Self-Induced Phase Matching in Parametric Anti-Stokes Stimulated Raman Scattering

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(Received 27 November 1996)

We show through the experiments of parametric anti-Stokes stimulated Raman scattering for solid hydrogen that the phase matching in a parametric process can be self-induced without the stringent restriction of the medium. We explain this self-induced phase matching as a consequence of strong coupling between radiation fields and medium spontaneously established through the stimulated Raman scattering process. [S0031-9007(97)03510-2]

PACS numbers: 42.50.Gy, 42.65.An, 42.65.Dr, 42.65.Sf

Since the invention of lasers, considerable effort has been directed towards the development of nonlinear optics, especially of optical parametric processes. A key issue of the development has been how to match the phase velocity of nonlinear optical polarization with that of generated radiation (phase matching). So far, it has been assumed that the phase velocity is rigorously restricted by the refractive index of the medium, and the phase-matching condition is stringently governed by the dispersion of the medium [1–4]. Here, we show through the experiments of stimulated Raman scattering (SRS) for solid hydrogen that the above assumption is not generally true, that is, the phase matching can be self-induced without the stringent restriction of the medium. We find that in terms of a parametric anti-Stokes SRS process the participating waves self-control their phase velocities to the appropriate values between vacuum and medium, and self-organize the phase-matching condition. We explain this self-induced phase matching as a consequence of strong coupling between radiation fields and medium spontaneously established through the SRS process. This finding will open new possibilities of optical parametric processes free from the restriction of the medium.

Solid hydrogen is a molecular crystal consisting of H_2 molecules, known as a quantum crystal [5]. Its remarkable feature is that the molecules have well-defined vibrational and rotational quantum states as in gas phase, and that their vibrational-rotational spectrum exhibits very narrow spectral widths [6]. Figure 1 illustrates the energy-level diagram for the SRS process for an H_2 molecule (parahydrogen). The present Raman process occurs at a transition of $\nu = 1 \leftarrow 0$, $J = 0 \leftarrow 0$ for the ground electronic state $X^1\Sigma_g^+$. The intermediate states are far detuned from resonances, since single-photon-allowed electronic states are located more than 90 000 cm^{-1} above the ground state. Hereafter, we denote the vibrational-rotational states of $(\nu = 0, J = 0)$ and $(\nu = 1, J = 0)$ as states 1 and 2, respectively. In terms of the spectral width, this Raman transition is known to show an extremely narrow width of less than 7 MHz (HWHM) [7], much narrower than those for Doppler-broadened gas-phase systems. This extremely narrow width for the 2-1 transition provides an opportunity to realize the strong coupling for this far-off resonant system. Strong coupling is established when the effective two-photon Rabi frequency for the 2-1 transition exceeds the dephasing rate for the transition, i.e., Raman width, and the condition is expressed as [8]

$$\Omega_s > \sqrt{2\gamma\Delta} \approx 6\ \mathrm{cm}^{-1}$$

for the Stokes-field Rabi frequency Ω_s. Here, γ and Δ are the width of the Raman transition and the detuning to the intermediate state, respectively, and we assumed $\Delta \approx 70\,000\ \mathrm{cm}^{-1}$. It should be mentioned that because of the extremely narrow Raman width the strong coupling

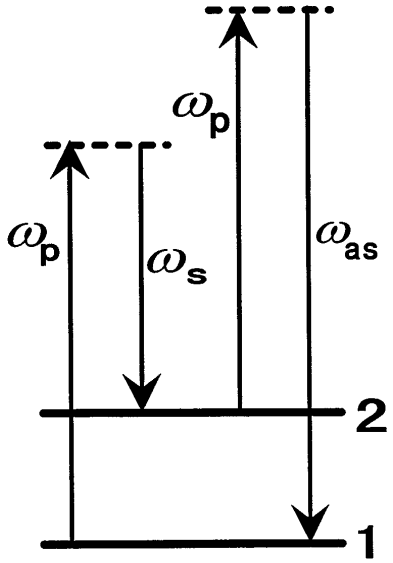

FIG. 1. Energy diagram for the SRS process. Subscripts of p, s, and as denote pump, Stokes, and anti-Stokes components, respectively. States 1 and 2 correspond to vibrational-rotational states designated by $\nu = 0$, $J = 0$ and $\nu = 1$, $J = 0$ in the ground electronic state $X^1\Sigma_g^+$, respectively. The energy separation between states 1 and 2 is 4149.7 cm^{-1}.

can be established with a Stokes Rabi frequency of 6 cm^{-1} which can be realized for an H_2 molecule with a reasonably low intensity of 7 MW/cm^2 for pulse-laser excitation.

