

Supporting Information

Derivation of compact formulae for linear and circular dichroic ratios in second harmonic generation.

Linear Dichroism. The linear dichroic ratio is defined to be the following.

$$R^{LD} = \frac{I_{-45}^{2\omega} - I_{45}^{2\omega}}{\frac{1}{2}(I_{-45}^{2\omega} + I_{45}^{2\omega})} = 2 \frac{|F+G|^2 - |-F+G|^2}{|F+G|^2 + |-F+G|^2} \quad (S1)$$

It is useful to separate the complex-valued parameters F and G into real and imaginary components (indicated by the subscripts), such that $F = F_R + iF_I$ and $G = G_R + iG_I$. Equation S1 can be expanded to the following.

$$R^{LD} = 2 \frac{(F_R + G_R)^2 + (F_I + G_I)^2 - (-F_R + G_R)^2 - (-F_I + G_I)^2}{(F_R + G_R)^2 + (F_I + G_I)^2 + (-F_R + G_R)^2 + (-F_I + G_I)^2} \quad (S2)$$

$$R^{LD} = 2 \frac{4(F_R G_R + F_I G_I)}{2(F_R^2 + F_I^2) + 2(G_R^2 + G_I^2)} \quad (S3)$$

The numerator can be simplified further to yield the following compact relation.

$$R^{LD} = 4 \frac{\text{Re}(FG^*)}{|F|^2 + |G|^2} \quad (S4)$$

Circular Dichroism. Derivation of the complementary equation for circular dichroism given in Eq. 1 of the manuscript has been presented previously,^{1,7,45,46} but is reproduced here for completeness.

$$R^{CD} = \frac{I_L^{2\omega} - I_R^{2\omega}}{\frac{1}{2}(I_L^{2\omega} + I_R^{2\omega})} = 2 \frac{|-iF+G|^2 - |iF+G|^2}{|-iF+G|^2 + |iF+G|^2} \quad (S5)$$

Separating the complex-valued parameters F and G into real and imaginary components yields the following expression.

$$R^{CD} = 2 \frac{(F_I + G_R)^2 + (-F_R + G_I)^2 - (-F_I + G_R)^2 - (F_R + G_I)^2}{(F_I + G_R)^2 + (-F_R + G_I)^2 + (-F_I + G_R)^2 + (F_R + G_I)^2} \quad (S6)$$

$$R^{CD} = 2 \frac{4(F_I G_R - F_R G_I)}{2(F_R^2 + F_I^2) + 2(G_R^2 + G_I^2)} \quad (S7)$$

The numerator can be simplified further.

$$R^{CD} = 4 \frac{\text{Im}(FG^*)}{|F|^2 + |G|^2} \quad (S8)$$

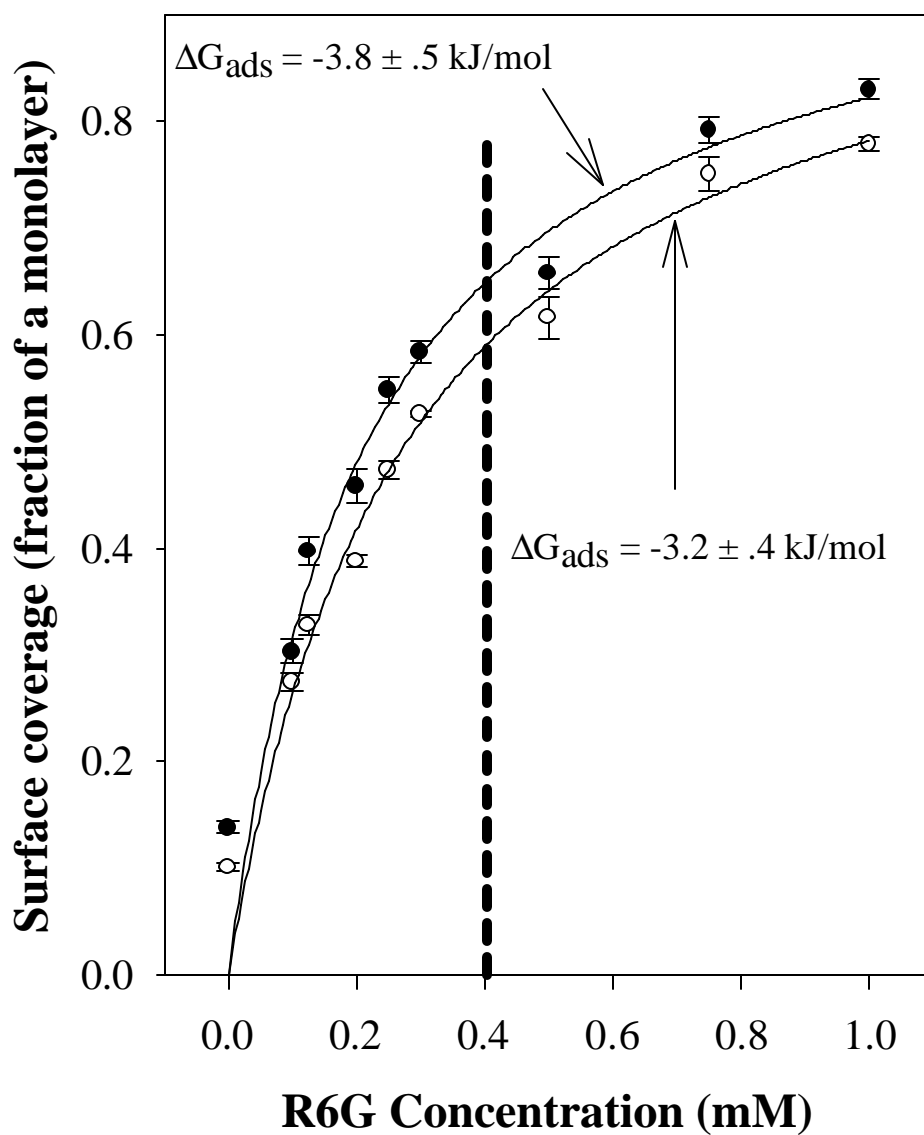


Figure S1. Adsorption isotherms for R6G at the glass / aqueous solution interface (open circles) and R6G at the glass / BSA / aqueous solution interface (i.e., after ~1 hr exposure to 1 μ M BSA / RhG, closed circles). Solid lines are fits of the data to Langmuir adsorption isotherms, with the corresponding Gibbs free energies for adsorption as indicated. A rhodamine concentration of 4×10^{-4} M was used for the SHG circular and linear dichroism for Systems 1 and 3, corresponding to an R6G surface coverage of ~60% of a monolayer (indicated by the dashed vertical line).