# Residual dephasing mechanism at low temperature in semiconductor quantum dots

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**Abstract.** We have systematically investigated low-temperature excitonic dephasing in CuBr and CdSe quantum dots by means of heterodyne-detected accumulated photon echo method. It has been clarified that two characteristic mechanisms determine the homogeneous broadening at low temperature: the small energy excitation in two-level system and the two-phonon Raman process of confined acoustic phonons. The former brings about the matrix-dependent behaviour of the temperature dependence between CuBr quantum dots in glass and CuBr quantum dots in NaBr below 15 K. Confined acoustic-phonon contribution becomes dominant above 15 K and 10 K for CuBr and CdSe quantum dots, respectively. These results closely resemble the previous report of CuCl quantum dots in weak confinement regime, showing the universal role of surface-related effects and confined phonon scattering as the residual dephasing mechanism of semiconductor quantum dots in matrices at low temperature.

## 1. Introduction

There is a growing interest in utilizing semiconductor quantum dots (QDs) for all-optical implementation of quantum information processing [1, 2]. Recently, dephasing time ranging from tens of picoseconds to a nanosecond of I-VII, II-VI and III-V semiconductor QDs has been reported [3-8], which is very long compared with femtosecond laser implementation. This offers remarkable opportunities for a number of implementations until the decoherence takes place. In order to realize the optical quantum information processing with semiconductor QDs, it is a pressing problem how to prolong dephasing time of QDs. From the fundamental physical point of view, full understanding of restriction to shorten the dephasing time of QDs is most important.

As a result of three-dimensional confinement effect, QDs exhibit characteristic discrete δ-function-like electronic energy spectrum. Discrete energy spectrum allows the one-phonon-assisted interlevel transition only for energy matching phonons. When the homogeneous linewidth of zero-phonon line is separated from the other lines by more than phonon energy, the temperature dependence of the homogeneous broadening is dominated only by inefficient two-phonon process. This brings forth the narrow linewidth, nonlinear temperature broadening and the long dephasing time in the time domain. However linear temperature broadening or activation type nonlinear temperature broadening was contradictorily reported in the self-assembled quantum dots [7, 8]. On the other hand, two-phonon Raman process of confined acoustic phonons was reported in

CuCl QDs in the weak confinement regime [4, 5], while smaller energy excitation process in the two-level-system (TLS) was proposed in [4] besides. Two-acoustic-phonon process alone was also reported in CdSe QDs embedded in the polymer [6]. Serious difference is present in the understanding of the temperature-dependent homogeneous broadening and dephasing mechanism of QDs. To clarify the phonon broadening mechanism of the zero-phonon line, we should investigate different kinds of QDs whose acoustic phonons are confined, because discrete phonon energy does not blur out temperature dependence of broadening. In this work, we studied CuBr QDs embedded in NaBr crystals and glass in the weak or intermediate confinement regime as well as CdSe QDs embedded in glass in the strong confinement regime at very low temperatures.

## 2. Experiment

The samples studied are CuBr QDs embedded in NaBr crystals (denoted by CuBr/NaBr QDs), CuBr QDs in potassium alminoborosilicate glass (CuBr/glass QDs) and CdSe QDs in $GeO_2$:$Na_2O$ glass (CdSe/glass QDs). The average radius of the QDs were estimated to be 2.9, 3.3, 3.8, 4.8 and 6.5 nm for CuBr/glass QDs, 2.9, 4.8 and 6.5 nm for CuBr/NaBr QDs, and 2.3, 2.5, 2.7 and 3.6 nm for CdSe/glass QDs. Homogeneous linewidth ($\Gamma_h$) was obtained from the inverse value of dephasing time, measured by the heterodyne-detected accumulated photon echo (HAPE) [9, 10]. Using this technique, we have carefully measured the temperature dependence of the homogeneous width from the time domain. For CuBr/NaBr and CuBr/glass QDs, a frequency-doubled beam of the output pulses of 130-fs temporal duration from a mode-locked Ti:sapphire laser was used to excite resonantly the $Z_{1,2}$ exciton in each sample. For CdSe/glass QDs, an optical parametric generator pumped by 200 kHz regeneratively amplified output of a femtosecond Ti:sapphire laser was used for the excitation of $1S_{3/2}1S_e$ transition. For a very low-temperature measurement ($T < 1.5$ K) one-shot $^3$He cryostat [4] was used, while an either liquid immersion or gas-flow type $^4$He cryostat was used above 1.5 K.

