Nondegenerate four-wave mixing participated by two-photon excited stimulated emission

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Two-photon excited stimulated emission at ~617 nm has been achieved in an organic chromophore solution pumped at 1064 nm. The stimulated emission can be observed only in the forward and backward directions and is characterized by its high directionality and spectral narrowing. The solution was illuminated simultaneously with a strong 1064-nm pump beam and a 1064-nm weak signal beam to form an induced Bragg grating. As the strong pump beam itself induced the backward two-photon excited stimulated emission, which was a reading beam, we observed backward nondegenerate four-wave mixing (NFWM). The newly generated wave by the NFWM has the same frequency as the reading beam. Due to the phase mismatching, this newly generated coherent wave was not exactly counter-propagating to the signal wave.

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Four-wave mixing (FWM) is a nonlinear process participated by four waves in a third-order nonlinear optical medium. FWM is one of the approaches that can efficiently produce the backward phase-conjugate beam[1]. specially in a nondegenerate four-wave mixing (NFWM) process, the frequency of the backward phase-conjugate beam is different from that of the signal beam, which can be used in the field of wavelength conversion, image processing etc. Although the nondegenerate four-wave mixing (NFWM) has been studied since 1979[2], here we reported to our knowledge for the first time a NFWM participated by two-photon excited stimulated emission.

In our experiment, the gain medium is a reported organic chromophore[3], trans-4-[p-(N-hydroxyethyl-N-methylamino)styryl]-N-methylpyridinium iodide (ASPI) dissolved in dimethyl sulfoxide (DMSO). There is no linear absorption for ASPI in the wide spectral range from 620 to 1600 nm[4]. However, when a focused pulse laser beam with wavelength of 1064 nm passes through an ASPI solution in DMSO, an orange-red fluorescence induced by two-photon excitation can be readily observed in any direction. Moreover, once the intensity value of the focused pump beam is increased above a certain threshold level, highly directional and coherent visible emission can be observed only in the forward and backward directions. The spectra of the two-photon excited forward and backward stimulated emission are shown in Fig. 1. The full-width at half-maxium (FWHM) of these two stimulated emission beams are about 11 and 13 nm, respectively. The central wavelength is about 617 nm.

We employed a 10-mm path-length quartz cuvette, filled with an ASPI/DMSO solution of concentration d_0 = 0.01 M, as the gain medium cell in our experiment. The laser source is a Q-switched Nd:YAG laser system (Continuum Precision II 8010) producing ~8 ns duration, 1.064-μm-wavelength, and ~1-cm^{-1} spectral-width laser output with a repetition rate of 10 Hz. Figure 2. shows the experimental setup. The laser beam is split into two beams I_1 and I_3 by beam splitter 1 with reflection/transmission ≈ 10/90 at 45° incidence. Both beams are focused via a lens (f = 10 cm) onto the center of the sample cell. The intensity of the beam I_3 is slightly lower than the threshold value. Thus no stimulated emission can be observed when the beam I_1 is blocked. The intensity of the beam I_1 is high enough to generate a stimulated emission along its forward and backward directions. However, when both I_1 and I_3 focused onto the same position inside the medium, two-photon excited stimulated emission (I_3') can also be observed along the backward direction of I_3 because part of gain path excited by I_1 can be shared by I_3.

As shown in Fig. 2, the signal wave I_3 is incident upon the medium at an angle θ with respect to the pump wave I_1. These two beams have the same frequency and the same polarization state, and therefore can produce an induced phase grating along the bisector direction of the crossing angle θ. However, the backward stimulated emission I_2 of pump beam I_1, as a reading wave with another wavelength (~617 nm), will create the diffracted wave I_4 with the same wavelength when the former passes through the induced Bragg grating. In this case the spatial information carried by the signal

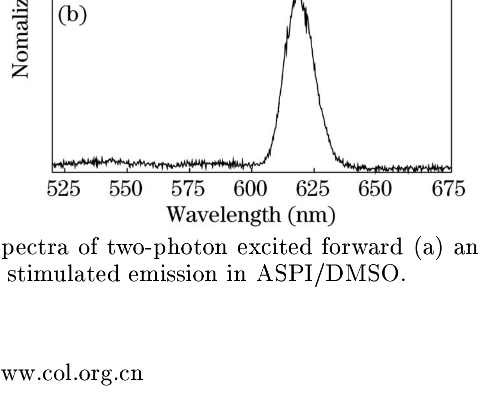

Fig. 1. Spectra of two-photon excited forward (a) and backward (b) stimulated emission in ASPI/DMSO.

