

## Charged exciton dynamics in GaAs quantum wells

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We study the dynamics of the charged and neutral excitons in a modulation-doped GaAs quantum well by time-resolved photoluminescence under a resonant excitation. The radiative lifetime of the charged exciton is found to be surprisingly short, 60 ps. This time is temperature independent between 2 and 10 K, and increases by a factor of 2 at 6 T. We discuss our findings in view of present theories of exciton radiative decay. [S0163-1829(98)03143-9]

The negatively charged exciton ( $X^-$ ), which consists of two electrons bound to a hole, is the semiconductor analogue of the hydrogen ion  $H^-$ . The existence of  $X^-$  was proposed nearly 40 years ago,<sup>1</sup> but it was only recently identified in semiconductor quantum wells (QW).<sup>2-6</sup> Charged excitons show up in the photoluminescence (PL) and absorption spectra of a QW as a sharp line at an energy below the neutral exciton ( $X$ ) line. The energy separation between the neutral and charged exciton lines ( $\sim 1$  meV in GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$  QW) represents the binding energy of the second electron to the exciton.

It is clear that an excess density of electrons, which could bind to the photo excited electron-hole pairs, is needed for the  $X^-$  to be formed. Indeed,  $X^-$  is observed in structures where extra electrons are created by modulation doping,<sup>2-4</sup> tunneling,<sup>5</sup> or optical excitation.<sup>6</sup> The appearance of  $X$  and  $X^-$  in modulation-doped QW is correlated with the electron gas conductivity.<sup>3</sup> This finding and recent studies by near-field spectroscopy<sup>7</sup> have led us to the conclusion that the  $X^-$  is created in modulation-doped QW by binding of a photo-excited electron-hole pair to a localized electron. The electrons are localized, separately one from another, in the minima of a random potential due to the remote ionized donors. This long-range potential does not alter significantly the internal structure of  $X^-$ .

In this work we present a comprehensive time-resolved PL study of the  $X$ - $X^-$  system in modulation doped GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$  QW. Recombination dynamics of excitons in semiconductor QW has been a subject of intense theoretical and experimental research over the last decade. It was found that the two dimensional (2D) nature of free excitons in QW suppresses the polariton splitting,<sup>8</sup> which precludes the radiative recombination of exciton polaritons in bulk semiconductors. Instead, each exciton state couples to a 1D continuum of photon states, resulting in its fast decay.<sup>9</sup> Indeed, short radiative decay times, of a few tens of picoseconds, were calculated<sup>10,11</sup> and measured<sup>12,13</sup> for excitons in a GaAs QW. The  $X$ - $X^-$  system is especially interesting, as it allows us to study and compare the radiative decay of free and bound excitons.

An experimental aspect of exciton lifetime measurements, which is often overlooked, is the importance of using the excitation light energy resonant with the exciton transition. Under nonresonant excitation a mixture of electron-hole

plasma and hot excitons is formed, and the dynamics of the excited system is governed by the exciton formation and cooling processes.<sup>14</sup> Only resonant excitation can create cold excitons directly in the radiative zone and gives direct access to the radiative lifetime.<sup>12</sup> In most of the measurements we have tuned the laser energy resonantly to the  $X$  or  $X^-$  transitions.

Our results can be summarized as follows. We find that the  $X$ - $X^-$  system at low temperatures does not reach thermodynamical equilibrium: if we create only the  $X^-$  population (by resonant excitation at the  $X^-$  energy) the  $X$  line does not appear in the PL spectra at any later times. Thus, by following the  $X^-$  PL decay we can measure directly its radiative lifetime. We find that it is surprisingly short, 60 ps, and increases monotonically with magnetic field to 120 ps at 6 T. We discuss these lifetimes in the context of present theories of exciton radiative decay. Finally, we show that charged excitons are created predominantly from cold excitons, with a typical formation time of the order of  $\sim 100$  ps.

