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Time-Resolved Photoluminescence Cross-Correlation Measurements on InAs Quantum Dots

L. M. HERZ[1]) (a), R. T. PHILLIPS (a), E. C. LE RU (b), and R. MURRAY (b)

(a) Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, UK

(b) Department of Physics, Centre for Electronic Materials and Devices, Imperial College, London SW7 2BZ, UK

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We have studied recombination and relaxation dynamics in InAs quantum dots by means of photoluminescence cross-correlation techniques with sub-picosecond time-resolution. We have determined the relative radiative efficiencies for carrier recombination from different energy levels of the quantum dots and show that the radiative efficiency decreases with increasing transition energy, indicating that carrier access to non-radiative recombination centres increases for higher-energy states inside the dots. Cross-correlation measurements with both excitation beams of the same circular polarization and with the beams having opposite circular polarization are shown to give direct insight into the spin relaxation dynamics in the quantum dots and the wetting layer.

The recombination and relaxation dynamics of carriers in self-assembled quantum dots have been studied extensively in the past few years [1, 2]. More recently, attention has been focussed on the spin relaxation in these structures [3]. Here we report the results of time-resolved photoluminescence cross-correlation measurements on self-assembled InAs quantum dots. We show that the cross-correlation technique gives insight into radiative and non-radiative recombination, and also into spin relaxation in these quantum dot systems.

The quantum dots were grown by molecular-beam epitaxial deposition onto GaAs of two monolayers of InAs followed by a GaAs capping layer. The sample was then annealed for 10 s at 780 °C in an inert atmosphere, in order to increase the ground-state emission energy of the dots by ≈ 350 meV with respect to the unannealed sample. Details of the growth and annealing procedure can be found in Ref. [2]. Time-resolved cross-correlation experiments were performed by exciting the sample with two identical beams of a mode-locked Ti:sapphire laser delivering 120 fs pulses with a repetition rate of 76 MHz. The two beams, whose inter-pulse delay could be varied, were chopped at frequencies $f_1 = 310$ Hz and $f_2 = 6/5f_1$, respectively, and focussed onto the same 130 μm diameter spot on the sample, which was held at 8 K. The photoluminescence (PL) from the central region of the Gaussian spot was dispersed in a double-grating spectrometer and detected by a Si-photodiode and a lock-in amplifier. The time-integrated PL was detected by coherent demodulation at f_1, and the cross-correlation signal at $f_1 + f_2$ [4, 5]. Two different sets of experimental conditions were applied: in the first, the sample was excited in the GaAs barrier with an excitation energy of 1.55 eV, and the excitation beams were polarized co-linearly. In the second, excitations were created only in the wetting layer (at an energy of 1.46 eV) and both beams were set to have either equal (σ^+, σ^+) or opposite (σ^+, σ^-) circular polarization.

[1]) Corresponding author; Fax: +44-1223-353397; e-mail: lmh33@cam.ac.uk

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Figure 1 shows the time-integrated (TI) PL spectra for the quantum dot sample at various excitation intensities for the first set of experimental conditions. As the excitation intensity is increased, Pauli blocking leads to a saturation of the emission from lower-lying dot states [1], so that at the highest excitation intensity employed, PL originates from all quantum dot transitions and the wetting layer. The same figure also shows cross-correlation spectra taken at 2 ps delay. In general, the cross-correlation signal at an inter-pulse delay δ is given by the difference between the TIPL created by two excitation pulses separated by a delay δ, and the TIPL created by two excitation pulses separated by an infinite delay [6]. Since the cross-correlation technique relies on a non-linearity of the PL with excitation density [6] it is well suited for application to quantum dots, whose non-linear state-filling effects should provide a pronounced cross-correlation signal. At an excitation intensity of 8.8 I_0 (where $I_0 = 1.3$ W cm^{-2} per beam) state-filling effects are starting to appear in the TIPL spectra. The cross-correlation spectrum at the same excitation intensity is characterized by a negative signal at the ground state (1–1) emission energy of the quantum dots (1.394 eV) and a positive signal at the emission energy (1.414 eV) of the first excited dot state (2–2). This can be explained as follows: if the first excitation pulse fills the ground state of the quantum dots, carriers created by the second pulse can no longer recombine from the ground state but instead recombine from the first excited state. The presence of the carriers created by the first pulse thus leads to a negative correlation signal at (1–1) and a positive signal at (2–2). As the excitation intensity is increased, more quantum dot levels saturate leading to an increasingly negative cross-correlation signal in the spectral

