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Polarization effect in impulsive rotational Raman scattering

Kenichi Ishikawa*
Department of Quantum Engineering and Systems Science, Graduate School of Engineering, University of Tokyo, Hongo 7-3-1, Bunkyo-ku, Tokyo 113-8656, Japan

Hiroyuki Kawano and Katsumi Midorikawa
Laser Technology Laboratory, RIKEN (The Institute of Physical and Chemical Research), Hirosawa 2-1, Wako-shi, Saitama 351-0198, Japan

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We theoretically study rotational Raman coherence excited impulsively in a hydrogen-gas-filled hollow fiber by an fs pump pulse with an arbitrary ellipticity. The results of our simulations based on a rotationally invariant formalism show that the use of an elliptically polarized pulse leads to more efficient phonon excitation than that of a linearly or circularly polarized one. The phonon amplitude dramatically depends both on ellipticity and propagation distance. The passage of a probe pulse in this excited media leads to the generation of high-order Raman sidebands in the pulse.

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In the past several years, a considerable amount of effort has been made to improve the technique of ultrashort-pulse generation. By use of a traditional method based on supercontinuum generation via self-phase modulation in a Kerr nonlinear medium and subsequent pulse compression, it is now possible to realize an optical pulse as short as 5 fs [1]. An alternative mechanism which has recently attracted growing interest is stimulated Raman scattering (SRS) [2–7]. Sokolov *et al.* [8] have generated near single-cycle pulses with pulse lengths of ~ 2 fs based on collinear generation of a wide spectrum of equidistant, mutually coherent Raman sidebands by achieving a near maximum Raman coherence with two nanosecond lasers. On the other hand, the recent experiment by Wittmann *et al.* [9] has demonstrated the frequency modulation of laser pulses via impulsive SRS using the rotation of hydrogen molecules and Fourier-synthesized short pulse trains from the compression of Raman sidebands. These latter authors have also studied the scheme using the vibrational SRS from SF_6 and reported the generation of 5.8-fs pulse trains. In this type of technique, an ultrashort pump pulse excites a Raman medium impulsively, and a weaker probe pulse, injected with a time delay, is scattered by the coherently oscillating molecular vibrations or rotations. This leads to the generation of Raman sidebands.

In view of its application to the pulse compression as well as for its fundamental interest, several theoretical investigations on the above-mentioned impulsive SRS have been done both for vibrational and rotational cases [7,10–12]. These studies were, however, concentrated on linearly polarized pump pulses. Although in the case of impulsive rotational SRS the phonon amplitude excited by the pump pulse is expected to depend on the ellipticity of the pump pulse, there exists no work on the polarization effect, to our knowledge.

In this Rapid Communication, we theoretically study the dynamics of rotational Raman coherence excited impulsively by a femtosecond laser pulse with an arbitrary elliptical polarization and subsequent scattering of a linearly polarized probe pulse in the excited medium. To be specific, we consider a hollow fiber with an inner diameter of 126 μm filled with 0.5 atm of H_2 as a Raman medium. We simulate the dynamics of pulse propagation by solving an extended nonlinear Schrödinger equation coupled with an equation for phonon creation. Our results reveal that an elliptically polarized pump pulse excites Raman coherence more efficiently than a linearly or circularly polarized pulses. Moreover, the phonon amplitude by an elliptically polarized pump largely variates with a propagation distance inside the medium as well as the ellipticity. By the passage of a probe pulse through the Raman medium excited by an elliptically polarized light, high-order Raman sidebands can be generated, comparably to the case of a linearly polarized pump pulse.