## 3. Results and discussion

Figure 1(a) shows the time-trace of HAPE signals of 6.5 nm-radius CuBr/NaBr QDs and CuBr/glass QDs at different temperatures. All the echo signals exhibit single-exponential decay with single time constants corresponding to $T_2/2$, where $T_2$ is the dephasing time of exciton in QDs [9]. Although no difference was seen between the time-trace signals of CuBr/NaBr and CuBr/glass QDs above 20 K, the former showed longer $T_2$ than the latter at 4 K. The difference of the homogeneous linewidth estimated by Fourier-cosine transformation [12] as a function of temperature is clearly seen in Fig. 1(b). Although they were similar to each other above 15 K, sudden narrowing occurred in CuBr/NaBr below 15 K, providing a large difference between them. This anomalous temperature dependence can be well reproduced by the expression,

$$\Gamma_h(T) = \Gamma_{h0} + A\cosh^{-2}\left(\frac{\delta}{2k_BT}\right) + B\sinh^{-2}\left(\frac{\hbar\omega}{2k_BT}\right) + C\left(\exp\left[\frac{\hbar\Omega_{LO}}{k_BT}\right]^{-1} - 1\right)^{-1} \quad (1)$$

for CuBr/NaBr QDs, while

$$\Gamma_h(T) = \Gamma_{h0} + AT + B\sinh^{-2}\left(\frac{\hbar\omega}{2k_BT}\right) + C\left(\exp\left[\frac{\hbar\Omega_{LO}}{k_BT}\right]^{-1} - 1\right)^{-1} \quad (2)$$

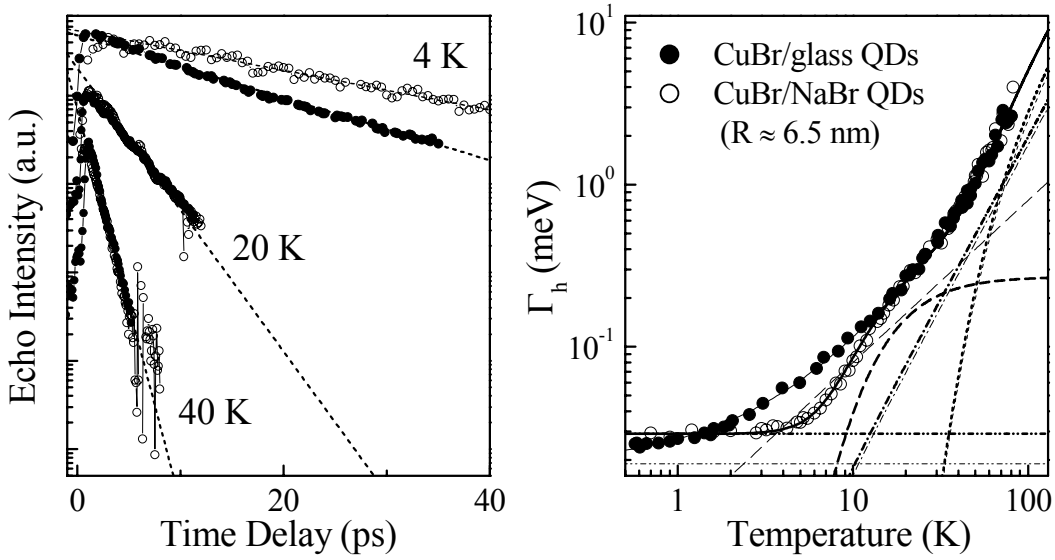


**Figure 1**. (a) Time-trace signals of HAPE for CuBr/NaBr QDs (open circles) and CuBr/glass QDs (closed circles) at 4 K, 20 K and 40 K. Dotted lines are the single exponential fits to the data. (b) The temperature dependent homogeneous linewidth of $Z_{1,2}$ exciton in these QDs. Open (closed) circles show the homogeneous width for CuBr/NaBr (CuBr/glass) QDs. Thick (thin) solid lines are fits to the data with following components: TLS (dashed line), confined acoustic phonons (dash-dotted line), and LO phonon scattering (dotted line). Dash-dot-dot lines show the temperature-independent width.

