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The spectral dependence of the photoluminescence recombination lifetime has been measured for individual self-assembled InGaAs/GaAs quantum dots, over the entire emission envelope. The measurements show a rising trend with increasing emission wavelength, increasing from 680 ps at 900 nm to about 1020 ps at 990 nm. Measurements of the out-of-plane diamagnetic coefficients for the dots show almost no correlation with wavelength. As a result, the rising trend in the lifetimes with wavelength is interpreted in terms of the emission energy being predominantly determined by the dot height, with higher dots exhibiting longer lifetimes. © *2010 American Institute of Physics*. [doi:10.1063/1.3293294]

The great scientific interest in quantum dots arises from two features, (i) they offer the opportunity to perform fundamental experiments on artificial atoms with a range of properties unavailable with real atoms, and (ii) they show considerable potential for application in functional devices, in areas such as light emission and quantum information processing. In optoelectronic applications, the emission time constant of a quantum dot is vital as it plays a large part in determining the device efficiency and, in some cases, the repetition rate. In quantum information processing, the balance between the carrier and spin lifetimes is crucially important for many envisaged implementations. It is thus extremely important to know and understand the relationship between the transient characteristics and the wavelength of a quantum dot.

Our single layer of self-assembled InAs quantum dots was grown by molecular beam epitaxy on an undoped GaAs substrate. 2.1 mono layers of InAs were deposited onto the GaAs at a temperature of 530 °C, followed by a 15 s growth interrupt before growth of the GaAs capping layer. The effects of the relatively high growth temperature are a low density of rather large quantum dots and an intermixing of Ga into the quantum dot layer.[1] The dot density was estimated from AFM measurements on a second layer of identically grown quantum dots on the top of the sample, giving about 4×10^9 cm^{-2}. The AFM images also revealed the heights of the dots to be 4.1 ± 0.9 nm, and the circular base to have a diameter of 72 ± 9 nm, where "±" indicates the standard deviation of the distribution. The luminescence from the quantum dot ensemble is peaked at about 955 nm, with a full width at half maximum of about 50 nm.

The sample was placed on a three-dimensional micropositioning unit, inside a He flow cryostat, and cooled to 4.1 K for all but the temperature-dependent measurements. Photoluminescence (PL) was excited with a mode locked Ti:Sapphire laser emitting at 765 nm, with a repetition rate of 76 MHz. A 100× objective lens focused the laser light onto the sample through a solid immersion lens mounted on the sample surface. Single dot PL was collected through the same objective lens as used for excitation, and sent to a 50 cm monochromator, followed by either a Si charge-coupled device camera or, for the time-resolved measurements, a Si avalanche single-photon detector, the output of which was connected to a time correlator card, triggered by a fast photo diode.

The temporal response of the setup has a full width at half maximum of approximately 650 ps. In spite of this, a deconvolution technique allows the ultimate temporal resolution to be governed by the bin width of the time correlator card, which is 37 ps. A function consisting of a rising exponential and a single exponential decay was fitted to each data set by the least-squares method. Correlation coefficients were calculated for each fit, and in each case were more than sufficient (>0.999 in nearly every case) to rule out the need to hypothesize any multiexponential decay.

Figure 1 shows the low-temperature PL recombination times for individual quantum dots as a function of emission wavelength (dots), obtained using a pump power of 50 nW. Also shown is the ensemble PL (solid line). The dependence of the exciton recombination time on emission wavelength has been reported before with high-density InAs quantum dots,[2,3] individual thickness fluctuations in GaAs quantum wires[4] and quantum wells,[5] and ensemble measurements on

