Efficient Carrier Relaxation Mechanism in InGaAs/GaAs Self-Assembled Quantum Dots Based on the Existence of Continuum States

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Comparison of near-field and far-field photoluminescence excitation (PLE) spectra gives new insight into the carrier relaxation process in InGaAs/GaAs self-assembled quantum dots. The near-field PLE spectra of single quantum dots clearly show 2D-like continuum states and a number of sharp lines, between a large zero-absorption region due to the quasi-0D density of states and the 2D wetting layer absorption edge. The results reveal an efficient intradot relaxation mechanism, proceeding as follows: The carriers can relax easily within continuum states, and make transitions to the excitonic ground state by resonant emission of localized phonons. [S0031-9007(99)09156-5]

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Carrier relaxation in defect-free self-assembled quantum dots (SAQDs) based on Stranski-Krastanow (SK) growth is an important subject of discussion not only because of device applications but for understanding the fundamental physics of zero-dimensional (0D) systems [1–5]. Because the confinement on length scales down to 10 nm in all three directions produces sublevel separations much larger than a few meV [6], therefore larger than the energy of LA phonons, it is predicted that a strongly reduced energy relaxation rate cannot be avoided, unless energy level separation equals the LO phonon energy, i.e., a "phonon bottleneck" is predicted [7]. Although relaxation through LO phonon scattering is expected to be very restricted due to the phonons' small energy dispersion, this phonon bottleneck effect seems unlikely to be occurring in SAQDs, since the optical measurements show intense photoluminescence (PL) from ground state recombination. Measurements on SAQDs have yielded characteristic features due to the discrete confined states with large energy separation. For example, with increasing the excitation power, PL spectra show additional peaks to high energy of the main peak, and these have been attributed to higher excited states [5]. On the other hand, the PL excitation (PLE) resonances do not directly reflect the higher excited states observed in PL but phonon features. This has been explained by the multiphonon filtering of inhomogeneously distributed excited states, that is, carrier relaxation being possible only when the energy separation of the confined states matches the energy of an integral number of phonons [2–4]. On the basis of this idea, interlevel relaxation is the dominant carrier relaxation mechanism in SAQDs. However, this mechanism is based on the density of states (DOS) expected in ideal 0D systems. In addition, the experimental results include inhomogeneous broadening due to measurements being of ensembles of dots.

The reduction of inhomogeneous broadening has been achieved by using spatially resolved spectroscopic techniques, such as diffraction limited confocal microscopy [8,9], micro-PL with a small mesa [1,10] and near-field scanning optical microscopy (NSOM) [11]. These microprobing methods allow for precise measurements of 0D systems. In this work, we investigate relaxation processes in InGaAs/GaAs SAQDs by using NSOM. Near-field PLE spectra strongly suggest the existence of continuum states in single dots. The density of states gradually increases from the end of a large zero-absorption region up to the wetting layer (WL) absorption edge. The data also suggest the breakdown of long-range order of the lattice, showing a number of spiky structures which can be explained as resonant Raman scattering associated with localized phonons. These results suggest that under nonresonant conditions the excited carriers can relax easily within continuum states and relax to the excitonic ground states by resonantly emitting phonons, thus efficient intradot carrier relaxation can occur.

Spatially resolved single dot spectroscopy was performed using NSOM at 8 K. In order to avoid carrier diffusion effects, both the excitation and collected luminescence light passed through the same fiber (i.e., illumination-collection mode NSOM) [11]. In the near-field measurements, the PL was excited by a Ti:sapphire laser at an excitation density of $\sim 10\ \mathrm{W/cm^2}$. The collected PL was dispersed by a 1 m long double monochromator with a charge-coupled device (CCD) detector. The InGaAs SAQD sample studied here was grown using the SK growth mode on a GaAs substrate as described in detail in Ref. [12]. In order to shift the PL peak to shorter wavelengths where a CCD camera with high quantum efficiency is available, the sample was annealed at 700 °C after the growth of a 140 nm thick GaAs cap. This changes only the luminescence energy and does not affect the intrinsic relaxation mechanisms of carriers in SAQDs [13]. The SAQDs have base length ~ 20 nm, height ~ 7 nm, and density $> 10^{10}/\mathrm{cm^2}$, from an atomic force microscopy study of an uncapped reference sample. In the magnetic field a single SAQD PL peak showed a diamagnetic shift of $\sim 4\mu\mathrm{eV/T^2}$, corresponding to the carrier wave function having lateral expansion ~ 5 nm. This result indicates that carriers are strongly confined in the lateral direction in excitonic ground states $|\mathrm{ex}\rangle$ of the SAQDs.

