Tunable stimulated Raman scattering by pumping with Bessel beams

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We report on a novel pumping scheme for stimulated Raman scattering (SRS) that uses a Bessel beam. We have used this scheme for SRS from molecular vibrations in acetone and from a polariton mode in a $LiIO_3$ crystal. A nearly diffraction-limited Stokes beam was observed along the cone axis of the Bessel beam. The generation mechanism of the Stokes beam is identified as being due to noncollinear scattering of the component plane waves that constitute the Bessel beam. Frequency tuning of SRS from polaritons in $LiIO_3$ by variation of the Bessel beam cone angle is demonstrated.

Stimulated Raman scattering (SRS) is a useful technique for frequency shifting of intense coherent light. As is well known, the Raman shift can be tuned continuously when SRS from polaritons is used and the Stokes emission is forced to include a finite angle with the pump by means of a resonator.[1,2] Here we present a novel pumping scheme that uses a Bessel beam that enables us to vary the angle between pump and Stokes light and to get tunable SRS without any resonator.

The field distribution of a Bessel beam propagating along the z axis is given by[3,4]

$$E = E_0 \exp[i(\beta z - \omega t)] J_0(\alpha r)\,, \qquad (1)$$

where $\alpha^2 + \beta^2 = k^2$, $k = 2\pi/\lambda$, $\alpha = k\ \sin(\gamma)$, $\beta = k\ \cos(\gamma)$, and J_0 is the zero-order Bessel function of the first kind. The Bessel beam can be considered as consisting of infinitely many plane waves whose wave vectors form a cone including an angle γ with the z axis. This cone angle is referred to as the steepness of the Bessel beam and is the characteristic parameter. The superposition of the plane waves enclosing an angle with the cone axis leads to a narrow propagation-invariant intensity maximum in the center, forming an intense line focus along the cone axis. This property is even more pronounced for nonlinear-optical processes such as SRS.

In our experiment we generated intense Bessel beams by illuminating a 30-mm-diameter fused-silica cone (axicon[5,6]) with a linearly polarized expanded laser beam of a frequency-doubled Q-switched Nd:YAG laser. The steepness γ of the resulting Bessel beam is determined by the wedge angle $\delta = 30°$ and the refractive index $n = 1.48$ of the axicon:

$$\sin(\gamma) = (n - 1)\tan(\delta)\,. \qquad (2)$$

After the axicon, a telescope consisting of two positive lenses was placed that projected the Bessel beam into the Raman medium. Varying the focal lengths of the lenses in the telescope permitted the steepness γ of the Bessel beam to be varied.[7]

For our first experiments we used acetone as a Raman medium that shows a Raman shift of 2921 cm^{-1} and relatively high gain. A Bessel beam with a steepness of 16.2° in air and of 11.8° inside the acetone was used. The line focus had a length of 5 cm in a 10-cm-long cell. We determined the laser pulse energy threshold for SRS in acetone to be 30 mJ with pulses of 10-ns width. This value was found to be ~20% higher than that obtained with a Gaussian pump beam focused in the liquid with a lens of 30-cm focal length. Figure 1 shows the resulting far-field pattern in the forward direction. It displays a circle of green pump light corresponding to the cone of plane waves constituting the Bessel beam. The Stokes light appears as an intense red spot in the center of the circle intrinsi-

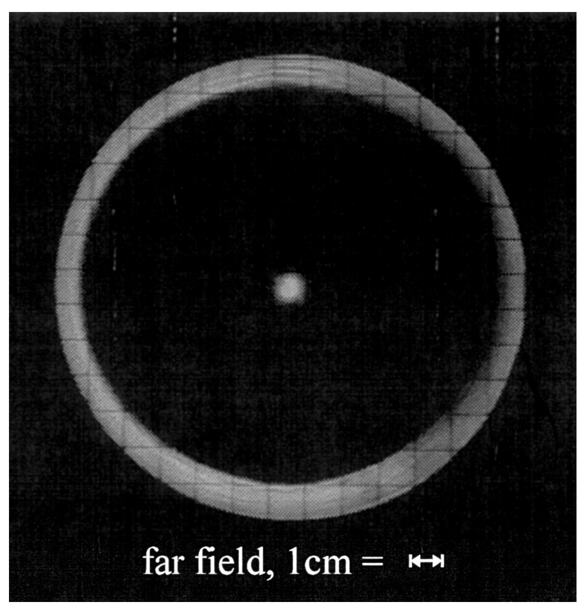

Fig. 1. Far-field pattern of SRS in acetone pumped with a Bessel beam. The outer ring is the far field of the pump Bessel beam at 532 nm. The central spot is the Raman-shifted beam at 629.9 nm.

