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Energy dependent carrier relaxation in self-assembled InAs/GaAs quantum dots

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We performed the selective excitation photoluminescence (SEPL) spectroscopy studies on InAs/GaAs self-assembled quantum dots (QDs). Under different excitation energies, different groups of QDs are selected and emit light. The excited carriers relax to the ground state through different mechanisms when excited at different energies. Three distinct regions with different mechanisms in carrier excitation and relaxation are identified in the emission spectra. These three regions can be categorized, from high energy to low energy, as continuum absorption, electronic state excitation, and multi-phonon resonance. The QDs' special joint density-of-state tail extending from the wetting layer peak facilitates the carrier relaxation and was suggested to explain these spectral results.

1 Introduction Self-assembled quantum dots (QDs) grown on lattice mismatched material systems using Stranski-Krastanov growth mode have found applications in many optoelectronic devices, such as quantum dot lasers [1] and quantum dot infrared photodetectors [2]. For these applications, it is important to understand the relaxation mechanism of carriers in such system with discrete energy levels. A lot of efforts have been made on the study of carrier relaxation mechanisms for self-assembled QDs by various spectral techniques [3-12].

It is well known that energy relaxation of carriers in QDs cannot be achieved by simply interaction with acoustic phonons due to large separation in quantized energy levels. Other channels such as Auger-type scattering [13] and/or multi-phonon scattering [14] are needed to facilitate the relaxation.

Carrier relaxation assisted by phonon scattering is based on the principle of weak coupling between carrier and phonon. In the weak coupling regime, phonons do not contribute to the energy states of the QD but only cause carrier scattering between states. So when inhomogeneous QDs are probed by selective excitation PL (SEPL) or PL-excitation (PLE) technique, the multi-LO-phonon filtered transition could be observed [8-12]. On the other hand, if the correlation between carriers and phonons becomes so strong that the energy spectrum of QD is enormously modified, strong coupling should be considered. In this regime, the new set of eigen-states associated with polaron is treated in the QD system. The relaxation mechanism of polarons, different from the principle of scattering, is a coherent flopping (Rabi flopping) between eigen-states [15]. In this case, the feature of anti-crossing has also been observed under magnetic fields [16-17].

Among the previous published results, carrier relaxations with or without multi-phonon filtering were both revealed [3-12]. These two types of behavior can be distinguished by the relaxation energy. When different QD subsets in a sample are selectively excited with energies smaller than the wetting layer (WL) absorption edge, the resonant electronic transitions should reveal the size dependent relaxation peaks due to the quantization. If the transitions undergo multi-phonon filtering, the relaxation spectra reveal the characteristic of certain LO-phonon replica. The possible reason for samples showing separately these two features has been attributed to the existence of competing non-radiative recombination channel [12].

In this study, with a single sample, both features were observed with different regions of excitation energies. This energy dependent relaxation mechanism is investigated and explained.

2 Samples and experiments The QD samples used in this study were grown by molecular beam epitaxy with valved cracker As source on (001) semi-insulating GaAs substrates. Single InAs QD layer with 150nm GaAs barrier layers and an additional QD layer with the same growth condition for atomic force microscope (AFM) measurement was grown in each sample. Two samples with different sizes were prepared. For sample A, 2.6MLs of InAs was deposited at 520 °C. It results in lens shaped QDs with average height of 9 nm and a diameter of 47 nm. For sample B, only 2.4MLs were used at 480 °C. The average height was only 1.5nm, and the base diameter was about 17nm. The SEPL and the PLE spectra of the QDs were measured at 13 K. Because of the different QD sizes, the two samples exhibit different transition energies. At 13 K, the ground-state emission peak of sample A is at 1.168 eV, while for sample B the transition energy is about 1.269 eV.

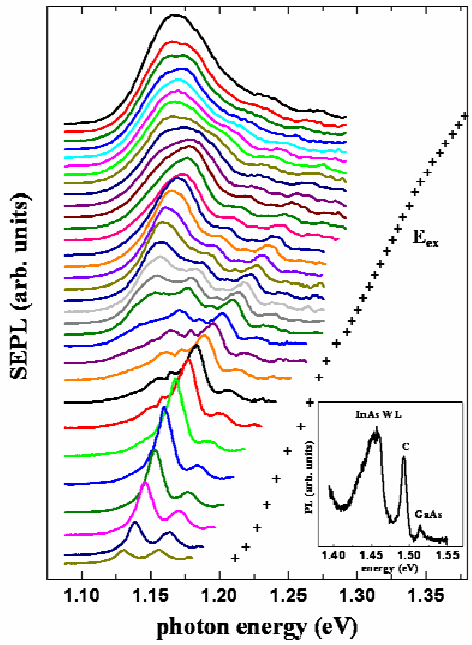

Figure 1 SEPL spectra of sample A taken at 13K. The cross on the right hand side of each spectrum shows the position of excitation energy.

