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ANOMALOUS EXCITATION INTENSITY DEPENDENCE OF PHOTOLUMINESCENCE FROM InAs SELF-ASSEMBLED QUANTUM DOTS

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Abstract—We have studied photoluminescence (PL) from single InAs self-assembled quantum dots (QDs) by micro-PL measurements and, in particular, studied the excitation intensity dependence. We have found strong quenching in both the peak and integrated intensity of narrow PL lines when the excitation power was increased. This suggests that the present behavior is mainly due to the change of carrier screening and relaxation between the InAs wetting layer and InAs QDs.

Carrier dynamics in quantum dots (QDs) is one of the critical issues in quantum nanostructures since the complete discretization of the energy levels due to three-dimensional quantum confinement greatly influences the phonon scattering of electrons[1]. This results, on one hand, in a slow energy relaxation of high energy electrons in the quantum dots (phonon bottleneck effect)[2]. Because this implies poor intrinsic luminescence efficiency, this casts a severe problem in the application of QD structures to optical devices. On the other hand, it also leads to a long coherence time for photoexcited electron–hole pairs as expected from analogy with atomic systems, which has advantages for the application of coherent control techniques[3] for ultrafast switching and quantum computation.

Although many investigations have been reported theoretically[4,5] and experimentally with various kind of QDs[6–8], the above points seem to be still controversial and more comprehensive study is indeed required. One of the obstacles to drawing clear conclusions is the large size fluctuations and thus large inhomogeneous broadening of QDs that can be realized by present state-of-the-art technology, for example, InGaAs/GaAs QDs of strain-induced self-assembling islands[9,10], InGaAs/AlGaAs quantum disks[11] and CdSe nanocrystal QDs[12]. To overcome such inhomogeneity, methods to achieve selective excitation[13–15] have been attempted and reported. One straightforward way is to limit the number of QDs that are to be characterized in real space. This can be carried out, for example, by using near field probes[16], by etching off[17] or masking[18] the region outside the range of interest, or by focusing an excitation beam and detecting PL from a extremely small area[19–23] and they all have succeeded in the observation of sharp PL lines expected from zero-dimensional nature of QDs and the excitons. In particular, the latter method is often referred as micro-photoluminescence (micro-PL) measurement and shown to be extremely effective for carrying out spectroscopic study on a single QD[19–22].

We report here on CW micro-PL investigations of carrier dynamics in single self-assembled InAs QDs. We find strong quenching of the micro-PL from QDs when the excitation intensity I_{exc} is increased. Possible mechanisms for the observed behavior described below implicate the controlling influence of electronic states in the nearby wetting layer.

The self-assembled InAs QDs were grown by Stranski–Krastanov growth mode on GaAs substrates by metalorganic vapor phase epitaxy. After the growth of GaAs buffer layer at 700°C, InAs with a nominal thickness of 1.8 ML was grown at 500°C. This amount of InAs is very near to the threshold for the dot formation. Then the QDs and WL were buried with successively overgrown GaAs and AlGaAs layers at 480 and 650°C, respectively. Finally, a GaAs capping layer was grown. Atomic force microscopy (AFM) studies were carried out on a sample without overgrown layers and revealed that the average size of the QDs is 3.28 nm in their height and 29.3 nm in their lateral size, with standard deviation of 45% and 42%, respectively. The density of QDs were also measured by AFM and was 5.2×10^8 cm^{-2}, though there were considerable density distribution in the sample used in the PL study. We took advantage of this to investigate density dependent relaxation and emission processes, as described later.

Micro-PL measurement was carried out with a ×25 microscope objective. The excitation beam was focused into ≤2 μm diameter spot on the sample in a variable temperature cold-finger cryostat. The PL collected through the same microscope objective was dispersed into a spectrometer with a CCD multichannel detection system. The spectral resolution in the measurement was mainly limited by the spectrometer and was 0.11 nm, which corresponds to about 0.16 meV around 900 nm. Argon ion, He–Ne and Ti:S lasers in cw operation were used as excitation sources. In these measurements, care was taken to achieve high positional stability and reproducibility of the excitation position on the sample at liquid helium temperatures.

Figure 1 shows typical PL spectra of the sample measured in different configurations. Spectra in (a), measured in the micro-PL configuration, show a number of sharp peaks in the energy range between 1.32 and 1.42 eV together with a broad feature at 1.44 eV. We confirmed that these sharp PL lines showed excellent reproducibility, and were observed up to temperatures of 60 K. The result of PL measurements with larger spots (~100 μm, macro-PL) are plotted in Fig. 1(b), showing broad peaks at 1.38 and 1.46 eV. From these results, we conclude that the sharp and broad peaks observed in Fig. 1(a) originate from the InAs QDs and InAs wetting layer (WL), respectively, as has already been reported by several groups[17,23]. The width of these peaks differs slightly from peak to peak down to a minimum of 0.16 meV and depends on the excitation condition. We believe that the measured peak widths are limited by the resolution limit of the present measurement setup and their intrinsic widths would be at least as narrow as previous reports[21].

