Coherent control over two-dimensional lattice vibrational trajectories in α-quartz using polarization pulse shaping

Masaaki Sato,[1] Takuya Higuchi,[2,3] Makoto Kuwata-Gonokami,[2,3,4] and Kazuhiko Misawa[1,5]

[1] *Department of Applied Physics, Tokyo University of A&T, 2-24-16 Naka-cho, Koganei, Tokyo 184-8588, Japan*
[2] *Department of Applied Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*
[3] *Department of Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan*
[4] *Photon Science Center, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*
[5] *Interdisciplinary Research Unit in Photon-nano Science, Tokyo University of A&T 2-24-16 Naka-cho, Koganei, Tokyo 184-8588, Japan*

kmisawa@cc.tuat.ac.jp

Abstract: We applied polarization pulse shaping to control the trajectory of two-dimensional vibrational motion in α-quartz. Polarization twisted pulses were used to impart peudorotational motion of the degenerate E-symmetry optical phonon mode selectively through impulsive stimulated Raman scattering.

OCIS codes: 320.5540, 320.7120, 300.6240.

1. Introduction

Femtosecond pulse shaping techniques are used to transform a single ultrashort pulse into tailored chemical and physical responses from molecules and materials. More recently, polarization shaped pulses with an arbitrary vectorial electric fields as a function of time have attracted considerable interest [1] for manipulating spatially anisotropic or chiral materials. One of the targets is optical control over two-dimensional vibrational trajectories along degenerate lattice vibrational modes. In earlier experiments [2], pairs of pulses with different polarizations were used instead of a single shaped pulse to impart momentum impulsively and selectively in orthogonal directions in the crystal of α-quartz. Pseudorotational motion of the atoms involved in the vibrational motion can be selected by variation of timing between two excitation pulses of different polarizations. However, the only adjustable parameter was the time delay between the pulses in a pair, which simultaneously determines both the relative phase between the vibrational motions along the two orthogonal axes of coordinate, and the beat frequency for repetitive excitation resonant with a particular mode. To achieve more flexible and arbitrary control, polarization-shaped pulses are required.

We recently implemented polarization pulse shaping for arbitrary vectorial fields by employing a Fourier-synthesis-based optical pulse shaper and quarter-wave plate. We controlled the instantaneous intensity and polarization state by the pulse shaper and converted such tailored pulses into terahertz waves, using a nonlinear crystal [3]. Wavelength conversion from the optical to terahertz frequency range was performed by optical rectification along the three-fold axis of the crystal. In the present study, we were successful in selectively driving a desired pseudorotational motion of a single vibrational mode in α-quartz crystal in a manner similar to the optical rectification for terahertz radiation. We used polarization-twisted pulses and discriminated degenerate E-modes by tuning the twist frequency.

2. Methods

Impulsive stimulated Raman scattering (ISRS) is used to study the mode selection by polarization-shaped pulses in the time domain. Pump-induced spectral phase modulation was probed. The α-quartz crystal has a C_3 axis of symmetry that yields three normal modes of oscillation. One is the breathing mode with A symmetry and the other two are degenerate E-modes. The displacements of the A-mode $q_{\mathrm{A}}(t)$ and E-modes $q_{\mathrm{E_a}}(t)$ and $q_{\mathrm{E_b}}(t)$ can be written in the following simple form owing to the three-fold crystal symmetry [3].

$$q_{\mathrm{A}}(t) = \int_{-\infty}^{t} R_{\mathrm{A}}(t-\tau)I(\tau)\mathrm{d}\tau = \int_{-\infty}^{t} R_{\mathrm{A}}(t-\tau)I_0\mathrm{d}\tau \tag{1}$$

$$q_{\mathrm{E_a}}(t) = \int_{-\infty}^{t} R_{\mathrm{E}}(t-\tau)I(\tau)\cos 2\eta(\tau)\cos 2\phi(\tau)\mathrm{d}\tau = \int_{-\infty}^{t} R_{\mathrm{E}}(t-\tau)I_0\cos 2\omega\tau\mathrm{d}\tau \tag{2}$$

$$q_{\mathrm{E_b}}(t) = -\int_{-\infty}^{t} R_{\mathrm{E}}(t-\tau)I(\tau)\cos 2\eta(\tau)\sin 2\phi(\tau)\mathrm{d}\tau = \int_{-\infty}^{t} R_{\mathrm{E}}(t-\tau)I_0\sin 2\omega\tau\mathrm{d}\tau \tag{3}$$

