Polarization effect in impulsive rotational Raman scattering in a hydrogen gas

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Abstract: We theoretically show that an elliptically polarized fs pulse impulsively excites rotational Raman coherence in a hydrogen gas more efficiently than a linearly polarized pulse. The phonon amplitude dramatically depends on ellipticity and propagation distance.

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Impulsive stimulated Raman scattering [1] is one of the methods of ultrashort-pulse generation: a femtosecond pump pulse excites a Raman medium impulsively, and a weaker probe pulse is scattered by the coherently oscillating molecular vibrations or rotations, leading to the generation of Raman sidebands. While existing work is concentrated on a linearly polarized pump pulse, in this study [2] we theoretically investigate the dynamics of rotational Raman coherence excited impulsively in a hollow fiber with an inner diameter of $126\,\mu$m filled with 0.5 atm of H_2 by a fs laser pulse with an *arbitrary ellipticity*.

We solve an extended nonlinear Schrödinger equation based on the rotationally invariant formalism [3],

$$\frac{\partial A_\gamma}{\partial z} + \sum_{\ell=2}^{\ell_{\max}} \frac{i^{\ell-1}\beta_\ell}{\ell!}\frac{\partial^\ell A_\gamma}{\partial t^\ell} + \frac{\sigma}{2}A_\gamma = \frac{2in_2k_0}{3}\left(1+\frac{i}{\omega_0}\frac{\partial}{\partial t}\right)\left[\left(|A_\gamma|^2+2|A_{-\gamma}|^2\right)A_\gamma\right] + i4\pi k_0 C_R \sum_{m\alpha} Q_m A_\alpha < m|\alpha\ -\gamma >, \qquad (1)$$

coupled with an equation for phonon creation:

$$\left(\frac{\partial^2}{\partial t^2}+\Omega_R^2\right)Q_n = C_R\sum_{\alpha\beta} A_\alpha A_\beta^* < -n|\alpha\ -\beta >, \qquad (2)$$

where A_γ denotes the electric field envelope of polarization $\gamma\,(=\pm1)$, Q_m the amplitude of the phonon of $m\,(=\pm2,0)$ units of angular momentum along the propagation axis z, 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. $< m|\alpha\,\beta >$ stands for the Clebsch-Gordan coefficient $< 2\,m|1\,1\,\alpha\,\beta >$.

The pump pulse has a sech^2 temporal profile with a peak intensity of $10^{13}\,\mathrm{W/cm^2}$ and a pulse duration (FWHM) of 40 fs. After the pump pulse has passed, Q_n is of the following form:

$$Q_n = q_n e^{-i\Omega_R t} + q_{-n}^* e^{i\Omega_R t} \qquad (n=\pm2,0). \qquad (3)$$

We plot q_n as a function of z for different values of e_p in Fig. 1. Although phonons can be impulsively excited even with a linearly ($e_p=0$) or circularly ($e_p=1$) polarized pulse, the phonon excitation is more efficient for an elliptical polarization, and is most efficient around $e_p=0.5$. The excitation of phonons with $m=\pm2$ largely depends on e_p, and its variation with z is remarkable.

We next consider 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. Figure 2 presents the spectrum of the probe pulse at $z=1$ m. In the case of an elliptically polarized pump pulse (thick solid and dotted curves), high-order Raman sidebands are efficiently

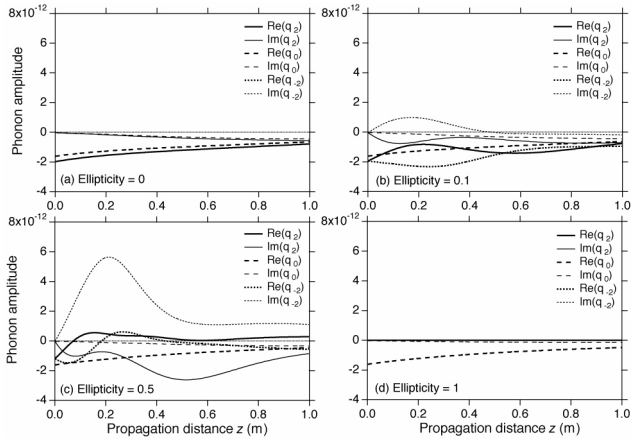

Fig. 1: The distribution of the phonon amplitude q_m $(m = \pm 2, 0)$ excited by a pump pulse with a duration of 40 fs and different values of ellipticity e_p indicated in the figure.

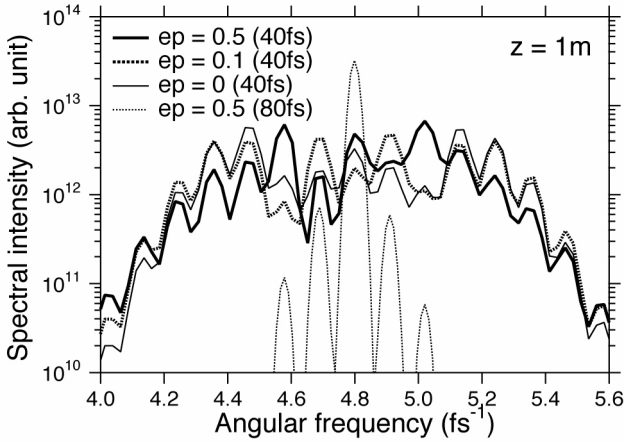

Fig. 2: Intensity spectrum of a probe pulse after 1m of propagation in the medium excited by a pump pulse with different values of ellipticity e_p and duration T_0. Thick solid curve: $e_p = 0.5$ and $T_0 = 40\,\text{fs}$, thick dotted curve: $e_p = 0.1$ and $T_0 = 40\,\text{fs}$, thin solid curve: $e_p = 0$ and $T_0 = 40\,\text{fs}$, thin dotted curve: $e_p = 0.5$ and $T_0 = 80\,\text{fs}$.

formed, and the bandwidth is at least comparable to the case of a linearly polarized pump pulse (thin solid curve).

In conclusion, the excitation of Raman coherence by the pump pulse depends strongly on its polarization and propagation distance, and is more efficient for an elliptically polarized pump pulse than for a commonly used linearly polarized pump. When a probe pulse is injected into thus excited media, high-order Raman components are efficiently formed in the pulse, and its spectrum is broadened to an extent at least comparable to the case of a linearly polarized pump.

References

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