

## Enhancement of Second-Order Nonlinear-Optical Signals by Optical Stimulation

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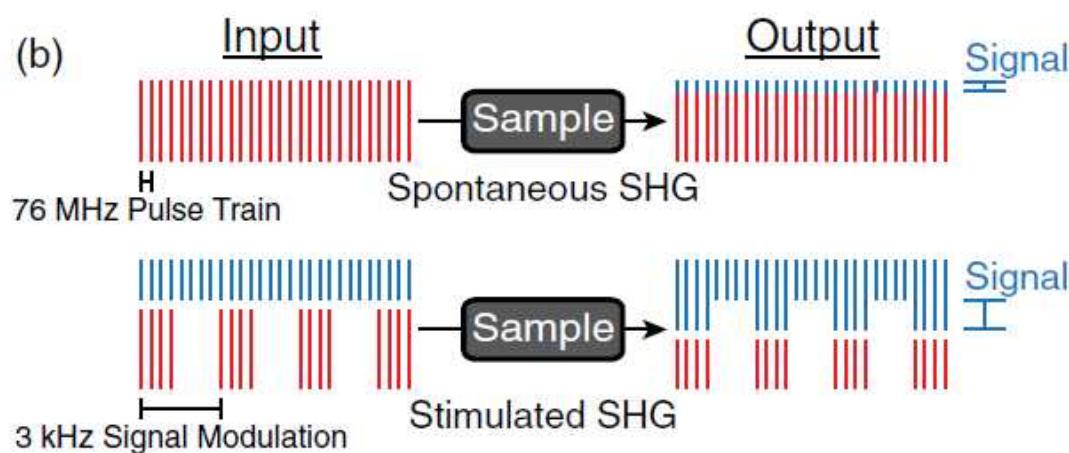
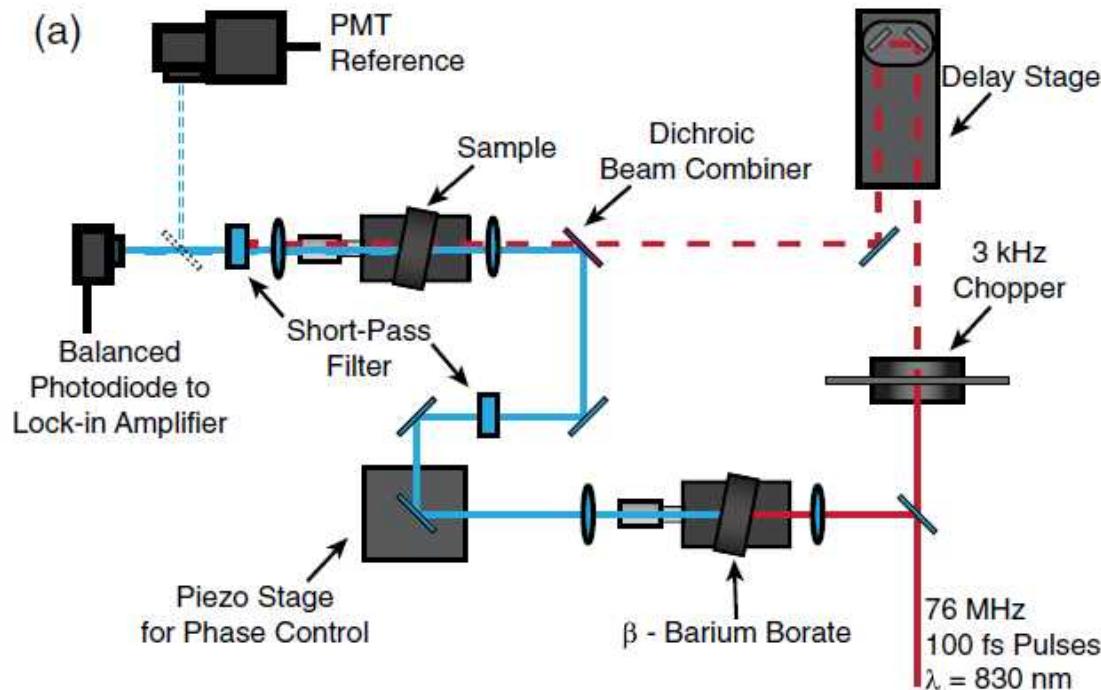
(Received 17 December 2014; published 6 May 2015)

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 ratio 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<sup>2</sup>, we obtain an SHG enhancement  $>10^4$  relative to the spontaneous signal. The stimulation 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 potential for this technique to improve signal-to-noise ratios in biological imaging and interfacial spectroscopy.

