Slow relaxation of excited states in strain-induced quantum dots

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We have studied photoluminescence from GaAs/$Al_xGa_{1-x}As$ strain-induced quantum dots in a magnetic field. These dots have high radiative efficiency and long (~ns) luminescent decay times. At low excitation intensities, corresponding to average carrier densities of less than one electron-hole pair per dot, excited-state (''hot'') luminescence due to slow interstate relaxation is observed. At intermediate intensities, where there are several electron-hole pairs per dot, the hot luminescence disappears, showing that the relaxation rate has increased. However, the excited-state emission reemerges at high excitation when the ground state is saturated. The interstate relaxation rate in the quantum dots under low excitation is at least two orders smaller than that of the host quantum well. The reduced rate is attributed to the discrete density of states in a quantum dot, which inhibits single-phonon emission because the excitons are spatially too large to couple to phonons with the required energy. When there are several electron-hole pairs per dot, carrier-carrier interaction accelerates relaxation. The magnetic field is used to separate the quantum dot states and allows us to probe how their relaxation depends on energy. We find that there is a strong increase in the relaxation rate when the sublevel energy exceeds about 20 meV. [S0163-1829(96)00124-1]

I. INTRODUCTION

Quantum dots are ''zero-dimensional'' physical systems in which particles are confined in all three dimensions so that their density of states is discrete, the separation between the states (''sublevels'') being determined by the confinement potential. Recent theoretical work suggests that in semiconductor quantum dots relaxation of excited states by phonon emission should be much slower than in nonzero-dimensional systems, which have a continuous density of states.[1–4] The reason is that phonons with energies matching the sublevel separations have wave vectors that are too large to couple to the confined states. This means that at low carrier density relaxation must depend on weak multiphonon processes. However, this theory is still the subject of considerable debate and other mechanisms for efficient relaxation have been proposed.[5–8]

Most of the theory developed thus far has been devoted to free-carrier relaxation in quantum dots and does not include excitonic effects. Bockelmann has pointed out that for undoped systems, these calculations assume high carrier densities where the Coulomb interaction is effectively screened.[3] However, in this regime Auger-like scattering among carriers should facilitate efficient cooling.[6] At low carrier densities, the excitonic character of the states must be considered. While excitonic levels are increasingly dense above the exciton binding energy, they are sparse below it.[3] Thus low-lying excitonic states are expected to cool slowly. On the other hand, Efros, Kharchenko, and Rosen[8] have recently suggested that Auger-type relaxation could also be important within a single exciton, with the slowly relaxing electron giving its energy to the quickly cooling hole. We find no experimental evidence for this intraexcitonic process.

As with the theoretical predictions, the experimental results are diverse. Many groups have observed high-energy structure in photoluminescence spectra from quantum dots at low temperatures and attributed it to hot luminescence from excited states due to slow interstate relaxation.[9–12] However, this assignment has not been verified conclusively, since such high-energy structure, more intense than expected from the Boltzmann factor, can arise from many causes. For example, it could originate in a different region of the sample from the lower-energy luminescence: only Ref. 10 ruled out this possibility by using photoluminescence excitation to confirm that the high-energy states do indeed relax to the low-energy states. It may come from saturation of the lower states at high excitation intensity, a process discussed below: again, only Ref. 10 used a sufficiently wide range of excitation intensity to rule this out. Hot luminescence can occur even if interstate relaxation is fast, if nonradiative decay of the excitons is also fast. There is evidence, which we will discuss later, that this may be the case in the work of Ref. 10.

These problems have arisen primarily because, as pointed out in Ref. 9, the usual techniques for fabricating quantum dots (ion implantation and direct etching) damage the semiconductor and produce a large number of nonradiative cen-

ters. These methods are used to impose a lateral potential profile in the plane of an ordinary semiconductor quantum well. An alternative way of modulating the band gap in the plane of a quantum well is by patterning microscopic arrays of a compressively stressed material (stressors) on the surface of the structure.[13,14] These stressors stretch the lattice beneath them, creating local regions of low energy in a near-surface single quantum well. The great advantage of this technique is that the original quantum well retains its structural integrity so that nonradiative losses are low and the recombination remains predominantly radiative up to temperatures above 50 K.[15] This means that a measurable signal can be generated by low excitation, and that the decay time is relatively long (~ns, compared with ~0.1 ns in the best previously reported material[12]) so that the presence of hot luminescence implies thermalization on this time scale.

