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Temperature-dependent micro-photoluminescence of individual CdSe self-assembled quantum dots

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We use micro- and nano-photoluminescence to study the temperature-dependent excitonic emission from CdSe quantum dots embedded in a ZnSe matrix. By varying the spatial resolution from 200 nm to 1.7 μm, we are able to study the temperature dependence of the ultranarrow ($\sim$200 μeV) emission from excitons confined to *single* quantum dots, as well as statistical ensembles of up to 200 dots. By measuring the quenching of the photoluminescence (PL) with temperature, we find compelling evidence that the PL emission from these samples results from two different kinds of states. Similar to previous work, we find that a broad PL line persists to 300 K with an activation energy of $\sim$40 meV. However, we find that the ultranarrow lines are quenched at about 60 K, indicating an effective activation energy of only 4.0 meV. © *1999 American Institute of Physics.* [S0003-6951(99)01228-0]

In recent years there has been intense interest in lower dimensional semiconductor nanostructures, particularly quantum dots, because of the subtle and profound changes that should occur as length scales become comparable to the exciton Bohr radii, and the dimensionality is reduced to nearly zero. One method for growing quantum dot structures utilizes the large lattice mismatch between two semiconductors (e.g., InAs/GaAs or CdSe/ZnSe) which is relieved to form islands, domes, or pyramids across the surface of the lower gap material (InAs or CdSe). While significant effort has been applied at the InAs/GaAs system, much less is known about the growth kinetics and nature of quantum dots in the II–VI systems.

One general finding in such structures is that the photoluminescence (PL) line shapes depend very much on the spatial scale which is sampled. While macro-PL experiments ($\sim$30 μm spatial resolution) exhibit a strongly inhomogeneously broadened line shape with a width as large as 80 meV,[1,2] a μ-PL experiment (1–2 μm resolution) resolves separate emissions from hundreds of individual dots with ultranarrow ($\sim$200 μeV) line shapes which reflect the delta-function like density of states of excitons confined to a box.[3–5] In a nano-PL (n-PL) experiment (100–500 nm) it becomes possible to study individual quantum dots.

Such experiments have provided us with our present knowledge of excitons in CdSe/ZnSe quantum dots: (1) macro-PL measurements show a broad 80 meV inhomogeneous PL line which exhibits a temperature-independent radiative lifetime of approximately 500 ps;[6–8] (2) this broad PL line persists to room temperature and quenches with an activation energy of 40 meV;[1,7] (3) a number of low-temperature μ-PL and n-PL experiments have resolved the ultranarrow emission from the quantum dots with estimates of the dot density ranging from 100 to 700 dots/μm^2.[3,4,7,9] An underlying assumption is that there is only *one* state with a single binding energy and radiative lifetime responsible for the PL from these structures.

In this letter, we report on the temperature dependence of ultranarrow emission from a single quantum dot using n-PL through fixed 200 nm apertures which shows that the PL from the quantum dot is quenched by 60 K with an activation energy of only 4 meV. In addition, we show that a statistical sample of ultranarrow emission from $\sim$200 quantum dots exhibits the *same* activation energy of 4 meV. Once the narrow line emissions disappear, the underlying broad PL line persists to room temperature with an activation energy of $\sim$40 meV. We argue that *two* distinct states must be responsible for emission in these CdSe dots: (1) a state responsible for the narrow (200 eV) PL lines which is only weakly bound with a 4.0 meV activation energy, and (2) a state responsible for the broad PL line which persists to higher temperatures which exhibits an activation energy an order of magnitude larger ($\sim$40 meV).

As previously described, our quantum dots are grown in the Stranski–Krastanow mode of strain-induced growth via molecular beam epitaxy (MBE).[1,2,4] The sample was fabricated on a (100)GaAs substrate using a Riber 32 R&D MBE machine equipped with elemental sources. First, a ZnSe buffer, approximately 1 μm thick, was grown at 300 °C. Three monolayers CdSe were then grown at a very slow rate of 0.025 monolayer per second for reproducibility and controlled deposition, resulting in the formation of dots on the sample surface. Finally, the dots were capped with a 50-nm-thick ZnSe layer.

