## Stimulated parametric four-photon mixing in glass fibers

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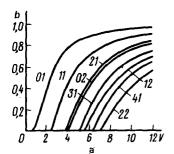
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The generation of Stokes-anti-Stokes pairs with frequency shifts up to 5000 cm<sup>-1</sup> has been observed in laser-pumped optical fiber. The observed effects are explained in terms of phase-matched four-photon mixing.

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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. Stimulated parametric four-photon mixing is a nonlinear process in which two pump photons  $\omega_{p1}$  and  $\omega_{p2}$  are converted into Stokes ( $\omega_{S}$ ) and anti-Stokes ( $\omega_{A}$ ) 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  $\beta_{mn}$  (Fig. 1a), so that if the photons  $\omega_{p}$ ,  $\omega_{A}$ , and  $\omega_{S}$  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



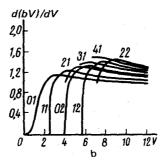


FIG. 1. a-Normalized effective refractive index b for various LP modes as a function of the characteristic parameter  $V=2\pi\overline{\nu}a\ (n_1^2-n_2^2)^{1/2}$ . Here a is the fiber radius,  $b_{mn}=(\beta_{mn}/k-n_2)/(n_1-n_2)$ ; and  $\Delta_n=n_1-n_2$  is the difference between the refractive indices 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  $\Delta \overline{\nu} < 400$  cm<sup>-1</sup> has been observed and analyzed<sup>4</sup> ( $\Delta \overline{\nu} = \overline{\nu}_A - \overline{\nu}_p = \overline{\nu}_p - \overline{\nu}_S$ ).

The phase-matching condition for a glass fiber is

$$\beta_{\Lambda} + \beta_{S} - \beta_{p1} - \beta_{p2} = \Delta K(\Delta \overline{\nu}) + f(\Delta \overline{\nu}) = 0, \tag{1}$$

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

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

For the present experiments we selected glass fibers with a SiO<sub>2</sub> + GeO<sub>2</sub> core and a SiO<sub>2</sub> cladding with  $\Delta n \approx 4.5 \times 10^{-3}$  and  $2a \approx 9~\mu m$ . 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  $\lambda_p$ , 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  $\sim$ 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  $\overline{\nu}_A > \overline{\nu}_p$  and a Stokes component (S) with  $\overline{\nu}_S < \overline{\nu}_p$ . The field distributions of these components correspond to  $LP_{01}$  and  $LP_{11}$  modes, respectively, and the frequency shift is  $\Delta \overline{\nu} = \overline{\nu}_A - \overline{\nu}_p = \overline{\nu}_p - \overline{\nu}_S \approx 105 \text{ cm}^{-1}$ . 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  $\Delta \overline{\nu}$  [in Fig. 2b,  $A(LP_{11})$  and  $S(LP_{21})$  with  $\Delta \overline{\nu} \approx 110 \text{ cm}^{-1}$ ; in Fig. 2c,  $A(LP_{21})$  and  $S(LP_{31})$  with  $\Delta \overline{\nu} \approx 67 \text{ cm}^{-1}$ ]. 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. In the cases studied, the Stokes component always corresponds to a mode of higher or-

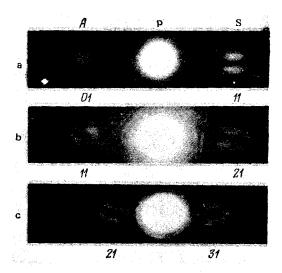


FIG. 2. Photographs of the stimulated, parametric, four-photon emission at the exit from a glass fiber ~6 m long after spectral decomposition by a diffraction grating. p, A, S-Near fields of the pump, anti-Stokes component, and Stokes component, respectively.

der 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:

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

Constructing the function  $\Delta K(\Delta \overline{\nu})$  [expression (1)] from data on the dispersion of fused quartz, <sup>6</sup> allowing for the very low  $\text{GeO}_2$  content in our fiber, and knowing the experimental values of  $\Delta \overline{\nu}$ , we found the corresponding value of  $\Delta K(\Delta \overline{\nu})$ , which is equal to  $-f(\Delta \overline{\nu})$  at phase matching, for each case (Fig. 2). Accordingly, by knowing only  $\Delta n$  and measuring  $\Delta \overline{\nu}$ , 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,  $\Delta \overline{\nu}_1 \approx 1950~{\rm cm}^{-1}$  and  $\Delta \overline{\nu}_2 \approx 2100~{\rm cm}^{-1}$  (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  $\Delta \overline{\nu}$  (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  $\sim$ 1 kW. Two cases may actually correspond to the given ex-

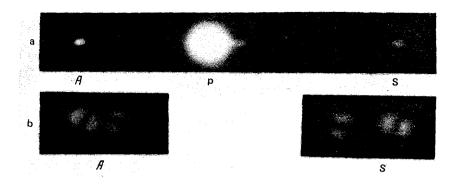


FIG. 3. Photographs of the stimulated, parametric, four-photon emission at the exit from a fiber 0.6 m long after spectral decomposition by a diffraction grating. p, A, S—Near fields of the pump, anti-Stokes, and Stokes components, respectively. a—General view (the spectrum of stimulated Raman scattering 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  $\Delta \overline{\nu}$  calculated from (2), with allowance for the functional dependence  $\Delta K(\Delta \overline{\nu})$  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.<sup>4</sup>

The generation of two Stokes—anti-Stokes pairs with different values of  $\Delta \overline{\nu}$  (Fig. 3) is evidently a consequence of different values of the phase constants  $\beta$  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. 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 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  $\lambda_{\rm S}\approx 696.7$  nm,  $\lambda_{\rm A}\approx 430.3$  nm ( $\Delta\overline{\nu}\approx 4400$  cm<sup>-1</sup>);  $\lambda_{\rm S}\approx 721$  nm,  $\lambda_{\rm A}\approx 421.5$  nm ( $\Delta\overline{\nu}\approx 4939$  cm<sup>-1</sup>). 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  $\Delta \overline{\nu}$  (~0.5 m), albeit much smaller than in the case of stimulated parametric four-photon mixing with small values of  $\Delta \overline{\nu}$  (more than 5 m).

We note in conclusion that by using fibers with different parameters this fourphoton 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.).

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