Coherent acoustic phonons in quantum dots

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Abstract. The authors studied confined acoustic phonons in semiconductor quantum dots by using coherent phonon spectroscopy. Time-resolved pump-and-probe method was applied to CuBr QDs embedded in glass matrices with the second harmonic pulses from a femtosecond Ti:S laser oscillator. A clear damped oscillation signal with frequency of sub-THz was observed at room temperature. The frequency of the oscillation increases with decreasing the mean radius of dots, but the size-dependence does not show a simple $1/R$ characteristics of confined acoustic phonon.

1. Introduction

In quantum dots (QDs), lattice vibrations are confined in nanometer-sized sphere and characteristic confined phonon modes appear. Recently, excitonic coherence in QDs is extensively studied because of the long coherence time in this system, which is desirable for device application such as quantum computation. Interaction with confined acoustic phonons is found to be responsible for the excitonic dephasing in QDs at low temperatures [1, 2] and should be understood in detail.

So far confined acoustic phonons in QDs have been studied mostly by Raman scattering [3-6], while little attention has been given to their dynamics. Moreover, in PbSe QDs, it was shown that coherent phonon experiment measured different confined acoustic modes from that in Raman scattering [7]. Studies on the coherent acoustic phonons in semiconductor QDs are limited to a couple of works (PbS QDs [8] and PbTe QDs [9]).

In this work, we study confined acoustic phonons in CuBr QDs embedded in glass matrices by applying coherent phonon spectroscopy. The size-dependence is investigated by using the samples with different mean radius. As an independent measurement of the phonon energy, persistent spectral hole burning (PSHB) was performed.

2. Experimental Setup

We performed a time-resolved pump-and-probe measurement at room temperature. The light source is a mode-locked titanium sapphire laser (Spectra Physics, MaiTai) which operates at 80 MHz. Typical pulse width is 80 fs. These pulses were converted into the second harmonics and were used as pump pulses and probe pulses. They were focused on the sample surface after passing through a delay line. The delay time between pump and probe pulses was modulated by introducing a shaker in the pump beam pass. A lock-in amplifier was used to extract the modulated signal. The modulation frequency was about 80 Hz, and the modulation depth was appropriately adjusted to obtain clear oscillation signals.

The samples used here were CuBr quantum dots embedded in silicate glass matrices. The mean radii of QDs were estimated by using an empirical formula between the excitonic

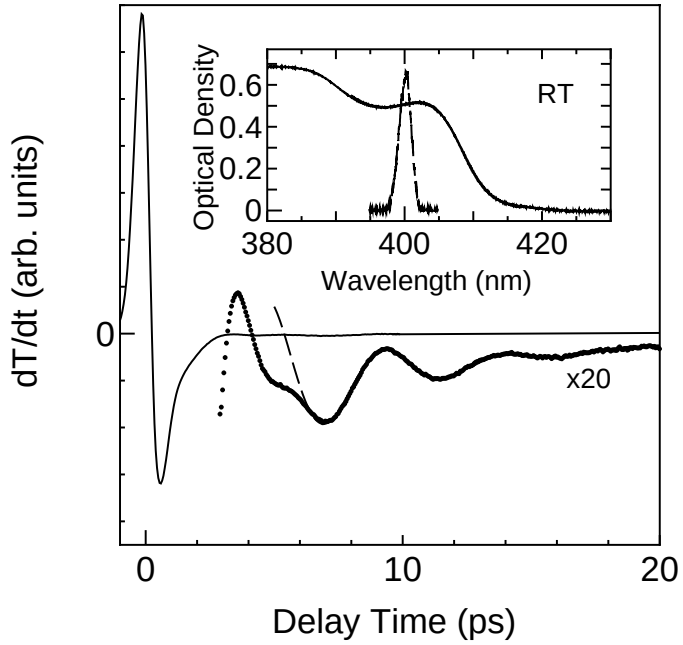

Figure 1. The results of pump-and-probe transmission measured by using delay time modulation. A clear damped oscillation is observed in the expanded plots. A dashed line represents the results of fitting by a damped cosine function. Inset : Absorption spectrum of CuBr QDs in glass at room temperature. Mean radius of the sample is 5.4 nm. A dashed line shows the spectrum of the pump and probe pulse.

absorption peak energy and the radius, that was determined by the small-angle X-ray scattering.

3. Results and Discussions

Absorption spectra of the CuBr QDs at room temperature are shown in the inset of Fig. 1. We can see two excitonic absorption bands near UV region. The mean radius of quantum dots is estimated to be 5.4 nm. The spectrum of the pump and probe pulses are shown in the inset as a dashed line, which locate at the high energy side of $Z_{1,2}$ exciton peak. The time trace of the transmission is shown in Fig. 1. Since the time modulation technique is utilized in this work, the data in this figure almost corresponds to the time-derivative of ordinary time trace. In this measurement, the amplitude of the time modulation was about 1 ps. We can see an abrupt change of transmission at zero time delay due to absorption saturation.