In this paper we report magnetoluminescence measurements on strain-induced quantum dots. The magnetic field sharpens and separates individual peaks and allows us to vary the interstate separations, so that the energy dependence of the relaxation rate can be determined. We show that the thermalization of electron-hole pairs in quantum dots depends strongly on the number of carriers in each dot. If the mechanism proposed by Efros, Kharchenko, and Rosen,[8] is weak and there is less than one electron-hole pair per dot, the exciton can only cool via phonon emission. Single-phonon emission is slow for the low-lying excitonic states in a quantum dot because the states are widely separated, and phonons with matching energy have a momentum that is too large to couple to the confined states. However, the states become increasingly dense at higher energy, so that high-energy excitons relax more quickly.[3] When there are several electron-hole pairs per dot, Auger-like carrier-carrier scattering leads to increasingly rapid cooling, even between low-lying states. Our observations are consistent with Bockelmann's predictions for the carrier density dependence of energy relaxation processes in quantum dots.

II. EXPERIMENT

The photoluminescence measurements described here were made on a sample consisting of a 15-nm-wide $GaAs/Al_{0.3}Ga_{0.7}As$ quantum well and separate arrays of nominally 200-, 300-, and 400-nm-wide square tungsten stressors. Details regarding the fabrication process can be found elsewhere.[14] Although these stressor sizes are large relative to the exciton Bohr radius (which is approximately 14 nm in bulk GaAs), the nearly parabolic potential due to the strain yields confined electron states that are typically 10–20 nm in extent at the bottom of the well.[15] The lateral confinement potential is 35–45 meV deep and electron sublevel splittings of approximately 2 meV are predicted.

The structures were excited in backscattering geometry with a 1.96-eV HeNe laser for cw measurements and either a 2.0-eV mode-locked dye laser or a 1.85-eV laser diode for transient measurements. The laser intensity was varied between 1 μW and 1 mW and focused to a spot size of approximately $(20\ \mu m)^2$. The spacing between dots is twice the stressor size and typically 400 dots lie within the excitation spot. The photoluminescence signal is dispersed in a 1-m single grating spectrometer and detected by a cooled GaAs photomultiplier tube via standard photon counting techniques. In the high excitation intensity transient measurements, the response time of the system was 300-ps full width at half maximum (FWHM) and the repetition rate was 81 MHz. At low excitation a very long decay component was detected, necessitating the use of a 2-ns FWHM pulsed laser diode where the repetition rate could be reduced to 4 MHz. All measurements were made at 30 K because excitation transfer from the interstitial quantum well increases the signal at this temperature. Magnetic fields are applied along the growth direction of the crystal. Since the energy level separations in which we are interested are at least 8 meV (since levels closer than this are not resolved), the Boltzmann factor in the excited states is always less than 0.05 and is not usually significant.

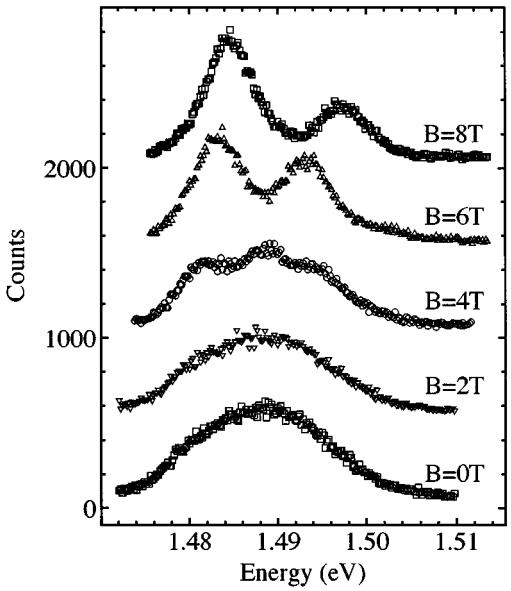

FIG. 1. Magnetic field dependence of the 300-nm dot photoluminescence spectrum at 30 K. The excitation intensity was 0.5 W/cm^2.

