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First observation of $E_2$ coherent phonon modes in CdS using a noncollinear optical parametric amplifier

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Abstract. We report the first observation of $E_2$ coherent phonon modes in CdS using a noncollinear optical parametric amplifier. We studied the intensity, temperature and excitation energy dependences of the high- and low-frequency $E_2$ modes. We obtain that generation mechanism under non- and near resonant regions is impulsive stimulated Raman scattering (ISRS) and $E_2$ modes are barely affected by Fröhlich interaction. As temperature increases, $E_2$ modes are affected by anharmonic effect. The observed phonon lifetime of the low- and high-$E_2$ modes in CdS is 243 ± 37 and 11.4 ± 0.4 ps, respectively, at 83 K. From fitting, we can estimate the lifetime of the high-$E_2$ mode at the low-temperature limit to be 49.3 ps.

1. Introduction

Coherent phonon means coherent lattice vibration. To excite coherent phonon, it is necessary that laser pulse duration is shorter than the oscillation period. Recent development in femtosecond laser technology has made it possible to excite coherent phonon.

Many studies of coherent longitudinal optical (LO) phonon in semiconductors have been reported [1, 2]. Among them, $E_2$ (non-polar) modes were studied only in ZnO and GaN [4, 5, 9]. To our knowledge, coherent phonon modes in CdS have not been reported, although optical properties of CdS have been studied extensively [6, 7, 8]. $E_2$ modes are expected to play a role in metrology, because the low-$E_2$ mode has a very long lifetime and enables highly accurate determination of frequency. There is also a possibility that we can observe the enhancement of amplitude of coherent phonon for $E_2$ modes even in intense excitation, because these two modes interact weakly with photoexcited carriers and are expected to show no saturation, contrary to the case for Bi [3].

In this study, we observed the high- and low-$E_2$ modes for the first time in the wide-gap semiconductor CdS, and studied the intensity and temperature dependence of dephasing for these modes.

2. Experiment

We performed a degenerative pump-probe experiment using a home-built noncollinear optical parametric amplifier (NOPA) that produces 35-60 fs pulses. The central pulse energy can be tuned from 1.94 to 2.38 eV and the typical spectral width is 70 meV.

The sample was a $10 \times 10 \times 1$ mm$^3$ single wurtzite structural crystal grown along the [0001] axis. Figure 1. shows relationship between pump energies and absorption spectra of our CdS sample at 83 K (a) and 300 K (b). Pump-induced transmittance changes were measured by polarization-sensitive
electro-optic sampling as a function of time delay between pump and probe. The electric field of the
pump was set along the [2110] axis and that of the probe was set 45 degrees with respect to the same
axis. The high- and low-E_2 modes are expected to be observed under these conditions [4]. All
experiments were performed at 83 K except for the temperature-dependence experiments.

![Figure 1](image1.png)

**Figure 1.** Relationship between pump energies and absorption spectra: (a) At 83 K and (b) At 300 K.

3. Result

3.1. Observation of E_2 modes

Figure 2. (a) shows typical transmittance change measured by the pump-probe method using the 2.21
eV pulse. We observe the two oscillating components on the slowly varying background, as shown in
the inset.

Figure 2. (b) shows fast Fourier transform (FFT) spectra for each time span. The two peaks at 1.27
and 7.67 THz correspond to the high- and low-E_2 mode, respectively [8]. The lifetime of the high-E_2
mode is less than 20 ps, whereas that of the low-E_2 mode is relatively long.

The frequency, amplitude, and decay time of the two modes was determined by fitting using the
following equation. The background was removed using a band-pass filter for low- or high-mode, respectively.

\[ \Delta T / T = A_{h,i} \exp(-t/\tau_{h,i}) \sin(2\pi f_{h,i} t + \alpha_{h,i}) \] (1)

The estimated phonon lifetimes of the low- and high-E_2 mode in CdS are 243 ± 37 and 11.4 ± 0.4 ps,
respectively. The former is longer than that in ZnO at 5 K (211 ps) [9].