Solid hydrogen of nearly pure parahydrogen (>99.9%) was prepared at helium cryogenic temperature in a copper cell, 2 cm long and 2 cm in diameter, by using the gas-phase-growing method [6]. Optical quality CaF_2 windows were attached to the cell, and aligned in parallel to form a cavity [9,10] for the Stokes radiation to control its propagation axis. A linearly polarized single-longitudinal-mode *Q*-switched Nd:YAG laser was used for pumping with a frequency-doubled output at 532 nm wavelength. The pulse duration of the pump laser was 10 ns (FWHM) with the maximum energy of 800 μJ. The laser beam was loosely focused to a parallel beam with a diameter of 0.4 mm in solid hydrogen. The SRS emission patterns were photographed in the forward direction, and temporal profiles were measured after dispersion with a monochromator. Measurements were carefully carried out by rotating the polarization of the pump beam, but we did not observe any effect on the polarization direction. All measurements were carried out at 4.2 K.

SRS emission was easily observed by eye when the pump intensity exceeded the threshold energy measured to be 70 μJ. Emission was observed to second order for both Stokes and anti-Stokes components. In this Letter, we restrict the discussion to the first order components. Figure 2(a) shows a typical SRS emission pattern observed for a pump-laser energy of 500 μJ. The Stokes axis was adjusted to coincide with the pump axis. The white spot at the center corresponds to the pump beam. The Stokes radiation appears as a red beam at 683 nm wavelength with a divergence angle of 30 mrad (HWHM). As expected from the conventional phase-matching condition, the anti-Stokes component is clearly observed as a blue ring at a wavelength of 436 nm, and the corresponding Stokes component is observed as a dark ring [4,10] in the red pattern. Respective emission angles for the blue and dark ring were 27 and 45 mrad, and are in good agreement with the values estimated from the dispersion [11] of solid hydrogen. However, particular attention must be directed to the emission color in the center; that is, the same color as the blue ring may clearly be recognized. This situations is definitely seen in the photograph of Fig. 2(b) obtained using a blue-pass filter. The central blue emission has the same wavelength as that for the blue ring and shows a beam divergence of 20 mrad exhibiting its origin as the coherent anti-Stokes emission. This observation cannot be explained by the conventional assumption for the SRS process that the phase matching is governed by the refractive index of the medium and its dispersion. The anti-Stokes emission at the center reveals a surprising fact that the phase matching can be established *not uniquely* under the condition governed by the medium dispersion.