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The inset of Fig. 1(a) shows far-field ensemble PL spectra in which emission is collected from thousands of SAQDs. PL at low excitation power has a Gaussian profile with a peak energy of ~1.26 eV and linewidth of ~80 meV (solid line in the inset). The peak at 1.41 eV is attributed to the WL. Upon increasing the excitation power, two additional peaks appear to higher energy (gray line). These may be attributed to the higher excited states of the SAQDs [5]. The observed energy separations of the higher excited states are estimated by Gaussian peak fitting to be about 50 meV. The main part of Fig. 1(a) shows an ensemble PLE spectrum. In this figure, the bottom horizontal axis represents relaxation energy (E_{rel}), that is, the difference between the excitation and detection energies ($E_{\mathrm{ex}} - E_{\mathrm{det}}$). As has been reported by many other groups [2–4], we also have observed PLE peaks whose energies are independent of the detection energy.

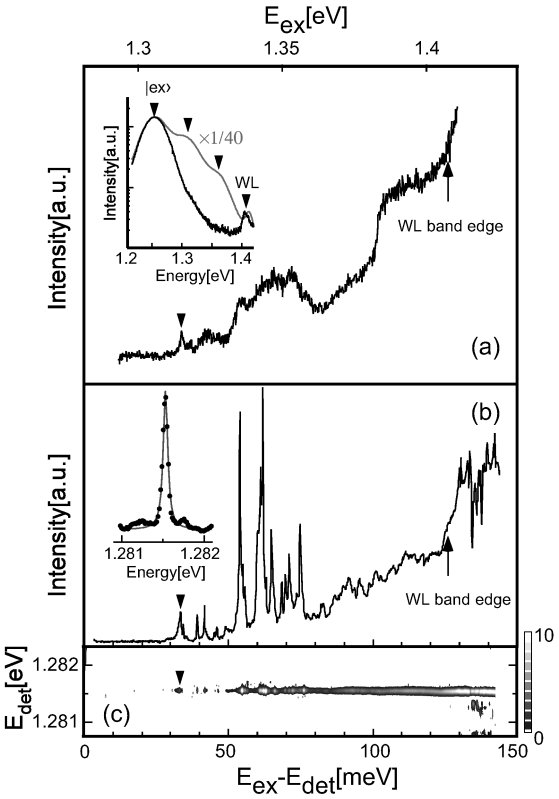

FIG. 1. (a) A typical far-field PLE spectrum from ensemble PL as shown in the inset. The detection energy $E_{\mathrm{det}} = 1.282$ eV. Inset: The solid and gray lines show ensemble PL at low power excitation $P_{\mathrm{ex}} = 5$ W/cm^2 and high power excitation $P_{\mathrm{ex}} = 70$ W/cm^2, respectively, with excitation energy $E_{\mathrm{ex}} = 1.959$ eV. The arrows on the high power spectrum indicate peaks at 1.254, 1.307, 1.358, and 1.41 eV determined by Gaussian fitting. (b) Near-field PLE spectrum of a PL feature as shown in the inset. The wavelength was scanned in 0.1 nm steps and the integration time at each wavelength was 30 sec. The solid line in the inset indicates a Lorenzian fit to the PL data with $E_{\mathrm{ex}} = 1.43$ eV. (c) Gray-scale PL intensities corresponding to the data in (b) are plotted as a function of detection and excitation energy. The 1LO phonon structure is seen at $E_{\mathrm{rel}} = 36$ meV as indicated by arrows.

From the far-field PL spectra, the WL absorption edge is estimated to be at $E_{\mathrm{ex}} = 1.41$ eV corresponding to $E_{\mathrm{rel}} = 120$ meV in Figs. 1(a), 1(b), and 1(c).