In Fig. 1(c), PL spectra are shown measured in the same micro-PL configuration as (a) with the same excitation intensity but obtained from a different position on the sample. One can readily see that the number of sharp lines is reduced greatly in (c), indicating that the density of quantum dots strongly differs in the two areas. One can also see that the luminescence from the WL is enhanced and red-shifted in (c) where the QDs are less dense. These results suggest that the carrier relaxation process depends on the density of QDs.

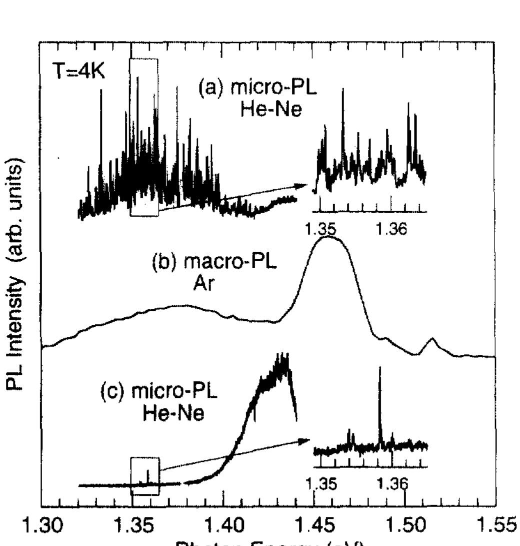

Fig. 1. PL spectra of InAs QDs and wetting layer (WL) measured in different experimental configuration.

In the present excitation condition, most of the absorption takes place in the GaAs barriers. We also have confirmed that absorption is small and negligible in the WL and QDs, respectively, by investigating the excitation energy dependence of the PL. The PL rate in the GaAs barriers is an order of magnitude weaker than PL from the WL, thus carriers first generated in the GaAs layer relax into the WL, and then into QDs. In addition, the existence of localized states in the WL is likely because of potential fluctuations induced by the thickness variations of the thin WL. This would also retard carrier diffusion in the plane of the WL. Thus our results on the energy dependent emission can be explained by the fact that carrier relaxation from WL to QDs is limited by the carrier diffusion in the WL.

Next, we show careful measurements of the excitation intensity dependence of QD and WL PL. Typical results are shown in Fig. 2(a) and (b) for QD and WL, respectively. Note that the spectra of Fig. 2(a) are offset for clarity, but all of them share the same vertical scale in the figure. One can clearly see that the PL intensity of the QDs shows strong nonlinear dependence and quenching behavior with I_{exc}. These effects are reversible, that is, the PL intensity does not depend on whether the excitation intensity is increased or decreased, within the present experimental range (maximum ~70 μW), and is reproducible and not hysteretic. Note that the WL PL breaks up into many peaks as the excitation density reduces. This clearly indicates the existence of localized states in the WL.

In Fig. 3(a), we plot the peak intensities of four peaks indicated by the arrows with label x in Fig. 2(a), and peaks associated with the WL [Fig. 2(b)], as a function of excitation intensity I_{exc}. Distinct quenching behavior of QD PL is evident, while that of WL linearly increases with I_{exc}. We also find their linewidths become broader as I_{exc} increases, as shown in Fig. 3(b). This may be the result of an increased spontaneous emission rate at high powers, or shifts of the emission peak within the integration time of the measurements (> 10 s). However, such broadening cannot explain the quenching behavior since the integrated PL intensity is also quenched.

In Fig. 2(a), we also see the appearance of several new peaks at intermediate excitation intensities, which can be classified into three types: broad background components, isolated sharp peaks appearing ~10 meV away from the original peaks, and sharp

