

Stimulated Raman gain scattering in planar dielectric waveguides

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We have measured the vibrational Raman spectrum of a 1.2- μm -thick polystyrene film by using this film as an optical waveguide in a waveguide-stimulated Raman gain experiment. A gain factor of 1.3% was measured for the strong benzene mode at 1002 cm^{-1} . This value is in reasonable agreement with theoretical predictions.
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In surface vibrational spectroscopy the combination of spontaneous Raman light scattering with optical waveguides yields a sensitive technique for characterizing thin films.¹ One can measure the Raman spectrum of either the waveguide itself or a thin (mono)layer on top of the waveguide by guiding the pump laser beam in the waveguide. This configuration leads to a large interaction volume even of very thin layers and hence to large signal levels. Monolayers can be detected with waveguide-resonant Raman spectroscopy,² and recently it was shown that monolayer sensitivity can be obtained without resonant enhancement when waveguide Raman spectroscopy is employed.³ Similar techniques have been developed for coherent anti-Stokes Raman scattering (CARS).⁴ Both pump and Stokes beams are guided by the waveguide with high electric field intensities and a large interaction length, resulting in strong nonlinear effects and thus in large CARS signal levels. A serious problem in applying waveguide CARS for the study of thin layers on top of a waveguide structure is the interference of the resonant thin-layer signal with the strong nonresonant background of the waveguide. Techniques,^{5,6} which are based on destructive interference among pump, Stokes, and CARS modes, to suppress this background are not always sufficient. We have shown theoretically⁷ that stimulated Raman gain spectroscopy can also lead to high signal levels of thin layers when pump and probe beams are guided by an optical waveguide. Because stimulated Raman gain spectroscopy is not hampered by a large nonresonant background it might be a good alternative technique for waveguide CARS. In this Letter we report what we believe are the first experimental results of such an approach.

In Fig. 1 a schematic of a waveguide-stimulated Raman gain spectroscopy setup is shown. Both pump and probe beams are guided by a specific mode of a waveguide. The probe beam experiences a gain if the energy difference between the pump and the Stokes photons equals the energy of a vibrational resonance of the waveguide or of a thin layer on top of this waveguide. In this study the waveguide consists of a polystyrene layer of which the stimulated Raman gain spectrum is measured.

In waveguide-stimulated Raman gain spectroscopy the gain can be expressed as⁷

$$G = (\omega_{\text{Stokes}}/2)\text{Re}(F^{kl})P_{\text{pump}}L/H. \quad (1)$$

In Eq. (1) F^{kl} is the overlap integral and contains all the information about the waveguide configuration and Raman cross sections and also depends on the applied guided pump mode TE_k and probe mode TE_l . The power in the pump mode is denoted P_{pump} , L is the interaction length between pump and probe beams, and H is the width of the beams inside the waveguide. In our situation, in which we have a 1.2- μm -thick polystyrene waveguide and the applied modes are of TE_0 character for the pump as well as for the probe beam ($k = l = 0$), the gain can be calculated to be⁷

$$G = 5.0 \times 10^{-5} P_{\text{pump}}L/H \quad (2)$$

for the peak gain of the 1002- cm^{-1} benzene ring breath vibration of polystyrene. The electric field distributions shown in Fig. 1 correspond to the TE_0 modes.

Polystyrene was dissolved in toluene, and this solution was spin coated onto a SiO_2 substrate to produce a 1.2- μm -thick polystyrene layer. The experimental configuration is schematically outlined in Fig. 2. A mode-locked Nd:YLF laser operating at 76 MHz is frequency doubled to 527 nm and is used to synchronously pump two dye lasers. The dye lasers are equipped with cavity dumpers to reduce the repetition rate to 3.8 MHz. One of the dye lasers provides the pump beam at 595 nm, whereas the other one operates as the tunable probe laser near 633 nm. The temporal widths of the beams are measured with an autocorrelator and are 5 ps. Both pump and Stokes beams

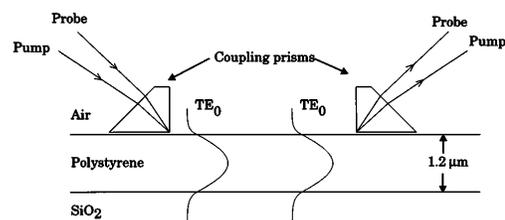


Fig. 1. Waveguide assembly and experimental configuration as used in a waveguide-stimulated Raman gain experiment. The electric field distributions of the TE_0 modes employed are shown for the 1.2- μm -thick polystyrene waveguide.

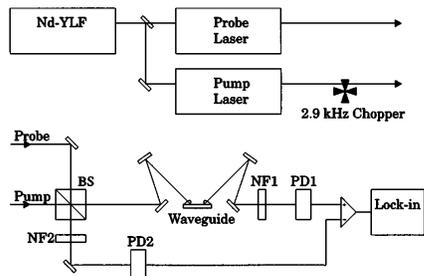


Fig. 2. Schematic representation of the experimental setup: BS, beam splitter; NF1, NF2, holographic notch filters for pump light rejection; PD1, signal photodiode; PD2, reference photodiode.