The inset of Fig. 1(b) shows a spatially and spectrally resolved nonresonant PL peak from a single SAQD at $E_{\mathrm{ex}} = \sim 1.43$ eV. The luminescence has a Lorenzian line shape with a linewidth of ~70 μeV. The corresponding PLE spectrum is shown in Fig. 1(b). There is virtually no signal below $E_{\mathrm{rel}} = 30$ meV [14]. This large zero-absorption region and the narrowness of the PL lines are attributed to the DOS expected in quasi-0D structures. A number of partly overlapping sharp resonant lines ($\Delta\nu < 1$ meV) are found above $E_{\mathrm{rel}} = 30$ meV, and will be explained in detail later. Except for these resonant peaks, we can find the continuum background. The background starting around $E_{\mathrm{rel}} = 50$ meV gradually increases up to the WL absorption band edge. Figure 1(c) shows a gray-scale plot of the PL intensity as a function of relaxation and detection energy. Horizontal and vertical cross sections of these data correspond to the PLE and PL signals, respectively. This plot indicates the absence of background emission of other wavelengths, and thus that the observed continuum is an *intrinsic* feature of the single SAQD. A similar background signal in this relaxation energy region is seen in almost all near-field PLE spectra from different single SAQDs; see Fig. 2. The observed continuum signal is also seen in InAs SAQDs grown by molecular beam epitaxy and consistent with polarized spectra, in which the suppression of spin relaxation was found to be increased at lower relaxation energies [11]. At energies where the density of continuum states is smaller, spin-flip scattering is more suppressed. It is important to note that such a continuum background has also been observed in many other reports on SAQDs [15], not only in far-field PLE but also in infrared absorption measurements [16]. However, little consideration has been given to this point because the results were from measurements of ensembles of dots, in which inhomogeneity obscures the contribution of the continuum background to intrinsic SAQD properties. Although it is surprising for us that the continuum of states penetrates as deeply into the 0D region as is observed in the PLE spectra, it provides for efficient relaxation in the SAQDs, as is discussed in detail below. In the present data, it is difficult to identify the origin of this continuum. Because SAQDs are connected with WL, there may exist a very gradual crossover from 0D to 2D nature in the DOS [17].

Around $E_{\mathrm{rel}} = 65$ meV, the far-field PLE spectrum exhibits a broad peak (~20 meV), the so-called 2LO resonance peak. In this energy range the near-field PLE spectrum shows various sharp resonance peaks as shown in Fig. 1(b). Figure 2 shows typical near-field PLE spectra from three different single PL peaks, whose energies correspond to the lower, center, and higher energy sides of the ensemble PL peak. Although in each spectrum the sharp resonance lines occur within the relaxation energy range of the broad resonance observed in far-field PLE, their number and energies are very different in each spectrum. Let

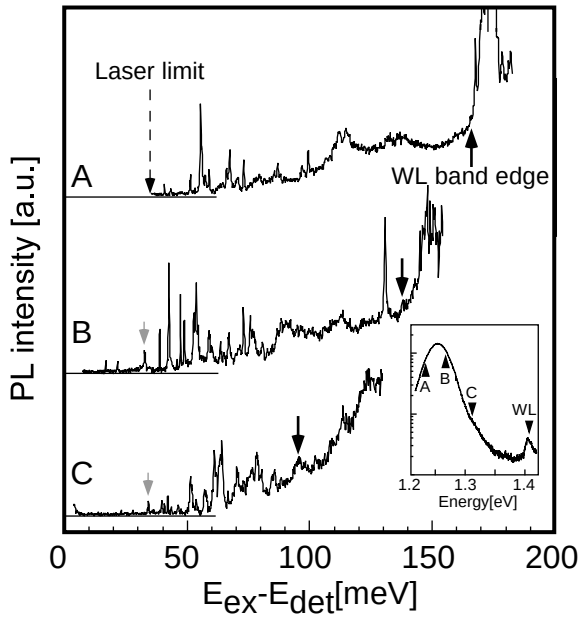

FIG. 2. Near-field PLE spectra for three different single SAQD features. Spectra *A*, *B*, and *C* correspond to detection at 1.2347, 1.2686, and 1.3132 eV, respectively (see inset). The solid arrows indicate the WL absorption edge, and thin gray arrows the 1LO feature. PL intensities are plotted as a function of relaxation energy ($E_{\mathrm{ex}} - E_{\mathrm{det}}$). In spectrum *A*, relaxation energies below $E_{\mathrm{rel}} = 40$ meV are outside the range scanned by the Ti:sapphire laser.

