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SECOND WORKSHOP ON

OPTICAL FIBRE COMMUNICATION

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SELECTIVE NONLINEAR SPECTROSCOPY OF INHOMOGENEOUSLY BROADENED PHONON RESONANCES IN A DISORDERED MEDIUM

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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 ($\sim 16\ \mathrm{cm}^{-1}$) of an inhomogeneously broadened phonon vibration ($\nu \approx 460\ \mathrm{cm}^{-1}$) in amorphous SiO_2.

PACS numbers: 42.65.Cq, 78.30.Gt

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 $\sim 1300\ \mathrm{cm}^{-1}$ (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

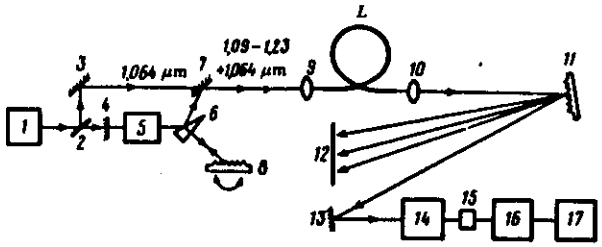

FIG. 1. Experimental arrangement. 1—LTIPCh-8 YAG:Nd^{3+} laser; 2—beam splitter; 3, 13—total-reflection mirrors; 4, 7—semitransparent mirrors; 5—LiF:F_2^- crystal; 6—expanding prism; 8, 11—diffraction gratings; 9, 10—lenses; *L*—fiber lightguide; 12—screen; 14—MDR-23; 15—FEU-28; 16—PAR-162 integrator;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

source (Fig. 1) consists of a pulsed YAG:Nd^{3+} laser ($\lambda_1 = 1.0642\ \mu\mathrm{m}$) and a pulsed LiF:$F_2^-$ tunable crystal laser ($\lambda_2 = 1.09$–$1.23\ \mu\mathrm{m}$, $\Delta\nu < 1\ \mathrm{cm}^{-1}$) (Ref. 4). Figure 1 shows the arrangement for visual observation and photography of the scattering spectra (the spectral width of the monochromator slit is $\leqslant 1\ \mathrm{cm}^{-1}$). Figure 2a is a panoramic scattering spectrum obtained during biharmonic pumping (ν_1, $\nu_2 = \mathrm{const}$) 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 ($< 1.5\ \mathrm{cm}^{-1}$) 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-

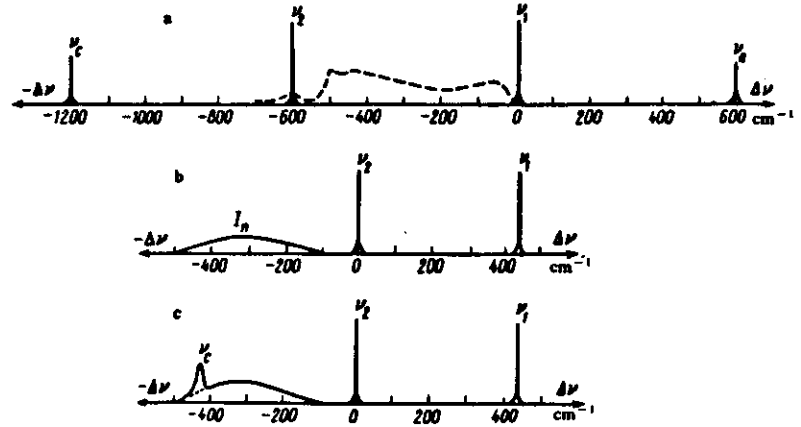

FIG. 2. Laser pumping and scattering spectra at the exit from a few-mode glass fiber lightguide (the dashed curve is the Raman spectrum of fused quartz).

tive phase matching for all three waves in fiber lightguides up to 30 m long with continuous tuning $\nu_a - \nu_1 = \nu_1 - \nu_2$ from 390 to 650 cm^{-1} ($\lambda_a = 1.0218$–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 $\nu_{ph} = \nu_1 - \nu_2$ (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.

