Selective nonlinear spectroscopy of inhomogeneously broadened phonon resonances in a disordered medium

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Panoramic Stokes and anti-Stokes scattering spectra have been measured in glass fiber lightguides with biharmonic monochromatic pumping. The results can be used to develop tunable narrow-band subnanosecond sources which emit in the Stokes and anti-Stokes spectral regions with respect to the pump. An arrangement for a selective stimulated-Raman gain is discussed. This arrangement has yielded the first measurements of the homogeneous broadening (~ 16 cm^{-1}) of an inhomogeneously broadened phonon vibration ($\nu \approx 460$ cm^{-1}) in amorphous SiO_2.

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The samples in these experiments, single-mode and few-mode glass fiber lightguides with the composition $SiO_2 + 3\%$ GeO_2 by weight, exhibit an extended, inhomogeneously broadened vibrational spectrum extending to ~ 1300 cm^{-1} (Fig. 2; see also Ref. 1). The glass fiber lightguide has the advantage of a high concentration of light without any limitation on the length of the sample; furthermore, intermode phase matching can be achieved over large interaction lengths.[2,3] The biharmonic pump

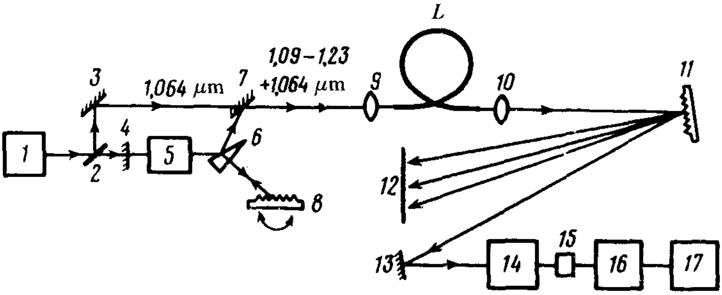

FIG. 1. Experimental arrangement. 1—LTIPCh-8 YAG:Nd^{3+} laser; 2—beam splitter; 3, 13—total-reflection mirrors; 4, 7—semitransparent mirrors; 5—LiF:F_2^- crystal; 6—expanding prism; 8, 11—diffraction gratings; 9, 10—lenses; L—fiber lightguide; 12—screen; 14—MDR-23; 15—FEU-28; 16—PAR-162 integrator; 17—chart recorder.

source (Fig. 1) consists of a pulsed YAG:Nd^{3+} laser ($\lambda_1 = 1.0642$ μm) and a pulsed LiF:F_2^- tunable crystal laser ($\lambda_2 = 1.09$–1.23 μm, $\Delta\nu < 1$ cm^{-1}) (Ref. 4). Figure 1 shows the arrangement for visual observation and photography of the scattering spectra (the spectral width of the monochromator slit is $\leqslant 1$ cm^{-1}). Figure 2a is a panoramic scattering spectrum obtained during biharmonic pumping (ν_1, $\nu_2 =$ const) of a few-mode lightguide ~ 3 m long. The intense anti-Stokes component with the frequency $\nu_a = 2\nu_1 - \nu_2$ and a narrow spectrum (< 1.5 cm^{-1}) results from the choice of phase-matching conditions $\mathbf{K}_a = 2\mathbf{K}_1 - \mathbf{K}_2$ during the selective excitation of two waveguide modes (LP_{01} and LP_{11}) with different propagation constants. Here the pump wave ν_1 is propagating in both the LP_{01} and LP_{11} modes, while the wave ν_2 and the anti-Stokes wave are propagating separately in a mode with a lower phase constant (LP_{11}) and a higher phase constant (LP_{01}), respectively.[3] Under these conditions we achieved effec-