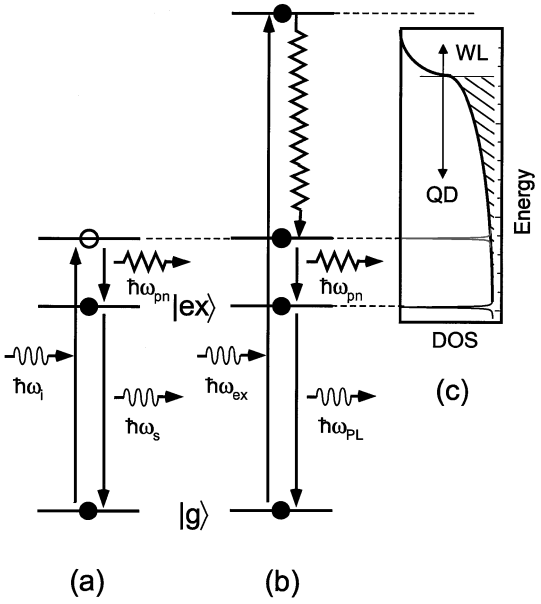

FIG. 3. (a) Schematic diagram of a resonant Raman process showing incident (ω_i) and emitted photons (ω_s) and an optical phonon (ω_{pn}). (b) Carrier relaxation mechanism under nonresonant conditions showing excited (ω_{ex}) and emitted photons (ω_{PL}). (c) Schematic diagram of the DOS with a phonon feature (gray line). $|g\rangle$ and $|\mathrm{ex}\rangle$ label the ground state and excitonic ground state, respectively.

us remember the process proposed to account for the PLE resonances [2–4]. It was predicted that only dots whose $|\mathrm{ex}\rangle$ is at the detection energy and whose excited states are separated from $|\mathrm{ex}\rangle$ by the energy of an integral number of LO phonons can relax to the $|\mathrm{ex}\rangle$ and contribute to the PLE signal. The resonant peaks observed in far-field PLE spectra were thus attributed to the multiphonon filtering of inhomogeneously distributed excited states. However, interpreting our data in terms of these phonon selected excited states, we observe *too many* excited states whose energy is separated by a few meV to be consistent with the confined states predicted for SAQDs [6]. Therefore, this interlevel relaxation mechanism cannot explain the observed single dot PLE spectra.

The sharp resonance lines observed in each near-field PLE spectra occur within almost the same relaxation energy region, showing an independence of detection energy as has been observed in far-field PLE spectra. Note that various studies of far-field PLE have identified PLE resonances as multiples of the LO phonon energies [2–4]. From the relaxation energy dependence, we conclude that the near-field PLE resonant peaks are predominantly due to resonant Raman scattering by integral numbers of phonons. Figure 3(a) is a schematic diagram of a resonant Raman process observed in PLE spectroscopy. When the relaxation energy of the incident light matches the energy of an integral number of available phonons, Raman scattering fulfils the resonance condition. Similar Raman features were observed for excitons localized in dotlike states in a quantum well (QW) [10]. Note that the relative intensity of the 2LO scattering is extraordinarily strong in comparison with 1LO. Generally, the intensity of Raman scattering changes drastically depending on the excitation conditions. Therefore, it seems likely that the phonon lines observed around 65 meV in our PLE spectra involve enhanced scattering cross sections. The enhancement may be due to the existence of the continuum states. Another possibility is that electronic excited states, which exist in the corresponding energy region, may provide the resonances. Further investigations are needed to clarify the resonance conditions. However, we stress that the observed 2LO lines involve strongly enhanced scattering cross sections due to strong phonon-electron interactions.

The sharpness of the resonance lines in near-field PLE allows us to clearly see the phonon spectra of individual SAQDs. The differences in the number and peak positions of the phonon lines in each spectrum may be attributed to inhomogeneity in the distribution of SAQDs. The broadness of far-field PLE peaks suggests that, even among dots with the same $|\mathrm{ex}\rangle$ energy, their phonon energies are scattered. It might be reasonable to attribute the observed Raman structures to *localized* phonons since these will be enhanced and strongly scattered in the individual SAQDs due to the lack of long-range order of the lattice structure. Inhomogeneity in the strain distribution and the In composition breaks the long-range order of the lattice and activates a number of phonons, even in symmetries theoretically forbidden in SAQDs. Therefore, the observed

spiky lines are predominantly attributed to integral numbers of localized phonons.

