Low frequency Raman gain measurements using chirped pulses

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Abstract: Two-beam coupling, attributed to Raman gain, is observed in dielectrics using chirped femtosecond pulses. A time resolved pump-probe geometry is used to vary the frequency difference between pulses in the terahertz frequency band. Stimulated Raman scattering couples the pulses transferring energy from the higher to the lower frequency beam, resulting in a dispersion shaped curve as a function of the temporal delay, dependent on the product of the pump and probe irradiances. The observed signal gives the Raman gain in SiO_2 and PbF_2 for detunings up to 10 THz (approximately 300 cm^{-1}) using mm-thick samples. This method may also be sensitive to the electronic motion responsible for bound-electronic nonlinear refractive index, which could yield the optical response time of bound electrons.

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The properties of stimulated Raman scattering (SRS)[1,2] and measurements of the Raman gain have been made in many materials and at various detunings.[3] Effects commonly observed in pulse propagation in optical fibers, such as the soliton self-frequency shift,[4] have their origin in low-frequency SRS making the low-frequency Raman gain spectrum of considerable interest. The limit in measuring the Raman gain at small frequency detunings is determined by the requirement that the DC pump beam must be removed from the detection system. Low-frequency Raman gain measurements have been reported down to 6 cm^{-1} in glass fibers with interaction lengths of several hundred meters. [5] Obviously this method is

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not applicable to other materials. Here we propose a novel method for measuring the Raman gain at low frequency with much shorter interaction lengths based on two-beam coupling of chirped pulses. This method relates the frequency difference between the pump and probe beams in a pump-probe experiment to the time-delay between the pulses.

The experiment essentially consists of a non-collinear pump-probe system.[6] The two-beam coupling between the pump and probe beams is due to a frequency difference between the beams. Because the pulses are identically chirped, this frequency difference occurs when the pulses are displaced in time with respect to each other. For a linearly chirped pulse, this frequency difference is constant in time at any given delay between the pulses. By varying the delay between the pulses, we vary the detuning between the pump and probe beams.

A noncollinear pump-probe geometry is used with pump and probe derived from the same source. Monitoring the probe beam energy, the gain of the SRS can be directly measured as a function of time-delay. Dogariu *et. al.*[6] recently reported a similar experiment for measuring stimulated Rayleigh-wing scattering in liquids using picosecond pulses. In that case, the signals are relatively large, with gains of several percent. However, Raman gains in thin dielectric samples of similar dimensions are several orders of magnitude smaller. Despite this, we demonstrate here that by using signal retrieval techniques, we can measure the Raman gain in dielectrics with interaction lengths of only a millimeter or so.

To measure very small changes in probe transmittance, we use a dual frequency modulation technique consisting of 1 MHz mechanical modulation of the pump beam, and 100 Hz modulation of the probe. The signal is detected by cascading high frequency and low-frequency lock-in amplifiers, resulting in a sensitivity to normalized changes in probe energy as low as a few times 10^{-6}. In measuring small Raman gain signals we have to eliminate competing signals such as three-photon absorption, scattering due to impurities, etc. We used a mode-locked Ti:Sapphire laser source providing purposely-chirped 100 fs (FWHM) pulses at 840 nm. We estimate the chirp by measuring pulse temporal and spectral widths. For linearly chirped pulses we can measure the linear chirp coefficient C, where, for Gaussian pulses, the input electric field is defined as:

$$E(\vec{r},t) = E_0 e^{i\left(\vec{k}\cdot\vec{r}-\omega t\right)} e^{-r^2/w_0^2} e^{-(1+iC)t^2/\tau_p^2}, \qquad (1)$$

with τ_p the pulse width (HW1/eM). For a given time-delay, τ, we can calculate the frequency difference between the pump and probe beam, Ω. For linearly chirped pulses,

$$\Omega = \omega_{pump} - \omega_{probe} = \frac{C\tau}{\tau_p^{\,2}}. \qquad (2)$$

We can immediately see the linear relationship between the time-delay and the detuning. The irradiance, I, of the probe beam (subscript p) sees the Raman gain, *g*, induced by the excitation beam (subscript e):

$$\frac{dI_p}{dz} = gI_e I_p. \qquad (3)$$

Integrating Eq. (3) over the pulse width τ_p and beam waists $w_{0p,e}$, we obtain the normalized change in the weak probe transmittance:

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$$\left(\frac{\Delta T(\tau)}{T}\right)_p = \frac{1}{\sqrt{2}\left(1+\frac{w_{0p}{}^2}{w_{0e}{}^2}\right)} I_{0e} L \exp\left(-\frac{1}{2}\left(\frac{\tau}{\tau_p}\right)^2\right) g(\tau), \tag{4}$$

where I_{0e} is the peak pump irradiance and L is the sample length.

