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Dynamics of exciton relaxation in $GaAs/Al_xGa_{1-x}As$ quantum wells

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We present an experimental study of the photoluminescence (PL) rise time of a single $GaAs/Al_xGa_{1-x}As$ quantum well. The PL rise time is investigated as a function of the excitation energy for different temperatures at low excitation densities ($\approx 10^{-8}$ cm^{-2}). A significant slowing down of the relaxation process is observed at low temperature when exciting the quantum well at the light-hole exciton energy; the slowing down disappears as either the temperature or the excitation power is increased.

The kinetics of exciton formation and relaxation in $GaAs/Al_xGa_{1-x}As$ quantum-well structures (QW) has recently received increasing attention, given the fundamental role that excitonic effects play in the optical properties of these structures even at room temperature.

In particular, the photoluminescence (PL) rise time at the heavy-hole exciton energy after picosecond or femtosecond excitation has been used in order to obtain information on the time required for exciton formation, on one hand, and for exciton cooling to center-of-mass kinetic energies within the homogeneous linewidth, on the other. We recall that, because of momentum conservation, only excitons within the homogeneous linewidth originated by inelastic scattering with acoustic phonons can directly couple to photons.[1]

Claims have been reported in recent papers that the PL signal shows a significant rise time for near-resonant photoexcitation of quantum-well structures. Different conclusions have been drawn; on the one hand, a long time for exciton formation has been claimed,[2] on the other, a slow exciton cooling has been invoked.[3,4] We believe that the question deserves clarification given the relevance of exciton relaxation in the optical properties of these structures, even if extrinsic effects may be, very likely, responsible for the behavior observed in Ref. 2. In fact, localization of excitons at crystal defects can modify the thermalization dynamics of the photogenerated carriers, giving rise to long photoluminescence rise times, depending on the localization kinetics.

We present, in this paper, time-resolved PL rise-time measurements in high-quality $GaAs/Al_{0.3}Ga_{0.7}As$ single quantum wells, as a function of photon energy, excitation intensity, and temperature.

As long as the excitation density n_{ex} is larger than approximately 10^9 cm^{-2} we find a slow increase of the PL rise time with increasing excitation photon energy in fair agreement with recent results from other authors.[3,4]

Moreover, a detailed measurement at 4 K of the spectral dependence of the rise time at low excitation ($n_{ex} \leq 10^9$ cm^{-2}) shows a strong increase of the PL rise time when the excitation photon energy equals the light-hole exciton energy. On the other hand, we find that the peak observed in the PL rise time for resonant excitation at the light-hole exciton is washed out when the sample temperature is increased to a few tens of degrees kelvin and/or the excitation density exceeds $(2-3)\times 10^9$ cm^{-2}.

The sample investigated consists of several single QWs of different thickness in the range 30–120 Å, grown in the same structure on a semi-insulating GaAs substrate with (001) orientation. The QWs are separated from each other by 500-Å-thick $Al_{0.3}Ga_{0.7}As$ barriers. We will only report here the experimental results for the 90-Å well; a similar phenomenology has been observed in the other QWs as well.

The PL dynamics was investigated using for excitation a Nd:YAG synchronously pumped dye laser (YAG is yttrium aluminum garnet), providing 5-ps pulses in the range 710–800 nm at a repetition rate of 76 MHz. The PL was dispersed through a 0.22-m double monochromator (energy resolution 1 meV) and detected by a synchroscan streak camera with an overall time resolution of 20 ps. A helium cryostat was used for varying the sample temperature between 4 and 50 K.

A comparison of the CW photoluminescence and photoluminescence excitation (PLE) spectra indicates, as shown in Fig. 1(a) for the 90-Å QW, that even at 4 K no significant Stokes shift is observed between the peaks corresponding to heavy-hole exciton recombination; exciton localization at crystal defects seems, therefore, to be absent in our sample.

Moreover, the sharp PL peaks measured (about 1 meV at 4 K for the 90-Å well at an excitation intensity of the order of 1 W cm^{-2}) clearly suggest smooth interfaces.

