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Carrier relaxation and quantum decoherence of excited states in self-assembled quantum dots

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We report systematic measurements of photoluminescence excitation spectra and dephasing times (T_2's) on various excited states of hundreds of individual quantum dots (QDs). From the variation of T_2's with the energy separation between excited states and the ground state (E_{rel}), we identified two distinct regions of E_{rel} where LO phonon emission and hole relaxation via LA phonon emission play as dominat dephasing mechanisms. We also found a clear evidence of significantly slow energy relaxation in the E_{rel} range where these phonon emission processes are suppressed due to the reduction of interaction phase space.

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In recent years, semiconductor quantum dots (QDs) become one of the focal points in condensed-matter physics. They are the key materials for next generation electronic and optoelectronic devices. These nanoscale quantum objects also provide an excellent ground to explore many fundamental physical phenomena such as quantum coherence/decoherence,[1–4] which is also critical for quantum information processing. Among various synthesis techniques, strain-driven growth of self-assembled QDs (SAQDs) is particularly promising for technological applications. However, despite intensive investigations, the understanding of the electronic structures and various electronic processes is still lacking. In particular, the mechanisms for inter-level carrier relaxation, which controls the quantum decoherence at low temperature,[1–4] have been very controversial. Due to a stronger confinement effect in SAQDs, there is a significant reduction in available phase space for the excited states to relax through phonon emission (the so-called phonon bottleneck).[5] Without alternative mechanisms, excited states of such systems should exhibit very long quantum dephasing times (T_2's). While earlier studies[6,7] suggested evidence for this phonon bottleneck effect, many recent studies[8–11] revealed evidence against this prediction and several alternative energy relaxation mechanisms have been proposed.[12–15]

In order to provide better understanding of electronic structures, carrier relaxation, and quantum dephasing phenomena in SAQD, we performed PLE spectroscopy and dephasing time measurements on a large number of individual SAQDs. The large statistical basis allows us to correlate the T_2's of excited states with their energy separations relative to the ground state [relaxation energy (E_{rel})]. This correlation together with the PLE spectra leads us to identify important dephasing mechanisms, each of which plays the dominant role in a different regime of E_{rel}. Furthermore, we have identified a regime where extra long dephasing time (40–90 ps) can exist, showing a clear evidence of suppressed phonon emission processes due to strong reduction in available phase space.

Our sample is a 6-ML-thick $In_{0.5}Ga_{0.5}As$ SAQDs grown by molecular-beam epitaxy. The sample was grown on (001) surface of an undoped GaAs substrate. The QDs and wetting layer were buried in the middle of 600-ML-thick GaAs matrix. The matrix was surrounded by two 300-ML-thick $Al_xGa_{1-x}As$ barrier layers on either side. Details of the sample growth processes were described elsewhere.[16] Cross-sectional scanning tunneling microscopy studies revealed that the QDs have average height, lateral dimensions and dot to dot separation of 4.5, 20–40, and 100 nm, respectively. It also showed nonuniform In distribution similar to the one discussed in Ref. 17. Photoluminescence (PL) emission spectra of our QD sample extend from 1.378 to 1.180 eV. Wetting layer absorption is around 1.433 eV. We used a mode locked Ti:sapphire laser with ~6 ps pulsewidth ($\Delta E = 300\,\mu$eV) as our excitation source and conventional μPL set up similar to those described in Ref. 18 for PL collection. The resolution of the PL emission spectra is ~100 μeV. Except for the temperature-dependent measurements, all measurements were done at 4 K.

We performed resonant excitation and collection of the PL signal on the cleaved edge of the sample. By using the PL imaging scheme described in Ref. 19, we can isolate and probe individual quantum dots with high spatial and spectral resolutions. Ground-state emissions of hundreds of individual QDs with one of their excited states in resonance with the excitation laser energy can be detected in parallel. We collected such a resonantly excited PL image at each excitation laser energy (E_{exe}) as we scanned E_{exe}. PLE spectra of individual QDs are extracted from the resulting stack of PL images.

Since the resonantly excited PL images recorded the ground-state PL emissions of selectively excited QDs, the distribution of the recorded PL peaks as a function of their relaxation energies will have a general profile similar to the PL spectra obtained from selective excitation of a large QD ensemble reported in Refs. 8 and 10. Figure 1(a) shows the distribution of 500 peaks with their emission energies ranging from 1275 to 1350 meV. The most distinct feature common to both our histogram and those selectively excited PL spectra is the peak appearing in the range of the LO phonon energies (E_{LO}'s) of $In_xGa_{1-x}As$QDs and GaAs matrix (30–37 meV). This prominent LO phonon related feature has