We have studied different single-side modulation-doped QW samples grown by MBE. All samples show a similar behavior and we have chosen to present the results for the highest-quality one. It consists of a buffer superlattice, a 20-nm QW, an undoped 50-nm  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  spacer layer, a Si  $\delta$ -doped region, another undoped 40-nm  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  layer and a 5-nm GaAs cap layer. The electron density and mobility after illumination are  $2 \times 10^{11} \text{ cm}^{-2}$  and  $4 \times 10^6 \text{ cm}^2/\text{V sec}$ , respectively. The sample is processed to form  $3 \times 3$  mm mesas. Ohmic contacts are annealed into the two-dimensional electron gas (2DEG) and a semitransparent Cr gate is evaporated on the top of the mesa. The electron density is varied by applying a voltage to the gate with respect to the contacts.

The sample is excited by a train of short light pulses ( $\sim 3$  ps) from a mode-locked Ti-sapphire laser with an average power density of  $\sim 30 \text{ mW/cm}^2$ . Each pulse creates  $\sim 10^7$  excitons/ $\text{cm}^2$ . The time resolved PL is recorded in cross polarization by a 2D streak camera (Photonetics streak scope), using a 0.25-m spectrometer with a 1200 grooves/mm grating. The overall time resolution is  $\sim 20$  ps. The temperature is varied between 2 and 10 K and the magnetic field from 0 to 6 T in an immersion cryostat placed inside a superconducting magnet.

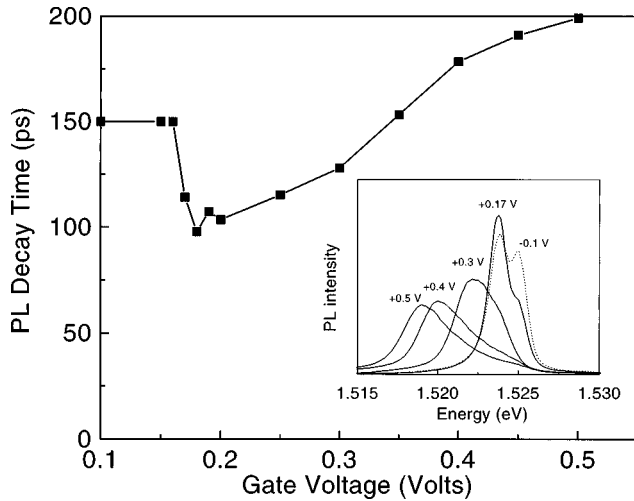


FIG. 1. The decay time of the spectrally integrated PL (excited off-resonantly at an energy of 1.61 eV) as a function of the gate voltage. Inset: the corresponding time-integrated spectra at several gate voltages.

The time-integrated PL spectrum of the sample exhibits a behavior that is characteristic for a gated 2DEG (inset of Fig. 1).<sup>3</sup> For gate voltages  $+0.2 < V_g < +0.5$  V the spectrum consists of a single broad peak, corresponding to a recombination of electron gas with photoexcited holes. With decreasing gate voltage the electron density decreases and as a result the PL line becomes narrower. At +0.2 V the spectrum abruptly changes and the exciton line,  $X$ , appears. The other low-energy line, which evolves from the 2DEG-related recombination, is the negatively charged exciton  $X^-$ . This abrupt change in the spectrum, which is due to electron localization, is clearly reflected in the PL dynamics. Figure 1 shows the dependence of the PL decay time on the gate voltage for a nonresonant excitation (in this case both  $X$  and  $X^-$  have the same decay time<sup>6</sup>). We can clearly observe that the decay time gradually decreases as the  $V_g$  is changed from +0.5 to +0.2 V, and increases abruptly at  $V_g < +0.2$  V. The gradual decrease in decay time at  $+0.2 < V_g < +0.5$  V is due to an increase of the electron-hole overlap with the change of the built-in field in the well.<sup>15</sup> The abrupt increase at  $V_g < +0.2$  V is due to electron localization and the appearance of excitons. The experiments which are described in the rest of this paper are performed at  $V_g = -0.1$  V, to ensure that the 2DEG is in the localized regime.

