Molecular orientation in Langmuir monolayers studied by surface second harmonic generation

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Introduction
We combined surface second harmonic generation (SHG) with depolarized light microscope to study molecular orientation in a smectic-C liquid-crystal Langmuir monolayer at an air-water interface. The molecules under investigation are 4-COOH-4'-carboxytrimenthyleneoxy)azoazobenzene (8AZ3). Monolayer exhibits a first-order phase transition from liquid expanded (LE) to liquid condensed phase (LC) with increasing surface pressure.

Depolarized reflected light microscope (DRLM)

In the DRLM setup, polarized light is incident onto the monolayer almost normal to the surface. The reflected light is imaged through a ×-polarized analyzer, which allows visualization of the anisotropy of the monolayer.

Surface second-harmonic generation (SHG)

As a second-order nonlinear process, optical SHG is highly surface specific. The polarization relates to the field as

\[ P_{2}(x,y,z) = \chi_{ij}^{(2)} E_{i}(x,y,z) E_{j}(x,y,z) \]

The first examples of SHG for Langmuir monolayers were reported in 1978 by Decker and coworkers. In our work, we used a He-Ne laser (632.8 nm) to generate SHG from LCs. The wave vector of the incident light is the same as the wave vector of the SHG light. The electric fields of the fundamental and second harmonic waves are parallel to the monolayer plane.

Results

We measured SHG as a function of Φ for different polarization combinations. By fitting the data with the theoretical model, we deduce all nonvanishing components of \( \chi \), and then determine the parameters related to molecular orientation.

For 8AZ3, the average tilt angle at the liquid condensed phase is ~65°. For the liquid expanded phase, it is ~59°.

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