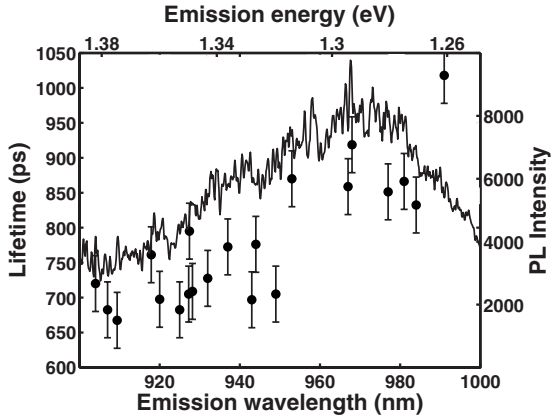

FIG. 1. Exciton lifetimes plotted as a function of the emission wavelength. The ensemble PL (continuous curve) is plotted also. All data were recorded at 4.1 K. Error bars mark the 95% confidence intervals.

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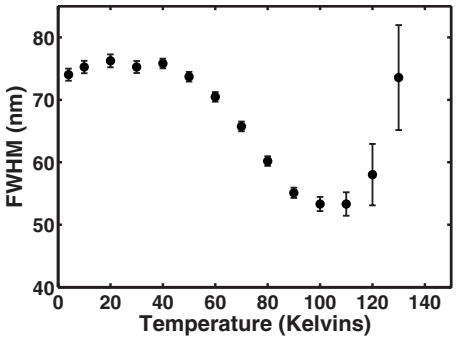

FIG. 2. Full width at half maximum for the ensemble luminescence as a function of temperature. Error bars mark the 95% confidence intervals.

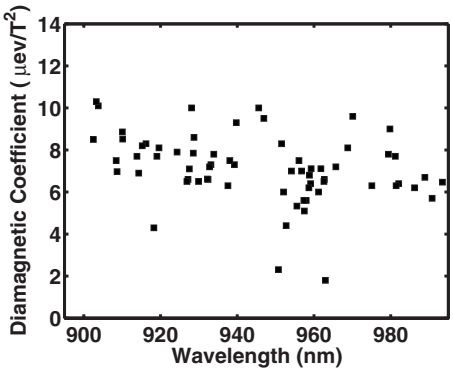

FIG. 3. Measured diamagnetic coefficients for the quantum dots using out-of plane magnetic fields up to 10 Tesla. The error bars on these data (not shown) are approximately the size of the plotted symbols. These data are described in more detail in Ref. 11.

self assembled InAs/GaAs quantum dots,[6] but not for individual self assembled InAs/GaAs dots.

An increasing oscillator strength with dot size was predicted theoretically by Takagahara[7] for spherical dots larger than the exciton Bohr radius. In Ref. 5, radiative lifetimes that decreased with emission wavelength were reported for "zero-dimensional" size fluctuations in quantum wells, where the structures were larger in the lateral dimension than the Bohr radius. In accord with the theory,[7,8] it is argued that in this regime of sizes, the oscillator strength of the exciton is proportional to the ratio of the quantum dot volume to the volume of the exciton. Since the oscillator strength is inversely related to the radiative time constant, then this argument accounts for their observed trend, which is opposite to the trend in our Fig. 1. Similar trends to that reported in Ref. 5 have also been observed in CuCl nanostructures[9] and colloidal CdSe nanocrystals.[10]

In Ref. 5, it is also shown that when the dot diameter is less than the Bohr radius, the opposite trend is observed; shorter wavelengths correspond to faster emission. This observation matches calculations for flat disklike dots by Andreani *et al.*,[8] predicting a minimum of the oscillator strength for a dot radius of about the Bohr radius. For our dots, however, assuming an equal mix of In and Ga, the bulk Bohr radius can be estimated to be roughly 16 nm, and we are clearly operating in the regime where the dot size (70 nm) is much larger. Even assuming pure InAs, the Bohr radius (30 nm) is still much less than the dot size. For such dots, we would therefore expect to see the opposite trend to that shown in Fig. 1.