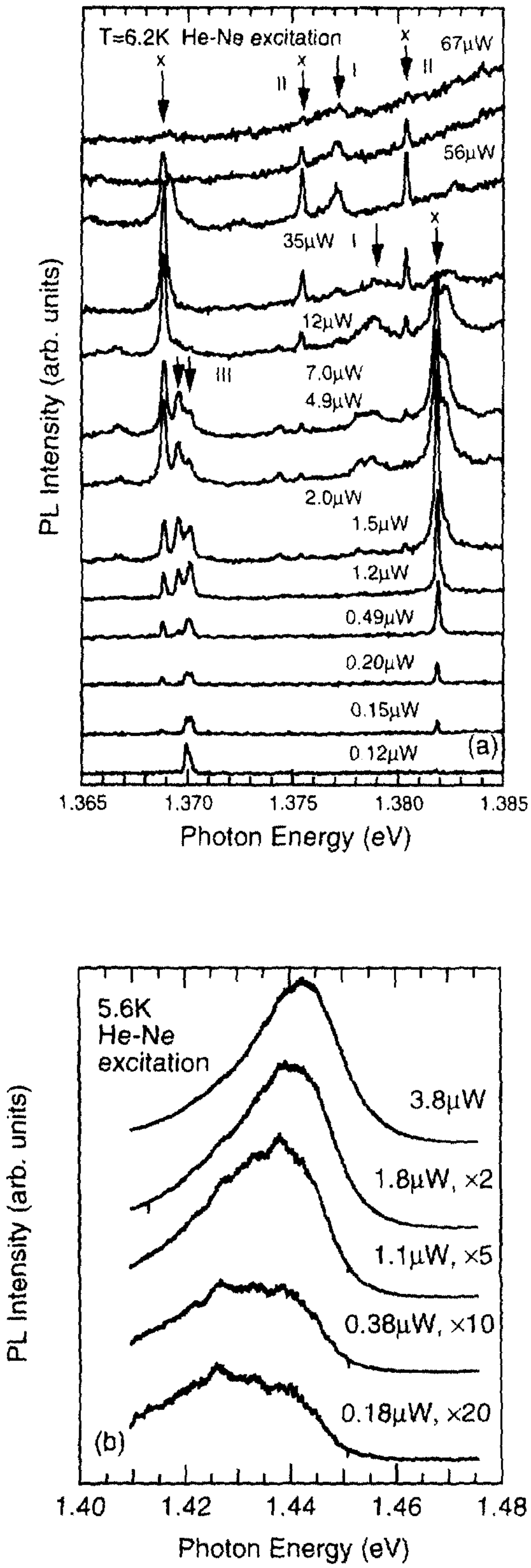

Fig. 2. PL spectra of InAs QDs (a), InAs WL (b) measured at various excitation intensity I_{exc}.

lines which appears very near (< 1 meV) to the original peaks. The first type of new peaks [labeled as I in Fig. 2(a)] can be seen in the broad peaks around

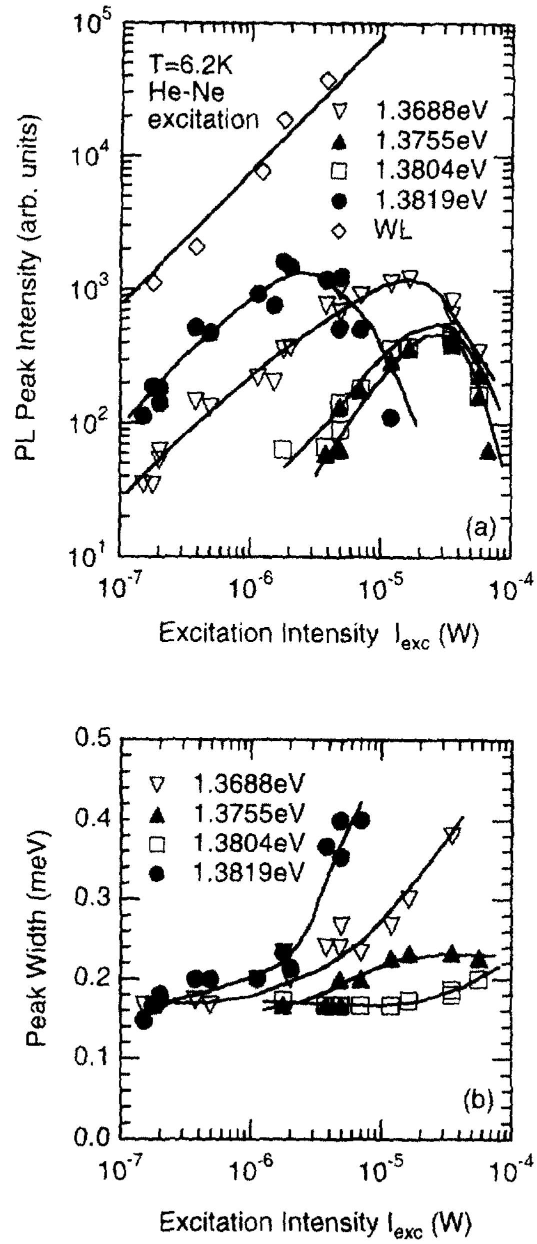

Fig. 3. Intensities (a) and widths (b) of QD PL for four peaks indicated by allows with label x in Fig. 2(a), plotted as a function of excitation intensity I_{exc}.