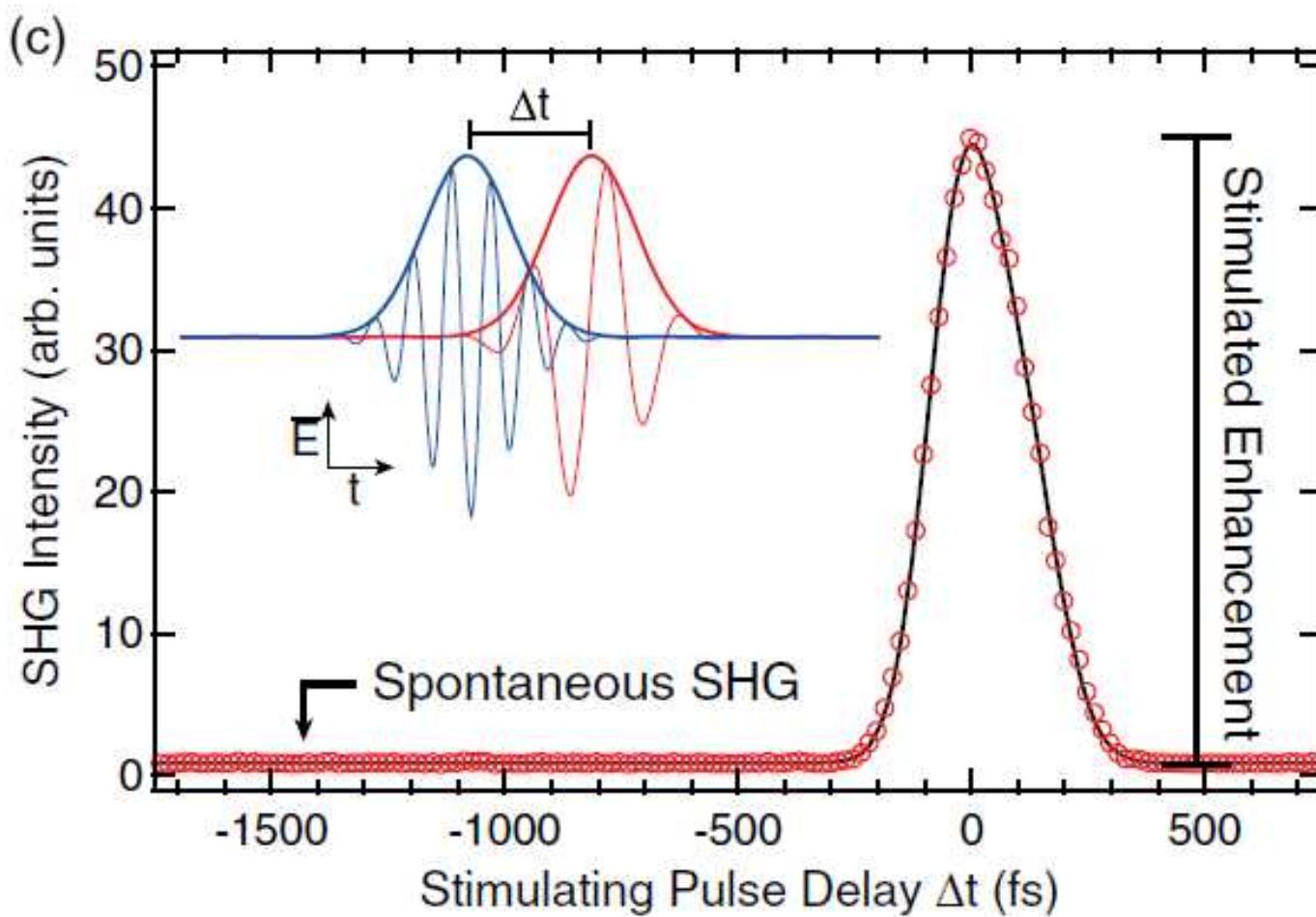
DOI: 10.1103/PhysRevLett.114.183902

PACS numbers: 42.65.-k, 73.20.-r, 87.64.-t

# Experimental Setup & Scheme



## SHG signal contrast - stimulated vs spontaneous

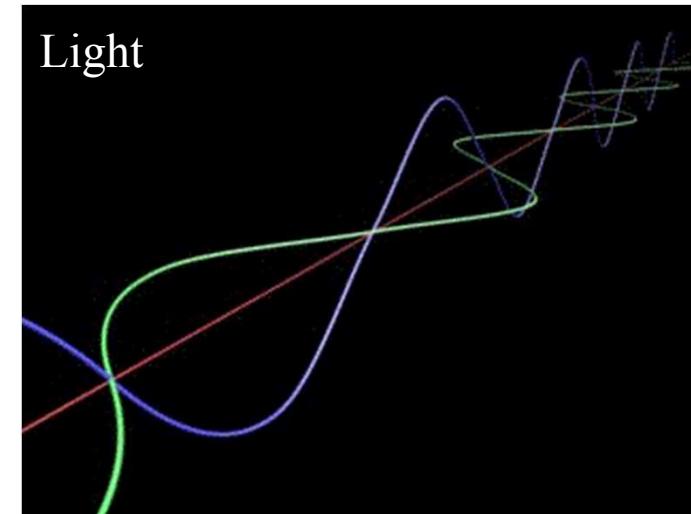
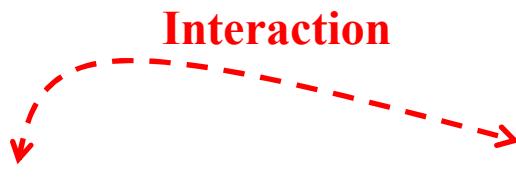


→ Why?

