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Dark-state coherence and stimulated Raman scattering in solid hydrogen

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We show through the experiments of stimulated Raman scattering in solid hydrogen that a strongly coupled dressed state, dark state, is spontaneously established as the eigenstate of the field–matter interacting system, and that in terms of the parametric anti-Stokes generation process the phase matching is self-induced as a consequence of the establishment of the dark state.

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1. Introduction

Optical processes with quantum interference or coherence are opening various new possibilities beyond the limit of conventional optical processes (Harris, this volume; Scully, this volume). The key physics of the processes is to establish a strongly coupled dressed state (typically, dark state) as the eigenstate for a Raman-driven three-level system. So far, it has been assumed that such processes can be realized only for low-density atomic systems. However, we show here using solid hydrogen as a medium that the strong coupling can be realized for a high-density solid-medium, even under far off-resonant conditions. It is shown through the experiments of stimulated Raman scattering (SRS) that in terms of the parametric anti-Stokes generation process the phase matching is self-induced without the stringent restriction of the medium as a consequence of the strong coupling spontaneously established through the SRS process. This is the first demonstration which has realized well-defined strongly coupled dressed state, dark state, in the solid medium.

2. Strong-coupling condition

Solid hydrogen is a molecular crystal consisting of H_2 molecules, and is known as a quantum crystal (Van Kranendonk 1983). Its remarkable feature is that the molecules have well-defined vibrational and rotational quantum states described by Bloch waves, and that the ground level designated by $v = 0$, $J = 0$ is described by the single Bloch wave of $\boldsymbol{k} = \boldsymbol{0}$, since it does not have any degeneracy. This single state nature enables the optical transitions from the ground level to pick up selectively the excited levels described by $\boldsymbol{k} = \boldsymbol{0}$, leading the vibration-rotational spectrum to very narrow spectral widths. Such nature of solid hydrogen can provide an opportunity to realize the strong-coupling condition even in the solid medium. The strong coupling for the far-off resonant SRS process shown in figure 1 requires the condition of $\Omega_S \succ \sqrt{2\gamma\Delta}$ for the Stokes-field Rabi-frequency Ω_S (Harris 1994*a*), where

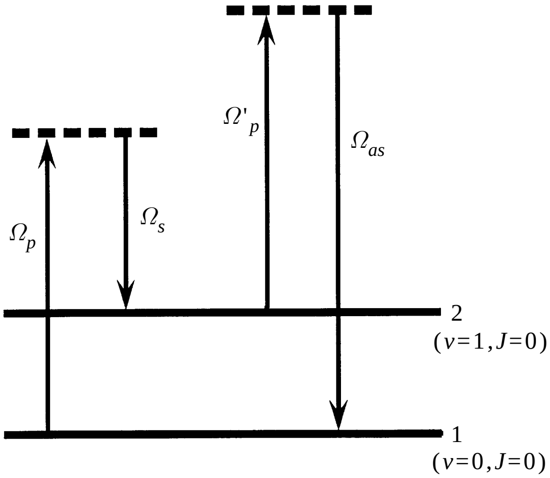

Figure 1. Energy diagram of the SRS process. Rabi frequencies are denoted by Ω_i (i = p, S, aS), where p, S and aS mean pump, Stokes and anti-Stokes, respectively.

γ and Δ are the width of the Raman transition and the detuning to the intermediate state, respectively. For the Raman transition of $v = 1 - 0$, $J = 0 - 0$ in the ground electronic state $X^1\Sigma_g^+$, the spectral width is known to show an extremely narrow width of less than 7 MHz (HWHM) (Momose *et al.* 1992). Because of this extremely narrow Raman width, it is expected that the strong coupling can be established with a Stokes Rabi-frequency of 6 cm^{-1} which is obtained with a reasonably low Stokes intensity of 7 MW cm^{-2}.

3. Experimental

Solid hydrogen of nearly pure parahydrogen (greater than 99.9%) was prepared at helium cryogenic temperature in a cell by using the gas-phase-growing method (Welky *et al.* 1994). Optical quality fused-quartz windows were attached to the cell, and aligned in parallel to form a cavity 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 dispersing with a monochromator. All measurements were carried out at 4.2 K.