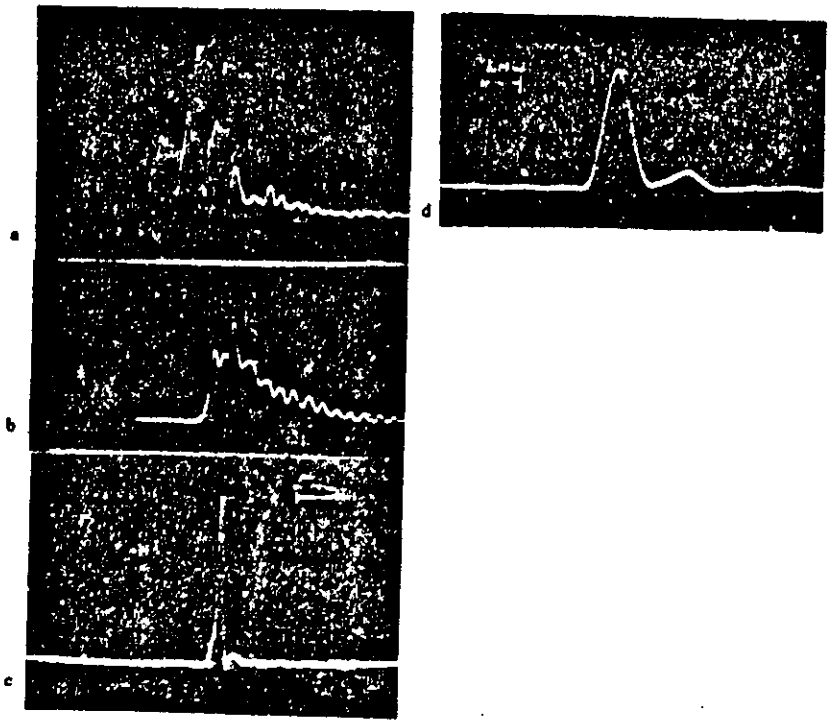

FIG. 3. Oscilloscope traces. a—The beam from the YAG:Nd^{3+} laser; b—that from the LiF:F_2^- laser; c, d—the stimulated-Raman amplification (Tektronix 7104, Ge avalanche photodiode).

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.

We wish to thank A. N. Gur'yanov and D. D. Gusovskiĭ for fabricating the lightguides.

[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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Parameters of the six-quark component of the deuteron

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New data are reported on the momentum distribution of the nucleons in the deuteron. The parameters of the six-quark component of the deuteron are determined. The use of this component leads to a good description of accurate data on deuteron stripping by nuclei (from Dubna) and on the electrodisintegration of deuterons (from Stanford).

PACS numbers: 27.10. + h, 25.10. + s, 25.45.Gh, 12.35.Ht

1. Our data on the momentum spectra of the protons emitted at a zero angle during the fragmentation of relativistic deuterons (8.9 GeV/c) by nuclei, obtained on the JINR proton synchrotron, were reported in Refs. 1 and 2 (preliminary results have been reported at international conferences[3]). These spectra can be used to find the momentum distribution of the nucleons in the deuteron, through the use of an expression derived by a relativistic approach:[4]

$$E\,d^3\sigma/d\mathbf{p} = \frac{1}{8\pi}\sigma_{NA}^{in}\frac{\sigma_{dA}^{in}}{\sigma_{dA}^{T}}\sqrt{\frac{m^2}{4\alpha(1-\alpha)^3}}\,F(\alpha)|\psi(k^2)|^2. \qquad (1)$$

Here σ_{dA}^{in} and σ_{NA}^{in} are the total inelastic cross sections for dA and NA interactions; σ_{dA}^{T} is the total dA cross section; $k^2 = (m^2 + p_1^{*2})/[4\alpha(1-\alpha)] - m^2$; α is the light-front variable, given by $\alpha = (E_p^* + p_\parallel^*)/M_d$; M_d and m are the masses of the deuteron and the proton; p^* is the momentum of the spectator proton in the rest frame of the deuteron; and $\psi(k^2)$ is the overlap integral of the deuteron wave function and the unbound np state (which is the same as the deuteron wave function in the momentum

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Stimulated parametric four-photon mixing in glass fibers

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The generation of Stokes–anti-Stokes pairs with frequency shifts up to 5000 cm^{-1} has been observed in laser-pumped optical fiber. The observed effects are explained in terms of phase-matched four-photon mixing.

