Influence of trapping on the exciton dynamics of Al$_x$Ga$_{1-x}$As films

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We present a systematic study on the exciton relaxation in high-purity AlGaAs epilayers. The time for the excitonic photoluminescence to reach its maximum intensity, defined as t_{max}, shows a nonmonotonic dependence on excitation density which is attributed to a competition between exciton localization and carrier–carrier scattering. A phenomenological four level model fully describes the influence of exciton localization on t_{max}. This localization effect is enhanced by the increase of the Al content in the alloy and disappears when localization is hindered by rising the lattice temperature above the exciton trapping energy. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1885173]

The simultaneous availability of commercial pulsed lasers and of nanostructured semiconductors in the late 1970's concentrated the attention of the optical studies on carrier dynamics on these heterostructures, diverting it from bulk III–V materials. GaAs and AlGaAs are the constituent materials of many heterostructures like VCSELs, superlattices, microcavities, cavity LEDs, or GRINSCH. The light emission characteristics of these heterostructures are fairly well understood as they have been the subject of intense study for the past decade.[1] A precise knowledge of the properties of their elemental constituents is needed for the complete understanding of the behavior of these devices and for the design of future systems based on these materials. However, little attention has been paid to the luminescence dynamics of bulk GaAs, and even less to that of AlGaAs. Only recently some works,[2–4] which complement but not complete older scattered studies,[5–8] have investigated the emission dynamics just in GaAs, since good alloy samples have not been available. However, important issues in III-V alloys, such as the influence of defects or aluminum content on the exciton dynamics, have not been addressed.

In this letter, we present a systematic study of the effect of exciton localization on the photoluminescence (PL) buildup in bulk GaAs and Al$_x$Ga$_{1-x}$As with x up to 0.05. In the past, exciton localization effects in bulk GaAs[7,8] and quantum wells (QWs),[9–11] have mainly been studied through their influence on the excitonic luminescence decay time. Still we have found that exciton localization has a stronger influence on the rise dynamics, which we characterize by the time the free-exciton PL takes to reach its maximum, t_{max}.

We investigated four high quality Al$_x$Ga$_{1-x}$As epilayers of 2.5 μm thickness, with Al concentration $x=0, 0.015, 0.03$, and 0.05, grown by molecular beam epitaxy.[12] All the samples, which were nominally undoped, showed p-type conductivity with hole concentrations in the range $1-8 \times 10^{14}$ cm^{-3}. The samples were mounted on a cold finger cryostat, which enabled precise control of the temperature between 5 and 45 K, and were nonresonantly photoexcited with a Ti:sapphire laser that produced 2 ps long pulses. The

PL was energy and time resolved by a synchroscan streak camera in conjunction with a spectrometer. The time and energy resolution of the overall setup is better than 12 ps and 0.4 meV, respectively. A low-temperature cw PL excitation characterization of the samples (not shown here), allowed a precise determination of the free and bound exciton emission peaks (*FX* and *BX*, respectively). Despite the presence of localization centers, the narrowness of the excitonic lines (full width at half maximum below 1 meV for the GaAs epilayer) guarantees the good quality of the samples.

Figure 1 shows the PL spectra at 5 K and low excitation density ($n=1.8 \times 10^{14}$ cm^{-3}) of the GaAs (black line) and $x =0.03$ (grey line) samples 1 ns after the excitation. The PL is dominated by the emission from *FX* and *BX* (A^0-*X*, D^0-*X*), as well as by electron-acceptor (e-A^0) recombination (peaks have been identified following Refs. 7 and 13). An exciton localization energy in acceptor related sites, of about 2.7 meV, can be extracted from the peaks' position. At low excitation densities, the *BX* luminescence overcomes that of the *FX* as the Al fraction in the alloy is augmented, due to the increase in the number of defects. However, at high excitation densities ($n \sim 7.5 \times 10^{15}$ cm^{-3}) the PL is completely dominated by the *FX* emission in all samples. In the analysis of the time-resolved PL we will concentrate on the *FX* and A^0-*X* lines.
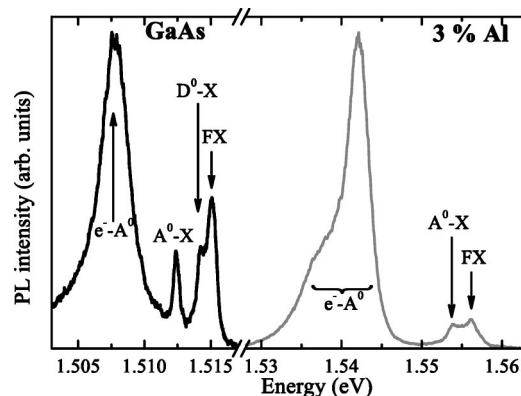

FIG. 1. Low temperature (5 K) PL spectra of the GaAs (black line) and $x =0.03$ (grey line) samples recorded at a delay of 1 ns after excitation (excitation density 1.8×10^{14} cm^{-3}; excitation energy 1.630 eV).