Figure 2 describes the measured PL signals when the sample is excited resonantly at the  $X$  and  $X^-$  energies at 2 K. We label the experimental curves with the notation  $A(B)$ , where  $A$  is the detection and  $B$  is the excitation energy ( $X$  or  $X^-$ ). In Figs. 2(a) and 2(b) we show the results obtained with excitation at the  $X$  energy,  $X(X)$  and  $X^-(X)$ , respectively. The  $X(X)$  signal rises almost instantaneously, due to the resonant creation of cold excitons.<sup>16</sup> The  $X^-(X)$  signal, on the other hand, rises gradually. This rise time corresponds to the process in which an electron captures an exciton to form  $X^-$ . While the  $X^-(X)$  signal exhibits a simple exponential decay, the decay of the  $X(X)$  signal is two exponential. The slow exponent of the  $X(X)$  decay is identical to that of the  $X^-(X)$  signal.

The equal decay rates of the  $X(X)$  and  $X^-(X)$  signals at

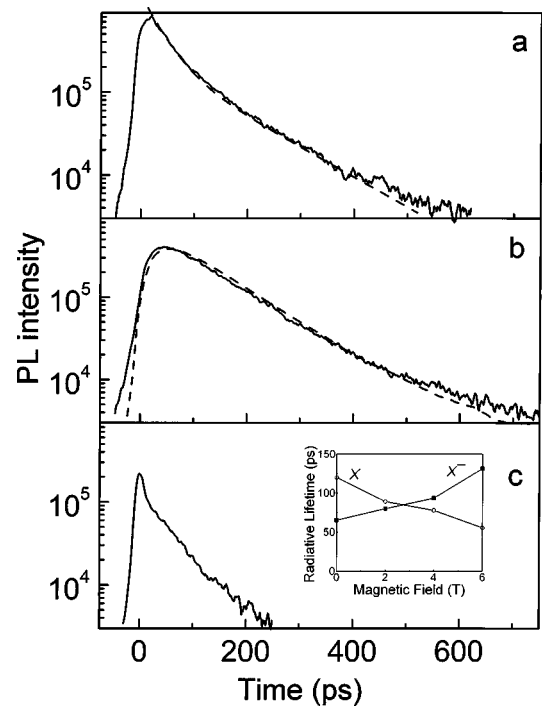


FIG. 2. The photoluminescence decay profiles at  $T=2$  K. (a)  $X(X)$ —the exciton signal for resonant excitation. (b)  $X^-(X)$ —the charged exciton signal for excitation at the exciton transition. (c)  $X^-(X^-)$ —the charged exciton signal for resonant excitation. Inset: The  $X$  and  $X^-$  radiative lifetime as a function of magnetic field.

late times might be interpreted as an indication of the existence of a thermal equilibrium between  $X$  and  $X^-$ .<sup>6</sup> However, the resonant excitation at the  $X^-$  energy clearly shows that the situation is different. Namely, in this case we observe a PL signal only at the  $X^-$  energy; *no exciton PL is observed at any later time*. One can thus conclude that the  $X^- \rightarrow X + e$  process is very slow at 2 K and the system cannot reach thermodynamical equilibrium. This situation persists up to 6 K. Only at 9 K a sizable  $X(X^-)$  signal starts to appear. We conclude, that the similarity of the  $X(X)$  and  $X(X^-)$  decay times is solely a consequence of  $X$  population feeding  $X^-$ . Indeed, the  $X^-$  radiative decay rate (see below) is faster than the decay of the  $X$  signal.