Figure 1 shows μ-PL spectra of our sample at three different temperatures. These data, obtained with 1.6 μm resolution, reveal the sharp emissions from individual quantum dots. At $T=6$ K, sharp peaks are superimposed over a broad emission; at $T=60$ K, most of the sharp peaks have disappeared while the broad emission remains intense. In order to facilitate measuring the temperature dependence of a single-

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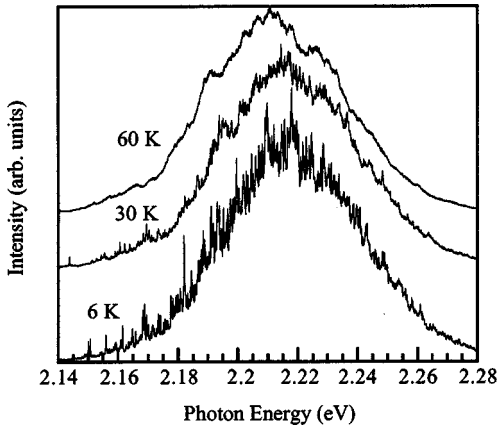

FIG. 1. μ-PL measurements displaying the broad peak width sharp structures superimposed. Note the disappearance of the sharp structures by 60 K.

dot emission, we have reduced the number of sharp peaks in our spectra by increasing our spatial resolution by fabricating 200 nm metal microapertures on the sample. Figure 2(a) shows the spectra measured through a microaperture as a function of temperature from $T=7$ to 60 K. This emission peak, occurring on the lower-energy side of the broad peak, has a width of 200 μeV at 7 K increasing to 900 μeV by 60 K. The sharpness of the emission line allows a sensitive measurement of the redshift behavior, known in bulk to be primarily due to the renormalization of the electron–phonon interaction. We find that the energy shift is different than expected for the bulk.[10]

Spectrally integrating the n-PL signal allows us to monitor the quenching of the emission strength with temperature, as displayed in Fig. 2(b). To extract an activation energy, we fit the intensity data to the form:

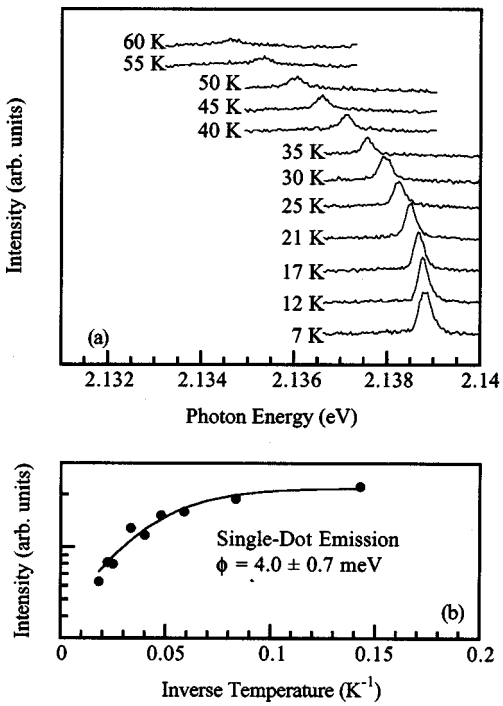

FIG. 2. (a) Photoluminescence measurements of a single dot as a function of temperature; the data were obtained through a 200 nm fixed aperture. (b) The spectrally integrated intensity of the quantum dot photoluminescence is plotted vs inverse temperature; this data yields an activation energy for the quantum dot of 4.0 ± 0.7 meV.