Our analysis is based on the rotationally invariant formalism developed by Holmes and Flusberg [13]. It follows from Eqs. (9)–(12) of Ref. [13] that

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2}-\frac{\partial^2}{\partial z^2}\right)E_\gamma=\frac{4\pi}{c^2}C_R\frac{\partial^2}{\partial t^2}\times\sum_{m\alpha}(Q_m E_\alpha+Q^*_{-m}E_\alpha)\langle m|\alpha-\gamma\rangle, \tag{1}$$

$$\left(\frac{\partial^2}{\partial t^2}+\Omega_R^2\right)Q_n=C_R\sum_{\alpha\beta}E_\alpha E^*_\beta\langle -n|\alpha-\beta\rangle, \tag{2}$$

where E_γ denotes the electric field of polarization $\gamma(=\pm 1)$, Q_m the amplitude of the phonon of $m(=\pm 2,0)$ units of angular momentum along the axis z of propagation, Ω_R the Raman frequency, and $C_R=N/\sqrt{2}(\partial\alpha/\partial Q)$, with N and $\partial\alpha/\partial Q$ being the molecular density and the differential polarizability, respectively. $\langle m|\alpha\beta\rangle$ stands for the Clebsch-Gordan coefficient $\langle 2m|11\alpha\beta\rangle$. Relevant coefficients are $\langle 2|1,1\rangle=\langle -2|-1,-1\rangle=1$ and $\langle 0|1,-1\rangle=\langle 0|-1,1\rangle$

*Electronic address: ishiken@q.t.u-tokyo.ac.jp

$=1/\sqrt{6}$. Thus the light is coupled to phonons that flip polarization more strongly than to those that do not change polarization. On derivation of Eqs. (1) and (2) we have neglected phonon damping and the last two terms of the sums in Eqs. (11) and (12) of Ref. [13]. We use the slowly varying envelope approximation (SVEA),

$$E_\alpha = A_\alpha \exp[i(k_L z - \omega_L t)], \tag{3}$$

for the electric field. For the phonon amplitude Q_m, on the other hand, we do not use the SVEA, since the laser pulse is in general shorter than the period $T_R(=57$ fs) of molecular rotation. Then Eqs. (1) and (2) are rewritten in a frame of reference moving with the group velocity of the pulse as

$$\frac{\partial A_\gamma}{\partial z} + \sum_{\ell=2}^{\ell_{\max}} \frac{i^{\ell-1}\beta_\ell}{\ell!} \frac{\partial^\ell A_\gamma}{\partial t^\ell} + \frac{\sigma}{2} A_\gamma = \frac{2in_2 k_0}{3}\left(1 + \frac{i}{\omega_0}\frac{\partial}{\partial t}\right)[(|A_\gamma|^2 + 2|A_{-\gamma}|^2)A_\gamma] + i4\pi k_0 C_R \sum_{m\alpha} Q_m A_\alpha \langle m|\alpha - \gamma\rangle, \tag{4}$$

$$\left(\frac{\partial^2}{\partial t^2} + \Omega_R^2\right) Q_n = C_R \sum_{\alpha\beta} A_\alpha A_\beta^* \langle -n|\alpha - \beta\rangle, \tag{5}$$

In the extended nonlinear Schrödinger equation (4) we have included the linear dispersion (the second term on the left-hand side), the fiber loss (the third term on the left-hand side), and the instantaneous Kerr effect [14] (the first term on the right-hand side). ω_0 denotes the carrier frequency of the pulse, and k_0 is the corresponding vacuum wave number. β_ℓ is defined as

$$\beta_\ell = \left(\frac{d^\ell \beta}{d\omega^\ell}\right)_{\omega=\omega_0}, \tag{6}$$

where $\beta(\omega)$ gives the dispersion relation of the wave number in the Raman medium including the waveguide contribution. It should be noted from Eq. (5) that

$$Q_m = Q_{-m}^* \quad (m = \pm 2, 0), \tag{7}$$

and, especially, Q_0 is real. This relation has been used in the derivation of Eq. (4). Equations (4) and (5) are valid both for the pump and the probe pulse if we use appropriate values of parameters. In the present study we solve the coupled equations Eqs. (4) and (5) by a split-step Fourier method with the fourth-order Runge-Kutta method for nonlinear steps. A_γ is normalized in such a way that $|A_\gamma|^2$ gives an intensity in W/cm^2 for convenience. The values of β_ℓ's are calculated with the linear susceptibility described in Ref. [3] and the waveguide contribution described in Ref. [15]. We set $\ell_{\max}$ to 5. The used parameters [10,11,16–18] are $\Omega_R = 0.1106$ fs^{-1}, $\sigma = 1.5$ m^{-1}, and $n_2 = 5.51\times10^{-20}$ cm^2/W both for the pump and probe pulse, $C_R = 1.2\times10^4$ for the pump with a wavelength of 785 nm, and $C_R = 1.3\times10^4$ for the probe with a wavelength of 392.5 nm, respectively.