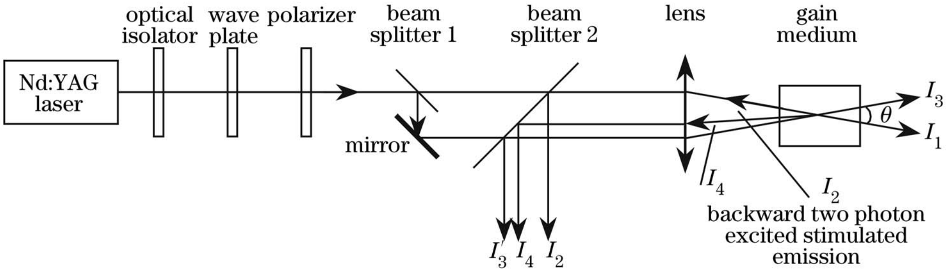

Fig. 2. Setup for nondegenerate FWM participated by two-photon induced stimulated emission.

wave I_3 can be restored in the wave I_4. The newly generated wave I_4 is not exactly counter-propagating to the signal wave I_3 because the wave vectors are different: $(\mathbf{k}_2 = \mathbf{k}_4) \neq (\mathbf{k}_1 = \mathbf{k}_3)$. In Fig. 2, I_3' is the very weak backward stimulated emission of I_3, which can identify the backward direction of the signal wave I_3. Only when the four wave vectors satisfy the phase matching shown in Fig. 3, the backward phase-conjugate wave of the signal beam can be produced efficiently. The angle α indicates the directional difference between I_3 and I_4. From Fig. 3, we can find the angle α can be described as

$$\alpha = \frac{\theta}{2} - \arcsin(\frac{k_1}{k_2}\sin\frac{\theta}{2}). \quad (1)$$

From it, one can see that the angle α increases with the difference of wave vectors of the two frequencies. Positive dispersion of the medium (the refractive index at 600 nm is greater than that at 1064 nm) makes α decrease while negative dispersion makes α increase. In this partially degenerate FWM process, $\alpha < \frac{\theta}{2}$.

In our experiment, the two-photon excited stimulated emission was obtained without cavity. The divergence angle for the stimulated emission is larger than that for the pump pulses, which makes this partially degenerate FWM more easily observed. The three backward output spots in the NFWM experiment are shown in Fig. 4. The crossing angle $\theta = 5.2°$. The energies of the pump wave I_1 and the signal wave I_3 is 2.3 and 0.26 mJ, respectively. The experimental angle $\alpha = 1.0°$ is slightly less than $1.1°$ derived from Eq. (1), which is reasonable since only the edge of the stimulated emission can approximately satisfies the phase-matching. The effect of dispersion can be neglected in this situation. The spectrum of the new generated backward wave I_4 is the same as that shown in Fig. 1.

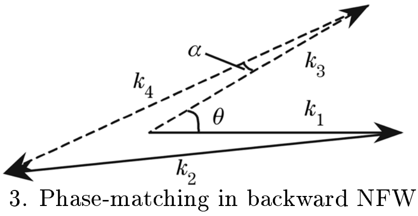

Fig. 3. Phase-matching in backward NFWM.

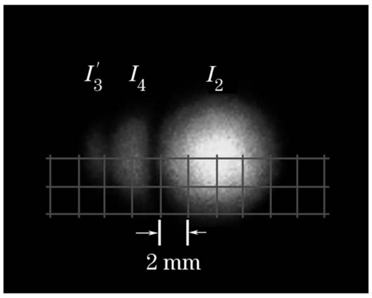

Fig. 4. Three backward output spots in our NFWM experiment.

In this paper, we presented a two-photon absorption enhanced partially degenerate FWM participated by two-photon excited stimulated emission. It is more convenient to be applied since only two beams (the pump and the signal beams) are needed in the experiment for this NFWM.

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