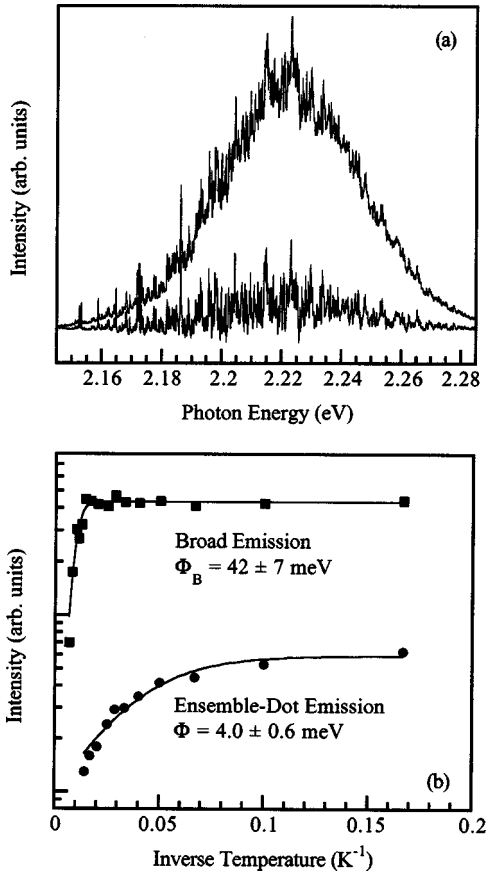

FIG. 3. (a) The original PL spectrum (upper) and the data isolating the sharp quantum dot spectral structures (lower). (b) A fit of the spectrally integrated intensity vs inverse temperature is displayed for the entire collection of sharp quantum dot spectral structures; a fit to the data yields an activation energy of 4.0 ± 0.6 meV.

$$I(T)=I_0[1+C\exp(-\phi/kT)]^{-1},$$

where ϕ is the activation energy of a single-dot exciton and C contains ratios of optical-collection efficiencies and effective degeneracies between the unbound and bound states. A careful fit to the data yields $\phi=4.0\pm0.7$ meV, a much smaller value than obtained from macro-PL data, and much smaller than the 80 meV spanned by the energies of the sharp-emission peaks. We have found that other single-dot emission lines behave similarly, yielding $\phi\sim4$ meV.

To investigate the entire collection of sharp emission lines, we isolate the sharp emissions from the broad emission by fast Fourier transforming the spectra, performing appropriate filtering on the transformed spectra to reduce the contribution from the broad-emission component and then back-transforming to recover only the sharp peaks.[10] This technique is reliable and possible because the broad-emission spectral width is at least two orders of magnitude larger than those of the sharp peaks. Figure 3(a) displays the dot-only spectra corresponding to the spectra in Fig. 1.

In the lower part of Fig. 3(b) we display a fit of the spectrally integrated intensity versus inverse temperature for the entire collection of sharp peaks displayed in Fig. 3(a). We obtain a value for the activation energy for this ensemble of lines of 4.0 ± 0.6 meV, suggesting that the dots in the ensemble have a common activation energy. This value is remarkably close to the value of 4.0 ± 0.7 meV obtained for a

single dot. The behavior of the integrated intensity of the broad emission line versus temperature up to 180 K [see top of Fig. 3(b)] suggests an activation energy of 42 ± 7 meV, distinctly different from that obtained for the sharp emission lines. The temperature dependence displayed here is similar to previous reports of macro-PL data where individual dots were not resolved. Preliminary measurements of the lifetimes of the excitons reveal a very long lifetime for the sharp emissions, typically several nanoseconds, in contrast to the broad emission where decays of $\sim$500 ps are observed.[11] Thus there is strong evidence that there are two distinctly different kinds of state, one associated with the sharp emission lines, and one associated with the broad emission line.

In summary, we have measured temperature-dependence photoluminescence (μ-PL and n-PL) from CdSe self-assembled quantum dots on ZnSe. We have observed sharp luminescence from *individual* quantum dots, typically 200 μeV in spectral width. By measuring the spectrally integrated intensity as a function of temperature, we have extracted a thermal-activation energy of the exciton(s) bound to the dot, $\phi=4.0\pm0.7$ meV. We have also measured the integrated intensity of the entire *ensemble* of dot emissions as a function of temperature and found a remarkably similar value of 4.0 ± 0.6 meV. This suggests a common activation behavior for the excitons in different quantum dots, even though their recombination energies span approximately the 80 meV width of the broad emission line. The contrasting behavior of the broad emission line, with a significantly different temperature dependence and a significantly different lifetime, is strong evidence that two different states are responsible for these observations.

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