Influence of parametric processes on the generation of Stokes components of stimulated Raman scattering under biharmonic pumping

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We have investigated experimentally and theoretically the mixed Raman-parametric generation of Stokes components under biharmonic pumping. We show that in the presence of a strong pump component the power of the weak component, which is needed for the observation of the stimulated Raman scattering, can be decreased by a factor of 10^4. The observed effect can be explained on the basis of phase locking in four-wave interaction.

1. Recently there have been many studies of four-wave resonant processes, a characteristic feature of which is the interaction of parametric (i.e., dependent on the phase relations) and Raman (two-photon) processes.[1] In all the investigated cases, the generated wave is produced because of parametric interaction, and the two-photon processes lead to absorption of this wave; the pump induces a transition of the medium from the ground state to an excited state, and the parametric process returned in the medium to the initial state. There is, however, a possible process in which the generated wave is increased both by the Raman and by the parametric interaction of the fields. This process is stimulated emission at Stokes frequencies by

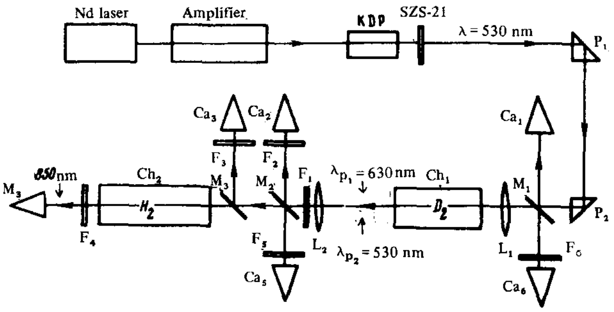

FIG. 1. Block diagram of experimental setup: Ch_1, Ch_2) Chambers with compressed D_2 and H_2; M_1—M_2) mirrors; P_1, P_2) prisms; L_1, L_2) lenses; Ca_1—Ca_6) calorimeters; F_1—F_6) light filters.

biharmonic excitation, in which case the medium goes over to an excited state. In this communication we present the results of a theoretical and experimental investigation of this phenomenon.

2. The interaction of a biharmonic pump $\sum_{j=1,2} \mathbf{e}_j A_j(z) \times \exp\{i[\omega_j t - K_j z - \phi_j(z)]\} + \text{c.c.}$ with a Raman-active transition (of frequency ω_{21}) can be investigated by the method described in[2]. The results of the theoretical analysis are presented below.

Let the intensity of one of the pump components, say $I_2 \sim A_2^2$, be sufficient to excite stimulated Raman scattering (SRS), i.e., let a Stokes component be produced with frequency ω_{2S} and amplitude $A_{2S}(z)$. This leads to parametric interaction of the waves at the frequencies ω_1, ω_2, and ω_{2S}, as a result of which a wave with frequency ω_{1S} is generated $(\omega_1 - \omega_{1S} = \omega_2 - \omega_{2S} = \omega_{21})$. Its amplitude A_{1S} increases like $A_1 A_2 A_{2S}$. The growth of A_{1S} produces also a stimulated Raman process $(\sim A_1^2 A_{1S}^2)$. As a result, mixed Raman-parametric generation of the field A_{1S} occurs in a certain region of interaction space, and phase locking of the interacting waves takes place in this entire region.[2,3]

Assume that $I_{10} = I_1|_{z=0}$ is much less than the threshold intensity $I_{10}^{\rm thr}$ sufficient to excite SRS in the absence of additional pumping at the frequency ω_2, and that the registered value of the Stokes component is $I_{1S} \approx 0.1 I_{10}$. It can then be shown that the necessary and sufficient condition for obtaining this conversion coefficient over the phase-locking length L is

$$10 A_{2S}^2|_{z=L} \geqslant A_{20}^2 > \delta c^2 k_{2S} \hbar^3 / 2\pi\omega_{2S}^2 N T r_1 r_2, \tag{1}$$

where $\delta = (k_1 - k_{1S}) - (k_2 - k_{2S})$ is the wave detuning, N is the particle-number density, T is the reciprocal of the working-transition line width, $r_j = r_j(\omega_j - \omega_{jS})$ is the second-order polarizability, which determines the SRS cross section at the frequencies ω_1 and ω_2, and $A_{20} = A_2|_{z=0}$.

When (1) is satisfied, the process in question is not critical to satisfaction of the linear-synchronism conditions $(\delta = 0)$, and the length L can greatly exceed δ^{-1}. Therefore the mixed Raman-parametric generation can be observed in collinear beams propagating in an isotropic medium, where $\delta \neq 0$.

The phase locking is bounded in space in this case, as in other four-wave processes.[2,3] An investigation shows that after the phase locking stops, under the condition $A_{10} \ll A_{20}$, the parametric interaction has little effect on the subsequent change of the Stokes component A_{1S}, which proceeds exclusively as a result of the Raman interaction, and almost total conversion of the pump A_1 into the component of frequency ω_{1S} is possible.