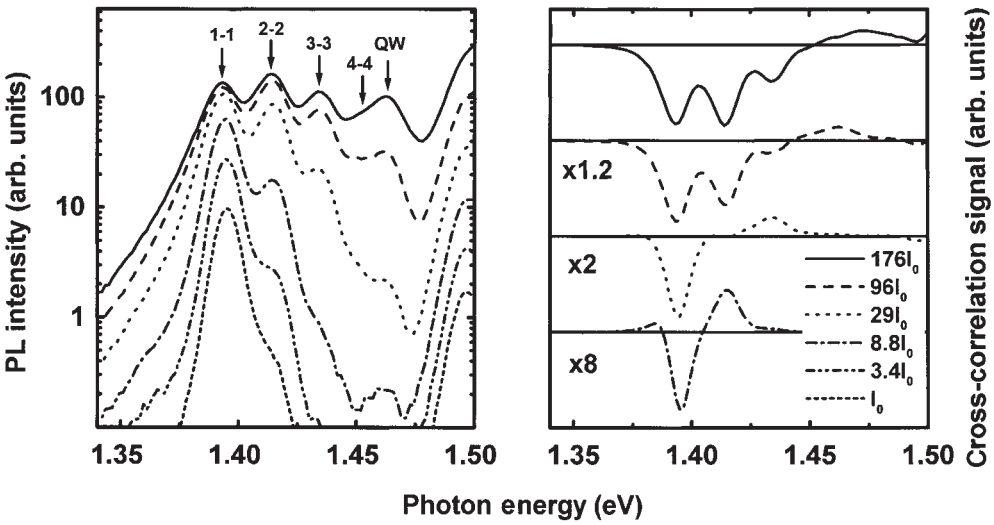

Fig. 1. Left: time-integrated photoluminescence spectra at 2 ps inter-pulse delay for various excitation intensities ($I_0 = 1.3$ W cm^{-2} per beam). Three quantum dot transitions are observed with peak energies of 1.394, 1.414 and 1.434 eV. The wetting layer transition (QW) can be found at 1.463 eV. The weak feature at 1.452 eV (labelled 4–4) may either be due to a fourth quantum dot transition or be part of the quantum well emission. Right: cross-correlation signal at 2 ps delay for various excitation intensities. The curves have been rescaled and offset for clarity; the additional axes indicate zero for each curve

region of the quantum dot emission. The important advantage of this technique is that unlike direct measurements of transient PL it gives insight into the non-radiative processes in the sample [6]. As varying the inter-pulse delay δ does not change the total number of carriers created in the barrier and captured into the wetting layer, the spectrally integrated cross-correlation signal should in principle be zero, since no carriers are gained or lost in the state-filling process. However, if the radiative efficiency for recombination from the excited states of the dots is lower than that from the ground state, the positive cross-correlation signal at the emission energy of the first excited state will no longer be able to balance the negative cross-correlation signal at the energy of the ground state, and the spectrally integrated signal will become negative. This is clearly the case for our sample: with increasing excitation intensity the spectral integral of the cross-correlation signal becomes increasingly negative, indicating that for the energetically higher-lying dot states and especially the wetting layer, non-radiative processes make a substantial contribution to the carrier recombination. We are able to estimate the relative radiative efficiencies η_i for recombination from the quantum dot states and the wetting layer from the amplitudes A_i of the cross-correlation signal for the i-th quantum dot transition peak and the QW emission peak, from

$$\frac{A_1}{\eta_1}+\frac{A_2}{\eta_2}+\frac{A_3}{\eta_3}+\frac{A_4}{\eta_4}+\frac{A_{\mathrm{QW}}}{\eta_{\mathrm{QW}}}=0\,. \tag{1}$$

To extract the cross-correlation amplitudes A_i from the spectra we have first determined the best fit to the TIPL spectra given by the sum of five Gaussians. We have then applied the same procedure to the cross-correlation spectra, with only the amplitudes A_i of the Gaussians as free parameters and the peak positions ϵ_i and widths w_i determined in the previous fit to the TIPL spectra as shown in Fig. 2. By repeating this for various inter-pulse delays and excitation intensities we were able to determine sets of parameters A_i and from the combination of the sets the relative radiative efficiencies to be $\eta_2/\eta_1 = 0.77$, $\eta_3/\eta_1 = 0.33$, $\eta_4/\eta_1 = 0.17$, and $\eta_{\mathrm{QW}}/\eta_1 = 0.08$. These results show that while the quantum efficiencies of the ground state and the first excited state of the dots are still similar, indicating that non-radiative recombination processes only play a minor role, these processes gain in importance for the energetically higher-lying states. This behaviour probably derives from the shallow confinement potential for these strongly annealed dots. The wetting