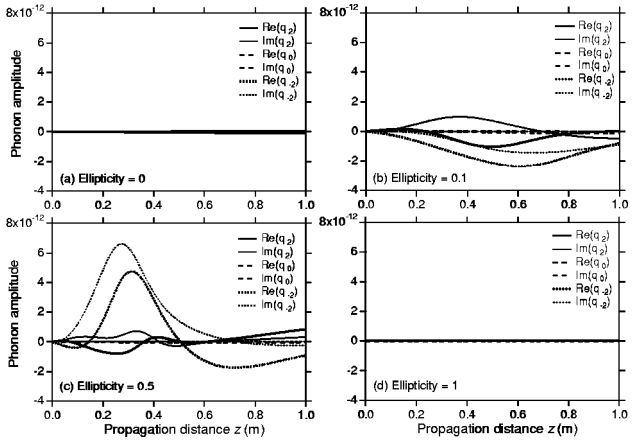

FIG. 1. The distribution of the phonon amplitude q_m ($m = \pm 2, 0$) excited by a pump pulse with a duration of 80 fs and different values of ellipticity e_p indicated in the figure.

There is another model to describe molecular rotation, i.e., the rigid rotor formalism [19]. In the present study, however, excited phonon density is much lower than molecular density as we will see below. In such cases the use of the rotationally invariant formalism is appropriate. The rigid rotor formalism would be necessary for the case of much more intense or longer pulses, which may cause molecular alignment and rotational wavepacket [19].

We first study how rotational Raman coherence excited impulsively by a pump pulse depends on the ellipticity e_p of the pump light and the propagation distance z. The pulse has a sech^2 temporal profile with a peak intensity of 10^{13} W/cm^2. For comparison we consider two different values of pulse duration T_0 (full width at half maximum), i.e., 40 fs and 80 fs. In the latter case, the phonon excitation is, strictly speaking, not impulsive, since the pulse duration is slightly longer than the molecular rotation period T_R. After the pump pulse has passed and before the arrival of the probe pulse, the phonon amplitude Q_n is of the following form:

$$Q_n = q_n e^{-i\Omega_R t} + q_{-n}^* e^{i\Omega_R t} \quad (n = \pm 2, 0), \tag{8}$$

where q_n depends on z in general. We plot q_n as a function of z for different values of pump pulse ellipticity,

$$e_p = \frac{|A_+| - |A_-|}{|A_+| + |A_-|} \tag{9}$$

in Figs. 1 ($T_0 = 80$ fs) and 2 ($T_0 = 40$ fs). In Fig. 1 we can see that the phonon is practically not excited when the pulse is linearly ($e_p = 0$) or circularly ($e_p = 1$) polarized. On the other hand, when the pulse polarization is elliptical, the phonon with $m = -2$ are efficiently excited, and that with $m = 2$ is also, though less, excited, while that with $m = 0$ is still hardly excited. These behaviors can be understood qualitatively as follows. For the linear polarization or for the phonon with $m = 0$, the gain is parametrically suppressed due to the Stokes–anti-Stokes (SA) coupling [20], as is well known