In the far-field PLE spectrum, in contrast to the broad peak at 65 meV, a narrower resonant peak is seen at a relaxation energy of around 36 meV, the so-called 1LO resonance peak. Although far-field PLE involves large ensembles of dots, this peak exhibits as narrow a linewidth as when it is observed in near-field PLE. As shown in Fig. 2, almost all near-field PL features show the resonance at the same relaxation energy. It seems to be reasonable to attribute this feature to a resonant Raman process in which a bulk GaAs phonon is emitted. Note that such a resonance is also observed in InAs SAQD samples, where the GaAs phonon exists in the cap layer [11]. Because the SAQDs confine electrons on length scales down to several tens of nanometers, the electron wave function penetrates the cap layer and couples with the phonon mode in the barrier.

Based on the observed continuum states and resonant Raman peaks, we can now propose a mechanism for the efficient carrier relaxation observed in SAQDs. Figure 3(b) is a schematic diagram of the relaxation process of excited carriers under nonresonant conditions. For excitation energies above the WL absorption edge, the carriers can relax very quickly through the continuum states by LA phonon scattering, and from appropriate energies reach $|\mathrm{ex}\rangle$ by resonantly emitting an integral number of localized phonons. As discussed above, because of the strong phonon-electron interactions, a number of localized phonons are available for the relaxation necessary before ground state emission. This efficient process explains the intense PL observed in SAQDs. Our results thus indicate that intradot relaxation of carriers through the continuum states is more important than interlevel relaxation through discrete confined states in SAQDs. The lack of long-range order results in the existence of a number of localized phonons, providing efficient relaxation channels to $|\mathrm{ex}\rangle$. So far, only the absence of a phonon bottleneck has been focused on in discussions of efficient relaxation in SAQDs. However, based on the mechanism discussed above, efficient relaxation of carriers can be achieved in SAQDs even in the presence of phonon selection.

Finally, we comment on the contribution of excited states to the PLE spectrum. Ensemble PL spectra [inset of Fig. 1(a)] suggest that there may exist two other excited states whose energies are separated by 50 and 100 meV from $|\mathrm{ex}\rangle$. Because there exists a continuum of states, the excited states should be visible in PLE. The near-field PLE spectra show various resonant peaks in the corresponding energy regions, so we cannot assign specific peaks to these excited states in the present data. Furthermore, unlike in higher dimensional structures, the PLE spectra do not directly reflect the absorption but also the ease of relaxation to $|\mathrm{ex}\rangle$, so absorption into excited states may be obscured by the continuum states from which efficient relaxation can occur. We stress again that the interlevel relaxation is not as important as relaxation through the continuum states.

In summary, we have performed PLE measurements on both single and ensembles of InGaAs/GaAs SAQDs. The near-field PLE spectra show a large no-signal region due to the dots' 0D character and gradually increasing continuum absorption connected with the 2D-like WL, indicating a crossover between 0D and 2D character. The spectra also show a number of sharp resonances, which can be attributed to resonant Raman processes associated with localized phonons due to the loss of long-range order of the lattice in SAQDs. These results reveal the efficient intradot relaxation process in SAQDs, in which the carriers can relax quickly within continuum states and make transitions to the excitonic ground state by phonon scattering, and thus account for the intense PL peak.

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[14] Some PLE spectra show very small spiky signals [e.g., Figs. 2(b) and 2(c)]. These may be (non-GaAs) 1LO features.
[15] For example, R. Heits *et al.* employed a gradually increasing background to evaluate the phonon energies by Gaussian fitting in Ref. [2]. Similar backgrounds are also seen in Ref. [3]. S. Sauvage *et al.* observed broad absorption in infrared absorption measurements [16].
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[17] The DOS of a real SAQD should be very different from that of the ideal one due to the WL, strain, etc. The potential of the WL outside the dots may be different from that of the WL within dots. Furthermore, the potentials of dots and WL will be affected by the strain in each other. Further investigations are needed to identify the origin of the continuum, however, we suggest that the relationship between the dots and the WL is a major consideration.