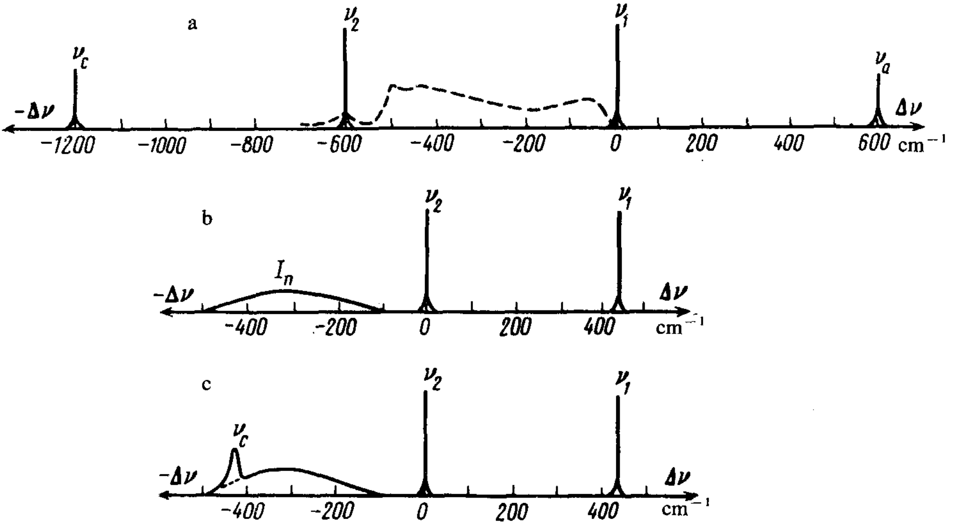

FIG. 2. Laser pumping and scattering spectra at the exit from a few-mode glass fiber lightguide (the dashed curve is the Raman spectrum of fused quartz).

tive phase matching for all three waves in fiber lightguides up to 30 m long with continuous tuning $\nu_a - \nu_l = \nu_l - \nu_2$ from 390 to 650 cm^{-1} (λ_a = 1.0218–0.9954 μm). In addition to the coherent anti-Stokes Raman scattering (CARS) which we have just described, the scattering spectrum (Fig. 2a) contains an intense narrow-band Stokes component$^{1)}$ with a frequency $\nu_S = 2\nu_2 - \nu_1$. This Stokes components is also observed in a single-mode glass fiber lightguide, in contrast with the CARS. The high efficiency of this transformation could not result from the satisfaction of the phase-matching condition[3]; it may be interpreted as either an ordinary stimulated-Raman amplification of selective Stokes scattering or an amplification of nonselective Raman scattering by selectively excited phonon modes $\nu_{\rm ph} = \nu_1 - \nu_2$ (selective stimulated-Raman amplification). The extended vibrational spectrum of the quartz allowed us to continuously tune the frequency of the Stokes component, ν_S, from 1.116 to 1.221 μm (the IR limit of the photographic system) by varying ν_2. This tuning band could easily be expanded beyond 1.3 μm into the region of the anomalous constitutive dispersion of SiO_2. Figures 3a and 3b are oscilloscope traces of the pump pulses at the entrance to the fiber.

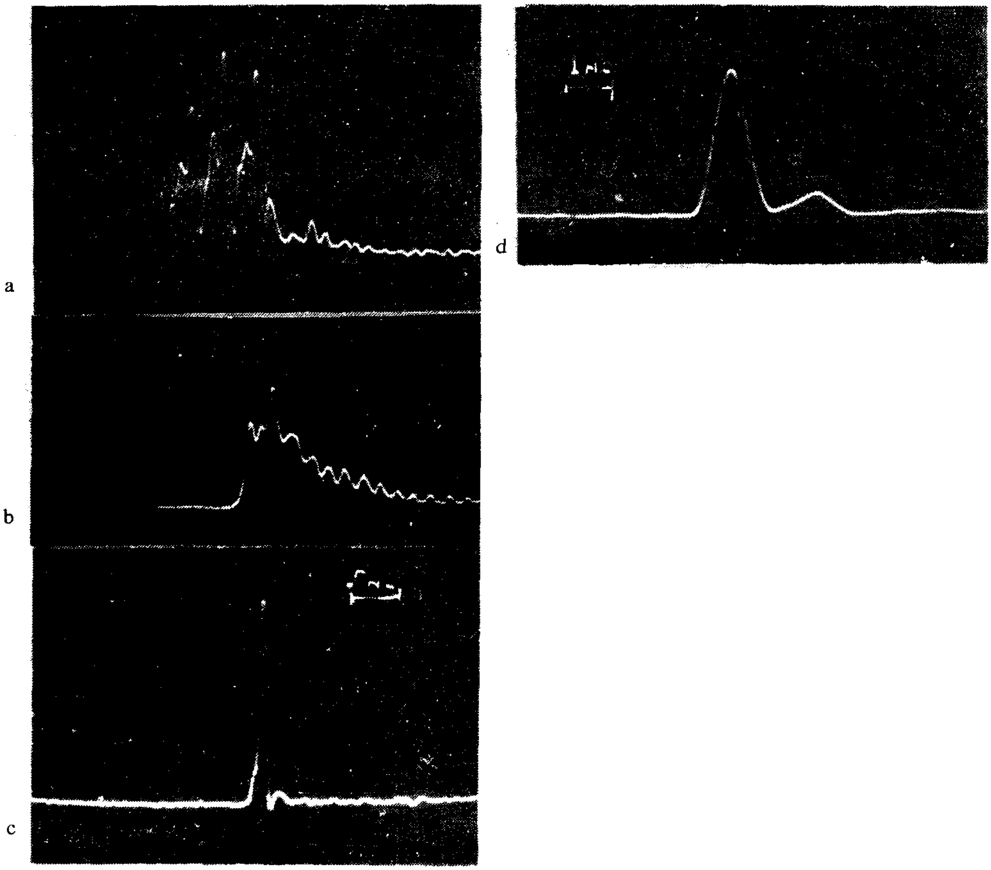

FIG. 3. Oscilloscope traces. a—The beam from the YAG:Nd^{3+} laser; b—that from the LiF:F_2^- laser; c, d—the stimulated-Raman amplification (Tektronix 7104, Ge avalanche photodiode).