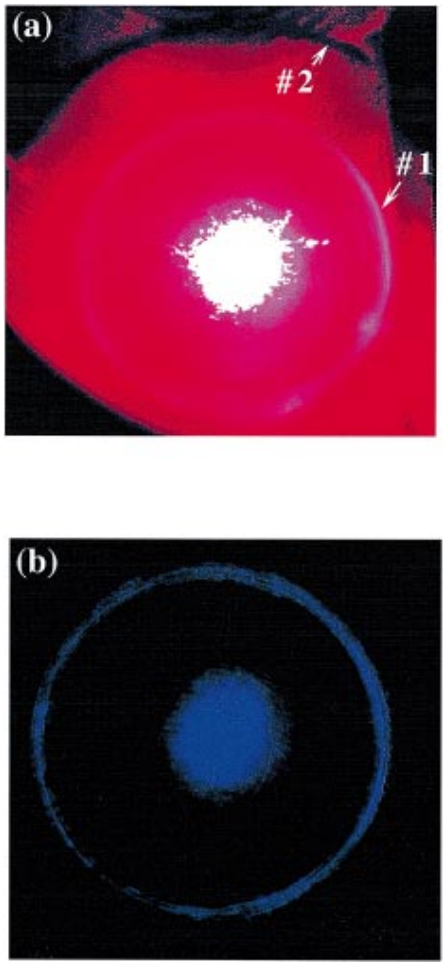

FIG. 2(color). Photograph of SRS emission in the forward direction. Anti-Stokes ring and corresponding dark ring in Stokes emission are marked in (a) by #1 and #2, respectively. Photograph in (b) was taken through a blue-pass filter, clearly showing, besides the ring pattern, the anti-Stokes emission at the center.

The observed phase matching for three on-axis waves of pump, Stokes, and anti-Stokes radiation means that the three waves propagate in the medium with the same phase velocity as in vacuum.

Figure 3 exhibits temporal profiles for the three beams in Figs. 2(a) and 2(b). The pump beam abruptly depletes and the Stokes beam quickly builds up, when the pump intensity exceeds the threshold. On the other hand, the anti-Stokes beam at the center builds up slowly and tends towards almost the same value as that of the depleted pump beam (shown in the inset), revealing a high conversion efficiency of nearly 50% from the pump beam to the anti-Stokes beam at the center. The Stokes peak intensity is about 20 MW/cm^2, which far exceeds the critical intensity for the strong coupling. Although the behavior of the blue ring seems briefly similar to that of the center blue spot with a weaker intensity of 20%, one can recognize a shoulder on the leading part of the profile marked by a solid arrow. This shoulder becomes

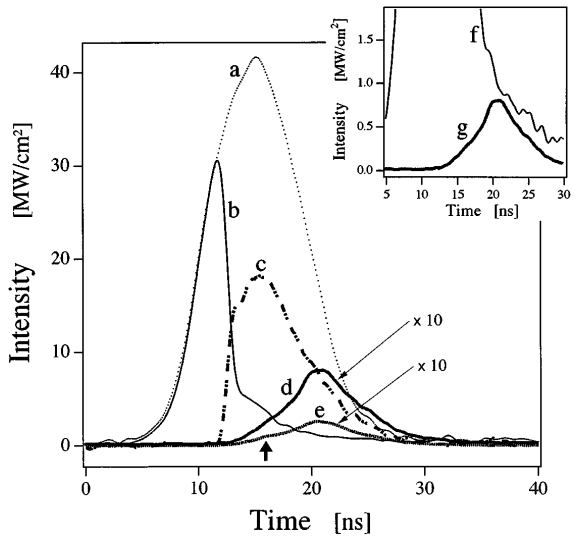

FIG. 3. Temporal profiles for SRS emission components: (*a*) incident pump beam, (*b*) transmitted pump beam, (*c*) Stokes beam, (*d*) anti-Stokes spot at the center, and (*e*) anti-Stokes ring. A shoulder on the leading part of profile (*e*) is marked by a solid arrow. Profiles of (*d*) and (*e*) are magnified by a factor of 10. In the inset, profiles are displayed with the same scale for transmitted pump (*f*) and anti-Stokes spot at the center (*g*).

a distinct peak with the decrease of pump intensity, revealing the blue spot due to a different physical origin from the blue ring.

Behaviors of the anti-Stokes blue spot were examined for various tilt angles of the Stokes axis to the pump axis. We found that the anti-Stokes blue spot appears for any Stokes tilt angle from 0 to 45 mrad, but the blue spot does not appear when the tilt angle has exceeded 45 mrad. The angle of 45 mrad corresponds to an angle where the phase matching based on the medium dispersion is satisfied. Typical emission patterns are displayed in Figs. 4(a) and 4(b). It is clearly seen that the blue spot appears off axis to the pump beam on the opposite side of the Stokes beam, and it shifts towards the blue-ring position as the increase of the Stokes tilt angle. These observations mean that in terms of the SRS process for solid hydrogen the participating waves self-control their phase velocities to the appropriate values between vacuum and medium, and self-organize the phase matching depending on the geometry between pump and Stokes beams.