# Nonlinear optical process

\*Polarization

Matter



$$U = -\int \tilde{F}_{res} dx = \frac{1}{2} m \omega_0 x^2 + \frac{1}{3} m \omega_0 x^3$$

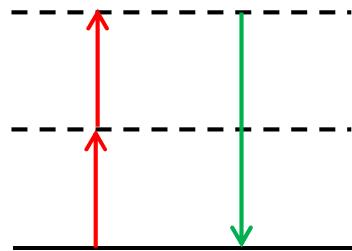
$$\left\{ \begin{array}{l} \ddot{x} + 2\gamma \dot{x} + \omega_0^2 x + ax^2 = -\frac{\lambda e E(t)}{m} \\ x = \lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + \dots \\ \tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c. \end{array} \right.$$

# 2<sup>nd</sup> Nonlinear optical process

Details – The Principles of Nonlinear Optics, Y. R. Shen, Chapter 1.3.  
Nonlinear Optics, Robert W. Boyd, Chapter 1.4.

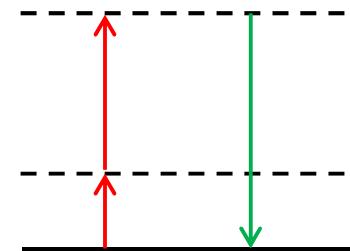
1) Second Harmonic Generation (SHG)

$$x^{(2)}(2\omega_1) = \frac{-a(e/m)^2 E_1^2}{D(2\omega_1)D^2(\omega_1)}$$



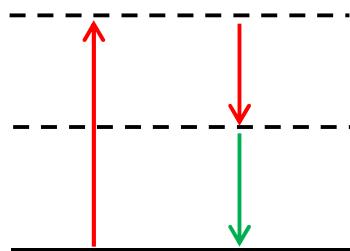
2) Sum-frequency Generation (SFG)

$$x^{(2)}(\omega_1 + \omega_2) = \frac{-2a(e/m)^2 E_1 E_2}{D(\omega_1 + \omega_2)D(\omega_1)D(\omega_2)}$$



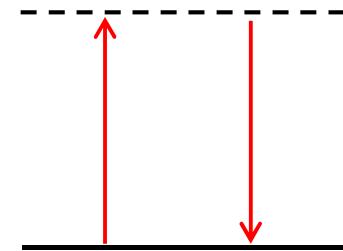
3) Difference-frequency Generation (DFG)

$$x^{(2)}(\omega_1 - \omega_2) = \frac{-2a(e/m)^2 E_1 E_2^*}{D(\omega_1 - \omega_2)D(\omega_1)D(-\omega_2)}$$



4) Optical Rectification

$$x^{(2)}(0) = \frac{-2a(e/m)^2 E_1 E_1^*}{D(0)D(\omega_1)D(-\omega_1)} + \frac{-2a(e/m)^2 E_2 E_2^*}{D(0)D(\omega_2)D(-\omega_2)}$$



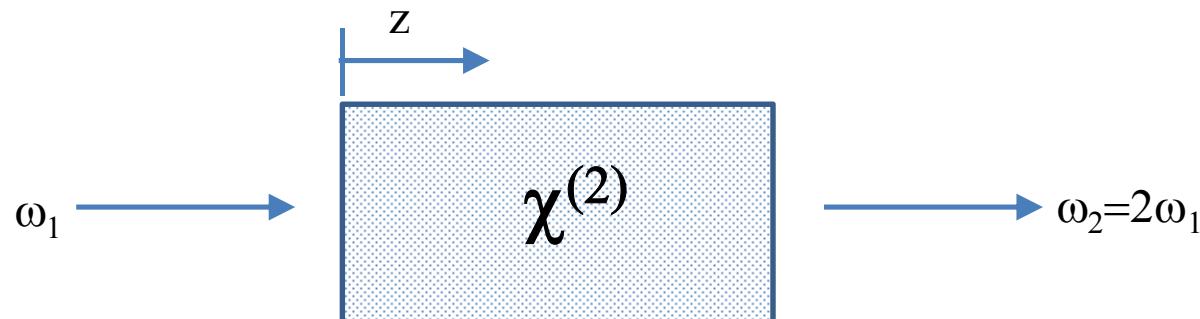
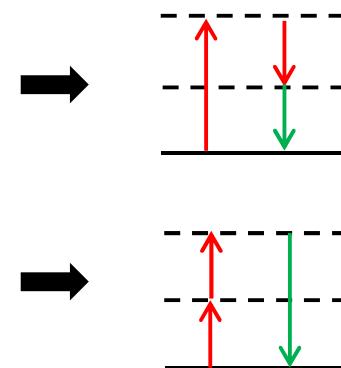
# Energy exchange - Coupled wave equation