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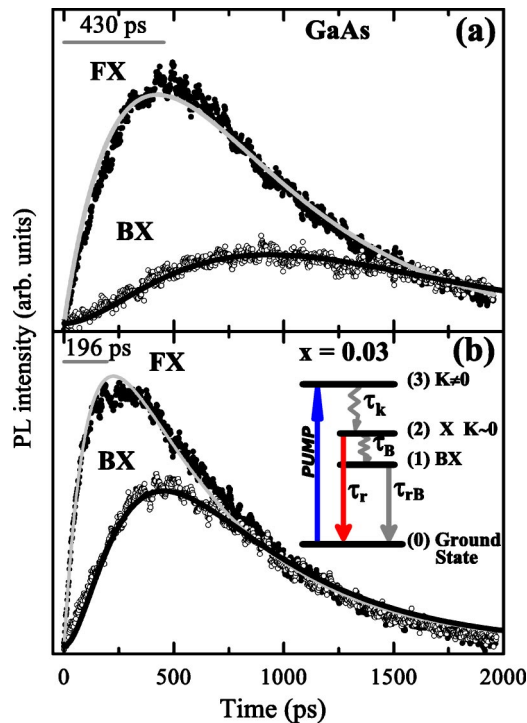
FIG. 2. (Color online) (a) Time evolution of the *FX* (solid dots) and the *BX* (A^0-X; open dots) for the GaAs sample (excitation density 1.8×10^{14} cm^{-3}, $T = 5$ K); (b) same as (a) for the Al$_x$Ga$_{1-x}$As $x = 0.03$ sample. t_{max} is indicated by horizontal bars. The solid lines are fits to the model described in the text ($\tau_k = 464$ ps, $\tau_B = 1240$ ps, and $\tau_r = \tau_{rB} = 565$ ps for GaAs). The inset shows the level scheme and the transitions considered in the model.

FIG. 3. (Color online) (a) Time for the free exciton to reach its maximum intensity, t_{max}, as a function of excitation density in the four investigated Al$_x$Ga$_{1-x}$As epilayers at a lattice temperature of 5 K. Aluminum content is indicated in each curve; (b) t_{max} as a function of excitation density for the GaAs sample at different lattice temperatures.

Figure 2 depicts the time evolution of the *FX* (solid points) and A^0-X (*BX*; open points) emissions for the GaAs and the $x = 0.03$ samples under the same conditions as in Fig. 1. t_{max}, which can be easily assigned, as indicated in the figure by the horizontal bars, is considerably longer for the GaAs than for the $x = 0.03$ sample. We have observed that t_{max} is also longer for the *BX* than for the *FX* (Fig. 2) in all investigated samples, revealing longer energy relaxation processes in the case of the *BX*. In addition, as can be clearly seen for GaAs in Fig. 2(a), the *BX* presents a concave time evolution at short times after the excitation, while that of the *FX* is convex. The concave curvature of the *BX* traces indicates that the buildup of the *BX* population results from a multistep relaxation process. Both facts, the shorter t_{max} for *FX* than for the *BX* and the concave curvature, evidence that the source of the *BX* luminescence is the trapping of *FX* with center of mass momentum $K \sim 0$, in a cascade process similar to that observed when carriers from the barrier are trapped in quantum dots.[3,14]

In order to account for this cascade process we have considered a four-level rate-equation model as depicted in the inset of Fig. 2(b). The excitation pulse creates electron-hole pairs which rapidly (< 20 ps)[15] form excitons with large K (level 3 in the inset). These *FX* relax their kinetic energy towards the radiative $K \sim 0$ states (level 2) via emission of acoustic phonons,[2] with a characteristic time τ_k. *FX* can then either radiatively recombine (τ_r) or get trapped in localization sites (τ_B) giving rise to a *BX* population (level 1), which can also radiatively recombine (τ_{rB}). The differential rate equations that describe the dynamics of such a four-level model can be easily analytically integrated.

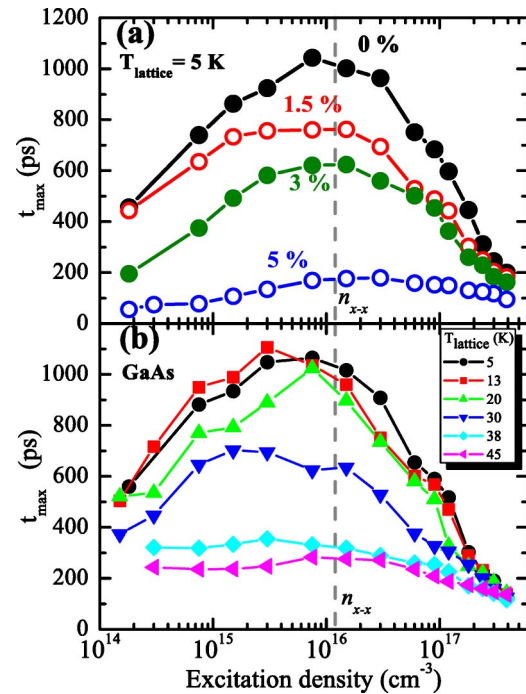Figure 2 also shows the *simultaneous* least squares fits of