III. RESULTS

The magnetic field dependence of the photoluminescence spectrum for an array of 300-nm dots is shown in Fig. 1. The excitation intensity is 0.5 W/cm^2, which corresponds to an average carrier density of approximately one electron-hole pair per dot. At low fields, the emission from the inhomogeneously broadened confined sublevels overlaps and the spectrum consists of a single broad luminescence band. However, this band splits at high fields into a weakly field-dependent ground state and excited states which resemble Landau levels and shift linearly with field. The integrated intensity of the excited-state emission decreases relative to the ground state as the magnetic field is raised. There is no evidence for population of the $n=2$ quantum well state, which is about 40 meV above the $n=1$ state, or of the light hole $n=1$ state, which may not be confined in these structures.[15]

In Fig. 2 the peak positions of the resolved states are plotted as a function of applied field. The vertical bars indicate the full width at half height of the luminescence peaks. The dotted lines are a theoretical fit to the data. The model

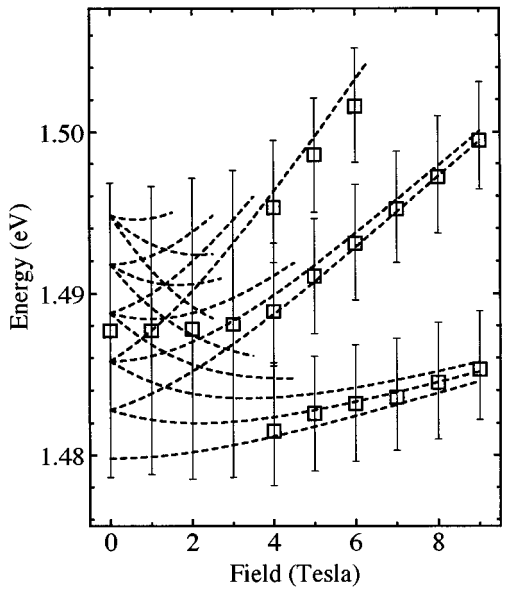

FIG. 2. Photoluminescence peak positions as a function of magnetic field. The solid vertical lines are FWHM linewidths and the dotted lines are a fit to the data as described in the text.

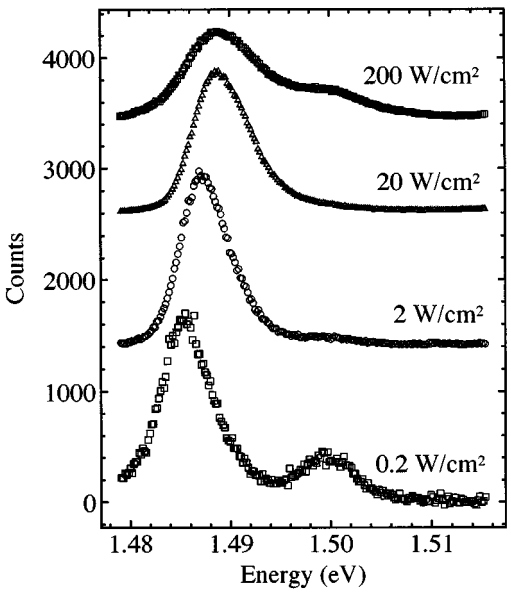

FIG. 3. Excitation intensity dependence of the 300-nm dot spectrum at 9 T, normalized by the excitation power. The value 0.5 W/cm^2 corresponds to approximately 1 exciton per dot.