![Figure 2](image2.png)

**Figure 2.** (a) Transmission change (raw data) as a function of the delay between pump and probe pulses. The inset shows appearance of phonon oscillations. (b) FFT spectra for each span.
3.2. Generation Mechanism and excitation intensity dependence of coherent E2 modes

Figure 3. (a) shows the pump polarization dependence of the high-E2 mode using 2.3 eV pulse. The observed phonon oscillation obeyed the Raman selection rule for E2 mode [4]. A similar result was obtained for the low-E2 mode using various excitation energies. We see that, in CdS, generation mechanism of coherent phonon below and near the band-gap is ISRS mechanism.

E2 modes are supposed to be little influenced by the carrier directly because E2 modes are not polar mode. Figure 3. (b) shows phonon dephasing rate for the high-E2 mode as a function of pump intensity using 2.3 eV pulse. Unlike the case for the LO modes [5], the dephasing rate does not increase. A similar result was obtained for the low-E2 mode and using various excitation energies.

![Figure 3. High-E2 mode excited by 2.3 eV: (a) Transient transmittance change as a function of time delay at the angle of pump polarization φ defined with respect to [2110]. (b) Dephasing rate as a function of pump intensity.](image)

3.3. Temperature dependence of dephasing rate

Figure 4. shows the effect of temperature on dephasing rate for the low-E2 mode using 2.07 eV pulse (a) and high-E2 mode using 2.21 eV pulse (b). Because of phonon-phonon interaction, dephasing times become shorter from 11 ps to 3 ps for the high-E2 mode and from 189 ps to 23 ps for the low-E2 modes with increasing temperature. This decrease in phonon lifetime with increasing temperature is seen at various excitation energies. It is thought that phonon lifetime is limited as a result of phonon-phonon interaction and related to phonon dispersion [10]. Because there are many possible branches of decay for the high-E2 mode but only a few for the low-E2 mode, the high-E2 mode is expected to be more affected by anharmonic effect, and thus to have a shorter lifetime, than the low-E2 mode. However, we cannot find the reason why the low-E2 mode shows superlinear dependence on temperature while the high-E2 mode shows linear dependence.

The temperature dependence of the dephasing rate for the high-E2 mode can be fitted using the following equation

\[ \Gamma(T) = \Gamma_0 + \Gamma_{DN}(1 + n_1 + n_2) + \Gamma_{UP}(n_3 - n_4) \]

\[ n_{1,2,3,4} = [\exp(hf_{1,2,3,4} / k_B T) - 1]^{-1} \]

Where \( \Gamma_0 \) is temperature-independent term related to defect and/or impurity scattering, and \( \Gamma_{DN} \) and \( \Gamma_{UP} \) are third-order anharmonic coefficients related to down-conversion process and up-conversion process, respectively [8]. From phonon dispersion curve for CdS [11], we take, for the high-E2 mode, the frequency that is \( f_1 = 3.19 \) THz, \( f_2 = 4.48 \) THz for the down-conversion channel and \( f_1 = 1.03 \) THz, \( f_4 = 8.7 \) THz for the up-conversion channel. We cannot identify the channel to fit for the low-E2 mode from our result, but the lifetime of that mode possibly approach nanosecond at the low temperature limit.

From this calculation, we can estimate that the lifetime of the high-E2 mode in CdS at low-temperature is 49.3 ps, which is longer than that in ZnO [12].
Although the lifetimes of both modes are affected by anharmonic effect, we did not observe frequency softening [9].

![Figure 4](image-url)  
**Figure 4.** Dephasing rate for E₂ modes as a function of temperature: (a) Low-E₂ mode; (b) High-E₂ mode

4. Conclusion
We studied the contribution of intensity, temperature and excitation energy to the high- and low-E₂ modes in CdS. We determined that generation mechanism under non-resonant region is ISRS and E₂ modes are not much affected by Fröhlich interaction. At high temperatures, E₂ modes are affected by anharmonic effect. The measured lifetime of the low- and high-E₂ mode in CdS are 243 ± 37 and 11.4 ± 0.4 ps, respectively, at 83 K in our measurement. From fitting, we estimate the lifetime of the high- E₂ mode at low-temperature limit to be 49.3 ps.

References