3 Discussions The SEPL spectra for sample A is shown in Fig. 1. The excitation energy varied from 1.378 eV to 1.211 eV with the corresponding spectra arranged from top to bottom. The position of excitation energy is indicated by the cross on the right hand side. They were all below the WL ground state energy (around 1.45 eV determined from the high power PL spectrum shown in the inset) to ensure that only the QD layer could absorb the pumping photons. Note that all spectra in Fig. 1 were excited with low power (~1 W/cm^2) to exclude any excited-state emission and also to suppress Auger scattering. When E_{ex} > 1.34 eV, the luminescence spectra show a broad peak indicating all QDs were excited. As the excitation energy decreased, fine structures with narrower peaks were observed in the spectra, because only certain sub-ensembles were resonantly excited due to the discrete density of states.

If we plot the spectra versus the relaxation energy, i.e., E_{ex} - E_{det}, detailed information about these peaks emerges. The result is shown in Fig. 2. The sharp peaks line up vertically in the same positions. The peak values of E_{ex} - E_{det} correspond exactly to the multi-LO-phonon energies. Only the QDs with the excited states at multiple phonon energies above the ground state are selectively excited. When the excitation energy increases above a certain value, a different excitation scheme was observed. Both the emission peak energy and E_{ex} - E_{det} varied with the excitation energy. Three distinct regions in the PL spectra under different excitation energies are designated as regions I, II and III respectively.

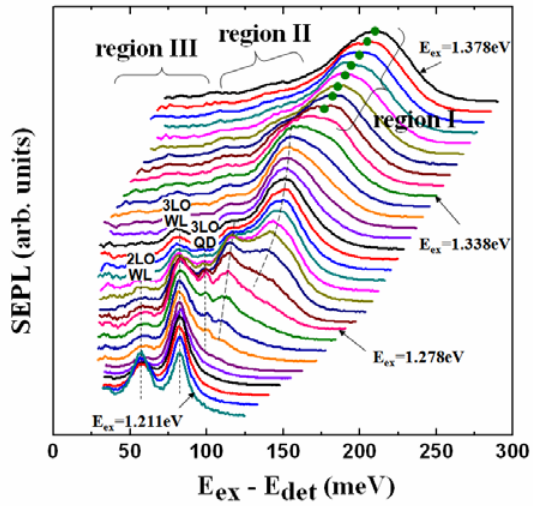

Figure 2 The SEPL spectra in Fig. 1 were plotted versus E_{ex} - E_{det} here, with intensity normalized to 1 for better clarity.

The broad emission spectra with peaks denoted by filled circles in region I indicate the whole QDs can absorb light and also have efficient carrier relaxation. For a self-assembled QD, the joint density of states between bound states and delocalized states [18] give rise to a quasi-continuous absorption region extending downward from the WL bandedge [19]. Therefore, when E_{ex} is lower than the WL peak but higher than the QD bound states, the excited carriers can relax energy to the ground state through the continuous tail, cascaded by multi-LO-phonon emission. The mechanism of energy rolling down can be primarily due to the interaction with acoustic phonons. In this excitation regime, although E_{ex} is lower than the WL bandedge, no selective excitation will occur because the excited electron and hole are not both localized. Thus the emission spectrum is basically the same as that from conventional PL measurement.

The density of states for such continuum decreases gradually towards the lower energy. Once the excitation energy of the pumping photons is low enough to reach the discrete levels of the QDs, the sharp levels enhance the absorption through resonant excitation. Note that the resonantly absorbed QDs still have a distribution in ground-state energy due to the fluctuations in shape, strain, and composition. At low excitation energies, when the carriers are resonantly excited to states below the continuum tail, the only way for them to relax to the ground state is through the multi-phonon process. The sharp peaks in the bottom few emission spectra are therefore signatures of various multi-phonon processes. Three phonon lines marked as vertical straight lines in Fig. 2 were clearly observed, and they were assigned as, from left to right, 2LO WL phonon peak (56 meV), 3LO WL phonon peak (83 meV) and 3LO QD phonon peak (100 meV). This assignment gives the energy for the WL phonon to be ~27.8 meV and the QD phonon to be ~33.3 meV. These values are very close to that in ref. 9. The strong phonon confinement in the thin WL lowers the energy of LO phonons, but in QDs, the strain effect dominates and increases the LO phonon energy [10].