$$-\nabla^2 \tilde{E}_n + \frac{\varepsilon^{(1)}(\omega_n)}{c^2} \frac{\partial^2 \tilde{E}_n}{\partial t^2} = -\frac{4\pi}{c^2} \frac{\partial^2 \tilde{P}_n^{NL}}{\partial t^2}$$

$$\tilde{E}_1 = A_1 e^{i(k_1 z - \omega_1 t)} + c.c.,$$

$$\tilde{E}_2 = A_2 e^{i(k_2 z - \omega_2 t)} + c.c.$$

$$\left\{ \begin{array}{l} \frac{d^2 A_1}{dz^2} \sim \frac{-8\pi i d_{eff} \omega_1^2}{c^2} A_2 A_1^* e^{i(k_2 - 2k_1)z} \\ \frac{d^2 A_2}{dz^2} \sim \frac{-4\pi i d_{eff} \omega_2^2}{c^2} A_1^2 e^{i(2k_1 - k_2)z} \end{array} \right.$$



# Energy exchange - Coupled wave equation

Details – The Principles of Nonlinear Optics, Y. R. Shen, Chapter 3.1-3.3.  
 Nonlinear Optics, Robert W. Boyd, Chapter 2.6.

$$\left\{ \begin{array}{l} A_1 = \left( \frac{2\pi I}{n_1 c} \right)^{1/2} u_1(z) e^{i\phi_1}, A_2 = \left( \frac{2\pi I}{n_2 c} \right)^{1/2} u_2(z) e^{i\phi_2} \\ u_1^2(z) + u_2^2(z) = 1 \\ \zeta = z/l \propto I^{1/2} d_{eff} \\ l = \left( \frac{n_1 n_2 c^3}{2\pi I} \right)^{1/2} \frac{1}{8\pi\omega_1 d_{eff}} \end{array} \right.$$

$$\frac{du_\omega}{d\zeta} = u_\omega u_{2\omega} \sin(\theta), \quad (4)$$

$$\frac{du_{2\omega}}{d\zeta} = -u_\omega^2 \sin(\theta), \quad (5)$$

$$\frac{d\theta}{d\zeta} = \frac{\cos(\theta)}{\sin(\theta)} \frac{d}{d\zeta} (\ln(u_\omega^2 u_{2\omega})), \quad (6)$$

$$l^{-1} = 2\omega^2 \left( \frac{2\pi d_{eff}}{c^2} \right) k_\omega^{-1} \sqrt{I_{total}}. \quad (1)$$