As a further check of the sample quality, we find a nearly linear dependence of the PL decay time τ_D as a function of temperature when T is raised from 4 to 50 K. As discussed in detail in Ref. 5, the whole set of observations, together with the observed approximate constancy of the PL integrated intensity in the same temperature interval, seems to indicate that free exciton recombination dominates the radiative processes in this sample and that extrinsic effects play a negligible role.

In the low excitation regime ($n_{ex} \leq 10^9$ cm^{-2}), under resonant excitation of the heavy-hole exciton, the PL de-

cay results in a fast monoexponential with a typical decay time $\tau_D = 120$ ps for the 90-Å well at 4 K [Fig. 1(b)]; a negligible rise time τ_R is observed within the experimental time resolution. Therefore no evidence of slow exciton formation is experimentally found, at least at low temperature, in agreement with Ref. 3. We shall then assume that, as a consequency of our limited time resolution, all the results presented have to be attributed to the exciton relaxation dynamics, and that the exciton formation time is within the instrumental time resolution.

The time evolution of the PL intensity at the heavy exciton peak (1.554 eV for the 90-Å well) is reported in Fig. 2 as a function of the excitation photon energy at 20 K, for an excitation density $n_{ex} \approx 10^8$ cm^{-2}. We have extracted the PL decay time τ_D and rise time τ_R at the heavy-hole exciton peak E_{HH} as a function of the excitation excess energy $\Delta E = E_{excl} - E_{HH}$ by fitting the experimental decay curves with a simple phenomenological three-level model based on single time constants for both the rise and the decay of the PL intensity. Note that, as expected, no dependence of the decay constant τ_D has been found when changing the excitation excess energy.

A fast rise time of the luminescence ($\tau_R \approx 40$ ps) is observed for near resonant excitation [Fig. 2(a); $\Delta E = 6$ meV]; moreover, a strong increase of the PL rise time is observed ($\tau_R = 200$ ps) when the excitation approaches the light-hole exciton energy [Fig. 2(b), $\Delta E = 11$ meV].

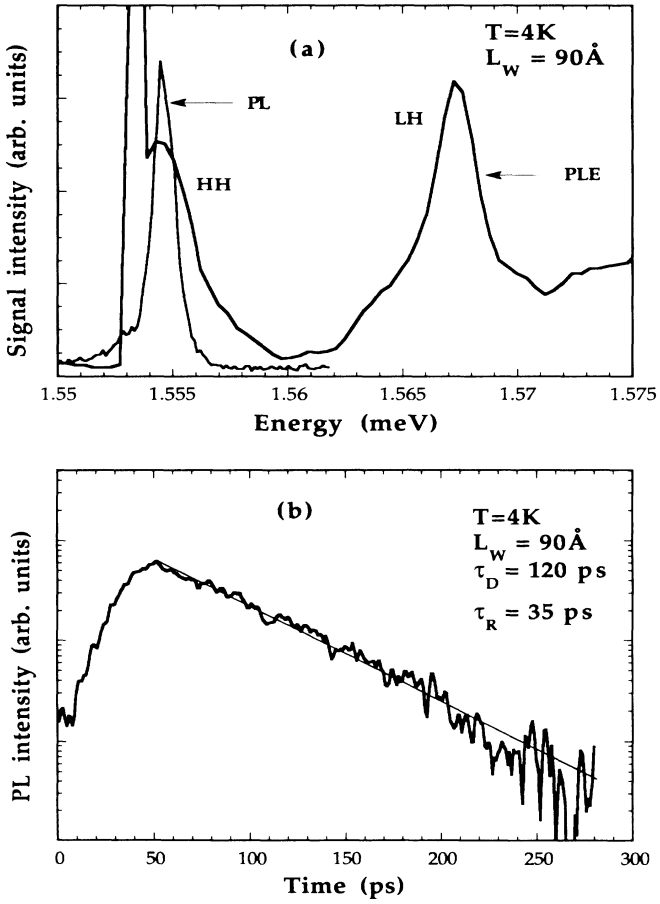

FIG. 1 (a) Comparison between the PL and PLE spectra of the 90-Å well at $T = 4$ K. Note the absence of a sizable Stokes shift between the heavy-hole exciton peaks. (b) PL time decay at the heavy-hole exciton energy E_{HH} of the 90-Å well, at 4 K. The values of the rise time and decay time, τ_R and τ_D, respectively, are the best-fit values within the model discussed in the text.