1.377 and 1.379 eV at intermediate intensity. The origin of these broad peaks and the background components are not clear at present and require further investigation. The peaks at 1.3755 and 1.3804 meV are of the second type (labeled as II) and quench at higher excitation intensity as shown in Fig. 3(a). On the other hand, the third type of new peaks (labeled as III) seems to showing a "pairing" behavior, as seen in peaks around 1.370 eV in Fig. 2(a). Similar behavior was also observed in the different set of excitation intensity dependence collected at a different sample position, as shown in Fig. 4. We can see clear inversions of

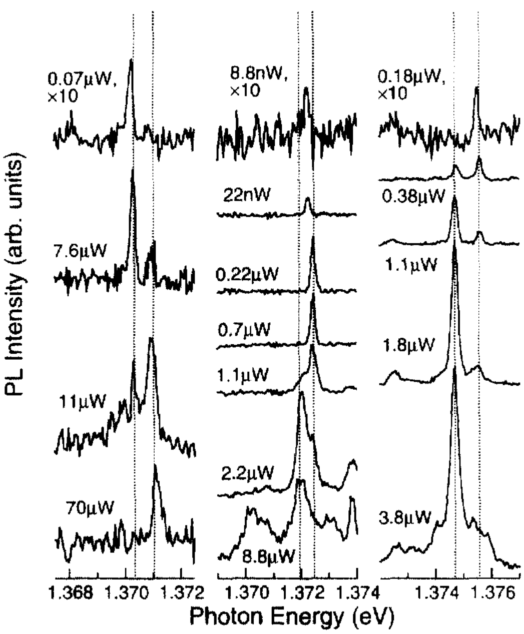

Fig. 4. PL spectra taken at three different positions and their intensity dependence.

the relative intensity of adjacent two peaks. Saturation of the PL intensity is expected and can be explained by band-filling effects, as reported by Hessman *et al.*[20] in self-assembled InP QDs, and by Kamada *et al.*[21] in strained InGaAs disks. The present quenching behavior, however, cannot be explained in this way. Although a full explanation is lacking at present, we believe that the third type of closely-spaced peaks are closely related to the present quenching behavior. They indicate that new luminescencing states become involved at high photoexcited carrier densities.

At high excitation, single narrow lines appear in parts of the spectrum which are at large energy differences (~10 meV) from the peaks observed at low intensities (induced peaks of type II). We believe these to be PL from different dots which are spatially separated from the first dot. Such behavior is expected if the carrier relaxation from wetting layer to QDs is modified by the high density of photoexcited carriers through enhancement of the diffusion length. At sufficiently low excitation, the number and excess energy of carriers in the wetting layer is not large. Carriers tend to become localized because of the large potential fluctuations within the WL, as mentioned earlier. Therefore, carriers trapped in this WL mostly have to recombine in the WL or QD in the vicinity of the excitation (and thus the detection) spot. However, if the WL diffusion length becomes longer either due to screening or carriers filling up states below the mobility edge, the carriers in the central region of the excitation spot are more mobile and diffuse to the outer region of the detection spot. Because of the random distribution of surrounding QDs and the Gaussian intensity distribution across the laser focus, this would lead to PL from other QDs turning on at various different excitation densities.

We now consider the sharp lines (type III) which appear at higher intensity around (~1 meV) the PL peaks from a single QD. In some cases, inversion of their relative intensity can be seen, as shown in Fig. 4. The new peaks appear on both the higher and lower energy side of the original peaks. These pairs of peaks seem to originate from the same QDs since all of the original peaks mentioned here are observed from the lowest excitation investigated, and quench at relatively weak excitation. Furthermore, the emergence of new peaks strongly correlates with the quenching of the original peaks. Possible origins of these states include exciton complexes, such as charged excitons, and biexcitons. However, we do not have any further evidence for such exciton complexes since the intensity dependence of the QD PL remains almost linear until quenching occurs, in contrast to the superlinear dependence expected from exciton complexes. Another new possibility is that as the carrier density in the WL around each QD increases, the energies of states within the QD change, through screening and exchange interactions. In this case it is possible to imagine that the exact spatial distribution of the wavefunctions can change the sign of the energy shift as indeed found in different QDs (Fig. 4). Quenching can then arise from a reduction in the overlap between WL and QD states through the same Coulombic interactions. So although Auger and carrier–carrier scattering effects might be expected to overcome any bottleneck in trapping carriers in the QD, this might be exceeded by the screening of the QD from the WL by surrounding weakly localized states. Additionally, as the WL states become more delocalized through the increased screening from the background carrier density, they may also be less efficiently scattered into lower energy QD states by phonons. A full theory to account for these observations must explicitly solve the QD and WL system in the presence of the Coulomb interaction. However, the results strongly suggest the influence of the Coulomb interaction between different occupied states in QD and WL.

In summary, we have investigated photoluminescence of InAs self-assembled QDs by micro-PL measurement. We have found extremely anomalous dependences of the QD PL on excitation intensity, including a quenching and pairing behavior of sharp spectral lines from a single QD. The importance of changes in the carrier relaxation from WL to QDs due to Coulomb interactions is suggested to explain the observed behavior.

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