$$u_\omega^2 + u_{2\omega}^2 = 1, \quad \theta = 2\phi_\omega - \phi_{2\omega}$$

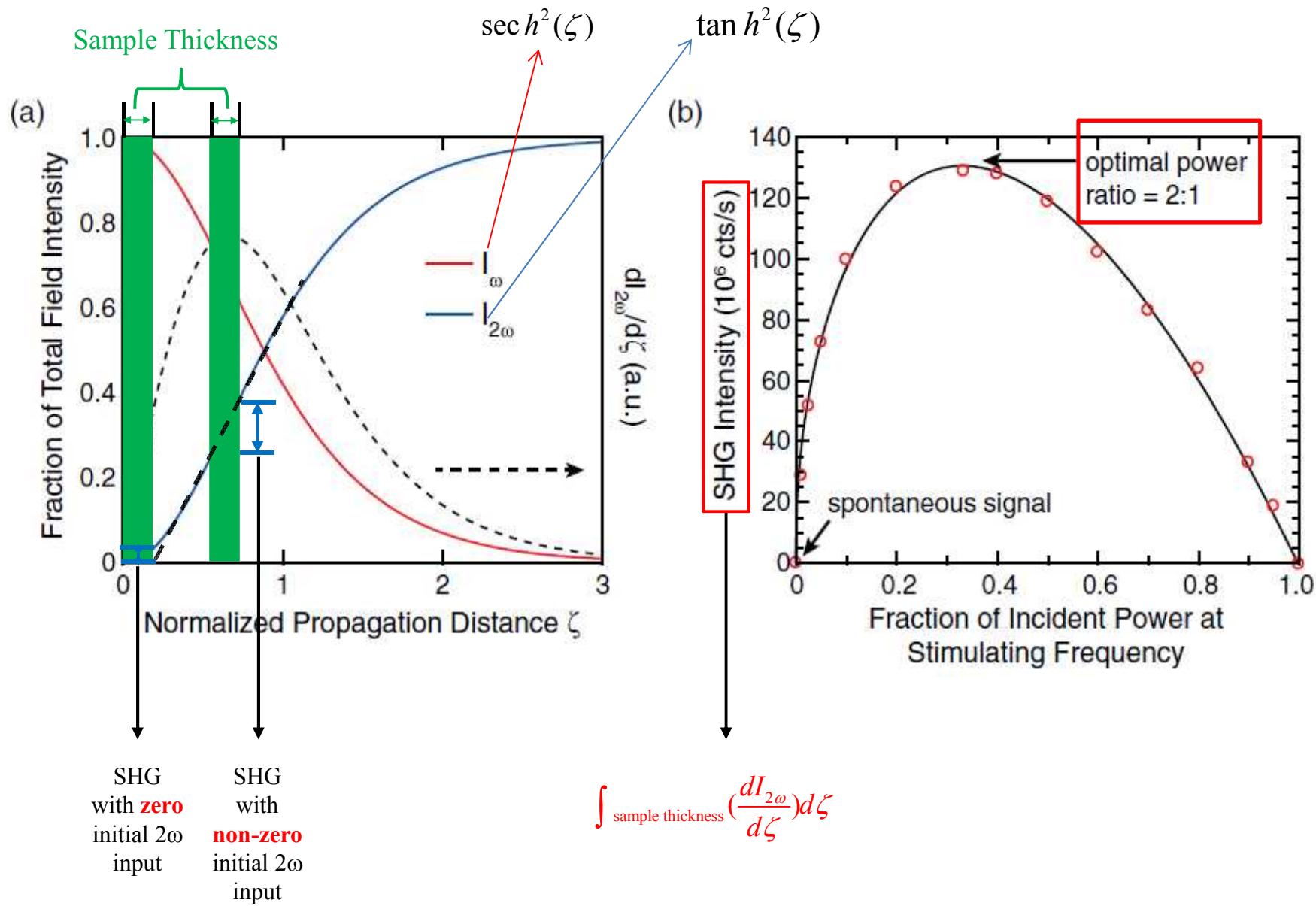
$$\zeta = z/l$$

# Specific case

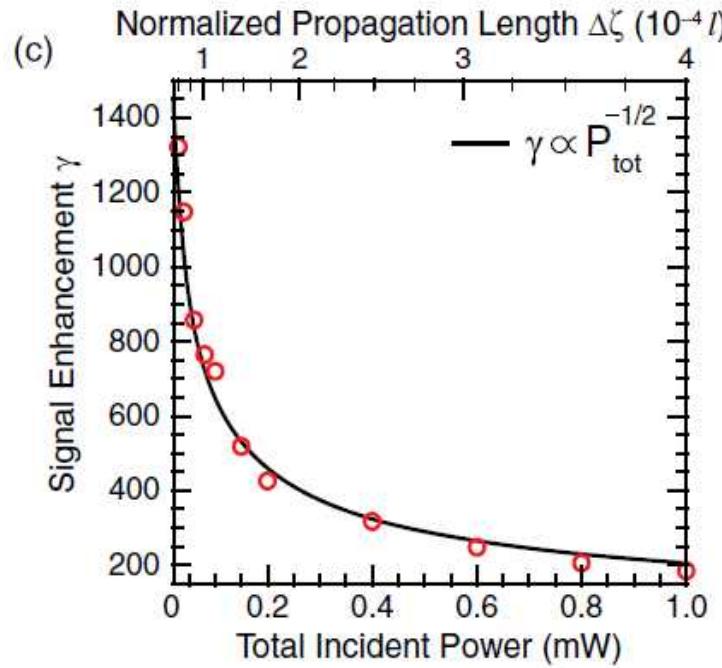
→  $\begin{cases} \Delta k = 2k_1 - k_2 = 0 : \text{perfect phase matching} \\ \theta = 2\phi_1 - \phi_2 = -\frac{\pi}{2} \end{cases}$

→ 
$$\begin{aligned} \frac{du_1}{d\zeta} &= -u_1 u_2 \\ \frac{du_2}{d\zeta} &= u_1^2 \end{aligned} \quad \longrightarrow \quad \begin{aligned} u_1(\zeta) &= \sec h(\zeta + \zeta_0) \\ u_2(\zeta) &= \tan h(\zeta + \zeta_0) \end{aligned}$$