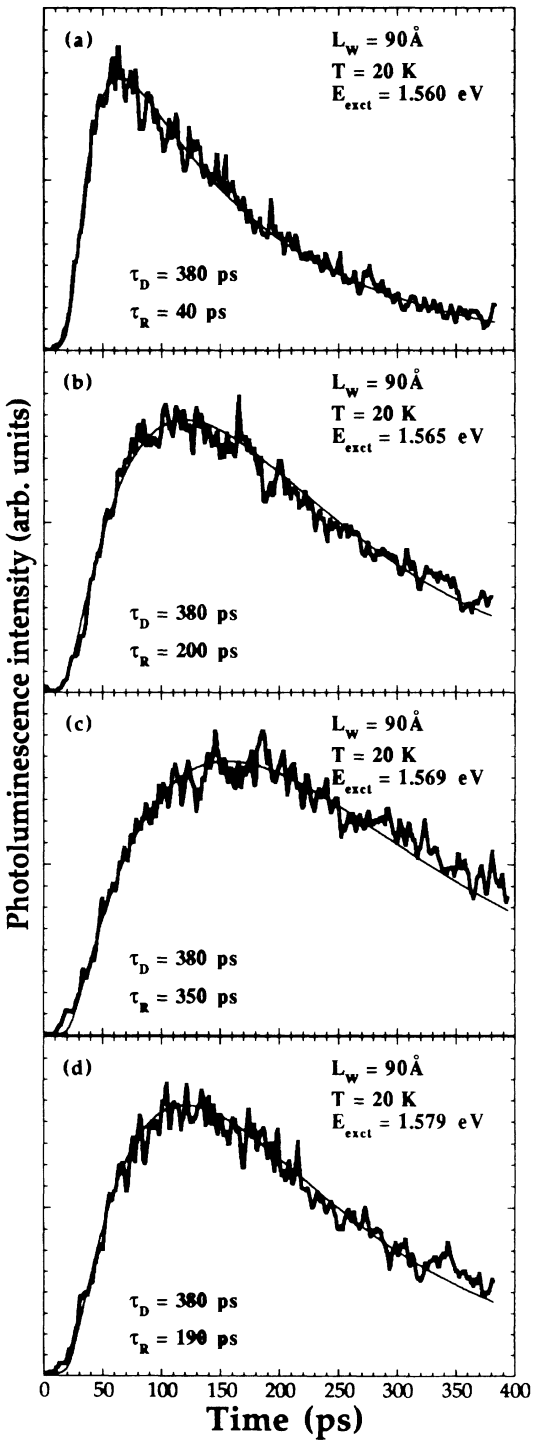

FIG. 2. PL time decay at E_{HH} for different excitation energies E_{excl}: (a) 1.560, (b) 1.565, (c) 1.569, and (d) 1.579 eV. The solid lines are the fits to the experimental curves obtained for the values of τ_D and τ_R reported in each case.

Finally, after reaching a maximum value ($\tau_R = 350$ ps) at the light-hole exciton energy [Fig. 2(c), $\Delta E = E_{LH} - E_{HH}$), τ_R starts decreasing ($\tau_R = 190$ ps) for a further increase of the excitation photon energy [Fig. 2(d), $\Delta E = 25$ meV].

It should be noted that all the measurements have been performed at constant PL peak intensity in order to keep a constant exciton density, under the assumption of an internal efficiency equal to 1.

The dependence of the PL rise time at the heavy-hole exciton peak of the 90-Å well as a function of ΔE is reported in Fig. 3 at three different temperatures, namely, 4, 20, and 50 K, for an excitation density $n_{ex} = 10^8$ cm^{-2}.

A slow continuous increase in the PL time with increasing excitation photon energy is observed at all temperatures; at the same time, for a given ΔE, a decrease of τ_R is found when increasing the temperature.