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The ability of glass fiber lightguides to withstand intense light over long distance makes them uniquely suitable for research on certain nonlinear effects. They are presently being used in very active research on such nonlinear effects as stimulated Raman scattering,[1,2] stimulated Brillouin (or "stimulated Mandel'shtam–Brillouin") scattering, and the optical Kerr effect.[3] Stimulated parametric four-photon mixing is a nonlinear process in which two pump photons ω_{p1} and ω_{p2} are converted into Stokes (ω_S) and anti-Stokes (ω_A) photons ($\omega_A > \omega_p > \omega_S$); from energy conservation we have $\omega_{p1} + \omega_{p2} = \omega_A + \omega_S$. This four-photon process requires a phase matching; the corresponding matching condition is written in terms of the wave vectors as $\mathbf{k}_{p1} + \mathbf{k}_{p2} = \mathbf{k}_A + \mathbf{k}_S$. In a medium exhibiting a normal "material" dispersion, phase matching is impossible in a collinear interaction of plane waves because the condition $\mathbf{k}_A + \mathbf{k}_S > 2\mathbf{k}_p$ always holds. In glass fibers the different modes have different propagation constants β_{mn} (Fig. 1a), so that if the photons ω_p, ω_A, and ω_S propagate in different modes it may be possible, in principle, to achieve phase matching by offsetting the dispersion of the medium by a modal dispersion. In a glass fiber, a

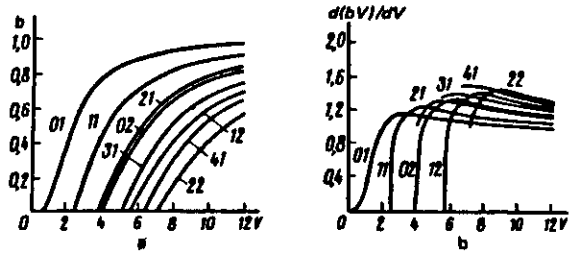

FIG. 1. a–Normalized effective refractive index b for various LP modes as a function of the characteristic parameter $V = 2\pi\bar{\nu}a(n_1^2 - n_2^2)^{1/2}$. Here a is the fiber radius,[7] $b_{mn} = (\beta_{mn}/k - n_2)/(n_1 - n_2)$; and $\Delta_n = n_1 - n_2$ is the difference between the refractive indices of the cladding and the core; b–the normalized group delay $d(bV)/dV$ for various LP modes as a function of the characteristic parameter V (Ref. 5).

stimulated parametric four-wave process of this type with frequency shifts $\Delta\bar{\nu} < 400$ cm^{-1} has been observed and analyzed[4] ($\Delta\bar{\nu} = \bar{\nu}_A - \bar{\nu}_p = \bar{\nu}_p - \bar{\nu}_S$).

The phase-matching condition for a glass fiber is

$$\beta_A + \beta_S - \beta_{p1} - \beta_{p2} = \Delta K(\Delta\bar{\nu}) + f(\Delta\bar{\nu}) = 0, \quad (1)$$

where $\Delta K(\Delta\bar{\nu})$ describes the dispersion of the medium, and

$$f(\Delta\bar{\nu}) = 2\pi\Delta n\left[(b_A + b_S - b_{p1} - b_{p2}) + \left(\frac{d(b_A V)}{dV} - \frac{d(b_S V)}{dV}\right)\Delta\bar{\nu}\right]. \quad (2)$$

For the present experiments we selected glass fibers with a $SiO_2 + GeO_2$ core and a SiO_2 cladding with $\Delta n \approx 4.5 \times 10^{-3}$ and $2a \approx 9$ μm. The fiber was pumped with a second harmonic from a neodymium garnet laser ($\lambda_p = 532$ nm) operating under periodic-pulse conditions with $F = 12.5$–25 Hz and $\tau_{pulse} = 15$ ns. For this wavelength λ_p, the parameter V of the fiber is $V \approx 6$. The laser beam was coupled into the fiber with the help of an objective; a second objective at the exit from the fiber sent the beam either to a monochromator, for spectral measurements, or to a diffraction grating and then to a screen or to photographic film.