# Characteristics of stimulated SHG intensity



# SHG signal contrast - stimulated vs spontaneous

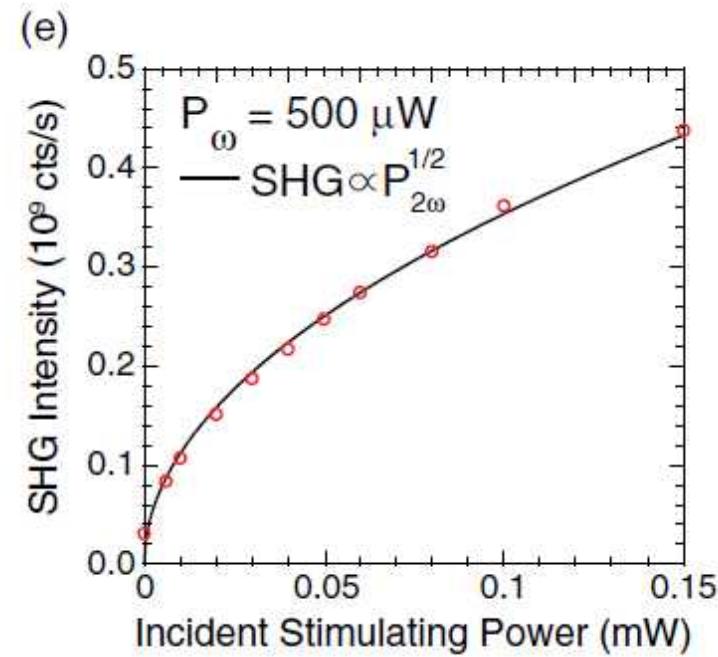
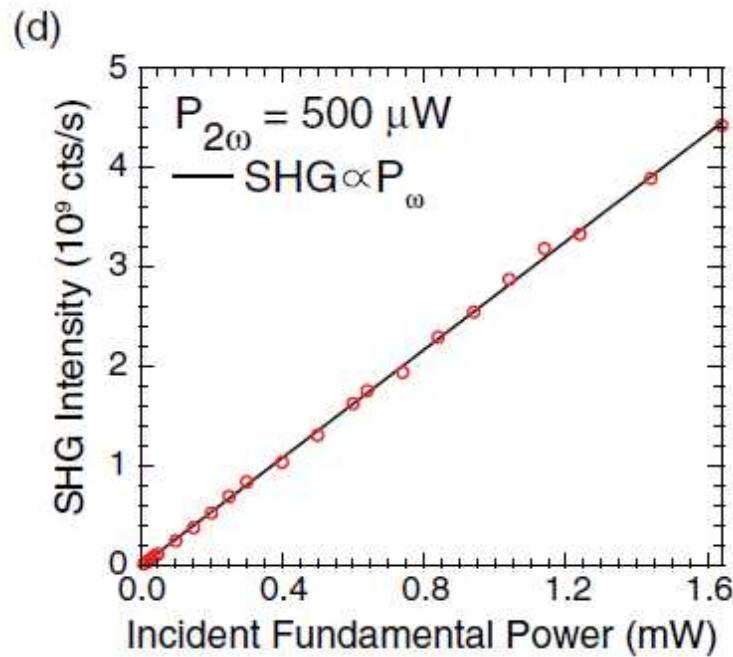


$$\left. \begin{aligned} A_1 &= \left( \frac{2\pi I}{n_1 c} \right)^{1/2} u_1(z) e^{i\phi_1}, A_2 = \left( \frac{2\pi I}{n_2 c} \right)^{1/2} u_2(z) e^{i\phi_2} \\ u_1^2(z) + u_2^2(z) &= 1 \\ \zeta &= z/l \propto I^{1/2} d_{eff} \\ l &= \left( \frac{n_1 n_2 c^3}{2\pi I} \right)^{1/2} \frac{1}{8\pi\omega_1 d_{eff}} \end{aligned} \right\}$$

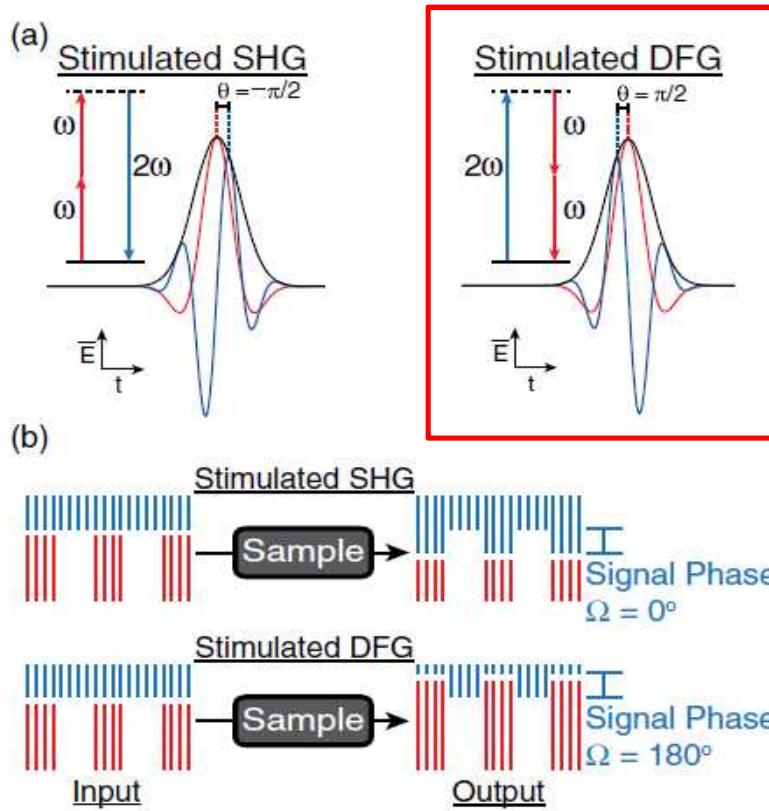
$$\left. \begin{aligned} u_2(\zeta) &= \tanh(\zeta) \sim 2\zeta \text{ for } \zeta \ll 1 \\ I_{2\omega, norm} &= u_2^2(\zeta) = a\zeta \text{ for } u_1(\zeta) = 2u_2(\zeta) \end{aligned} \right\}$$