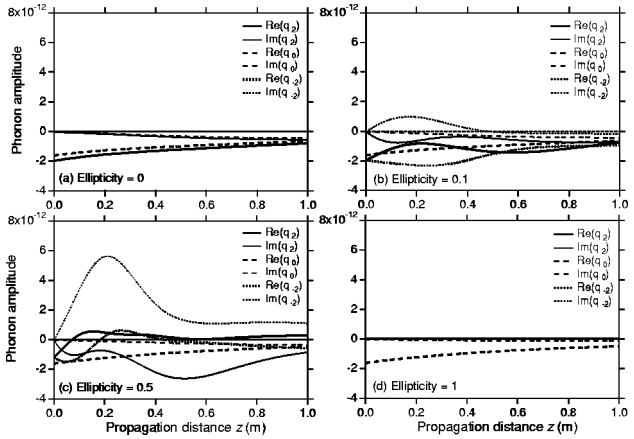

FIG. 2. The distribution of the phonon amplitude q_m ($m=\pm 2,0$) excited by a pump pulse with a duration of 40 fs and different values of ellipticity e_p indicated in the figure.

in vibrational SRS. On the other hand, for the phonons with $|m|=2$ in the case of an elliptically polarized pulse, the SA coupling is only partial, so that these phonons can be excited. For a circularly polarized pulse, no phonon with $|m|=2$ is excited due to the lack of the field of the opposite polarization. In Fig. 2, unlike in Fig. 1, phonons can be impulsively excited even with a linearly or circularly polarized pulse, and the phonon with $m=0$ can also be generated, since the pulse is shorter than the period T_R of the molecular rotation. It should be noted in Fig. 2, however, that the phonon excitation for $m=\pm 2$ is more efficient for an elliptical polarization than for a linear or circular polarization, and is most efficient around $e_p=0.5$. q_0 is nearly independent of e_p and does not vary much with z except for a gradual decrease. It follows from Eq. (5) that

$$\left(\frac{\partial^2}{\partial t^2}+\Omega_R^2\right)Q_0=\frac{C_R}{\sqrt{6}}\sum_\gamma |A_\gamma|^2. \tag{10}$$

Neglecting linear dispersion and self-steepening (the term containing $i/\omega_0\partial/\partial t$), we can obtain the variation of $\Sigma_\gamma|A_\gamma|^2$ by use of Eq. (4) as, after some algebra,

$$\sum_\gamma |A_\gamma(t,z)|^2 \approx e^{-(\sigma/2)z}\sum_\gamma |A_\gamma(t,z=0)|^2, \tag{11}$$

or an exponential decrease due to a fiber loss, independent of pulse ellipticity e_p. Although linear dispersion and self-steepening are not completely negligible, the resulting dependence of q_0 on e_p is quite small. On the contrary, the excitation of phonons with $m=\pm 2$ largely depends on e_p, and its variation with z is remarkable, as can be seen both from Figs. 1 and 2. The highest value of phonon amplitude in Fig. 1 is approximately 7×10^{-12} in such a system of units that $|A_\gamma|^2$ gives an intensity in W/cm^2. This value corresponds to a phonon density of 2.2×10^{16} cm^{-3}, which is much lower than the molecular density (1.3×10^{19} cm^{-3}). This observation confirms the validity of the rotationally invariant formalism [13] in the present study.

Let us now turn to the investigation of the Raman sideband formation in a linearly polarized weaker probe pulse by the phonons excited by the pump pulse. At this stage, it is instructive to discuss briefly the probe pulse propagation obtained by rewriting its envelope A_α as the sum of different frequency components, $A_\alpha=\Sigma_j A_{j\alpha}$, where $A_{j\alpha}$ is the envelope of the component of frequency $\omega_j=\omega_L-j\Omega_R$. If we neglect the dispersion and the Kerr response and assume that the probe is sufficiently weak that Q_n can be described by Eq. (8), the propagation of each component j is described by

$$\frac{\partial A_{j\gamma}}{\partial z}=-4\pi k_0 C_R(\bar{q}_m A_{j+1,\alpha}-\bar{q}^*_{-m}A_{j-1,\alpha})\langle m|\alpha-\gamma\rangle, \tag{12}$$

$$A_{0,+}=A_{0,-}=A_0, \text{ and } A_{j\alpha}=0 \text{ if } j\neq 0 \text{ at } z=0, \tag{13}$$