for CuBr/glass QDs. The equations (1) and (2) are the same except the second term which originates from the matrix-dependent TLS contribution. The first term, $\Gamma_{h0}$, is the temperature-independent width. The third term corresponds to the two-phonon Raman process of confined acoustic phonons, whose energies (0.96 and 0.86 meV, respectively) were determined by PSHB. The fourth term is the contribution of one LO-phonon scattering, which is important above 40 K. The LO-phonon energy of bulk CuBr, $\hbar\Omega_{LO} \approx$ 21.0 meV, was used. These results are similar to the previous report of CuCl QDs [4] except the LO-phonon contribution.

Now, let us discuss the dephasing mechanism of the exciton in CuBr/NaBr and CuBr/glass QDs below 15 K. Carrier trapping at the surface of dots produces more than one ground states of the total system consisting of QDs and host materials [11]. Many ground-state configurations can be simplified into the double welled potential representation. This is equivalent to the two-level system (TLS) model [13] that has successfully accounted for many anomalous low-temperature properties of glasses. When the energy of TLS is represented by the characteristic energy δ, the homogeneous broadening is in proportion to $\cosh^{-2}(\delta/2k_BT)$, while $T$-linear dependence is derived for widely distributed TLS energy [4]. In our case, δ = 2.8 meV is obtained for CuBr/NaBr QDs as a fitting parameter. The contribution of TLS and confined acoustic phonon was also observed for CdSe/glass QDs.

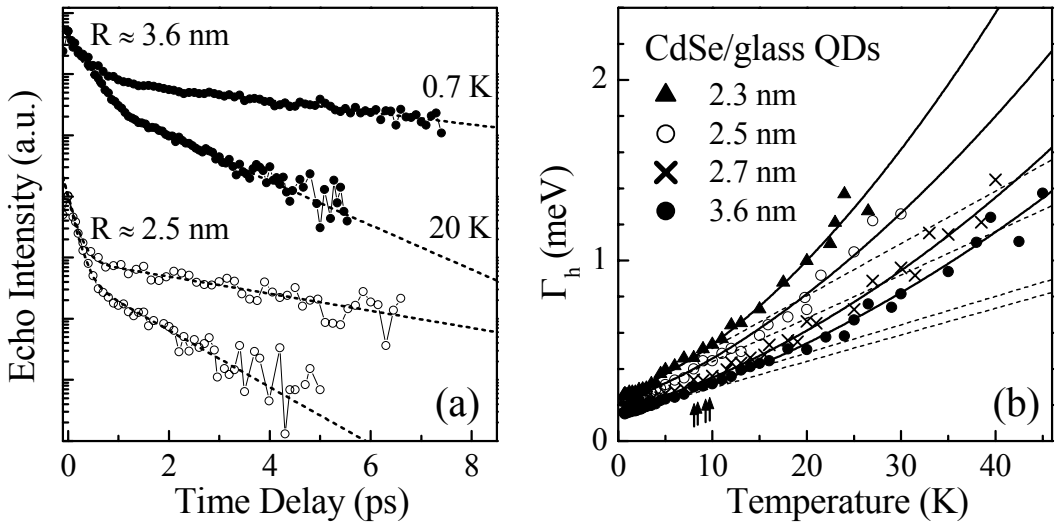


**Figure 2**. (a) Typical HAPE signals for CdSe/glass QDs at 0.7 K and 20 K. Closed (open) circles correspond to QDs with 3.6 nm (2.5 nm) radius. Dotted lines show the double exponential fit to the data. (b) Homogeneous linewidths as a function of temperature for CdSe/glass QDs ranging from 2.3 to 3.6 nm radius. Solid curves show the least squares fits considering the contributions of TLS (dotted lines) and confined acoustic phonons. For the vertical arrows, see the text.