assumes free-carrier states in a two-dimensional parabolic potential and a magnetic field, and can be solved analytically.[16] At zero field, the sublevels due to the potential are evenly spaced with a degeneracy that increases linearly with energy. When a magnetic field is applied, these sublevels split into orbitally nondegenerate states of different angular momentum. As the field increases these states cross, and converge to Landau levels at high field. Even though the model neglects some important terms in the Hamiltonian, in particular the Coulomb interaction between electrons and holes, the fit is quite good. We use the effective mass of electrons in bulk GaAs ($0.07m_e$), so that the only fitting parameter is the quantum dot sublevel splitting, which is chosen to be 3 meV. Actually, any value less than 3 meV would fit the data. However, the intensity dependence at high field described below, which depends on the splitting of the sublevels within the Landau levels, suggests that the zero-field sublevel splitting is at least 2.5 meV. These numbers are reasonably consistent with the calculated splitting for electrons of 2 meV.[14]

The intensity dependence of the 300-nm dot photoluminescence at 9 T is shown in Fig. 3. The initial disappearance and eventual reemergence of the high-energy peak with increasing excitation is striking. It shows that population of this state at low intensity is nonthermal. We attribute the peculiar intensity dependence to changes in interstate relaxation with carrier density. In particular, the relaxation rate of the excited state is slow in the low-intensity regime, but rapidly increases when we begin to put more than one electron-hole pair in each dot. At higher intensity still the ground state becomes filled and excited-state luminescence reappears. This interpretation is supported by the blueshift of the ground-state peak at intermediate intensities, where electron-hole pairs are sequentially filling the ladder of closely spaced sublevels within the Landau level, whose degeneracy is proportional to magnetic field.[17] This degeneracy is lifted by the parabolic potential, as indicated by the dotted lines in Fig. 2. Since the ground level has finite degeneracy, it saturates at high excitation, leading to excited-state luminescence even if relaxation is fast.

The field dependence of the energy of the first excited state in the high-intensity regime is similar to that of the low-intensity regime, but the slope of the energy vs field is smaller. To fit these data with the same procedure described above requires an effective mass of $0.09m_e$, an enhancement of 30%. This increase in the effective mass at high carrier density is similar to what has been observed in ordinary quantum wells, and is a well-known and understood phenomenon.[18]

We have confirmed that the high-energy emission comes from states closely associated with the states responsible for the low-energy emission in two ways. We find that the field-dependent splitting of the photoluminescence peaks is independent of dot size, while the strain-induced lattice dilation and hence the energy of the ground-state luminescence varies considerably. Furthermore, as the excitation intensity is increased and the ground level saturates, the integrated emission intensity remains constant. These results show that the high- and low-energy luminescence come from the same part of the sample.

We have also investigated arrays of 200- and 300-nm-wide quantum wires, where the particles are free to move in one direction and so have a continuous density of states. Even though the fabrication procedure and the magnitude of the strain for the wires are essentially identical to those of the dots, only a single narrow luminescence peak from the ground-state exciton is present at low excitation, even at high field. This result confirms the previous observation of rapid relaxation in strain-induced wires[19] and shows that the hot luminescence from excited states is a feature unique to quantum dots, where the density of states is discrete.

In order to gain insight into the relaxation process, we have measured the transient response and time-resolved spectra from the quantum dots after excitation with a short

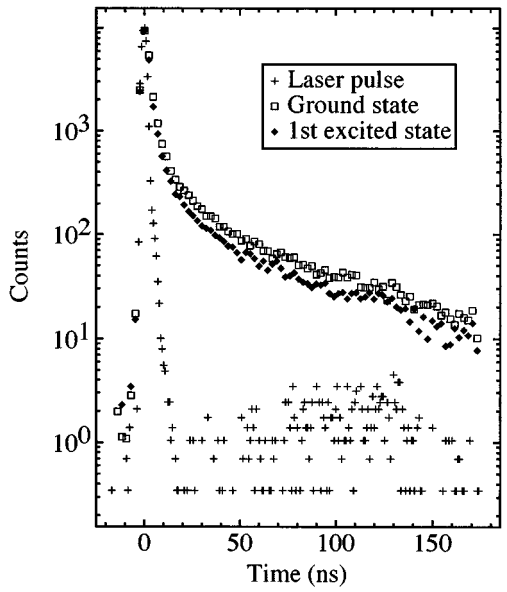

FIG. 4. Time dependence of the 200-nm dot luminescence after pulsed excitation at 7 T. The laser intensity was 50 nJ/cm^2 per pulse with a repetition rate of 4 MHz.

