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 ($E_{\text{rel}}$), we identified two distinct regions of $E_{\text{rel}}$ where LO phonon emission and hole relaxation via LA phonon emission play as dominant dephasing mechanisms. We also found a clear evidence of significantly slow energy relaxation in the $E_{\text{rel}}$ 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, 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, 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). Without alternative mechanisms, excited states of such systems should exhibit very long quantum dephasing times ($T_2$'s). While earlier studies suggested evidence for this phonon bottleneck effect, many recent studies revealed evidence against this prediction and several alternative energy relaxation mechanisms have been proposed.

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 ($E_{\text{rel}}$)]. 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 $E_{\text{rel}}$. 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 a 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$_x$Ga$_{1-x}$As barrier layers on either side. Details of the sample growth processes were described elsewhere. 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 PL set up similar to those described in Ref. 18 for PL collection. The resolution of the PL emission spectra is ~100 $\mu$eV. 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 ($E_{\text{exc}}$) as we scanned $E_{\text{exc}}$. 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 of In$_{0.5}$Ga$_{0.5}$AsQDs and GaAs matrix (30–37 meV). This prominent LO phonon related feature has
been discussed much in the literatures, however, with conflicting interpretations regarding its origin. One interpretation prefers the phonon-assisted absorption to the ground state while the other favors the efficient energy relaxation of carriers created in the excited states with as required by this process. Instead, their 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. In addition to these features, we observed that some of the QDs have 1 to 2 sharp absorption peaks in low region marked as region 1.

Since 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 states of hundreds of QDs 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 the laser pulse as required by this process. Instead, their energy locations vary systematically toward lower as their 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 as. Since the of this peak group falls around the probability of finding one or more peaks with open phonon relaxation channels becomes very high for most of the QDs and finally resulting in the accumulation of peaks around .21

According to the recent theoretical calculations, 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 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. In addition to these features, we observed that some of the QDs have 1 to 2 sharp absorption peaks in low region marked as region 1.

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Figure 2 shows oscillation amplitudes of the PL signal (solid diamonds) emitted from the ground state (exciton 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...
of the spectra shown in Fig. 1(a). 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 of threshold 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\(_{\text{em}}\) ranging from 1275 to 1350 meV. The strong dependence of T\(_2\)’s upon nature of the excited states and their E\(_{\text{em}}\)’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\(_{\text{rel}}\) regimes. Further detailed analysis reveal information on inter-level carrier relaxation processes which control the dephasing at low temperatures.

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\(_{\text{rel}}\):

1. Almost all the excited states with E\(_{\text{rel}}\approx E_{\text{LO}}\) 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\(_{\text{rel}}\neq E_{\text{LO}}\) 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\(_{\text{rel}}\approx E_{\text{LO}}\). On the other hand, relatively longer T\(_2\)’s of the other states with E\(_{\text{rel}}\neq E_{\text{LO}}\) point to the existence of another dephasing mechanism. Toda et al. have suggested that the RRS mechanism is responsible for similar value of T\(_2\)’s.

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\(_{\text{LO}}\). 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. Comparable magnitudes of these T\(_2\) values and our results (7–30 ps) support this interpretation. For an exciton state with E\(_{\text{rel}}\approx E_{\text{LO}}\), it will relax repeatedly via this LA phonon emission process until it reaches a state with E\(_{\text{rel}}\approx E_{\text{LO}}\) and then decays rapidly to the ground state via the LO phonon emission.

On the other hand, for a first excited state with E\(_{\text{rel}}\approx E_{\text{LO}}\), 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, these interactions should be strongly enhanced 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\(_{\text{rel}}\)<20 meV [see gray region of Fig. 3(a)] clearly indicates slow energy relaxation in this E\(_{\text{rel}}\) 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. 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-
Inter-level hole relaxation among closely spaced valence-band states via the emission of LA phonons for the higher excited states with $E_{\text{rel}}<E_{\text{LO}}$. (3) Relaxation via the continuum states for the excited states with high $E_{\text{rel}}$. 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 $E_{\text{rel}}$ 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 $E_{\text{rel}}$ range where the efficient phonon emissions are suppressed by the phonon bottleneck effect.

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21. If there were no excited states with $E_{\text{rel}}<E_{\text{LO}}$ phonon assisted absorption peak could become observable at high excitation intensity as shown in Ref. 20.
24. Energy separations of higher excited hole states calculated in Ref. 22 range from 3–10 meV. Since our QDs are bigger and have less In concentration than those QDs considered in the calculations, it is reasonable to expect more energy levels with closer energy levels separation.