The main feature in Fig. 3 is the sharp peak in τ_R observed at 4 K when the excess energy is equal to the light-heavy exciton energy splitting. A smoothing of this peak for increasing temperature is clearly observed.

The strong increase in τ_R at $\Delta E = E_{LH} - E_{HH}$ does not seem to agree with similar measurements recently reported by Damen *et al.*;[3] in fact, the authors conclude that the time evolution of exciton luminescence is independent of the excitation energy up to 100 meV above the heavy-hole exciton energy. Similar findings are also reported in Ref. 6. On the other hand, an increase of the PL rise time at the light-hole exciton energy, for decreasing excitation density, has already been reported by Eccleston *et al.*,[4] but no detailed analysis of the spectral and temperature dependence of τ_R is given.

We believe that the origin of the discrepancy between our results and those in Refs. 3 and 6 lies in the fact that, as already indicated in Ref. 4, the time evolution of the exciton luminescence is significantly affected by the excitation intensity. We report in Fig. 4 the PL rise time at the heavy-hole exciton peak for the 90-Å well at 4 K as a function of the excitation density for three different excitation energies: $E_{exct} = E_{LH}$, $E_{LH} + 10$ meV, $E_{LH} - 6$ meV. We see that for excitation densities in the range $2 \times 10^9 < n_{ex} \leq 10^{11}$ cm^{-2}, as reported in Ref. 3, no dependence of τ_R on n_{ex} is observed; on the other hand, as n_{ex} is decreased below 2×10^9 cm^{-2}, while no significant variations in τ_R are found for excitation above or below E_{LH}, a strong increase in the PL rise time is measured for resonant excitation at E_{LH}.