We will now show how several straightforward explanations for the tendency in Fig. 1 can be excluded. In our PL spectra, the intensity measured on either side of an exciton line is typically only marginally higher than the detector dark count, so we rule out any influence of background emission on the lifetimes. The emission times can conceivably be modified by radiative coupling between quantum dots. In this case, however, any enhancement of the emission rate is determined by the density of dots emitting at the same energy. This means that the emission rate would peak at the same wavelength as the luminescence, so this form of coupling is ruled out. Furthermore, our dot density is far too low for tunnel coupling between dots to modify the exciton survival times.

Evidence against phonon-assisted carrier escape and subsequent recapture can be found in Fig. 2, showing temperature-dependent measurements on the ensemble. The emission linewidth was found to remain constant for temperatures below 40 K, before decreasing to a minimum at 100 K. The decrease in linewidth at temperatures above 40 K is explained by phonon-activated thermalization of the exciton population over the entirety of the available quantum dot states. We are led to infer that the redistribution of carriers does not occur at 4 K, and therefore cannot explain our observed temporal response. Measurements of the linewidth of individual dots at different temperatures support this conclusion; there is no change in the linewidth at temperatures below 40 K. Figure 2 also provides empirical evidence against any tunnel coupling between dots, as tunneling would eliminate the need for thermal activation of the carrier redistribution.

The observed rising trend in emission times with wavelength also occurs in our sample repeatably in macro-PL measurements at various pump powers, though the range of time constants is typically less for the macro-PL experiments (about 650 ps at 900 nm to 750 ps at 975 nm). This may be related to the higher pump powers used for the macro-PL measurements. While the single-exciton lifetime measurements in some cases indicated typically weak temperature dependencies that were unique to each dot, the macro-PL experiments showed no temperature dependence up to 50 K. It has been observed[6] that nonradiative recombination channels can have stronger effects on shorter wavelength dots but the absence of any multi-exponential decays and the absence of any consistent temperature dependence of the decay constants rule out a great many of the known nonradiative processes.

Data on the out-of-plane diamagnetic shifts of these quantum dots, described elsewhere,[11] are shown in Fig. 3, and are seen not to vary much as a function of wavelength. Since the diamagnetic coefficient is determined by the exciton diameter, generally considered to fix the spectral position within the emission envelope, Fig. 3 should show a strong positive trend. The conclusion reached, therefore, is that the dispersion of emission energies from the ensemble is not due to the dispersion of dot diameters. Furthermore, if the emission wavelength was determined by the dot composition, then the diamagnetic coefficient would increase with wavelength, due to changes to the exciton reduced mass, which is again opposite to the weak negative trend in Fig. 3. We argue instead that the height of the dot controls its emission wavelength, and that the dots emitting at longer wavelengths are those with greater heights.

Our dots have a diameter almost 20 times their height, and are also several times wider than the bulk Bohr radius. We feel, therefore, that the oscillator strengths for our excitons are essentially those of very narrow quantum wells, with slight modification due to the weak lateral confinement. Keldysh[12] has provided the theoretically derived oscillator strength for such a narrow quantum well, showing that it is inversely proportional to the well width. As the dot height increases, therefore, the oscillator strength would be expected to decrease, and the radiative lifetime would get longer, matching with our observations.

It may also be that there is a height-dependent permanent dipole moment in these dots, affecting the carrier overlap, and therefore the lifetime, but we find no evidence in the literature that higher dots exhibit larger dipole moments. Regarding the scatter in Fig. 1, it is likely that the variability of results at any given wavelength is governed by a spread of lateral sizes, with the lifetimes in that case possibly conforming to the trend observed elsewhere,[5,6] and predicted theoretically.[7,8]

We have measured the emission times for individual InAs/GaAs quantum dots over the entire ensemble emission range, and found a rising trend with wavelength. This trend is opposite to that typically observed for similar structures. Our observations are well explained if the emission wavelength is predominantly fixed by the dot height, which is indicated by measurements of the dot diamagnetic coefficients.

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