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Stimulated Second Harmonic Generation for High-Sensitivity Interfacial Spectroscopy and Imaging AARON GOODMAN, Department of Chemistry, Massachusetts Institute of Technology, WILLIAM TISDALE, Department of Chemical Engineering, Massachusetts Institute of Technology — Second-order nonlinear optical interactions such as sum- and difference-frequency generation are widely used for bioimaging and as selective probes of interfacial environments. However, inefficient nonlinear optical conversion often leads to poor signal-to-noise ratios and long signal acquisition times. Here, we demonstrate the dramatic enhancement of weak second-order nonlinear optical signals via stimulated sum- and difference-frequency generation. We present a conceptual framework to quantitatively describe the interaction and show that the process is highly sensitive to the relative optical phase of the stimulating field. To emphasize the utility of the technique, we demonstrate stimulated enhancement of second harmonic generation (SHG) from bovine collagen-I fibrils. Using a stimulating pulse fluence of only 3 nJ/cm^2, we obtain an SHG enhancement of $> 10^4$ relative to the spontaneous signal. The stimulated enhancement is greatest in situations where spontaneous signals are the weakest - such as low laser power, small sample volume, and weak nonlinear susceptibility - emphasizing the importance of this technique for improving signal-to-noise ratios in biological imaging and interfacial spectroscopy.

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