The above findings show that the conventional perturbation approach is not adequate for the present SRS process; in other words, the SRS process spatially and temporally evolves the eigenstate of the whole fields-medium system to the strongly coupled state between medium and fields. As known for Raman-type two-photon interaction in an atomic system, such a strongly coupled state [12] can be expressed by an antisymmetric linear combination of states 1 and 2, termed as dark state.

$$|\text{dark}\rangle = \frac{1}{\Omega}\left(\Omega_s|1\rangle - \Omega_p|2\rangle\right),$$

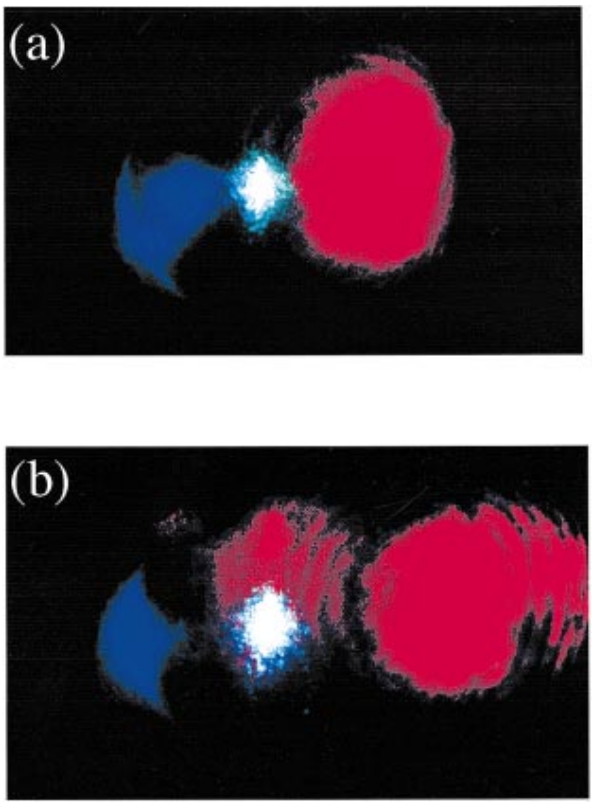

FIG. 4(color). Photographs of SRS emission taken by changing the tilt angle of the Stokes axis to the pump axis. (a) was taken for the tilt angle of 25 mrad; the blue spot is observed at the middle between the center and the blue ring. (b) was taken for the tilt angle of 45 mrad; the blue spot overlaps with the blue ring.

where Ω_s and Ω_p correspond to Rabi frequencies for Stokes and pump fields, and $\Omega^2 \equiv |\Omega_s|^2 + |\Omega_p|^2$. When the dark state has been established, both pump and Stokes fields propagate in the medium with the same phase velocity as in vacuum [13–16]. In the context of the strongly coupled eigenstate, the present anti-Stokes generation process can naturally be understood due to the establishment of the dark state; the center blue spot in Fig. 2(b) is a clear demonstration of the establishment of the dark state which satisfies the phase-matching condition for the vacuum. On the other hand, the off-axis blue spot may be explained through the spatial and temporal evolution of the eigenstate from the weak coupling to the strong coupling. Through the evolution process, the fields effectively experience refractive indices changing from the medium value to the vacuum value. Such an effective change of refractive indices enable one to realize the off-axis phase matching on the inside of the blue ring as observed in Figs. 4(a) and 4(b).

In conclusion, we have found a new phenomenon of self-induced phase matching in the parametric anti-Stokes SRS process for solid hydrogen. We have explained the self-induced phase matching from the viewpoint of strongly coupled eigenstate, dark state, spontaneously

established through the SRS process. The present finding in solid hydrogen may open new types of optical parametric processes which are free from the restriction of the medium in a broad wavelength range from the infrared to the vacuum ultraviolet.

The authors thank B. P. Stoicheff and S. E. Harris for critical comments. They also thank R. S. D. Sihombing for his help in the experiments. This work was partly supported by a Grant-in-Aid for Scientific Research in a Priority Area, "Mutual quantum manipulation of radiation field and matter," from the Ministry of Education, Science, Sports, and Culture of Japan.

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