Figure 2a is a photograph of the near field of the beam emerging from an ~6-m-long fiber after spectral decomposition by a diffraction grating. In addition to the pump frequency (p), we see an anti-Stokes component (A) with $\bar{\nu}_A > \bar{\nu}_p$ and a Stokes component (S) with $\bar{\nu}_S < \bar{\nu}_p$. The field distributions of these components correspond to LP_{01} and LP_{11} modes, respectively, and the frequency shift is $\Delta\bar{\nu} = \bar{\nu}_A - \bar{\nu}_p = \bar{\nu}_p - \bar{\nu}_S \approx 105$ cm^{-1}. By varying the angle at which the beam entered the fiber, and thereby exciting different groups of modes preferentially, we obtained different Stokes–anti-Stokes modal compositions with different shifts $\Delta\bar{\nu}$ [in Fig. 2b, A(LP_{11}) and S(LP_{21}) with $\Delta\bar{\nu} \approx 110$ cm^{-1}; in Fig. 2c, A(LP_{21}) and S(LP_{31}) with $\Delta\bar{\nu} \approx 67$ cm^{-1}]. In all the photographs in Fig. 2 the Stokes components are slightly more intense than the anti-Stokes components, and there is some spectral "smearing," apparently due to a variation of the diameter of the fiber along its length.[4] In the cases studied, the Stokes component always corresponds to a mode of higher order than that corresponding to the anti-Stokes component. One of the pump photons is propagating in the same mode as the Stokes photon, while the other pump photon is propagating in the same mode as the anti-Stokes photon. In this case, Eq. (2) simplifies:

$$f(\Delta\bar{\nu}) = 2\pi\Delta n\left(\frac{d(b_A V)}{dV} - \frac{d(b_S V)}{dV}\right)\Delta\bar{\nu}. \quad (2')$$

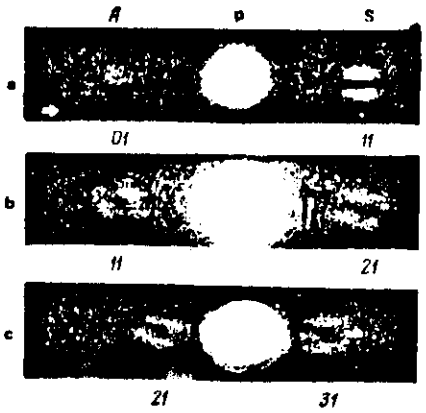

FIG. 2. Photographs of the stimulated, parametric, four-photon emission at the exit from a glass fiber ~6 m long after spectral decomposition by a diffraction grating. p, A, S–Near fields of the pump, anti-Stokes component, and Stokes component, respectively.

Constructing the function $\Delta K(\Delta\bar{\nu})$ [expression (1)] from data on the dispersion of fused quartz,[6] allowing for the very low GeO_2 content in our fiber, and knowing the experimental values of $\Delta\bar{\nu}$, we found the corresponding value of $\Delta K(\Delta\bar{\nu})$, which is equal to $-f(\Delta\bar{\nu})$ at phase matching, for each case (Fig. 2). Accordingly, by knowing only Δn and measuring $\Delta\bar{\nu}$, we were able to calculate the difference between the group delays for the different modes from (2') and thus estimate the parameter V from Fig. 1b. For cases a, b, and c in Fig. 2, we found values of 6, 5.5, and 6.1, respectively, for V. The apparent reason for the differences in these values of V is a discrepancy between the actual index profile of our fiber and the stepped profile for which the results in Fig. 1 were calculated.

In fibers with lengths in the range 0.3–1 m we observe the generation of two Stokes–anti-Stokes pairs corresponding to LP_{11} modes with approximately equal frequency shifts, $\Delta\bar{\nu}_1 \approx 1950$ cm^{-1} and $\Delta\bar{\nu}_2 \approx 2100$ cm^{-1} (Fig. 3). The direction of the lobes of the LP_{11} modes of one pair is orthogonal to the lobe direction of the other pair. In contrast with stimulated parametric four-photon processes with small values of $\Delta\bar{\nu}$ (Fig. 2), there are no significant differences between the intensities of the Stokes and anti-Stokes components within each pair. The pump power corresponding to the generation threshold was measured at the exit from a fiber 0.6 m long and found to be ~1 kW. Two cases may actually correspond to the given ex-