Both pulses have the characteristic peak structure, and the ν_2 pulse is delayed ~ 10 ns with respect to the ν_1 pulse. The time at which ν_S appears (Fig. 3c) is determined by the overlap of the pump pulses, and in several cases there is a single peak <0.7 ns long (Fig. 3d; the resolution of the detection system). The oscilloscope trace of the anti-Stokes scattering is similar in shape. We may thus conclude that biharmonic pumping of few-mode and single-mode glass fiber lightguides can be used to develop a continuously tunable narrow-band subnanosecond light source which operates in the anti-Stokes parts of the spectrum with respect to the pump.

Under the assumption that the narrow width (<2 cm^{-1}) of the ν_S line results from distortion of the homogeneously broadened phonon-resonance line at large selective stimulated-Raman gain values $\alpha(\nu) \gg 1$ for the weak spontaneous scattering (large values of I_1 and I_2), we introduced a wide-band probe signal $I_p \ll I_1$, I_2 (Fig. 2b), obtained through generation at the faces of the LiF:F_2^- crystal (Fig. 1). This signal was used, in addition to the light at the frequencies ν_1 and ν_2, in order to satisfy the condition[5] $\alpha(\nu) < 3$ in a lightguide ~ 2 m long. As a result, we observed at the exit from the fiber a narrow selective stimulated-Raman-gain component $I_p(\nu)$ (Fig. 2c) against the background of the nonselectively amplified wide-band probe signal. The shape of the narrow component apparently corresponds to homogeneous broadening ($\delta\nu \approx 16$ cm^{-1}) of an inhomogeneous phonon resonance (with $\nu \approx 460$ cm^{-1}) excited by the biharmonic field. With increasing $\alpha(\nu)$ (with increasing I_1 and I_2) we observe a characteristic contraction of the narrow component, confirming that selective stimulated-Raman amplification is occurring and indicating that saturation has not been reached. Saturation would also lead to a spectral broadening of both the Stokes and anti-Stokes scattering.

We might note that, by analogy with the linear spectroscopy of the selective laser excitation of inhomogeneous resonances,[6,7] the use of selective biharmonic excitation in the nonlinear spectroscopy of disordered media can make it possible to study in more detail the structure of inhomogeneous broadened spectra in measurements of panoramic spectra of coherent and incoherent scattering (in the Stokes and anti-Stokes regions). Specifically, it becomes possible to distinguish the homogeneous broadening, the frequencies and relationships between the various modes, and the velocities and relaxation channels for the excitations in the medium.

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[1)] The energy of the Stokes wave, E_S, was measured by a pyroelectric detector (Molectron J3-05) and found to range up to 38% of the energy $E_1 \approx E_2$.

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[1] G. E. Walrafen and P. N. Krishnan, Appl. Opt. **21**, 359 (1982).
[2] R. H. Stolen, J. E. Bjorkholm, and A. Ashkin, Appl. Phys. Lett. **24**, 308 (1974).
[3] E. M. Dianov, É. A. Zakhidov, A. Ya. Karasik, P. V. Mamyshev, and A. M. Prokhorov, Pis'ma Zh. Eksp. Teor. Fiz. **34**, 40 (1981) [JETP Lett. **34**, 38 (1981)]; Zh. Eksp. Teor. Fiz. **83**, 39 (1982) [Sov. Phys. JETP **56**, 21 (1982)].
[4] T. T. Basiev, Yu. K. Voron'ko, S. B. Mirov, V. V. Osiko, A. M. Prokhorov, M. S. Soskin, and V. B. Taranenko, Kvant. Elektron. (Moscow) **9**, 1741 (1982) [Sov. J. Quantum Electron. **12**, 1125 (1982)].

[5] N. Bloembergen, Nonlinear Optics, Benjamin, New York, 1965 (Russ. Transl. Mir, Moscow, 1969).
[6] T. T. Basiev, Yu. K. Voron'ko, and A. M. Prokhorov, Spektroskopiya kristallov (Spectroscopy of Crystals), Nauka, Moscow, 1978, p. 83.
[7] T. T. Basiev, Yu. K. Voron'ko, A. Ya. Karasik, V. V. Osiko, and I. A. Sherbakov, Zh. Eksp. Teor. Fiz. **75**, 66 (1978) [Sov. Phys. JETP **48**, 32 (1978)].

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