Figure 2(a) shows the HAPE signals for CdSe/glass QDs with the radii of 2.5 and 3.6 nm. The Fourier-cosine transformation of the two-exponential decay in the HAPE signal gives a narrow Lorentzian line superimposed on a broader one. The faster decay component is ascribed to the confined acoustic phonon sideband whose temperature dependence is consistent with that of the Debye-Waller factor [14]. From here on, the temperature dependence of the zero-phonon linewidth obtained from the slower decay component is discussed. Between 0.7 and 10 K, homogeneous widths show almost linear dependence on temperature. However, they deviate from the *T*-linear dependence above 10 K as indicated by arrows in Fig. 2(b) and show quadratic increase up to 45 K. The overall data are fitted by the expression,

$$\Gamma_{\mathrm{h}}(T) = \Gamma_{\mathrm{h0}} + AT + B\sinh^{-2}\left(\frac{\hbar\omega}{2k_{\mathrm{B}}T}\right). \tag{3}$$

Equation (3) shows the dominant contribution of exciton-TLS and exciton-confined-phonon interaction similar to CuBr/glass QDs system. The temperature, where the hyperbolic-sine-term becomes effective, increases with decrease of dot size (arrows in Fig. 2(b)). The confined phonon energies were experimentally observed by Raman scattering, which agree well with our fitting values, $\hbar\omega$. According to the hole-burning measurement for interband [15] and intraband [16] excitation, the coupling to the LO phonon is little below 50 K. Thus it is naturally understood that the remarkable broadening above 10 K comes from the two-phonon Raman process of the confined acoustic phonon, as in polymer system [6].

The size dependence of the homogeneous widths extrapolated to 0 K for CuBr and CdSe QDs are displayed in Fig. 3. The experimental data are characterized well by a

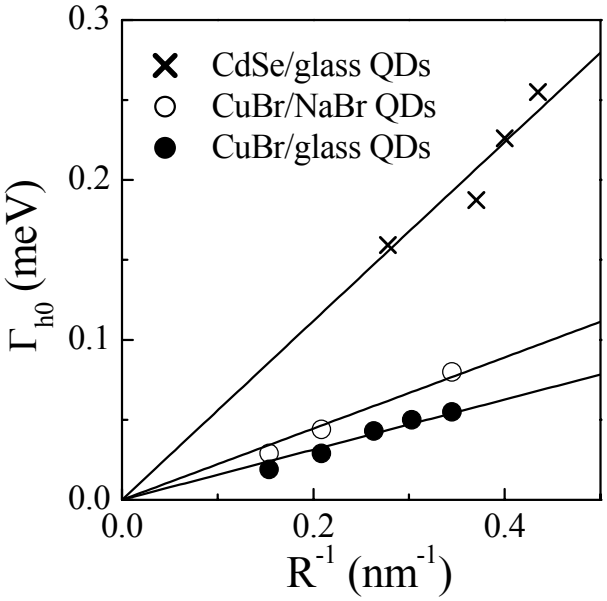


**Figure 3**. The size dependence of the homogeneous linewidth extrapolated to 0 K, $\Gamma_{h0}$. Closed circles, open circles and crosses correspond to CuBr/glass, CuBr/NaBr and CdSe/glass QDs, respectively. The solid lines indicate the fits by linear least squares method.

linear inverse size dependence, i.e., $\Gamma_{h0} \propto R^{-1}$. The temperature-independent width $\Gamma_{h0}$ is broader than the inverse of nonradiative lifetime, e.g., 100 ps for 3.6 nm CdSe/glass QDs, measured by luminescence decay and pump-and-probe decay as the fast-decay component [14, 17]. This indicates that the residual broadening mechanism at low temperature is not dominated by the lifetime broadening but by the surface-related dephasing process. If the carrier trapping driven by wave function overlap at the surface of QDs mainly causes the excitonic dephasing at low temperature, the trapping rate should vary as the surface-to-volume ratio, $R^{-1}$ [18]. For all the samples, not only $\Gamma_{h0}$ but also the coefficients of the TLS term, $A$, were found to increase linearly depending on $R^{-1}$. This suggests that the surface-driven dephasing is the dominant mechanism for both CuBr and CdSe QDs embedded in matrices at low temperature.

## 4. Conclusion

Using heterodyne-detected accumulated photon echo, we have systematically investigated the temperature broadening process of quantum dots ranging from weak confinement regime to strong confinement regime. The anomalous temperature dependence at very low temperature can be explained well in terms of a universal model based on the residual dephasing mechanisms: two-phonon Raman process of confined acoustic phonons and two-small-energy-excitation process in two-level-system.

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