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cally separated from the pump because the Raman gain is highest for Stokes photons traveling along the cone axis owing to the extended and narrow line focus of the Bessel pump beam. The generation mechanism of the Stokes beam is illustrated in Fig. 2. Passing through the Raman medium, all the component plane waves of the Bessel beam transfer energy to the on-axis Stokes wave through noncollinear interaction, the scattering angle for the forward SRS being equal to the Bessel beam steepness γ.

Stokes light was observed in forward and backward directions, its divergence being less than two times the diffraction limit at an energy conversion level of 10%. The high spatial quality of the Stokes beam can be explained as follows. The Raman medium is excited in a cylinder-shaped region with the Fresnel number $F = d^2/(\lambda L)$, where λ is the wavelength and d and L are the diameter and the length, respectively, of the pumped region. L is given by $L = R/\tan(\gamma)$, where R is the radius of the laser beam illuminating the axicon.[3] For an ideal Bessel beam the diameter d is determined by the size of the central lobe of the Bessel function, $0.766 \times \lambda/\sin(\gamma)$, which is approximately 2 μm in our case. For a realistic Bessel beam produced with a laser of finite divergence $\Delta\Theta$, there is an additional broadening given by[8] $d = \Delta\Theta \times R/\sin(\gamma)$; thus $d \approx 25$ μm in our setup ($R = 10$ mm, $\Delta\Theta = 5 \times 10^{-4}$ rad, and $\gamma = 11.8°$). The Fresnel number of the pumped region is thus much less than unity: $F \approx 0.02$. This means that the geometric angle d/L subtended by the cylinder is less than the diffraction angle λ/d, and therefore such a region supports only a single transverse spatial mode,[9] which is achieved, of course, at the expense of significant diffraction losses; consequently the SRS threshold is somewhat higher than with a Gaussian pump beam.

Because the Stokes beam is generated through noncollinear scattering of the plane waves that constitute the Bessel beam (Fig. 2), one can expect that in SRS from polaritons one can tune the Raman shift by varying the Bessel beam steepness γ. We demonstrated this by using a $LiIO_3$ crystal as the Raman medium. We used a y-cut 1.4-cm-long crystal with antireflection-coated surfaces ($R \le 2\%$ for $\lambda = 530-560$ nm). Both spontaneous and stimulated Raman scattering in this material have been examined extensively.[10–13] One of the strongest Raman active modes[14] that can be observed in SRS with the pump propagating along the x or the y crystallographic axis is the polariton mode associated with the phonon of symmetry A at 795 cm^{-1}. Through the strong competition between the modes in a stimulated process under long-pulse excitation with an extended interaction length one can expect only the mode with highest gain to be observed. The wavelength of the scattered Stokes light was measured by a monochromator with a spectral resolution of 6 cm^{-1}.

Figure 3 shows the results obtained with the polarization of the pump beam perpendicular (ordinary polarization) and parallel (extraordinary polarization) to the optical axis of the crystal. Collinear scattering with $\gamma = 0$ was performed with the usual Gaussian beam focused into the crystal. The threshold of SRS pumped with the Bessel beam was comparable with that pumped with a Gaussian beam and was near the damage threshold of the crystal. With the Bessel pump beam at 532 nm used in our experiments the Raman shift can be tuned from 760 to 795 cm^{-1} for ordinary incident light and from 738 to 795 cm^{-1} for extraordinary light. The error bars include the finite resolution of the monochromator (6 cm^{-1}) and the statistical error that is due to the pulse-to-pulse energy variations. Within this accuracy the wavelength of the Stokes output was determined from the monochromator readout. The solid curves represent what one would expect from the known polariton dispersion relation.[10] To calculate the expected values of the Stokes frequency, we used the condition of both energy and momentum conservation:

