[3]. SHG is a special case of SFG. It has been noted that because of degeneracy of the input frequencies, SHG is forbidden in an isotropic chiral liquid, even though SHG from the chiral response of a molecular monolayer can be detected. From symmetry consideration, it can be shown that if in a fluid medium, the chiral molecules are oriented uniaxially, chiral (optically active) SHG can become allowed. This could happen in nematic and smectic-A phases of chiral LC and in flow-aligned chiral molecular liquids. Here, we report the first study of the process using a chiral smectic-A LC as the uniaxial fluid medium. In our experiment, the LC material used was *o*-nitroalkoxyphenyl biphenylcarboxylate (W314). We were able to observe phase matching of chiral SHG in the medium and deduce the chiral nonlinear susceptibility from the results (Fig. 3). The SHG output vanished when the LC moved into the isotropic liquid as expected (Fig. 4). This work provides a method for us to measure chiral nonlinearity of chiral LC molecules, or more generally, chiral molecules that can be uniaxially aligned in a fluid medium.

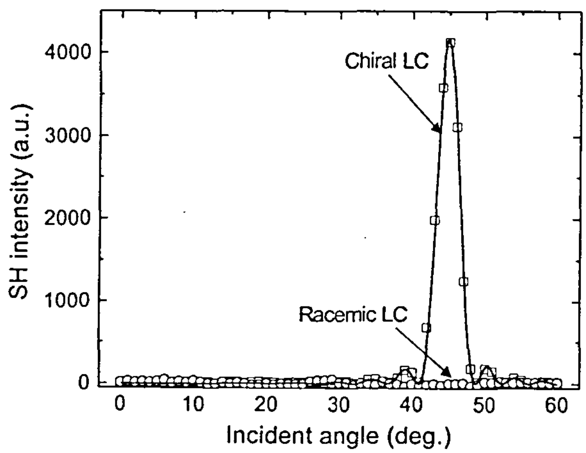

Fig. 3. Angular phase matching dependence of W314 as a function of external incident angle.

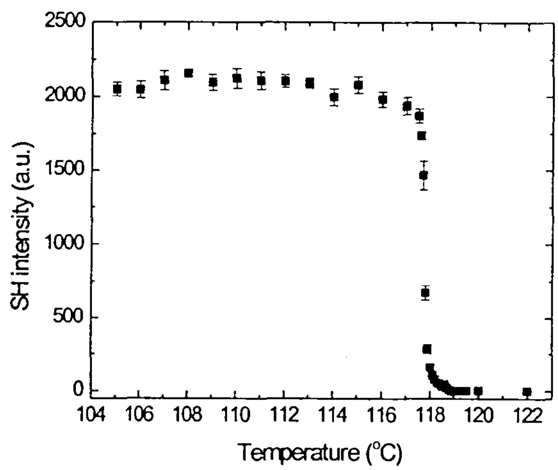

Fig. 4. Temperature dependent SHG of W314 from smectic-A phase to isotropic at the phase matching angle.

References

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3. S. H. Han and Y. R. Shen, "Chiral nonlinear optical response of a chiral liquid crystal by second-harmonic generation" , Opt. Lett. Submitted (2003).