the temporal traces extracted from the four-level model to the *FX* and *BX* data. For the sake of reducing the number of fitting parameters, the fits were performed assuming the same recombination times for *FX* and *BX* ($\tau_r = \tau_{rB}$). Good agreement is found between the experiments and the fits. From the fitting parameters we have found a trapping time τ_B four times shorter in the $x = 0.03$ epilayer than in the GaAs one, evidencing a density of traps about four times greater in the $x = 0.03$ sample ($\tau_B \propto [\text{traps}]^{-1}$).[7]

Figure 3(a) shows the dependence of the *FX*-t_{max} on excitation density at 5 K for the four investigated epilayers. All the samples show a similar nonmonotonic behavior,[16] with a maximum t_{max} at a carrier density $n \sim 1.2 \times 10^{16}$ cm$^{-3} \equiv n_{x-x}$. This density corresponds to a mean distance between excitons of ~ 40 nm, which is of the same order of magnitude as the exciton Bohr radius (a_B) in GaAs (11.2 nm). Hence, for $n > n_{x-x}$, exciton-exciton elastic scattering is an important source of scattering events and results in a fast relaxation of *FX* with large K toward the radiative states with $K \sim 0$, as previously reported in QWs.[15,17] Exciton-free carrier collisions also take place for the highest excitation densities considered in this work, and take part in the fast relaxation of carriers as also demonstrated in the case of QWs.[17] The higher the carrier density in the sample the more efficient these processes become, resulting in a decrease of t_{max} with increasing excitation density. The threshold density n_{x-x} only depends on a_B and thus is expected to be independent of the Al content for the low concentrations considered in this letter, as borne out by our experiments.

For $n < n_{x-x}$, *FX*-t_{max} increases with increasing excitation density. This behavior can be understood if trapping of *FX* in localized states is taken into account in the framework of the proposed four-level model. The PL spectra in all investigated samples are dominated by *BX* emission for densities below

$n_{x\text{-}x}$. Therefore, at the lowest studied densities, there are two mechanisms for the depletion of excitons from the $K \sim 0$ *FX* level: (i) exciton radiative recombination, and (ii) trapping of *FX* with $K \sim 0$ into localized *BX* states. As the excitation density is increased, *BX* trapping states are gradually filled up to their saturation. When n is high enough, the number of available localization sites is small compared to the *FX* population, and the trapping channel (ii) has very little effect on the dynamics of the *FX* population. The direct consequence of the saturation of trapping centers, and therefore, the closing of one of the depleting channels, is the increase of t_{max}. This situation is reproduced by the model, and its extreme case corresponds to the elimination of the *BX* level: fixing the values of τ_k and τ_r obtained from the fits of Fig. 2(a), and taking $\tau_B \rightarrow \infty$, the model would give a *FX* temporal trace with t_{max} considerably longer (512 ps) than that obtained in the presence of the localization channel (430 ps). This is in qualitative agreement with the increase of t_{max} with carrier density shown in Fig. 3(a).

The increase of the Al content in the alloy results in two effects: (i) an acceleration of the dynamics due to the enhancement of alloy scattering, which results in the reduction of t_{max} for any n; (ii) a larger number of alloy defect-related traps. As already mentioned above, a higher density of localization centers produces a reduction of the trapping time τ_B which, according to the four level model, results in an additional decrease of t_{max} for $n < n_{x\text{-}x}$.

In order to gain insight into the influence of trapping on the *FX* dynamics, time resolved experiments have been performed at different lattice temperatures. The results for GaAs are shown in Fig. 3(b), which depicts t_{max} versus excitation density for temperatures up to 45 K. For $T \lesssim 30$ K, the observed behavior follows the trends discussed above for low T (nonmonotonous dependence of t_{max} on n). In contrast, if the lattice temperature is raised above the exciton localization energy (2.7 meV $\leftrightarrow$ 31.3 K), the *BX* are ionized and trapping is hindered. In the absence of the trapping depletion channel, according to the model no dependence of t_{max} on excitation density for $n < n_{x\text{-}x}$ is expected, as found experimentally for $T = 38$ K and $T = 45$ K [Fig. 3(b)].

In conclusion, we have presented a systematic study of the effect of exciton localization on the PL rise time in AlGaAs. For high excitation densities, exciton–exciton and exciton–carrier scattering dominate the dynamics. For low excitation densities, t_{max} is strongly influenced by the trapping of *FX* into *BX* states. The competition between localization and carrier–carrier scattering obtains a nonmonotonic dependence of the rise time, t_{max}, on excitation power. A four-level phenomenological model fully describes the excitation-density, aluminum-content, and lattice-temperature dependence of the rise time.

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