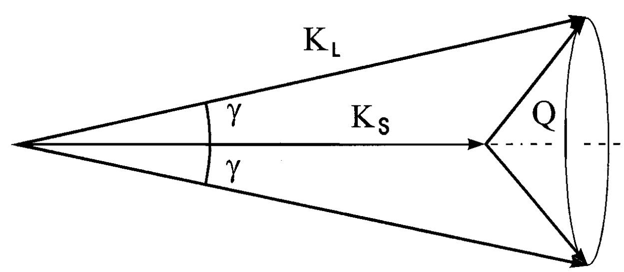

Fig. 2. Wave-vector diagram of forward Stokes beam generation. $\mathbf{K}_L$, $\mathbf{K}_S$, and $\boldsymbol{Q}$ are the wave vectors of the pump, Stokes, and material excitation waves, respectively.

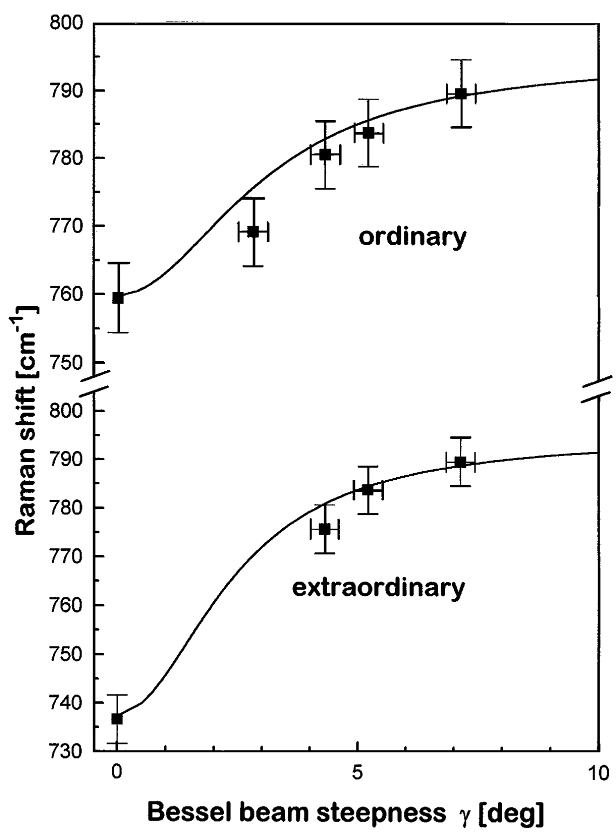

Fig. 3. Tunable SRS from polaritons in a $LiIO_3$ crystal. The Raman Stokes shift is plotted relative to the steepness γ of the Bessel beam for ordinary and extraordinary polarizations of the pump beam. The solid curves are the calculated tuning curves.

$$\omega_S + \Omega = \omega_L\,, \tag{3}$$

$$\mathbf{k}_S(\omega_S) + \mathbf{Q}(\Omega) = \mathbf{k}_L(\omega_L)\,, \tag{4}$$

where Ω is the polariton mode frequency. Equations (3) and (4) can be rewritten as

$$Q^2(\Omega) = k_L^2(\omega_L) + k_S^2(\omega_L - \Omega) - 2k_L(\omega_L)k_S(\omega_L - \Omega)\cos\,\gamma\,, \tag{5}$$

where γ is the angle between pump and Stokes light. Using the refractive indices and the known polarition dispersion,[10] we can solve Eq. (5) for γ.

The available tuning range is determined by the difference between the phonon frequency of 795 cm^{-1} and the Raman shift for straightforward scattering with usual pump beam ($\gamma = 0$). The tuning range is not large because of the nearly flat region of the polariton dispersion for 532-nm pump wavelength. It might be extended by use of a Raman medium with stronger dispersion for the given pump wavelength.

In conclusion, a novel pumping scheme for SRS has been demonstrated. The noncollinear scattering of plane waves that constitute the Bessel beam is identified as the mechanism for the Stokes light generation. The main advantages of this scheme are the simple optical setup, the geometrical separation of pump and Stokes, the nearly diffraction-limited Stokes output, and the possibility of tuning without the use of resonators.

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