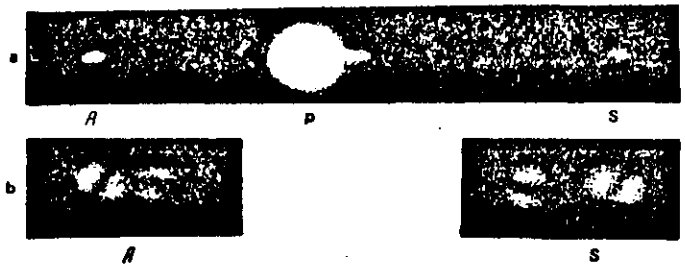

FIG. 3. Photographs of the stimulated, parametric, four-photon emission at the exit from a fiber 0.6 m long after spectral decomposition by a diffraction grating. p, A, S—Near fields of the pump, anti-Stokes, and Stokes components, respectively. a—General view (the spectrum of stimulated Raman scattering is seen at the right of the pump); b—Stokes and anti-Stokes components with a better spectral resolution than in photograph a.

perimental situation, in which the Stokes and anti-Stokes photons are propagating in the LP_{11} mode: 1) Both of the pump photons are in the LP_{01} mode; 2) one of the pump photons is in the LP_{01} mode, while the other is in the LP_{11} mode. The frequency shift $\Delta\bar{\nu}$ calculated from (2), with allowance for the functional dependence $\Delta K(\Delta\bar{\nu})$ and the data in Fig. 1a, agrees well with the experimental shift for case 2) but is approximately twice as high as the experimental value for case 1). It should be noted, however, that in case 2) the overlap integral of the fields of the four modes involved in the process (the polarization of the mode is taken into account) is zero; this integral determines, in particular, the efficiency of the stimulated parametric four-photon mixing in a glass fiber.[4]

The generation of two Stokes–anti-Stokes pairs with different values of $\Delta\bar{\nu}$ (Fig. 3) is evidently a consequence of different values of the phase constants β for the LP_{11} modes, which may in turn be a result of birefringence in the fiber, i.e., the lifting of the degeneracy between the two orthogonal polarizations of the LP mode because of an ellipticity or anisotropy of n_1 in the lightguide core.[7] Further evidence for this conclusion comes from the fact that we were able to achieve preferential generation of one of the two Stokes–anti-Stokes pairs by using a $\lambda/2$ plate to rotate the polarization direction of the laser beam at the entrance to the fiber. Even further, when we used a fiber with a definitely low birefringence[7] we did not observe a splitting of the LP_{11} mode. We do note, however, that the polarization of the LP_{11} modes generated in our case was elliptical in all cases.

We also observed generation of Stokes–anti-Stokes pairs with $\lambda_S \approx 696.7$ nm, $\lambda_A \approx 430.3$ nm ($\Delta\bar{\nu} \approx 4400$ cm^{-1}); $\lambda_S \approx 721$ nm, $\lambda_A \approx 421.5$ nm ($\Delta\bar{\nu} \approx 4939$ cm^{-1}). In this case four or five modes, distributed over the spectrum, participated simultaneously in the generation. In the anti-Stokes region, for example, we found the LP_{41} and LP_{22} modes, which, incidentally, do not propagate in this fiber at the pump frequency because of the cutoff condition (Fig. 1a). The coherence length of the stimulated parametric four-photon mixing, which is limited by the inhomogeneity of the properties of the fiber along its length, turned out to be extremely large for large values of $\Delta\bar{\nu}$ (~0.5 m), albeit much smaller than in the case of stimulated parametric four-photon mixing with small values of $\Delta\bar{\nu}$ (more than 5 m).

We note in conclusion that by using fibers with different parameters this four-photon mixing could be exploited to generate coherent light with new frequencies. From a different standpoint, the method is convenient for selective excitation of individual modes, and it furnishes valuable information on the characteristics of the fiber (the phase constants, the group delays, the birefringence, etc.).

We thank A. N. Gur'yanov and D. D. Gusovskiĭ for fabricating the fibers.

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