where $\bar{q}_m=-iq_m$. Equation (12) can be solved analytically for the following two cases if we assume that $\bar{q}_m$ is independent of z: In case $\bar{q}_2=\bar{q}_{-2}=\bar{q}$, we can write the solution of Eq. (12) as,

$$A_{j,+}=A_{j,-}=A_0 J_j(8\pi k_0 C_R|\bar{q}_2+\bar{q}_0/\sqrt{6}|)e^{-ij\phi}, \tag{14}$$

where $J_j(z)$ denotes the Bessel function of the first kind of order j, and $\phi=\arg(\bar{q}_2+\bar{q}_0/\sqrt{6})$. This behavior is qualitatively similar to the case of vibrational Raman scattering. On the other hand, in case $\bar{q}_{-2}=|\bar{q}_{-2}|e^{i\phi}\neq 0$ and $\bar{q}_0=\bar{q}_2=0$, the solution of Eq. (12) reads

$$A_{0,+}(z)=A_{0,-}(z)=A_0\cos 4\pi k_0|\bar{q}_{-2}|z, \tag{15}$$

$$A_{1,-}(z)=A_0 e^{-i\phi}\sin 4\pi k_0|\bar{q}_{-2}|z, \tag{16}$$

$$A_{-1,+}(z)=-A_0 e^{i\phi}\sin 4\pi k_0|\bar{q}_{-2}|z, \tag{17}$$

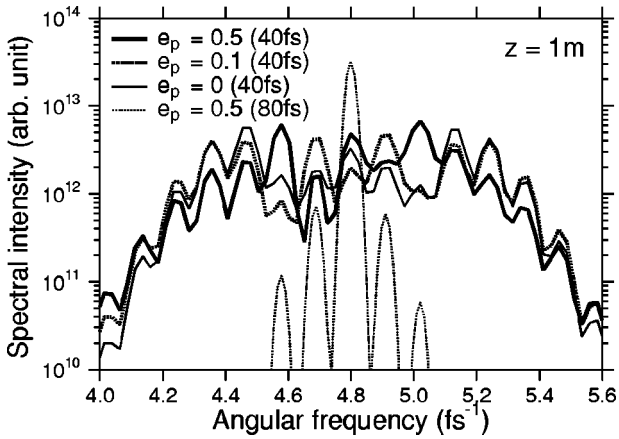

FIG. 3. Intensity spectrum of a probe pulse after 1 m of propagation in the medium excited by a pump pulse with different values of ellipticity e_p and duration T_0. Thick solid curve: $e_p=0.5$ and $T_0=40$ fs; thick dotted curve: $e_p=0.1$ and $T_0=40$ fs; thin solid curve: $e_p=0$ and $T_0=40$ fs; thin dotted curve: $e_p=0.5$ and $T_0=80$ fs.

and all the other components remain zero. Thus, if q_2 or q_{-2} has been selectively excited by the pump pulse, Raman components only of the first order can be formed in the probe pulse.

Bearing the above discussion in mind, let us investigate the Raman component formation in a probe pulse with a peak intensity of $10^{11}\mathrm{W/cm}^2$ and a duration of 80 fs, by solving of the coupled equations (4) and (5). The group velocity matching [21] between the pump and the probe is achieved for the fiber diameter and the gas pressure considered in the present study. Figure 3 presents the spectrum of the probe pulse at $z=1\mathrm{m}$. In case $e_p=0.5$ and $T_0=80$ fs, the pump pulse is elliptically polarized and has a duration longer than T_R (thin dotted curve), the spectrum is hardly broadened. Since the phonon with $m=-2$ has been selectively excited by the pump as can be seen from Fig. 1(c), the dynamics of the probe pulse is essentially described by Eqs. (15)–(17): most of the pulse energy is exchanged only among the fundamental and the first-order Raman components. On the other hand, in the case of an elliptically polarized pump pulse with $T_0=40$ fs (thick solid and dotted curves), high-order Raman sidebands are formed, and the bandwidth is at least comparable to the case of the linearly polarized pump pulse (thin solid curve). In this case, in contrast to the previous case, Figs. 2(b) and 2(c) show that a considerable amount of phonons with $m=0$ and 2 are excited in addition to that with $m=-2$. This allows an efficient formation of high-order Raman components in the probe pulse.

