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Investigation of vibrational characteristics in BBO crystals by femtosecond CARS

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ABSTRACT

Femtosecond time-resolved coherent anti-Stokes Raman spectroscopy (CARS) is utilized to study the ultrafast vibrational dynamics in BBO crystals at room temperature. Time-resolved two-beam and three-beam CARS are detected. The vibrational dephasing time is analyzed and the changes of vibrational mode intensities with the polarization of pump pulses are observed.

1. Introduction

Coherent anti-Stokes Raman spectroscopy (CARS) has many advantages such as high spatial and temporal resolutions, high sensitivity, etc. Femtosecond time-resolved CARS is a powerful spectroscopic tool for studying the vibrational dynamics of high frequency Raman modes in time domain since the 1980s for some decades [1,2]. This technique has been widely used in different samples, such as polydiacetylene [3,4], porphyrin [5,6], toluene [7], methanol–water [8], ethanol [9], and dipicolinic acid [10–12]. Recently some scientists used the femtosecond time-resolved CARS to study the coherent vibrational process in crystals [13,14]. Liu et al. [15] gained as broad as 12,000 cm^{-1} CARS signals using two crossing femtosecond laser pulses. The effect of the crossing angle between two input beams on the spectrum and emitting angle of the Raman sidebands was studied in detail.

BBO crystals are the abbreviation of β phase barium metaborate crystals and have a wide transmission range, a remarkable nonlinear coefficient, a high damage threshold, etc. They are often used for frequency doubling or in optical parametric amplifiers. The research of BBO crystals focuses on the mechanical and optical properties, and its applications in laser generation; however, research for the ultrafast dynamics process in BBO crystals is relatively rare.

In this paper, two-beam CARS is used to get duration of the laser beams, and then three-beam CARS is used to study the coherent vibrational dynamics process of BBO crystals. The intensities of vibrational modes are controlled by using the polarization of pump pulses.

2. Experimental

The experimental setup used for the time-resolved CARS is shown in Fig.1. The pulses from a commercial femtosecond laser system Ti:sapphire regenerative amplifier (Micra+Legend, Coherent) at 800 nm (2.5 mJ, 1 kHz and 40 fs) were divided into two parts by a 1:1 beam splitter. One beam was used as the Stokes beam ($\mathbf{k}_3$) and the other was used to pump an optical parametric amplifier (OPA, TOPAS, Light Conversion). The output of OPA was split into two parts by a 1:1 beam splitter to obtain the pump and the probe beams ($\mathbf{k}_1$ and $\mathbf{k}_2$). The three beams were attenuated with power of 5–10 mW by a variable neutral-density (ND) filter. The generation of CARS requires spatial and temporal overlap of the pulses in the samples. The relative timing among the different beams was varied by computer controlled delay stages (Jump Star, mmt32-150 with resolution of 0.625 μm). The three beams were aligned parallel to each other, and overlapped in the folded-BOXCARS beam geometry (4 mm spot diameter, square with 10 mm sides). The three beams were focused on a 0.1-mm-thick BBO (θ=29.5°) using a 250 mm focal lens at room temperature. This folded-BOXCARS beam geometry ensures that the CARS signal propagates in a direction different from those of the incoming beams and can therefore be background-free collected. The CARS signal was filtered by a spatial filter (a pinhole) and were detected by two methods. First, it was spectrally dispersed in a monochromator (Omni-λ500, Zolix) and detected by a photomultiplier tube (PMT) with a lock-in amplifier (SR830,

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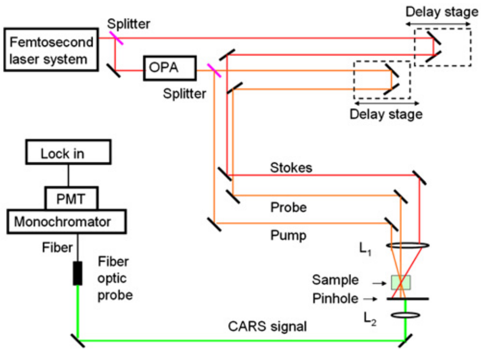

Fig. 1. Femtosecond time-resolved CARS experimental device.

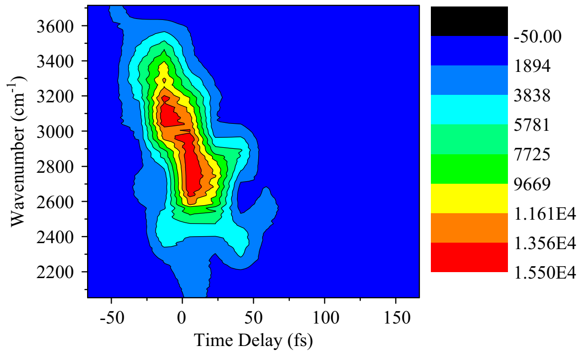

Fig. 2. Time-resolved two-beam CARS of BBO crystals (675 nm pump, 800 nm Stokes).

Stanford Research Systems). Second, it was directly recorded with a fiber optic spectrometer (HR4000, Ocean Optics).