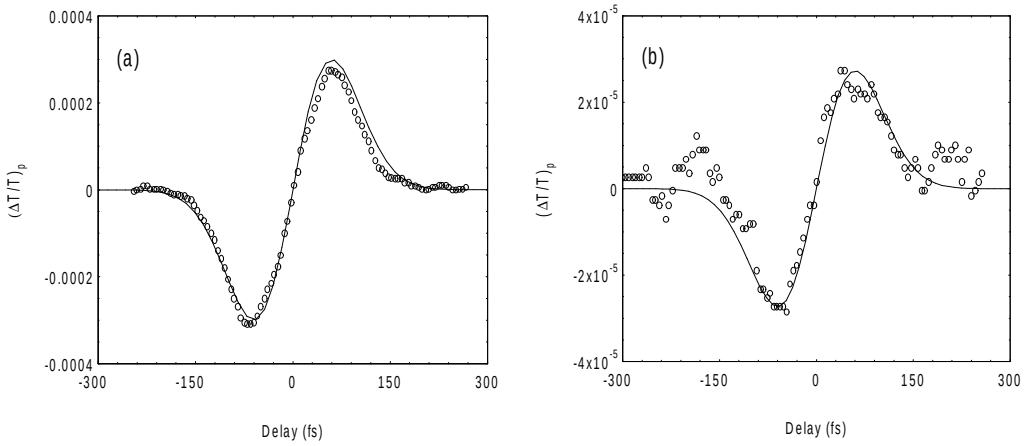

Fig.1 Two-beam coupling data (circles) (a) in PbF_2 and (b) in SiO_2 with theoretical fits (solid lines) assuming a Raman gain linearly dependent on time-delay. The pulses are linearly chirped with C = 1.3. Here C is measured to be 1.3± 0.1.

Figure 1 represents the measured normalized changes in transmittance (circles) for PbF_2 (a) and SiO_2 (b) as functions of the time-delay τ. The solid lines are fits using Eq. 4, which give a Raman gain, $g(\tau)$ that is linearly dependent on τ. We obtain $g(\tau) = 5.5\cdot10^{-14}\ \tau$ for PbF_2 and $g(\tau) = 4.8\cdot10^{-15}\ \tau$ for SiO_2, where g is in cm/W and τ is in fs.

For linear chirp we can use Eq. 2 to express the time-delay dependence in terms of frequency $\nu=\Omega/2\pi$. In this case we can directly extract the linear frequency dependence of the gain from Eq. 4 by removing the time-correlation term, $\exp\{-½(\tau/\tau_p)^2\}$, from the data shown in Fig. 1. By fitting the so-transformed data shown in Fig. 2 for PbF_2 (squares) and SiO_2 (circles) we obtain $g(\nu)$.

The lines fitting the data in Fig. 2 give the following frequency dependence for the Raman gain:

$$g(\nu)_{PbF_2} = (9.6 \pm 1.9)\cdot 10^{-13}\nu \tag{5.a}$$

$$g(\nu)_{SiO_2} = (8.4 \pm 1.7)\cdot 10^{-14}\nu, \tag{5.b}$$

where g is in cm/W and ν in THz.

Equation 5.b is in good agreement with the previously reported measured Raman gain spectra of silica glass in fibers,[5,7] if we account for the fact that our measurements are performed with 100 fs pulses, within the 400 fs turn-on of the vibrational Raman effect.[8] However, those methods are limited to glass since long fibers are required to achieve detectable stimulated Raman gain. Using our method, any sample can be used as long as the laser frequency is chosen to lie in a region of very low nonlinear absorption.

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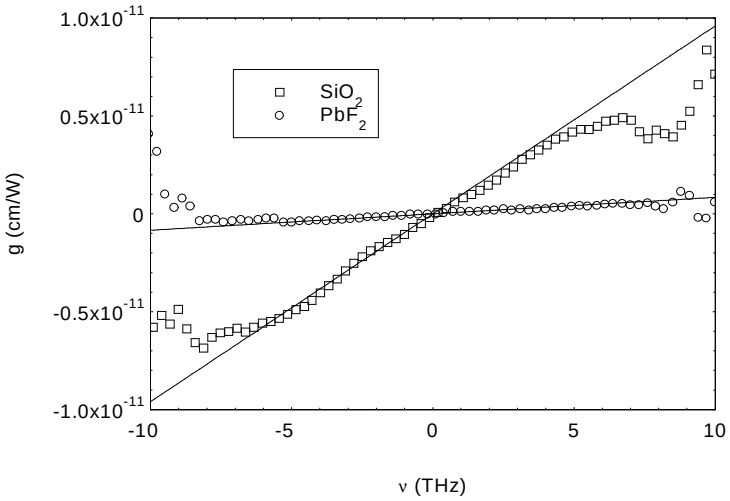

Fig. 2. Low frequency Raman spectra for PbF_2 (squares) and SiO_2 (circles) obtained using the chirped-two-beam coupling method, along with linear fits.

So far, the chirped pulse two-beam coupling technique has been used to measure the effects of molecular rotation (stimulated Rayleigh-wing scattering),[6] and of nuclear vibration (stimulated Raman scattering). However, this method may also be sensitive to the electronic motion responsible for bound-electronic nonlinear refractive index, n_2. Typically this response time is predicted to be ~1 fs or less. A simple calculation assuming 0.1 ~ 1 fs Debye relaxation times for the bound electrons reveals that signals similar in magnitude to the Raman signals should be measurable. The challenge that remains here is to find combinations of materials and detunings for which the nuclear signals would not dominate the electronic signals.

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