$$\begin{aligned} I_{sig}^{stim} &\propto I_{total}^{3/2} \times d_{eff} \times z, \\ I_{sig}^{spont} &\propto I_{total}^2 \times d_{eff}^2 \times z^2, \\ \gamma &= I_{sig}^{stim} / I_{sig}^{spont} \propto I_{total}^{-1/2} \times d_{eff}^{-1} \times z^{-1}. \end{aligned} \quad (7)$$

# SHG signal contrast - stimulated vs spontaneous

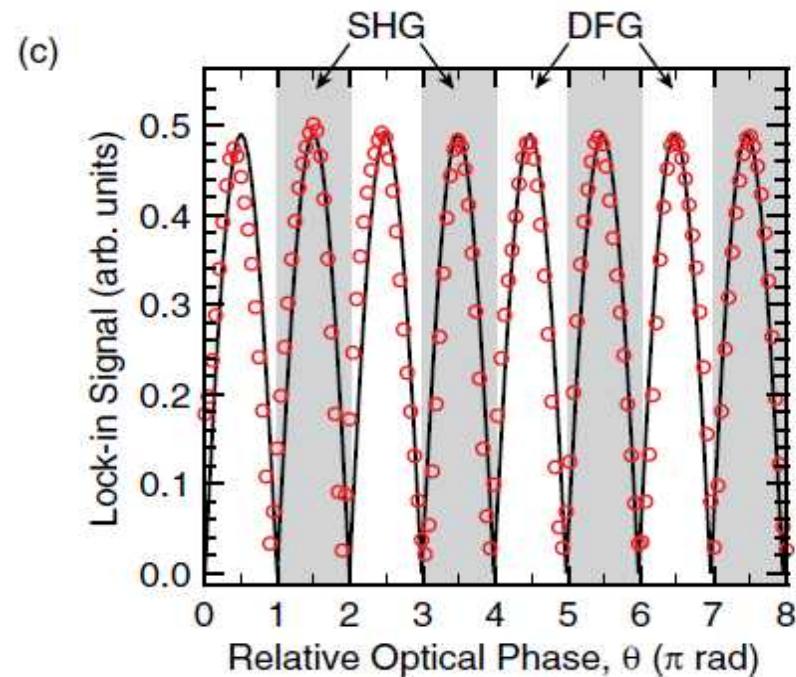


# SHG signal contrast - stimulated vs spontaneous



for  $\theta = +\frac{\pi}{2}$

<http://www.wolframalpha.com/input/?i=dy%2Fdx%3D-%281-y%5E2%29>



$$\frac{du_\omega}{d\zeta} = u_\omega u_{2\omega} \sin(\theta), \quad (4)$$

$$\frac{du_{2\omega}}{d\zeta} = -u_\omega^2 \sin(\theta), \quad (5)$$

$$\frac{d\theta}{d\zeta} = \frac{\cos(\theta)}{\sin(\theta)} \frac{d}{d\zeta} (\ln(u_\omega^2 u_{2\omega})), \quad (6)$$