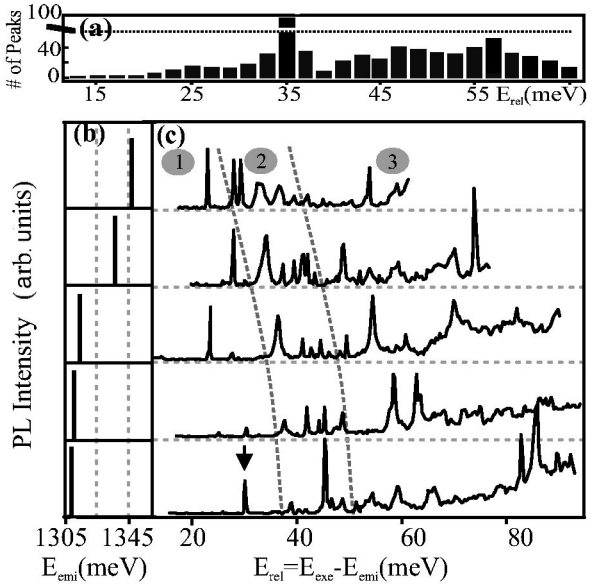

FIG. 1. (a) Total number of PL peaks in 2-meV energy bin vs E_{rel}. (b) Vertical lines show the position of PL peaks where the PLE spectra on (c) are collected. (c) Typical PLE spectra of five individual SAQDs.

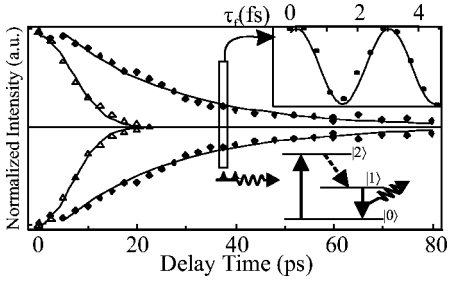

FIG. 2. Wave-function autocorrelation of an excited state marked by the arrow in 1(c). Top inset: expanded view at $\tau_c = 37.5$ ps showing the coherent oscillation in the PL intensity. Bottom inset: Interaction of ps pulse pair with three-level system.

been discussed much in the literatures, however, with conflicting interpretations regarding its origin. One interpretation prefers the phonon-assisted absorption to the ground state[20] while the other favors the efficient energy relaxation of carriers created in the excited states with $E_{\mathrm{rel}} \approx E_{\mathrm{LO}}$.[10] As we discuss below, our systematic studies clearly show that the latter is the dominant process.

Figure 1(c) displays five typical PLE spectra of individual SAQDs located at different spatial and spectral locations as examples. Their ground-state emission energies (E_{emi}'s) are given in Fig. 1(b). These spectra reveal several interesting features obscured in other conventional PLE studies[8–10] due to ensemble averaging effects. One can roughly divide these spectra into three regions as indicated by the curved-dashed lines in Fig. 1(c).

We first discuss region 2 where most of the QDs show a group of four to six absorption peaks with close (<5 meV) energy separations. The intensities and linewidths of these peaks increase dramatically when their E_{rel}'s fall in the E_{LO} range. These peaks have been interpreted as the results of resonant Raman scattering (RRS) with localized LO phonons.[15] However, they do not appear in the fixed energy range around E_{LO} as required by this process. Instead, their energy locations vary systematically toward lower E_{rel} as their E_{emi}'s increase. This variation is consistent with the behavior of excited states of QDs when the quantum confinement is reduced due to the increase of QDs Ga concentration. Therefore, these peaks are indeed originated from the different excited states of QDs. With this interpretation, dramatic increase of strength and linewidths mentioned earlier can be explained as the results of efficient relaxation of carriers created in the excited states with $E_{\mathrm{rel}} \approx E_{\mathrm{LO}}$. Since the E_{rel} of this peak group falls around E_{LO} the probability of finding one or more peaks with open LO phonon relaxation channels becomes very high for most of the QDs and finally resulting in the accumulation of peaks around $E_{\mathrm{rel}} \approx E_{\mathrm{LO}}$.[21]

According to the recent theoretical calculations,[22,23] QDs can have 2–3 bound states and several closely spaced quasibound states in the conduction band and many (>6) hole bound states with close energy-level separations of a few meV in the valence band. Energy-level separations of the peak group in region 2 match qualitatively[24] with level separations among the hole bound states. Therefore it is reasonable to interpret these peaks as the results of absorption, which creates an electron-hole pair with the hole in one of those excited hole states.

In region 3, nearly all the QDs show continuumlike absorption tails extending from the lower energy edge of the wetting layer (1.429 eV) down to the edge of region 2. The local variation of the wetting layer may be responsible for this absorption band. Many sharp features are riding on the tail of this absorption band and they may originate from quasibound states of the QDs. Features similar to those appearing in regions 2 and 3 have also been observed in other single dot PLE spectra.[15,20] In addition to these features, we observed that some of the QDs have 1 to 2 sharp absorption peaks in low E_{rel} region marked as region 1.