The tail part of the time trace is expanded by 20 and is plotted by dots. A clear oscillation of transmission intensity is observed on the tail part of the bleaching recovery. The change of transmission by this oscillatory component is small as $\Delta T/T \leq 10^{-5}$. The oscillatory component is well fitted by a damped cosine function whose period is 4.6 ps and damping constant is 3.4 ps. As seen later, this oscillation corresponds to the confined acoustic phonon that was coherently generated by the ultrashort pump pulses. The observed damping constant is comparable to the so-called dephasing time coming from the size-distribution of the sample, so we do not discuss the damping constant here.

In order to investigate the size-dependence of the oscillation, the same measurement was performed for the samples with different mean radii. Figure 2(a) shows the absorption spectra of the samples at 2 K. The mean radii of these samples are 2.0, 2.7, 4.2, 4.9, 5.4 nm, from top

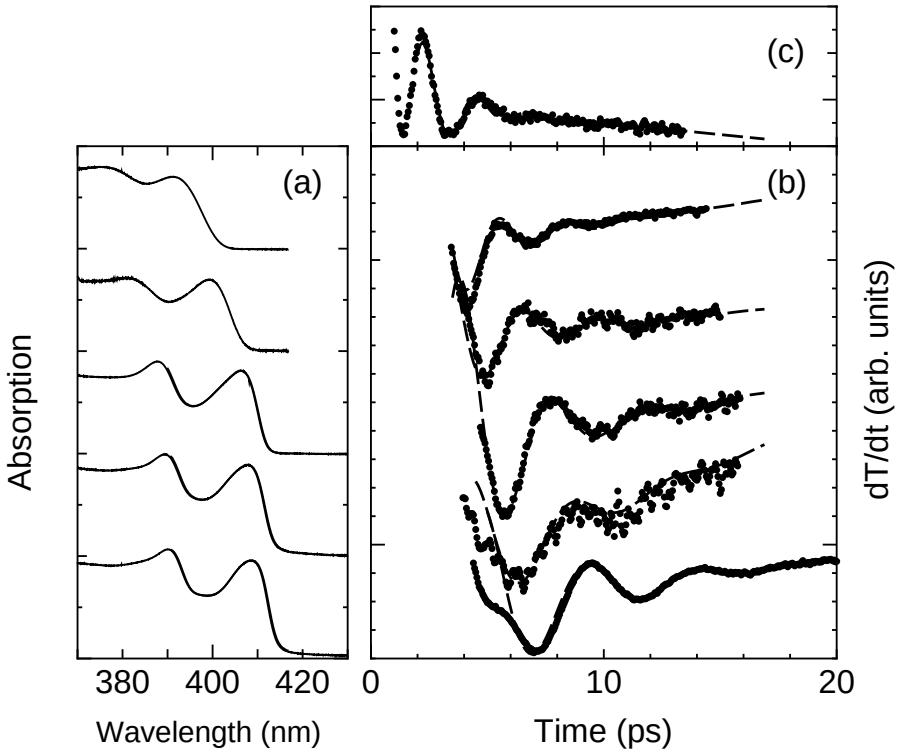

Figure 2. (a) Absorption spectra of CuBr QDs at 2 K. The mean radii of these samples are 2.0, 2.7, 4.2, 4.9, 5.4 nm, from top to bottom. (b) The tail part of the time trace for corresponding samples. The lowest curve is the same data that shown in Fig. 1. (c) Another oscillatory component that was observed in 4.9 nm sample.

to bottom. The results of pump-and-probe measurement for corresponding samples are shown in Fig. 2(b). The wavelength of the laser is shifted for each sample in order to observe the most clear oscillation signal. Since the signals around zero time delay are similar among them, only the tail part of time traces are plotted here. It is easily seen that the oscillation period decreases with decreasing dot radius. All data are well fitted by a damped cosine function as shown by dashed lines.

The frequency of the confined phonon is known to be proportional to the inverse of the dot radius. We plotted the frequency of the oscillation as a function of the inverse of dot radius in Fig. 3. This feature seems to be roughly reproduced in our case, but the plotted points can not be on a line that pass through the origin of the coordinate axes.