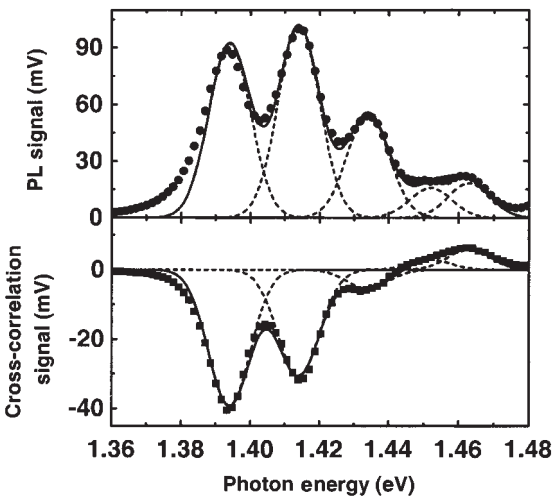

Fig. 2. Time-integrated photoluminescence spectrum (top) and cross-correlation spectrum (bottom) at an excitation intensity $I = 96I_0$ and 2 ps inter-pulse delay. The solid lines represent best fits to the data by the sums of five Gaussians

layer in this sample contains a relatively high number of non-radiative traps, as indicated by its low emission efficiency. Higher-energy dot states will increasingly have access to these centres since their wavefunction extends further into the wetting layer than that of lower states. As shown by Sercel [7] such traps may also provide an efficient means of inter-level relaxation in quantum dots. Our results may therefore partly explain the absence of any phonon-bottleneck for this sample [2].

In a second set of experiments, the excitation energy was set to be resonant with the wetting layer emission and the two beams were chosen to have either equal (σ^+, σ^+) or opposite (σ^+, σ^-) circular polarization. Since Pauli blocking depends strongly on the spin orientation of the excited carriers, the comparison between the two cases should give direct insight into the spin-relaxation in this system. This can be seen in Figs. 3A and B where the decay of the cross-correlation signal at the energy of the first excited dot transition is plotted for the two cases at two different excitation intensities. For the (σ^+, σ^+) excitation a stronger cross-correlation is observed than for (σ^+, σ^-) since spin-statistics enhance the state-filling effects for the former case. However, if the second excitation pulse arrives at a time when any spin-orientation of the carriers created by the first pulse has decayed, no difference should be observed between the two cases. The difference between the cross-correlation decay curves for the two excitation conditions therefore relates to the lifetime of the spin-orientation in the system, and this is plotted in Fig. 3C for two excitation intensities. We find that the difference signal decays with an initial fast (≈ 50 ps) component relatively independent of the excitation intensity, which we attribute to the electron spin relaxation in the wetting layer and through capture into and relaxation in between the quantum dot levels [3, 8]. The initial decay is followed by a longer decay whose lifetime increases from 380 ps at an excitation density $I = 40I_0$ to 1200 ps at $I = 121I_0$. This long-lived decay is most likely caused by a stacking of spin-oriented electrons in the quantum dots due to Pauli-block-

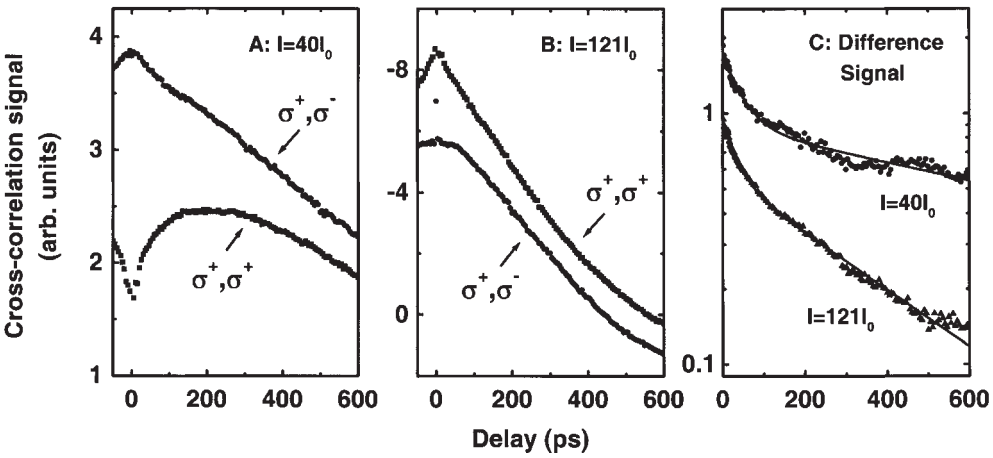

Fig. 3. A) and B) Cross-correlation delay curves taken at the peak of the first excited state transition (2–2) for excitation with (σ^+, σ^+) and (σ^+, σ^-), with excitation intensities A) $I = 52$ Wcm^{-2} and B) $I = 157$ Wcm^{-2} per beam. Note that the correlation signal in A) is smaller for the (σ^+, σ^+) than for (σ^+, σ^-) since the correlation signal changes from positive to negative with increasing excitation intensity (see B). C) Absolute value of the difference in the cross-correlation signal between the two cases, given for the excitation intensities in A) and B)

ing [9, 10]. However, as the excitation intensity is increased further, the relative magnitude of the difference signal decreases (not shown) because the available states for both spin orientations become increasingly filled.

In summary, we have shown that time-resolved cross-correlation spectroscopy is a sensitive tool to give an insight into radiative and non-radiative recombination and spin-relaxation in InAs quantum dots.

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