Since E_{rel}'s and inter-peak energy separations of these absorption peaks differ significantly from one region to another, dephasing mechanisms of the excited states are also expected to be different. To determine these mechanisms, we measured the T_2's of excited states of hundreds of SAQDs by adapting the wave-packet interferometry technique used in Ref. 1 to our PL imaging scheme. In this experiment, the phase locked picosecond pulse pair is generated by a standard Michelson interferometer. Time delay between two pulses is controlled by the coarse and fine control of $\tau_c > 1$ ps and $\tau_f \sim 0.3$ fs steps, respectively. We took a resonantly excited spectral image at each time delay step as we scanned the time delay without changing the excitation energy. The wave-packet autocorrelation signals of various excited states of hundreds of QDs were extracted later from the stacks of PL images.

Figure 2 shows oscillation amplitudes of the PL signal (solid diamonds) emitted from the ground state (exciton $|1\rangle$ state shown in lower inset) of a particular QD, together with the cross correlation of the laser pulse (open triangles) as a function of coarse pulse delay. This autocorrelation trace was measured at the PLE peak marked by a vertical arrow on one

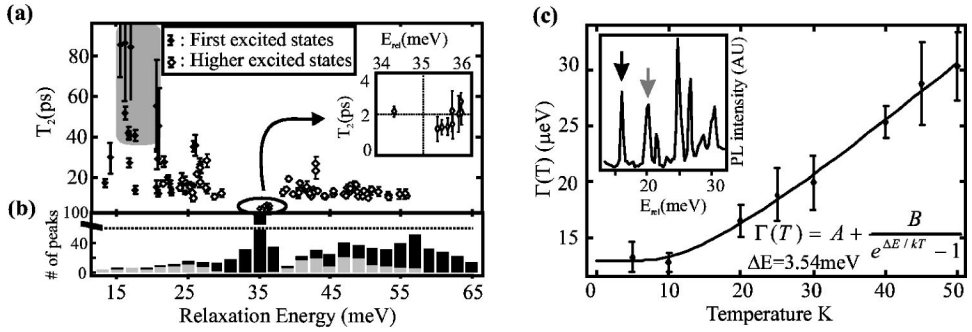

FIG. 3. (a) Scatter plot of T_2's of 110 excited states of different QDs vs their E_{rel}. Inset: Magnified view around $E_{\mathrm{rel}}=35$ meV. (b) Histogram of 500 PLE peaks; black columns, total number of peaks observed in a 2 meV energy bin [same as 1(a)]; gray columns, number of peaks with $T_2>7$ ps in the same energy bin. (c) Temperature dependence of decay rate measured from the PLE peak marked by the black arrow in PLE spectra shown in the inset.

of the spectra shown in Fig. 1(c). The inset displays an expanded view at $\tau_c=37.5$ ps showing the coherent oscillation in the PL intensity. The decay of the oscillation amplitudes reflects the loss of coherence in the excited states during the time delay. This technique allows us to measure the T_2's longer than 7 ps. Lorentzian linewidth of a PLE peak is used to measure T_2's shorter than 3 ps.

We performed similar measurements on various excited states of hundreds of other QDs with their E_{emi} ranging from 1275 to 1350 meV. The strong dependence of T_2's upon nature of the excited states and their E_{rel}'s is shown in Fig. 3(a). Histograms of Fig. 3(b) indicate the manifestation of fast ($T_2<7$ ps) and slow ($T_2>7$ ps) dephasing processes at distinct E_{rel} regimes. Further detailed analysis reveal information on inter-level carrier relaxation processes which control the dephasing at low temperatures.[1–4]

The T_2's of higher excited states [their absorption peaks appear in region 2 of Fig. 1(c)] are plotted as ''open diamonds'' in Fig. 3(a). This plot shows the following correlations between T_2 and E_{rel}.

(1) Almost all the excited states with $E_{\mathrm{rel}}\approx E_{\mathrm{LO}}$ have T_2's shorter than 7 ps [See gray columns in Fig. 3(b)]. Lorentzian linewidths of some of their PLE peaks give T_2 as short as 1 ps [See inset of Fig. 3(a)].