There are some interesting facts in our data as seen in the following. First of all, let us consider the size-dependence of the confined acoustic phonon based on an elastic continuum model[10, 11]. According to this model, vibration modes of elastic sphere can be categorized into two groups; spheroidal modes and torsional modes. We discuss here the spheroidal mode under stress-free boundary condition. Each modes can be specified by two indices, that is, the angular momentum number l and the branch number n. For example, the eigen frequency of the spherical modes (l=0) are obtained by solving the following equation.

$$\tan\left(\frac{\omega a}{V_l}\right) = \frac{\omega a / V_l}{1 - \frac{c_{11}}{4c_{44}}\left(\frac{\omega a}{V_l}\right)^2}$$

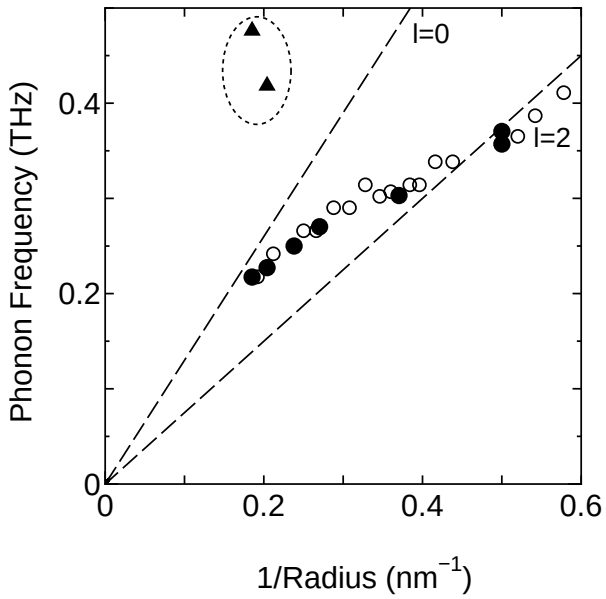

Figure 3. Size-dependence of the frequency of oscillatory component (solid circles). Dashed lines represent the size-dependence of the lowest l=0 mode and l=2 mode calculated by using elastic continuum model. Solid triangles represent the frequency of the high-frequency component (see text). Open circles show the results of PSHB spectroscopy. The data of two different measurements coincide quite well.

Here, V_l and a are the longitudinal sound velocity and the radius of the sphere, respectively. c_{11} and c_{44} are elastic constants of the material.

Finally, eigen frequency can be expressed as ξ_{ln}/R, and ξ_{ln} depends on the mode and material parameters. The lowest frequency is realized for the l=2 spheroidal mode with the smallest branch number. If we assume that the elastic constants and density of CuBr QDs are the same as those in bulk CuBr, we can calculate the coefficient ξ_{ln} of confined acoustic phonons in CuBr QDs. These results are presented in Fig. 3 as dashed lines for (l=0, n=1) and (l=2, n=1) mode. The observed data are close to the l=0 mode for larger radius, while they are close to l=2 mode for smaller radius.

We would like to mention that the same behavior was observed in PSHB spectroscopy. In that experiment, the size-dependence of the confined acoustic phonons was measured very precisely based on the site-selective excitation technique utilizing ns-pulsed dye laser at 2 K. Confined acoustic phonon appears as a small sideband near the main hole in PSHB spectrum, and the energy separation from the main hole is plotted in Fig. 3 as a function of selectively excited radius by open circles. These two data coincides quite well, indicating the high reliability of our measurements. At the moment, we do not have reasonable explanation for this strange behavior in the size-dependence. To the best of my knowledge, all of the experimantally observed coherent acoustic phonon in quantum dots are assigned to the breathing mode (l=0, n=1) and not the mode (l=2, n=1). A systematic study of polarized Raman scattering on the confined phonon in CuBr QDs may be valuable, because probably we can distinguish these two modes by the difference of polarization dependence[13].

In addition to the above mentioned mode, we have observed another oscillatory component whose frequency is about twice high. The signal was obtained in CuBr QDs with mean radius of 4.9 nm by using smaller modulation amplitude, and plotted in Fig. 2(c) and Fig. 3. Unfortunately, this component is not systematically studied yet, but it is possible that the fast oscillation component comes from the mode with higher branch number, for

example (l=0, n=2) mode. Recently, such overtone modes are reported in Raman scattering of CdS_xSe_{1-x} QDs[12].

Sometimes the initial phase of coherent phonon oscillation is useful to discuss the generation mechanism of coherent phonons. In our case, all of the coherent phonon signals were well fitted by a cosine function. This fact means that the real phonon oscillation is sine-like function, because in this work we measured time-derivative signal by using delay time modulation. It is quite different situation from other systems such as PbSe QDs[7], PbS QDs[8], and PbTe QDs[9] where acoustic phonon oscillation has a maximum amplitude at t=0 and the displacive excitation mechanism of coherent phonon [14] is suggested.

4. Conclusion

Confined acoustic phonons in CuBr quantum dots was studied by using coherent phonon spectroscopy. A clear damped oscillation signal with frequency of sub-THz was observed at room temperature. The size-dependence of the coherent phonon frequency agreed with that of PSHB measurement. It was compared to the calculation based on the simple elastic continuum model, and found that the size-dependence could not be simply explained by one of the eigenmodes.

References

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