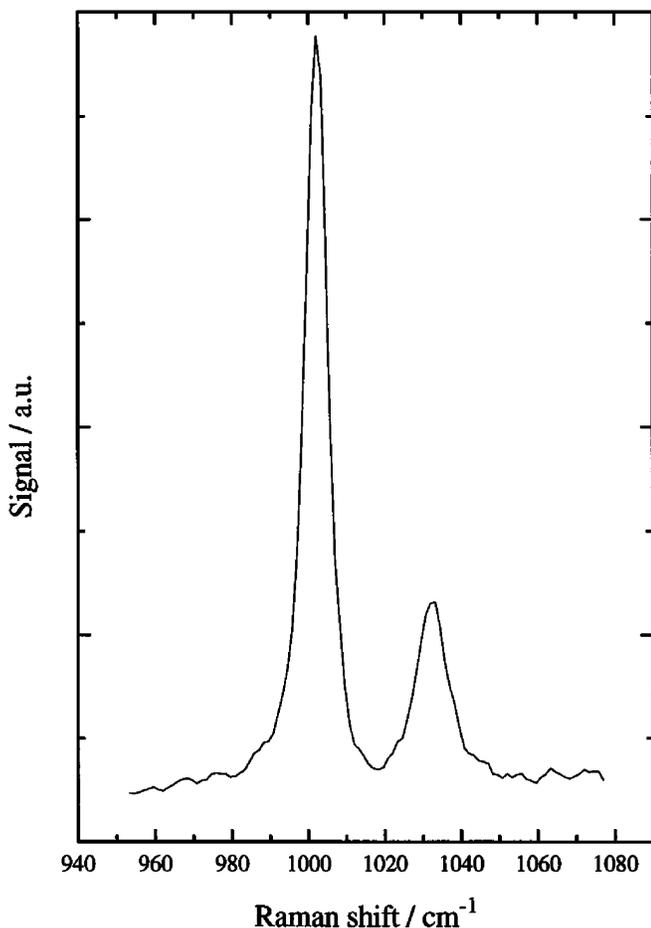


Fig. 3. Stimulated Raman gain spectrum of a 1.2- μm -thick polystyrene waveguide. The peak gain is 1.3×10^{-2} .

were made collinear and coupled to the TE_0 mode of the waveguide by an SF6 prism. Another SF6 prism, separated 1 cm from the incoupling prism, was used to couple the beams out of the waveguide. The light at the probe wavelength was filtered out by a holographic notch filter and collected onto a photodiode. Spatial filtering of the probe beam was not possible because the outcoupling angles of pump and probe beams are nearly identical. We achieved separation of the stimulated Raman gain signal from the probe signal by modulating the pump laser at 2.9 kHz and using a lock-in amplifier tuned to 2.9 kHz. The main source of noise originates from jet stream fluctuations of the probe

dye laser. This noise was reduced with a reference photodiode to monitor the intensity fluctuations of the probe beam. This reference signal is subtracted from the signal in a differential amplifier with a high common mode rejection factor. With this configuration a signal-to-noise ratio of 1 can be achieved for a 1-s integration time if the gain is as small as 1×10^{-5} . A higher sensitivity, down to a gain of 1×10^{-8} , and thus a significant signal-to-noise ratio improvement can be obtained if a megahertz modulation frequency is used together with a dye laser stabilizer for the probe beam.⁸

The average power in the waveguide was estimated to be 1.0 mW for the pump beam and 0.4 mW for the probe beam. This corresponds to a peak power of $1 \text{ mW}/(5 \text{ ps} \times 3.8 \text{ MHz}) = 52 \text{ W}$. The widths of the beams were $\sim 100 \mu\text{m}$, corresponding to a pump intensity of $44 \text{ MW}/\text{cm}^2$. Because the pump and the probe beams are collinear the interaction length is determined by the distance between the two SF6 prisms and is 1 cm. The spectrum was measured from 950 to 1080 cm^{-1} in 400 steps. The accumulation time was 2 s for each spectral point. As shown in Fig. 3, the 1002-cm^{-1} vibration of the benzene ring is clearly visible, as is the vibration of 1030 cm^{-1} , which is smaller in intensity by a factor of 4. Band positions and relative peak heights correspond to spontaneous Raman data. The spectral width of 6.7 cm^{-1} of the vibration is, however, larger than the true value known from the literature. This is caused by the spectral width of 3.5 cm^{-1} of the probe and pump beams. The measured peak gain is 1.3×10^{-2} . The calculated gain from Eq. (1) is 2.5×10^{-1} . This is ~ 20 times larger than the measured value. At this point, however, it should be noted that the calculated gain is the steady-state gain, whereas the measured gain is in the transient regime because the pulse widths are comparable with the vibrational relaxation time. It is argued⁹ that the transient gain is smaller than the steady-state gain. For Gaussian-shaped pulses the transient gain G_t is given by

$$G_t/G_{ss} = \sqrt{\pi/2\sigma} \exp(\sigma^2) \text{erfc}(\sigma), \quad \sigma = \frac{1}{4} \frac{\tau}{\tau_v}, \quad (3)$$

with G_{ss} the steady-state gain, τ the width of the pulses, and τ_v the vibrational relaxation time. In our case ($\tau \sim 5 \text{ ps}$, $\tau_v \sim 5 \text{ ps}$) this results in $G_t/G_{ss} \sim 0.25$. With this factor taken into account the calculated gain is reduced to 6.3×10^{-2} , which is in reasonable agreement with the observed gain. The discrepancy of a factor of 5 is possibly due to uncertainties in the experimental parameters such as pump power and beam widths inside the waveguide and the exact value of the Raman cross section of the 1000-cm^{-1} vibration of polystyrene.

We have demonstrated the possibility of measuring Raman spectra of thin films by using waveguide-stimulated Raman gain spectroscopy. From these measurements we can conclude that waveguide-stimulated Raman gain spectroscopy is a promising method for the study of thin layers. The reasonable agreement between measured and calculated gain factors lends credence to the theoretical predictions of monolayer sensitivity for molecules on top of a thin-film waveguide structure.

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