laser pulse. The transient response is intensity dependent and the decay is nonexponential, with an average radiative lifetime of approximately 2 ns. At high intensity, where occupation of the excited levels is due to saturation of the excitonic ground state, the spectra evolve rapidly in time. After a few nanoseconds, only ground-state emission is present in the spectrum. However, the transient behavior under relatively low excitation is very different (see Figs. 4 and 5). First of all, a very slow process with a decay time of almost 70 ns is observed in this regime. It is associated with slow transfer from the interstitial quantum well via a long-lived

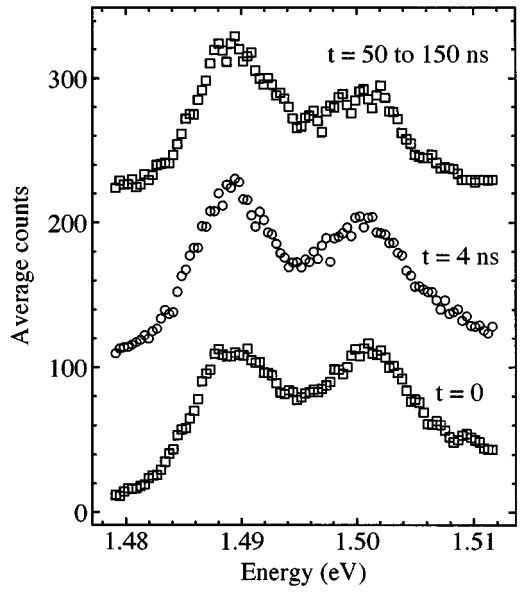

FIG. 5. Time-resolved photoluminescence from the 200-nm dots under the same experimental conditions as those given in Fig. 4. The average cound number is scasled so that the three spectra can be compared.

nonradiative intermediate state, probably analogous to the state found in strain-induced dots by Gu *et al.*[20] Secondly, we find that the shape of the photoluminescence spectrum is nearly independent of time, even if we wait 100 ns after pulsed excitation.

IV. DISCUSSION

The hot luminescence and the transient behavior at low carrier densities show that for the first excited state the energy relaxation time and radiative recombination time are comparable. The recombination time is approximately 2 ns, while the relaxation time depends on the energy level separation as discussed below. Following pulsed excitation, the ground-state population rises initially as the system reaches steady state. After this condition is met, the spectrum remains unchanged in shape while the overall intensity decays. Since the long decay component is due to transfer from the inter-dot quantum well region, it should be viewed as a source that slowly feeds excitons into the dots. Hence the presence of hot luminescence in the spectrum at very late times does not imply that the cooling time is this long, but merely that it is of the same order as the radiative recombination time.

Since the observation of hot luminescence is inconsistent with the mechanism proposed by Efros, Kharchenko, and Rosen,[8] we assume that the dominant relaxation mechanism is single-phonon emission.[3] For this process, conservation of energy requires that the emitted phonon energy $\hbar c_s q$ match the energy difference between the initial and final confined states. Here, c_s is the speed of sound and q is the phonon wave vector. When the splitting exceeds the energy $\hbar c_s/a$, where a is the smallest length scale of the exciton wave function, the exciton-phonon matrix element decreases rapidly.[3] At zero magnetic field, excitons in our quantum well are smallest along the growth axis of the crystal, where they have an effective radius of approximately 7 nm. Assuming a bulk GaAs-like linear phonon dispersion with a longitudinal sound velocity $c_s = 3700$ m/s, the corresponding threshold energy is 0.3 meV for LA phonons, and even less for TA phonons. Hence the estimated sublevel splitting of 3 meV is well within the slow relaxation regime. Two-phonon processes, in which large q phonons can take part, will normally be much weaker, unless the energy splitting is so large that zone-boundary phonons with their large density of states can take part (we will return to this point).