Thus, the conversion coefficient in the process considered here, in contrast to other types of four-wave interaction,[2,3] is quite insensitive to the choice of thickness of the medium, which can be much larger than L.

It should be noted that all the foregoing results are valid also in the case when the SRS pumping A_2 is replaced by two-photon absorption between working levels.

3. We investigated experimentally the excitation of Stokes components by biharmonic pumping of compressed hydrogen. The experimental setup is shown in Fig. 1. The strong pump component of frequency ω_2 was the second harmonic of a frequency-stabilized single-mode neodymium laser $(\lambda_2 = 530$ nm); the source of the pump of frequency ω_1, the Stokes component of which was investigated, was the SRS Stokes component $(\lambda_1 = 630$ nm) of the radiation with $\lambda_2 = 530$ nm, obtained in a deuterium chamber Ch_1.

The radiation with λ_1 and the nonconverted part of the second harmonic with $\lambda_2 = 531$ nm were focused by lens L_2 into chamber Ch_2 with compressed H_2. To verify the theoretical conclusion that the process is not critical to satisfaction of the synchronism conditions, the length of the focal region in Ch_2 was of the order of 5 cm, which was much larger than the linear-synchronism length $\delta^{-1} = 0.18$ cm (the H_2 pressure in Ch_2 was 60 atm). The ratio of the entrance pump intensities $I_{10}(\lambda_1)/I_{20}(\lambda_2)$ was varied by a set of filters F_1 and registered with calorimeters Ca_2 and Ca_3. The presence of the Stokes components $I_{1S}(\lambda_{1S} = 850$ nm) in Ch_2 was registered with calorimeter Ca_4 having a threshold sensitivity $(I^{\rm reg})_{\min} = 0.1$ W.

The energy scatter of the pulses of the initial generator (which led also to scatter of I_{10} $(\lambda_1 = 630$ nm)) was such that at an average value $I_{20} = I_{20}^{\rm thr}$ the response sig-

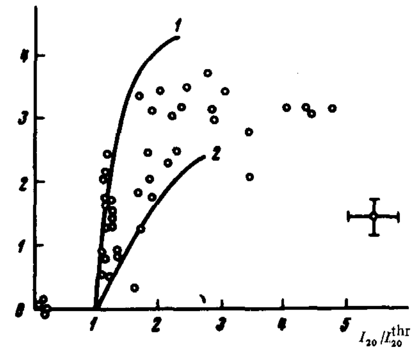

FIG. 2. Decrease of threshold intensity I_{10} as a function of I_{20}: curves 1 and 2 are calculated for $(I_{1S}^{\rm reg})_{\min}$ equal to 0.1 and 5 W, respectively, the SRS threshold is $I_{20}^{\rm thr} = 50$ kW.

nal $(I_{1S}^{\mathrm{reg}})_{\min}$ at $\lambda_{1S} = 850$ nm oscillated between 0.1 W and 5 W. Figure 2 shows the theoretical plots 1 and 2 of $I_{10}(I_{1S}^{\mathrm{reg}})/I_{10}^{\mathrm{thr}} = f(I_{20}/I_{20}^{\mathrm{thr}})$ and the corresponding experimental points registered for a signal with $\lambda_{1S} = 850$ nm at the levels 0.1 and 5 W, respectively. We see that practically all the experimental points lie in the region between these curves.

With increasing I_{20}, practically the entire wave with λ_1 was converted into the Stokes component with $\lambda_{1S} = 850$ nm at powers $I_{10} \approx 0.1$ kW $\ll I_{10}^{\mathrm{thr}} \approx 12$ kW and $I_{20} \approx 150$ kW.

In experiments with opposing beams I_1 and I_2 at $< I_{10}^{\mathrm{thr}}$ the Stokes component A_{1S} was not observed even at $I_{20}/I_{20}^{\mathrm{thr}} = 30$. This proves that the effect is due to parametric interaction of the waves. On the basis of the data presented in this section, we can state that the "lowering of the threshold" of the SRS in[4] is also due to Raman-parametric generation.

The foregoing results show that Raman-parametric generation can ensure production of Stokes components from weak sources (with power density on the order of hundreds of W/cm^2), for example from dye lasers. Thus, new frequency-tunable IR sources can be obtained. By virtue of the much lower sensitivity to the satisfaction of the wave-synchronism conditions (see[1]) than in the other four-wave processes employed for similar purposes, one can expect such devices to have wider frequency-tuning ranges in systems with invariant beam geometry. By way of example of a system suitable for the discussed purposes, we cite KI vapor, in which it is possible to obtain a Stokes component with $\lambda_{1S} = 9000$ nm from a source with $\lambda_1 = 455$ nm via two-photon resonance of an additional pump with $\lambda_2 = 951$ nm; the latter may be a dye laser or a parametric light generator.

A detailed article dealing with mixed Raman-parametric generation under biharmonic pumping will be published by us later.

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