We think that some nontrivial insight on the dynamics

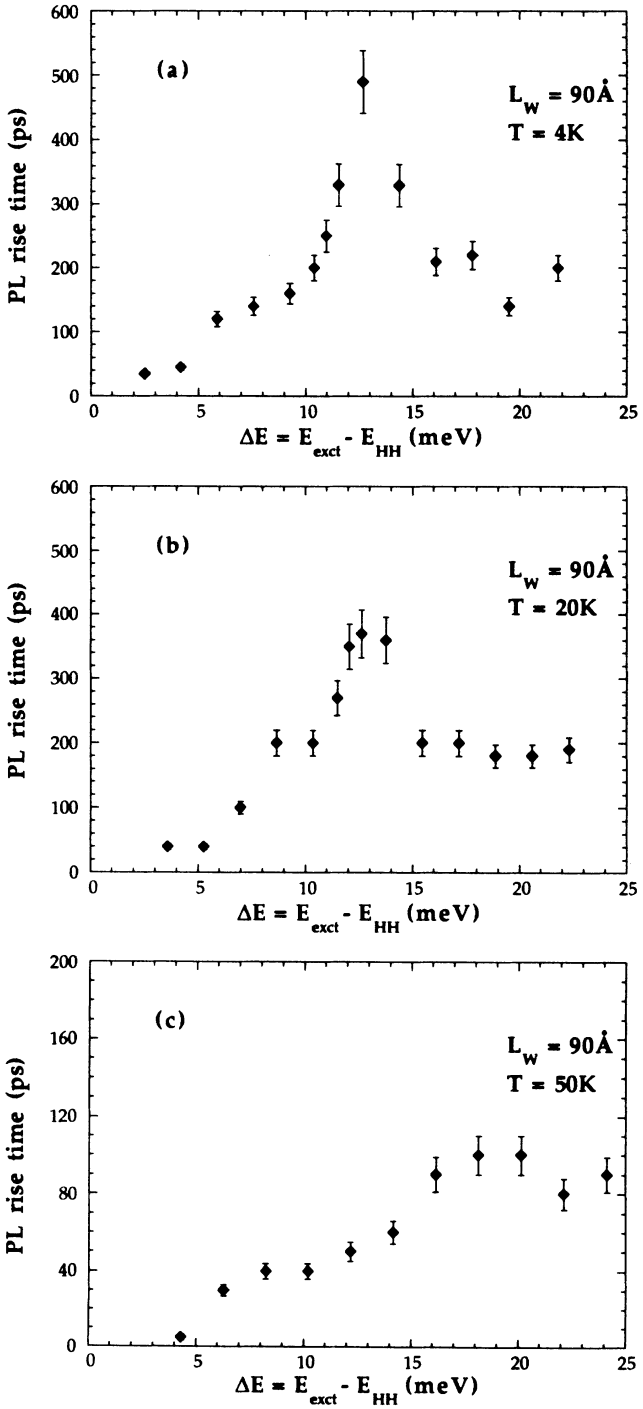

FIG. 3. PL rise time at E_{HH} as a function of $\Delta E = E_{exct} - E_{HH}$, for three different temperatures: (a) 4, (b) 20, and (c) 50 K. Note the change of vertical scale in (c).

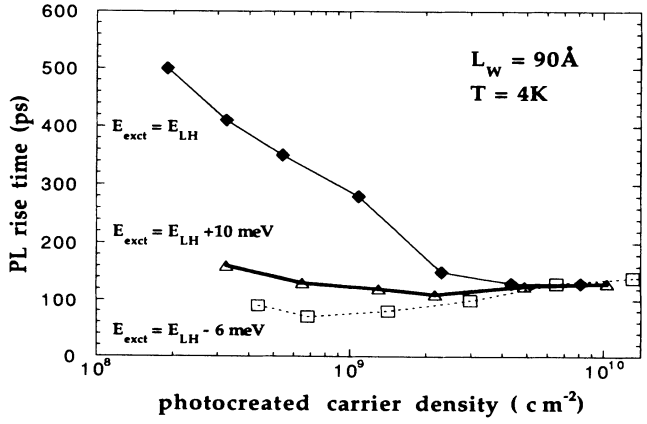

FIG. 4. PL rise time at E_{HH} of the 90-Å well ($T = 4$ K) as a function of the excitation density for three excitation photon energies: $E_{exct} = E_{LH}$, ◆; $E_{LH} + 10$ meV, △; and $E_{LH} - 6$ meV, □.

of exciton relaxation in GaAs quantum wells can be obtained by the analysis of the data presented.

If we limit the discussion to values of the excitation excess energy ΔE smaller than E_{LO}, the LO phonon energy, the relaxation of the photogenerated carriers into $K=0$ radiative excitations can only occur by exciton-acoustic phonon interaction, and exciton-exciton, exciton-free carrier, and exciton-defect collisions. We will assume that the last process plays a minor role in our sample, given the fact that no exciton localization has been observed in the low-temperature PL spectra.

As far as exciton-exciton scattering is concerned, while it is a very efficient process in producing thermal equilibrium within the excitonic gas, it should not be as much effective as an energy-loss mechanism at least for $n_{ex} \leq 10^9$ cm^{-2}.[7] Only exciton-free carrier and exciton-acoustic phonon scattering should provide the main relaxation mechanism at densities $n_{ex} \leq 10^9$ cm^{-2}, where the strong features in τ_R are observed.

In fact, exciton formation under resonant excitation is very likely to proceed without significant free carrier generation as opposed to the case of nonresonant excitation, as confirmed by the PLE spectrum in Fig. 1(a). We believe that the strong increase in τ_R observed at E_{LH} reflects the less efficient exciton-exciton scattering probability as compared to free carrier scattering,[8] on the one hand, and the reduced exciton-acoustic phonon scattering rate compared to free carrier–acoustic phonon scattering, on the other.[9] Both cooling of the photogenerated excitons to the heavy-hole exciton energy and momentum relaxation to $K=0$ are then disfavored for resonant excitation, giving rise to a sharp increase of τ_R around E_{LH}.

The smoothing of the peak in τ_R for increasing temperature and excitation density is consistently expected as a consequence of the increased scattering rates of the processes dominating exciton relaxation.

We refer to a forthcoming paper for the presentation of a wider set of data on exciton relaxation at low density, showing the occurrence of sharp features in τ_R as a function of the excitation excess energy at other band-edge energies as well, together with a detailed discussion of the implications on the exciton relaxation dynamics in these structures.

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