Bockelmann has calculated the spectrum of excitonic states in cylindrically symmetric parabolic quantum dots.[3] He finds that while level separations are large near the bottom of the well, the states become increasingly dense at high energies. This phenomenon is most easily understood by rewriting the electron-hole pair Hamiltonian in center-of-mass and relative coordinates. The low-lying states can then be seen as $1s$ excitons quantized by the center-of-mass part of the Hamiltonian. At high energies, particularly above the exciton binding energy, the relative Hamiltonian becomes more important and generates a dense distribution of states, most of which have nonzero angular momentum and hence are nonradiative.

In this picture we would expect quantum dot excitons to cool efficiently in the high-energy region where the states are dense and slowly in the low-energy region where levels are well separated. When the magnetic field is increased, we

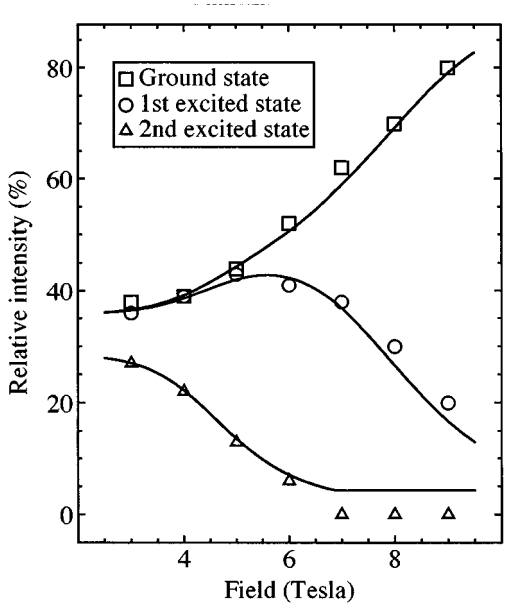

FIG. 6. Relative integrated intensity of resolved peaks in the 300-nm dot low excitation spectra as a function of field. The solid lines are a fit to the data as described in the text.

expect to see an enhancement in the relaxation rate of low-lying radiative sublevels as they increase in energy and move into the high-density region. This field dependence, which is very different from what has been reported for ordinary quantum wells,[21] was recently predicted by Bockelmann[22] and is what we observe. Figure 6 shows the relative integrated intensities of the three resolved states in the 300-nm dot spectra as a function of magnetic field. With increasing magnetic field, the intensity of the excited states weakens considerably relative to the ground state.

We have made this interpretation more quantitative by fitting the field dependence of the relative intensities to a simple four-level model. We assume that all three radiative exciton states have the same recombination rate ω_r which is independent of field.[23] We also assume that all excitation cools rapidly to the highest of the three radiative levels and that further relaxation only occurs between adjacent levels. The relaxation between levels has two components: a slow energy-independent part ω_s and a fast component ω_f that turns on as the level crosses a threshold energy E_0. The slow component determines the intensity distribution at low fields. The fast component models the effect of level density on relaxation. Since the changeover from low to high density is not sudden, we assume that the threshold energy E_0 has a Gaussian broadening ΔE. Thus the total relaxation rate is given by $\omega_p(E)=\omega_s+f(E)\omega_f$, where

$$f(E)=\begin{cases}\displaystyle\int_0^E e^{-(E'-E_0)^2/2\Delta E^2}dE' & \text{for } E<E_0\\ 1 & \text{for } E\geq E_0.\end{cases}$$

The solid lines in Fig. 6 represent such a fit, where we have again used $0.07m_e$ and 3 meV for the effective mass and zero-field sublevel spitting. The low-field data require slow initial cooling rates of $1\omega_r$ and $2.5\omega_r$ for the first and second excited states, respectively. The fast cooling rate that becomes active when the level energy exceeds E_0 is $20\omega_r$. The threshold energy E_0 is found to be approximately 20 meV above the centroid of the broad low-field photoluminescence band, with a Gaussian broadening ΔE of 5 meV. While this simple model ignores many important effects including the complex changes in the exciton-phonon matrix element with the changing size of the magnetoexcitonic states, it reproduces the data remarkably well.

