Investigation of Sum-Frequency Generation of Chiral Molecules in Solutions

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Abstract

A main purpose of this paper is to examine the origin and mechanism of non-linear optical activity in solution. For this purpose, within the dipole-dipole formalism of SFG, we study the non-Condon effect and the effect of the breakdown of the Born-Oppenheimer approximation. We find that in order to determine which mechanism is responsible for this SFG it is necessary to compare the SFG band-shape functions on surfaces and in solutions.

Quite recently, visible-visible Sum Frequency Generation (SFG) signals from 1,1'-Bi-2-naphthol (BN) in a bulk BN solution have been observed. [1,2] To understand the origin of the SFG activity of BN in solution, we shall theoretically examine a SFG mechanism of chiral molecules in a bulk solution. For this purpose, we shall employ the Herzberg-Teller theory to study the non-Condon effect; we shall also study the effect of the breakdown of the Born-Oppenheimer adiabatic approximation on this nonlinear optical activity in solution. The SFG beyond the dipole-dipole approximation like the dipole-magnetic dipole approximation and the magnetic dipole-magnetic dipole approximation will also be explored in this paper.

[1] M. A. Belkin, S. H. Hau, X. Wei, and Y. R. Shen, Phys. Rev. Lett. **87**, art. no. 113001 (2001).

[2] P. Fischer, K. Beckwitt, F. W. Wise, A. C. Albrecht Chem. Phys. Lett. **352**, 463 (2002).