(2) The other excited states in this region with $E_{\mathrm{rel}}\neq E_{\mathrm{LO}}$ have T_2's of 7–30 ps. It is very clear that energy relaxation via the LO phonon emission is responsible for very short T_2's of the excited states with $E_{\mathrm{rel}}\approx E_{\mathrm{LO}}$. On the other hand, relatively longer T_2's of the other states with $E_{\mathrm{rel}}\neq E_{\mathrm{LO}}$ point to the existence of another dephasing mechanism. Toda *et al.*[25] have suggested that the RRS mechanism is responsible for similar value of T_2's. However, we have shown that the PLE peaks are the results of absorption, which creates an electron-hole pair with the hole in one of the valence-band states. We therefore proposed an alternative dephasing mechanism based on inter-level carrier relaxation.

Due to the close energy-level separations (<5 meV) between hole bound states, holes trapped in higher excited states can relax to the next lower level by emitting LA phonons. This hole inter-level population relaxation rate will be much faster than that of electrons if electronic inter-level separations do not match E_{LO}. Therefore, the T_2's of the higher excited states are mainly determined by this inter-level population relaxation of holes. LA phonon emission processes are also responsible for the T_2's of 30–40 ps in QDs with weak confinement potential.[1–4] Comparable magnitudes of these T_2 values and our results (7–30 ps) support this interpretation. For an exciton state with $40<E_{\mathrm{rel}}<55$ meV, it will relax repeatedly via this LA phonon emission process until it reaches a state with E_{rel} around E_{LO} and then decays rapidly to the ground state via the LO phonon emission.

On the other hand, for a first excited state with $E_{\mathrm{rel}}<E_{\mathrm{LO}}$, it has to relax through either emission of a single high-energy LA phonon or multiple phonons if no other alternative relaxation mechanisms are available. According to the theoretical predictions,[5,13] these interactions should be strongly suppressed for a first excited state with its absorption peak in region 1 of Fig. 1(c). Indeed, the observation of extra long (40–90 ps) T_2's of some of the first excited states with $15<E_{\mathrm{rel}}<20$ meV [see gray region of Fig. 3(a)] clearly indicates slow energy relaxation in this E_{rel} regime. However, this time scale is still shorter than the radiative relaxation time and quenching of ground-state PL is not observed. In addition to these states, we also observed many other states with T_2's of 7–40 ps in this region [See Fig. 3(a)]. This large variation of T_2's indicates the existence of other relaxation mechanisms. For example, it has been theoretically predicted that, the presence of deep level traps could provide an efficient relaxation channel with its strength sensitively depending upon the spatial separation between the trap site and the QD.[12] This mechanism provides a possible explanation for the large variation of T_2's.

These ultralong T_2 states also show interesting temperature dependence behavior. Figure 3(c) displays the plot of decay rate ($\Gamma=\hbar/T_2$) vs temperature for one of the excited states, together with the PLE spectrum of the QD. The black arrow marked the peak where the measurements were made. The result can be fitted with the equation shown in Fig. 3(c), which describes the activation of exciton to a higher state by absorption of a LA phonon. The fitted value of $\Delta E=3.5$ meV is in reasonable agreement with the 4.07-meV energy-level separation to the next absorption peak marked by the gray arrow. This result indicates that phonon absorption process becomes a dominant dephasing mechanism at higher temperatures, a natural consequence since the relax-

ation channel via phonon emission to the ground state is restricted. Dephasing rates of other excited states also show similar temperature dependence.

Most of the excited states whose absorption peaks appear in region 3 of Fig. 1(c) have T_2's shorter than 7 ps. Since these absorption peaks are riding on the continuumlike absorption bands, there are more available final states for a carrier to relax its energy. Furthermore, in this energy range, the excited state wave functions of a QD can overlap energetically as well as spatially with those of the continuum states of other QDs and wetting layers. Therefore, a trapped exciton can also relax its energy via the release to other carriers in the continuum states. This Auger-like mechanism and availability of more final states may be responsible for the short T_2's of the excited states in this region.

In summary, our studies clarify the following major energy relaxation mechanisms and their active energy regimes: (1) Energy relaxation via the emission of LO phonons for very fast dephasing of the excited states with $E_{\mathrm{rel}} \approx E_{\mathrm{LO}}$. (2) Inter-level hole relaxation among closely spaced valence-band states via the emission of LA phonons for the higher excited states with $E_{\mathrm{rel}} \neq E_{\mathrm{LO}}$. (3) Relaxation via the continuum states for the excited states with high E_{rel}. Relative efficiencies of these three dephasing mechanisms are compared in the histograms shown in Fig. 3(b). The superior efficiency of the LO phonon emission mechanism is highlighted by the coincidence of the sharp peak of black columns and the valley of gray columns. Gradual decrease in height of gray columns with the increase of E_{rel} indicates the slow transition of the dephasing mechanism from the inter-level relaxation of holes to the relaxation via the continuum states. Furthermore, we also observed a significant slow down of energy relaxation in low E_{rel} range where the efficient phonon emissions are suppressed by the phonon bottleneck effect.

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