Here, $I(t)$, $\phi(t)$, and $\eta(t)$ denote the intensity envelope of the excitation pulse, polarization azimuth angle, and ellipticity, respectively. In the present study, we used polarization-twisted pulses in which the direction of linear polarization was rotated at a prescribed angular frequency over the intensity envelope, as depicted in Fig. 1(a). Polarization-twisted pulses correspond to conditions as $\phi(t) = \omega t$ and $\eta = 1$. A sufficiently large linear chirp was introduced so that the temporal duration of the intensity envelope was long enough than the vibrational periods assumed a quasi-continuous excitation $I(t) \approx I_0$. Therefore, the breathing A-mode cannot be excited. On the contrary, the direction of the E-mode vibration was circularly rotating with an angular frequency of the electric polarization vector of $\Omega = 2\omega$.

3. Results

The red and blue curves in Fig. 1(b) represent the Raman spectra calculated from ISRS transients for excitations with polarization-twisted pulses at 128 and 402 cm^{-1} of the tuning frequency Ω, respectively. The Raman spectrum obtained with Fourier-transform limited (FTL) pulses is also shown in green. By FTL pulses A- and E-modes are simultaneously excited, and owing to the broad bandwidth of the pump pulse several frequency modes within the pulse bandwidth are observed. The measured frequencies of 128, 203, 357, and 465 cm^{-1} are in good agreement with the reported values of 128, 207, 356, and 464 cm^{-1} [4].

A well defined spectral peak was observed in each Raman spectrum just around the tuning frequency shown on the red and blue curves. Either peak shows resonance to the pseudorotational motion in α-quartz. All other A-modes were eliminated as designed. With 402-cm^{-1} polarization-twisted pulses, a small peak appeared at 401 cm^{-1} on the blue curve. This 401-cm^{-1} mode has a very weak Raman cross section, and it is usually hidden behind the tail of the much stronger A-mode at 464 cm^{-1} .

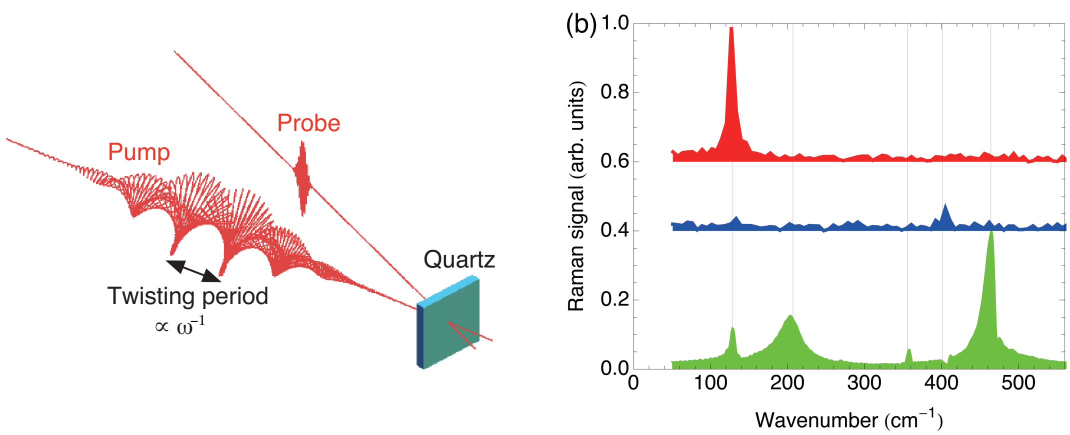

Fig. 1. (a) ISRS configuration by excitation with polarization-twisted pulse (b) Raman spectra for excitations with polarization-twisted pulses at Ω= 128 cm^{-1} (red) and 402 cm^{-1} (blue), as well as with Fourier-transform limited pulses (green)

4. Discussion and conclusion

With use of a polarization-twisted pulse, the two-dimensional pseudorotation of the E-mode was selectively enhanced, while the A-mode was strongly suppressed. If arbitrarily-shaped excitation waveforms are used, more complex vibrational trajectories are also controlled. This technique is applicable to a broad class of degenerate modes, such as electron spin coherence, which can also be induced in different polarization directions by pulses of different polarization.

References

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