In conclusion, we have investigated the effect of the pump pulse polarization on the impulsive excitation dynamics of rotational phonons in a hollow fiber filled with a hydrogen gas and Raman sideband formation in an accompanying weaker probe pulse. We have considered an arbitrary initial ellipticity of the pump pulse and based our analysis on the rotationally invariant formalism [13]. The excitation of Raman coherence by the pump pulse depends sensitively on its polarization and is more efficient for an elliptically polarized pump pulse than for a linearly or circularly polarized pump. In the case of an elliptically polarized pump pulse shorter than the period of the molecular rotation, phonons with all the three values of $m(=\pm 2,0)$ are excited and the amplitude of those with $m=\pm 2$ strongly depends on the pump ellipticity and the propagation distance. When a probe pulse is injected into thus excited media, high-order Raman components are formed in the pulse, and its spectrum is broadened to an extent at least comparable to the case of a commonly used linearly polarized pump pulse.

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[1] S. Sartania, Z. Cheng, M. Lenzner, G. Tempea, Ch. Spielmann, and F. Krausz, Opt. Lett. **22**, 1562 (1997).
[2] S. Yoshikawa and T. Imasaka, Opt. Commun. **96**, 94 (1993).
[3] H. Kawano, T. Mori, Y. Hirakawa, and T. Imasaka, Phys. Rev. A **59**, 4703 (1999).
[4] S.E. Harris and A.V. Sokolov, Phys. Rev. Lett. **81**, 2894 (1998).
[5] A.V. Sokolov, D.D. Yavuz, and S.E. Harris, Opt. Lett. **24**, 557 (1999).
[6] Fam Le Kien, Nguyen Hong Shon, and K. Hakuta, Phys. Rev. A **64**, 051803(R) (2001).
[7] M. Wittmann, A. Nazarkin, and G. Korn, Appl. Phys. B: Lasers Opt. **70**, S261 (2000).
[8] A.V. Sokolov, D.R. Walker, D.D. Yavuz, G.Y. Yin, and S.E. Harris, Phys. Rev. Lett. **87**, 033402 (2001).
[9] M. Wittmann, A. Nazarkin, and G. Korn, Opt. Lett. **26**, 298 (2001).
[10] V.P. Kalosha and J. Herrmann, Phys. Rev. Lett. **85**, 1226 (2000).
[11] V.P. Kalosha and J. Herrmann, Opt. Lett. **26**, 456 (2001).
[12] V. Kalosha, M. Spanner, J. Herrmann, and M. Ivanov, Phys. Rev. Lett. **88**, 103901 (2000).
[13] R. Holmes and A. Flusberg, Phys. Rev. A **37**, 1588 (1988).
[14] G. P. Agrawal, *Nonlinear Fiber Optics*, 3rd ed. (Academic, San Diego, 2001), p. 219.
[15] A. Rundquist *et al.*, Science **280**, 1412 (1998).
[16] A.L. Ford and J.C. Browne, Phys. Rev. A **7**, 418 (1973).
[17] R.W. Minck, E.E. Hagenlocker, and W.G. Rado, Phys. Rev. Lett. **17**, 229 (1966).
[18] E.T.J. Nibbering, G. Grillon, M.A. Franco, B.S. Prade, and A. Mysyrowicz, J. Opt. Soc. Am. B **14**, 650 (1997).
[19] J. Ortigoso, M. Rodriguez, M. Gupta, and B. Friedrich, J. Chem. Phys. **110**, 3870 (1999).
[20] N. Bloembergen and Y.R. Shen, Phys. Rev. Lett. **12**, 504 (1964).
[21] A. Nazarkin, G. Korn, M. Wittmann, and T. Elsaesser, Phys. Rev. A **65**, 041802(R) (2002).