4. Results and discussion

The coherent first Stokes emission was readily observed as a red beam at a wavelength of 683 nm when the pump intensity exceeded the threshold energy of 70 μJ. Above the pump energy of 200 μJ, the first Stokes intensity showed a saturation behaviour, and the temporal profile of the pump beam was strongly depleted. Figures 2*a*–*d* exhibit typical SRS emission patterns for a pump-laser energy of 500 μJ. Photographs in figures 2*a*, *b* were taken by adjusting the Stokes axis to coincide with the pump axis, while those in figures 2*c*, *d* by tilting the Stokes axis 30 mrad from

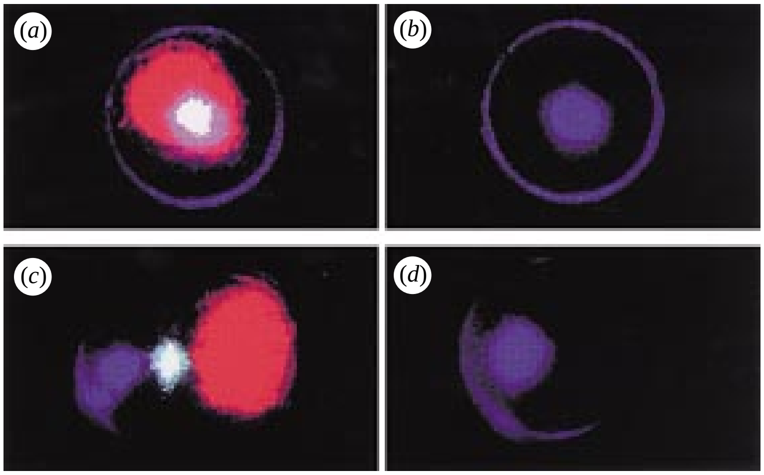

Figure 2. Photographs of SRS emission in forward direction. (*a*) and (*b*) were taken by adjusting the Stokes axis on the pump axis. (*c*) and (*d*) were taken by tilting the Stokes axis 30 mrad to the pump axis. (*b*) and (*d*) were taken with a blue-pass filter.

the pump axis. Photographs in figures 2*b*, *d* were taken through a blue-pass filter for observing the anti-Stokes emission pattern at 436 nm. For this pump energy of 500 μJ, the Stokes intensity was measured to be 20 MW cm^{-2}, much higher than the expected intensity to realize the strong coupling. In figure 2*a*, the white spot at the centre corresponds to the pump beam, and as expected from the conventional phase-matching condition, the anti-Stokes component is clearly observed as a blue ring. However, particular attention must be directed to the emission colour in the centre; that is, the same colour as the blue ring may clearly be recognized. This situation is definitely seen in the photograph of figure 2*b*. The central blue emission has the same wavelength as that for the blue ring exhibiting its origin as the coherent anti-Stokes emission. The anti-Stokes emission at the centre means that the phase matching is established for on-axis three waves of pump, Stokes and anti-Stokes radiation. Furthermore, when the Stokes axis is tilted to the pump axis, the anti-Stokes blue spot appears on the opposite side of the Stokes red spot as shown in figures 2*c*, *d*. By tilting the Stokes axis more, the blue spot shifts towards the blue ring, but it never exceeds the blue ring. For the geometry where the blue spot overlaps with the blue ring, the conventional phase-matching condition is satisfied. Thus, it has been found in the SRS process for solid hydrogen that the refractive indexes are strongly modified from the conventional values, and that the phase matching is self-induced for the parametric anti-Stokes generation depending on the geometry for the pump and Stokes beams without the strict restriction of the medium.

The above finding of self-induced phase matching for the parametric anti-Stokes generation reveals that the eigenstate of the whole field–medium system has been spontaneously evolved from the conventional weak-coupling system to the strongly coupled system through the present SRS process, and that the idea of phase matching must be completely changed to meet for situation of the strongly coupled system. As known for Raman-driven system in figure 1, the strongly coupled eigenstate can be expressed by anti-symmetric linear combinations of states 1 and 2, termed as dark state. For the dark state, the participating waves experience effectively the refractive index as in vacuum (Harris 1993*b*, 1994; Eberly *et al.* 1994, 1996). This means that the blue spot at the centre, that is, the phase matching for on-axis three waves, would be a clear manifest of the establishment of the dark state; in other words, the on-axis anti-Stokes generation is due to the dark-state coherence. Here, it should be mentioned that the waves may effectively experience refractive indices changing from the conventional medium values to the vacuum value through the evolution process, since the strong-coupling regime is temporally and spatially evolved from the weak-coupling regime in which the waves experience the conventional refractive indices. This turns out that the off-axis phase matching observed in figures 2*c*, *d* may be understood due to the effective refractive indices through the evolution process. Apparently, such effective refractive-indices cannot satisfy the phase matching on the outside of the blue ring determined by the conventional refractive indices.

5. Conclusions

We have shown using solid hydrogen that the dark state can be established as the eigenstate in the Raman-driven three-level system even for the high-density solid medium, and that due to the dark-state coherence the phase matching is self-induced for the parametric anti-Stokes generation process at any condition between vacuum and medium. The present finding in solid hydrogen may open a way to a new class of nonlinear optical processes free from the restriction of the medium.

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