The spectra in region II showed different characteristics due to a different carrier relaxation scheme. With such excitation energy, the QDs are still selectively excited through resonant excitation to the discrete states of the QDs. These states, however, are high in energy and are within the continuum tail of the joint density-of-states. The relaxation of the excited carriers can therefore still be achieved by the help of the continuum states in addition to the multi-LO-phonon scattering. With the help of the continuum states, the energy separation of the excited state and the ground state does not have to be exactly the same as the multi-LO-phonon energies. Because the states are within the continuum tail, the excited electron-hole pairs can change their discrete natures in density-of-state by part with energy conserved, say, the electron can transfer energy to the hole and push the hole to the continuous hole states. The efficiency of this electron-hole scattering has been confirmed [20-22]. As soon as the hole is pushed to the continuous states, the efficient relaxation channel mentioned in region I works. Therefore, the process of electron-hole scattering cascaded by phonon scattering excludes phonon bottleneck and explains the spectra in region II in Fig. 2. The difference between the excitation energy and the emission energy, therefore, reflect the size dependent excited-state separations in the QDs. We notice that the emission peaks become narrower as we enter the resonant excitation regions (region II and III). This is because the resonant excitation selects certain QDs with specific energy levels. The narrowest peaks belong to those at very low excitation energies because the selected QDs need to satisfy not only the resonant excitation condition but also an additional requirement of multi-phonon resonance.

As described above, if the excited states of QDs are high enough in energy and are within the continuum tail, the resonantly excited carriers have efficient channels to relax energy to give ground-state emission. The spectra under selective excitation technique will thus not be filtered by LO-phonon energies. To further prove this idea, the sample with ultra small InAs QDs was studied. The reason for choosing small QDs is: the closeness of the high energy quantized states to the WL state ensures the presence of the continuum tail, which assist carrier relaxation to the ground state.

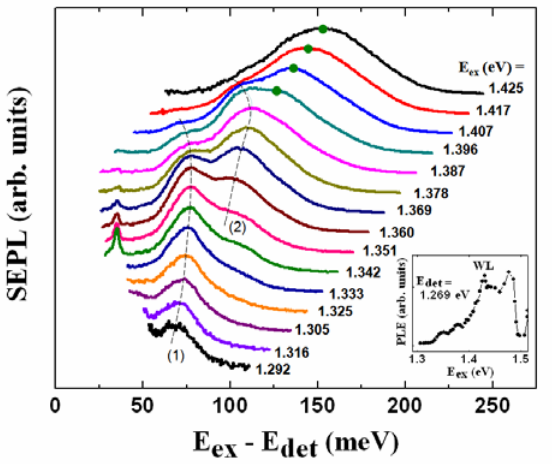

Figure 3 SEPL spectra of sample B taken at 13 K. The x-axis refers to E_{ex} - E_{det}.

The SEPL spectra of sample B, which has small QDs, are shown in Fig. 3. Each spectrum was plotted versus E_{ex} - E_{det}, and the intensity was normalized to 1 for better clarity. All excitation energies were also below the WL bandedge, which was 1.43 eV in this case (determined by the PLE spectrum shown in the inset). The excitation scheme of the highest few curves in Fig. 3 belongs to region I as previously mentioned. When E_{ex} is lower than around 1.41eV, two peaks associated with resonant excitation emerge, and they become prominent when E_{ex} is even lower. These two peak series are labeled by (1) and (2) in Fig. 3. It is clear that the two peaks do not line up vertically, although they are within the energy range achievable by triple of common LO-phonons. This indicates that they have high efficiency in relaxation without suffering any phonon bottleneck. The result agrees with our previous assumption that the excited-states of QDs in sample B, of which the dot size is very small, are high enough in energy to touch the continuum tail. For the sharp peaks that are located rigidly at 35 meV, because of this characteristic energy, they were tentatively attributed to the Raman process associated with the interface phonons in the QD structure [10, 23].

4 Conclusions The energy spectra of InAs/GaAs self-assembled QDs have been studied using selective excitation photoluminescence. The dependence of the emission

spectra on the excitation energy provides fruitful information on the carrier relaxation mechanism. Three distinct regions can be identified as the excitation energy changes. At high excitation energies, all quantum dots are excited and participate in the emission with the help of the continuum states. At low excitation energies, because of the absence of the continuum states, carrier relaxation has to be facilitated by a multi-LO-phonon process. So only the QDs with excited states at certain multi-LO-phonon energies above the ground state can absorb and emit light. At medium energy, the QDs are resonantly excited through discrete electronic states. But the relaxation of the carriers does not need to satisfy the multi-phonon resonance condition with the help of continuum tail. The electron-hole scattering provides an efficient channel for the electron to lose its large quantized energy first and then the hole can release energies in cascade to the ground state by emitting phonons.

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