# SHG image - collagen fibers

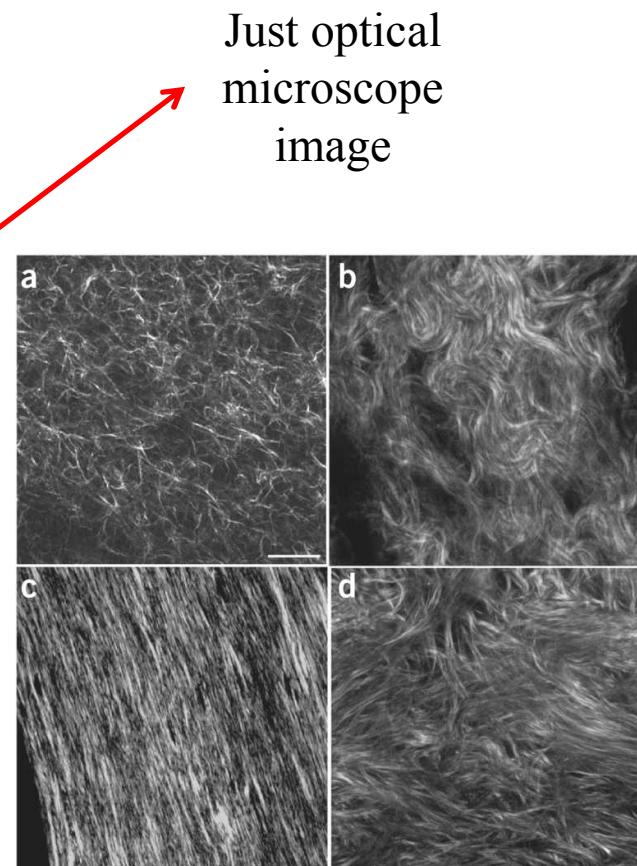
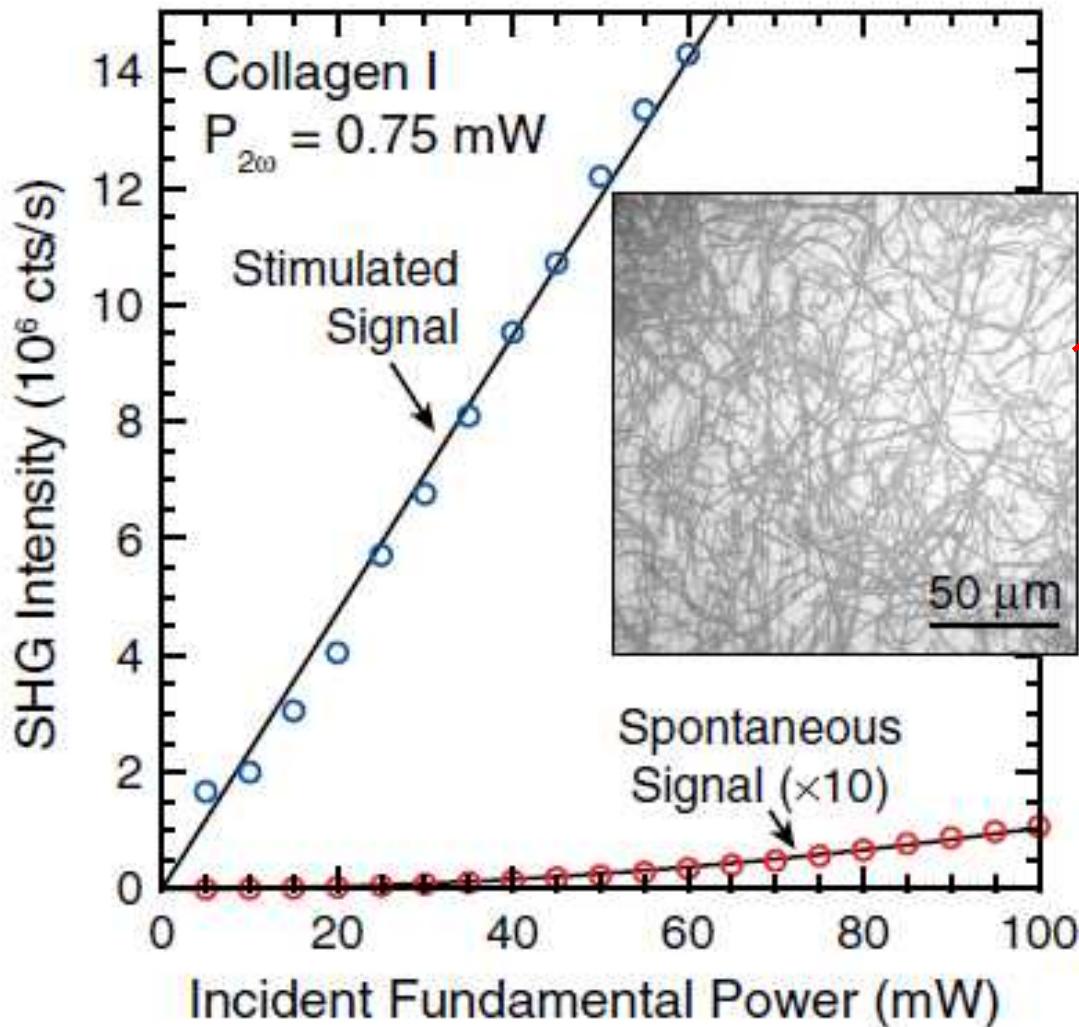


Figure 1.  
Montage of SHG imaging of collagen tissues. (a-d) The single optical sections show representative images of self-assembled collagen gel (a); mouse dermis (b); mouse bone (c); and human ovary (d). Scale bar, 30 μm.

Campanola and coworkers, Nat Proctoc 7, 654 (2012).