Interpretation of the threshold energy E_0 is not unambiguous. Bockelmann's theory suggests that E_0 should be close to the exciton binding energy, where the onset of excitonic p states leads to an increasingly dense distribution of states.[22] However, our measured threshold is well above the exciton binding energy (approximately 8 meV for a 15-nm quantum well, increasing with lateral confinement and magnetic field).[24] On the other hand, while E_0 is much less than the optical phonon energies of GaAs, it is in a region where the two-phonon density of states for acoustic phonons is increasing rapidly.[25] Thus the increased relaxation rate may be due to the onset of two-phonon relaxation processes, rather than to the increase in exciton density of states. While this interpretation is appealing, there are problems with it. In particular, it fails to account for the observed increase in relaxation from the second to the first excited state when the separation of the former from the *ground* state reaches E_0.

We now turn to the excitation intensity dependence of the interstate relaxation. Contrary to the quantum well case,[26] quantum dot excitations cool more quickly at high carrier density. In order to elucidate the mechanism for this phenomenon, we analyze the data in the context of another simple model. Once again, we assume that the relaxation of excited states has two components. Here, the slow, intensity-independent part describes the relaxation of single excitons in quantum dots via phonon emission, i.e., the rate ω_p above. The faster, intensity-dependent component models the relaxation when more than one electron-hole pair is present in the same dot. Since this situation corresponds to effective local carrier densities larger than 10^{17} cm^{-3}, we might expect the relaxation to be dominated by Auger-like scattering processes among carriers. The calculated relaxation rate of carriers in quantum dots by Auger scattering initially increases linearly with density,[6] but it should be noted that the physical picture for this calculation is somewhat different from the one we are using.

Nevertheless, we follow Ref. 6 and assume that the total relaxation rate is given by $\omega_T(I)=\omega_p+I\omega_A$ where I is the excitation intensity. An example of the fit is given in Fig. 7 with $\omega_p=3\omega_r$ and $\omega_A=2\omega_r$. The success of the fit indicates that our assumption of a linear relationship between the Auger scattering rate and carrier density is reasonable. A stronger carrier density dependence will not fit the data. The model does not take into account the discrete nature of the problem, which is expected to be particularly important for the range of excitation intensities that we are using here. For example, the Auger scattering process that we are considering requires at least two excited electron-hole pairs in the same dot, but the average carrier density in each dot is less than one electron-hole pair under low excitation. Hence, even if we use a Poisson distribution for the number of carriers in each dot, Auger scattering should be weak in this regime.

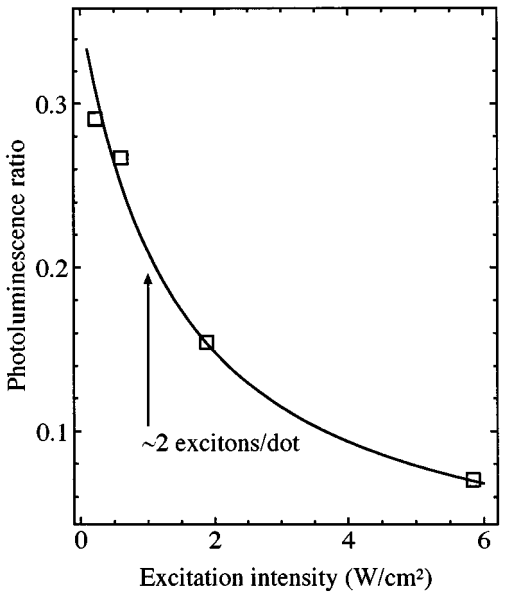

FIG. 7. Excitation intensity dependence of the ratio of integrated emission from the first excited and ground states in the 300-nm dots at 9 T. The solid line is a fit to the data as described in the text.

