Supporting Information for

Significant Chiral Signal Amplification of Langmuir Monolayers Probed by Second Harmonic Generation

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Experimental Setup: In this work, a mode-locked femtosecond Ti/ sapphire laser (Tsunami 3960C, Spectra-Physics) with a repetition rate of 82 MHz and a pulse width of 80 fs was used for the SHG-LD measurements. The incident light wavelength was 800nm and the incident angle was 70° with a laser power of 600 mW or 150 mW. The polarized light was focused around a 30-50 μm diameter spot at the air/water interface. The incident polarization angle was adjusted using a rotating half-wave plate driven by a computer-controlled stepper motor. SHG signals were collected with a high-gain photomultiplier tube (Hamamatsu) and single photon counting system (SR400, Stanford Reasearch System) at different incident polarization angles. The supporting stage below the trough could be translated, and we chose several positions of the monolayer to detect the SHG-LD signal. In this way, we were able to obtain dependence curves of the relationship between SHG intensity and the incident polarization angle for different positions of the monolayer and at different times. The SHG-LD experimental setup was similar to that previously reported.[1-2]

Chemicals: TyrC18 and TrpC18 were synthesized and purified according to the procedure in the literature.[3-4] Chloroform solution of 0.5 mM was used for spreading. We spread 10μL on the subphase in a Teflon trough with an area of 80 cm^2. Before detection, the solvent was allowed to evaprate from the air/water interface for more than 15 min to allow the molecules on the surface forming the monolayer and the monolayer to achieve thermodynamic equilibrium by means of free diffusion and intermolecular interactions.

False Chirality Discussion: The differences between the SHG signals do not necessarily indicate that the monolayer is chiral because in-plane anisotropy also brings about similar differences in SHG-LD experiments. This is called false chirality. In plane anisotropy, which can be classified into C_{1h}, C_{3v} and C_{2v} symmetry, may give rise to false chirality in SHG-LD experiments.[5] C_{1h} or C_{3v} symmetry leads to nonzero s-in/s-out signal, and this is not case in our experiments. Now consider the C_{2v} symmetry. Let δ_{45s} be the variation of $\chi_{eff,45s}$ at different positions and δ_{chiral} be the variation of $\chi_{eff,chiral}$. If the chiral signals originated from C_{2v} anisotropy, the ratio $\delta_{45s}/\delta_{chiral}$ would have been 1.46.[2] The fitting results of $\chi_{eff,45s}$ and $\chi_{eff,chiral}$ at different positions of L-Tyr/TPPS monolayer are listed in

Table S1. It can be seen that δ_{chiral} is larger than δ_{45s}, indicating no C_{2v} anisotropy or false chirality.

Table S1 Fitting results of $\chi_{eff,45s}$ and $\chi_{eff,chiral}$ at different positions of L-Tyr/TPPS monolayer

Position	1	2	3	4	5
$\chi_{eff,45s}$	29.55 ±1.89	28.43 ±1.64	28.38 ±1.81	27.72 ±1.76	26.27 ±2.03
$\chi_{eff,chiral}$	55.16 ±1.46	51.61 ±1.28	55.77 ±1.34	59.43 ±1.23	58.30 ±1.38

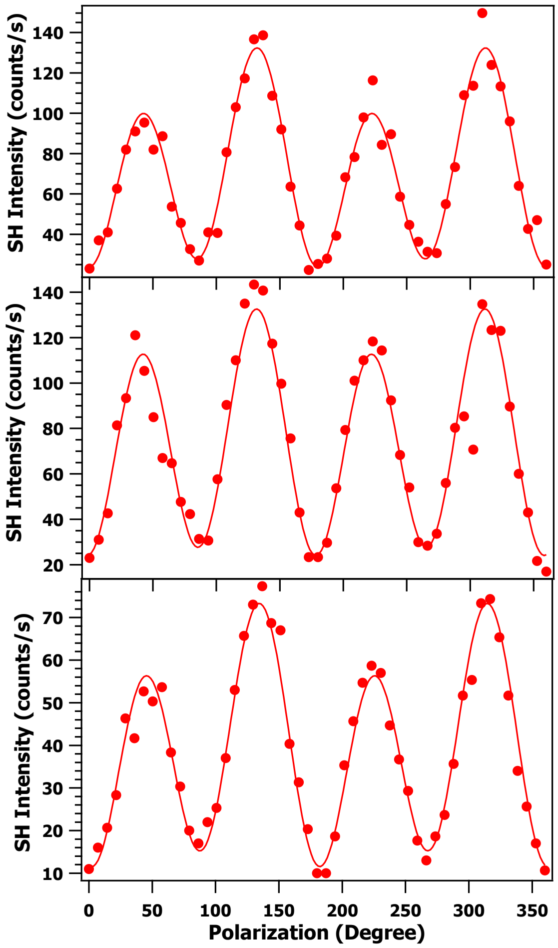

Figure 1S. Dependence curves of the SHG intensity versus the incident polarization angle for L-TyrC18 (up), D-TyrC18 (middle) and rac-TyrC18 (down) monolayers on the pH=5.4 TPPS solution subphase fitted using eq 1. The circles indicate experimental values and the solid curve is the fitted result. The laser power for the pH=5.4 TPPS solution subphase was 600 mW.

Reference:

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