3. Results and discussion

3.1. Two-beam CARS

The time-resolved two-beam CARS of BBO crystal (675 nm pump, 800 nm stokes) is shown in Fig. 2. The signal is collected in $2\mathbf{k}_3-\mathbf{k}_1$ direction.

There are two islands in the contour plot which can be seen clearly in Fig. 2. We can see that two laser beams first stimulate the vibrational mode of 3100 cm^{-1} and then after 40 fs stimulate the vibrational mode of 2750 cm^{-1} from Fig. 2. Raman vibrational modes can be distinguished in time domain by using pulse chirp characteristics.

Furthermore the duration [intensity full width at half maximum (FWHM)] of pump and Stokes beams can be got from time-resolved two-beam CARS. Rocha-Mendoza et al. [16] clearly show that CARS response for Gaussian pulses $\propto \exp(-4t_0^2/4.14\tau^2)/\tau^2$, where t_0 is the time delay and τ is FWHM of the Gaussian pulses. The dependence of two-beam CARS signal on pump delay of BBO crystal (670 nm pump, 800 nm stokes) is shown in Fig. 3. Fitting the dependence of CARS signal on time delay using CARS response $\propto \exp(-4t_0^2/4.14\tau^2)/\tau^2$, we can get FWHM of pump and Stokes beams as 42 ± 1.2 fs. This result is similar to FWHM of the legend laser pulse.

3.2. Three-beam CARS

The time-resolved three-beam CARS of BBO crystal (675 nm pump, 800 nm Stokes) is shown in Fig. 4. The signal is collected in $\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3$direction.

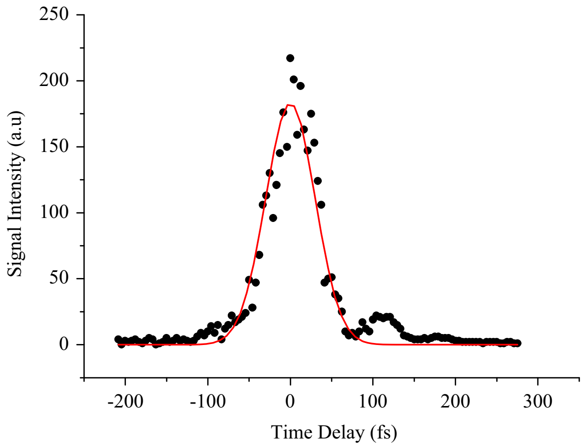

Fig. 3. Dependence of two-beam CARS signal on pump delay of BBO crystals (670 nm pump, 800 nm Stokes).

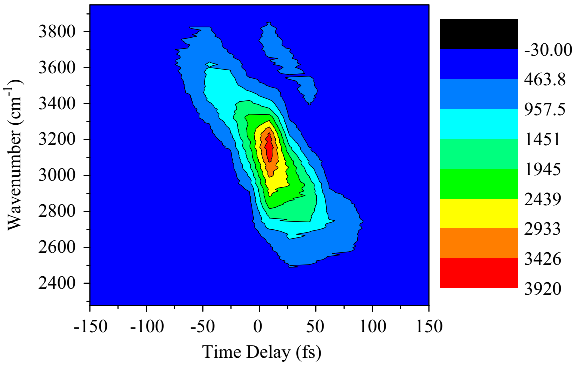

Fig. 4. Time-resolved three-beam CARS spectrum of BBO crystals (675 nm pump, 800 nm Stokes).

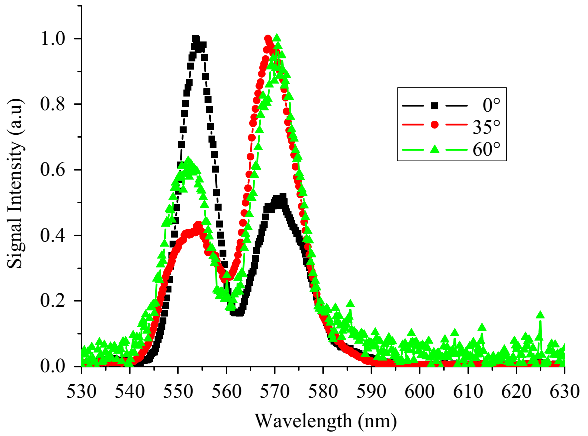

Fig. 5. CARS of BBO crystals under pump pulse of different polarizations (675 nm pump, 800 nm Stokes).

Three-beam CARS is different from two-beam CARS. Only the vibrational mode of 3100 cm^{-1} appears clearly in three-beam CARS. The vibrational mode of 3700 cm^{-1} is weaker. The vibrational mode of 3700 cm^{-1} is not detected in the two-beam CARS, which may be due to different chirps between the pump and the probe beams.

The polarization of pump pulses will influence the intensity of the vibrational mode. The CARS of BBO crystals under pump pulses of different polarizations is shown in Fig. 5. The degree is polarization difference between the pump and Stokes pulses. From Fig. 5 we can see that the polarization of pump pulses will