We estimate that 1 W/cm^2 corresponds to an average density of two electron-hole pairs per dot. This estimate is based on absorption coefficients at our exciting wavelength of 4×10^4 cm^{-1} for GaAs and 2×10^4 cm^{-1} for the $Al_{0.3}Ga_{0.7}As$ barriers, where the $Al_{0.3}Ga_{0.7}As$ absorption is approximated by shifting the GaAs spectrum.[27] We assume complete excitation transfer from the barriers to the quantum well and from the interdot region of the well to the dots.[15] For comparison with the fitted data, this density is indicated in Fig. 7. The lack of agreement between experiment and theory below this intensity can be attributed to the discrete threshold for Auger scattering described above. Finally, although we have not made a careful study of the effect, we note that the Auger scattering rate increases with field.

V. CONCLUSION

Our main conclusion is that our observation of hot luminescence from excited quantum dot states under low excitation intensity confirms the prediction of Bockelmann and Egeler[6] of slow energy relaxation between these levels. For the relative population of excited and ground states that we have observed in this work, ordinary quantum wells have cooling rates on the order of 10^{10} eV/s.[26] This rate would give relaxation times on the order of 1 ps for our level separations at high field. Similarly, relaxation is found to be fast relative to recombination in quantum wires.[19] In contrast, in dots we find cooling times on the order of 1 ns, which means that the quantum dot relaxation is three orders of magnitude slower. While the application of a magnetic field lowers the relaxation rate in quantum wells, Ryan *et al.* find that even at 8 T carriers cool to an effective temperature of 100 K in approximately 50 ps,[28] and their measurements were made at a high carrier density where quantum well carrier relaxation is relatively slow because of screening. We conclude that relaxation rates in our quantum dots are at least two orders slower than the host quantum well.

Identification of the hot luminescence as emission from the dots, as opposed to another radiating center in the sample, is unambiguously verified by its dependence on magnetic field and stressor size. The slow relaxation rate is attributed to the discrete density of states in these structures. This idea is supported by the observed absence of slow relaxation in strain-induced quantum wires, where the structures are very similar but the density of states is continuous. At first glance, the inhomogeneous broadening of the quantum dot sublevels (the unpatterned quantum well itself has a FWHM of about 5 meV, presumably due to well-width fluctuations) might suggest that the density of states is continuous. However, our results indicate that sublevels within individual dots are in fact discrete and reasonably well described by parabolic lateral confinement. While a three-dimensional confinement potential always produces discrete states regardless of how rough the walls are, the consistency of our data with Bockelmann's calculations suggests that individual dots are reasonably smooth and well behaved, and that the inhomogeneous broadening arises primarily from dot to dot fluctuation.

We have also seen that the hot luminescence weakens with magnetic field and disappears with increasing excitation intensity. To explain these effects we propose a model, based on the theory of Bockelmann and Egeler,[3,6] that incorporates two relaxation mechanisms which dominate at different carrier densities. When there is less than one exciton per dot, excitons cool primarily via phonon emission. Since this mechanism depends on the sublevel density and the phonon density of states, both of which increase at high energies, the relaxation rate becomes more efficient as the confined levels shift into the high-energy regime with field. When the excitation intensity is increased so that there are several electron-hole pairs per dot, the local carrier density is high and carrier-carrier interaction is strong. Intersublevel Auger scattering between carriers then accelerates relaxation and electron-hole pairs cool more quickly. Our results in this density regime disagree with those of Brunner *et al.*,[10] who found little change in the relative strength of the hot luminescence when there are several pairs in the dot. This discrepancy may be due to rapid nonradiative decay in their samples. Our data are consistent with the theory of Bockelmann and Egeler,[3,6] and are inconsistent with the mechanism proposed by Efros, Kharchenko, and Rosen,[8] which predicts rapid relaxation at all densities. As the magnetic field is increased, the interstate relaxation rate increases rapidly when the upper level reaches a threshold energy of about 20 meV above the ground state. While qualitatively consistent with Bockelmann's prediction, this increase can possibly be attributed to two-phonon decay.

While our results were obtained on dots in which the confining potential for electrons is parabolic, they should in principle apply to dots with square well confinement. However, in practice such dots tend to have very short luminescence decay times because of the method of fabrication, and only in samples of exceptionally high quality will the relatively slow thermalization processes discussed here be observable.

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