Figure 2(c) shows the  $X^-(X^-)$  signal: it rises within the experimental resolution and then decays exponentially with a short time constant of 60 ps (the spike near  $t=0$  is due to reflections from the sample surface). This decay time is temperature independent between 2 to 9 K [Fig. 3(b)], and we interpret it as the radiative lifetime of the  $X^-$ . The nonradiative decay channels may safely be neglected within this temperature range, because the time and spectrally integrated PL intensity under nonresonant excitation was measured to be constant between 2 and 40 K. The inset of Fig. 2(c) shows the dependence of the  $X^-$  radiative lifetime on the magnetic field applied normal to the layers. We observe a gradual increase in lifetime from 60 ps at zero field to 120 ps at 6 T. The measured  $X^-$  radiative lifetime agrees well with recent measurements of the radiative lifetime of impurity bound excitons in GaAs QW.<sup>17</sup> A decay time of 120 ps is observed when an exciton bound to a donor, with 2.2 meV binding energy, is resonantly excited. Since the radiative lifetime of a bound exciton in 2D is proportional to its binding energy,<sup>18</sup>

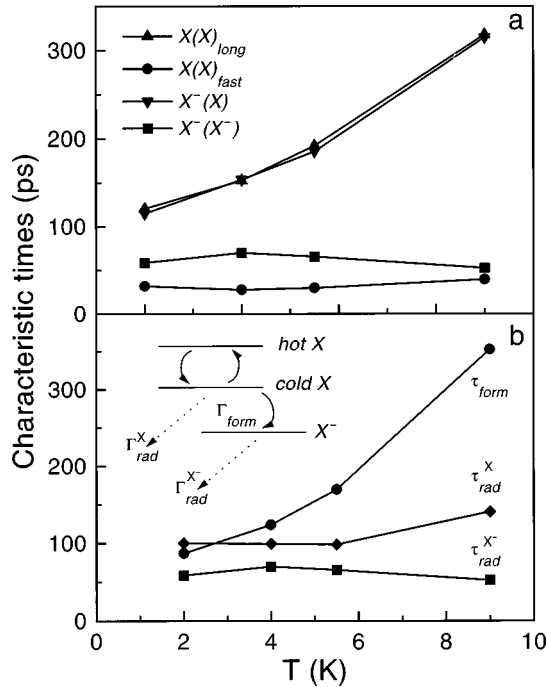


FIG. 3. (a) The decay times of the  $X(X)$  and  $X^-(X)$  signals as a function of temperature. (b) The  $X^-$  and exciton radiative recombination times and the  $X^-$  formation time. The latter two times are deduced from the two-level model shown in the inset. The data at 9 K are less reliable, because the neglected  $X^- \rightarrow X + e$  process becomes important.

one gets a very good agreement between these two measurements. Furthermore, this dependence on binding energy can explain the observed behavior in magnetic field: it was shown that the  $X^-$  binding energy grows from 1.1 meV at zero field to 1.7 meV at 6 T.<sup>19</sup>

The short radiative lifetime of the  $X^-$  seems surprising, however, in a view of a recent theories on the radiative decay of localized excitons and biexcitons.<sup>20</sup> Namely, it was shown that the radiative lifetime of these bound complexes is  $\sim (\lambda/l)^2 \tau$ , where  $\lambda$  is the light wavelength in the medium,  $l$  is the localization length or the size of the complex, and  $\tau \sim 25$  ps is the radiative lifetime of a free exciton. The radius of  $X^-$  can be estimated to be  $\sim 20$  nm and it is expected to be localized in potential fluctuations with a typical size of  $\sim 50$  nm, determined by the spacer width in our sample.<sup>3</sup> Either one of these two lengths then implies a huge increase in lifetime with respect to the free exciton, by a factor of  $(\lambda/l)^2 > 20$ . This is clearly not the case in our experiment:  $X^-$  radiative lifetime is similar to that measured for free excitons in an undoped QW (Refs. 12 and 13) and in the present work (see later in the paper). A possible explanation, which might resolve the contradiction between the theory and our experiment is as follows. We consider  $X^-$  as an exciton bound to a localized electron. Each electron forms a potential well for the exciton, with a depth equal to the  $X^-$  binding energy of  $\sim 1$  meV. We argue that the exciton may effectively tunnel through this shallow potential barrier into another electron localization site, which is  $\sim 50$  nm away. Thus, we propose to consider the  $X^-$  as a delocalized exciton, tunneling between a few localized electrons.

Let us turn to discussing the dynamics of the  $X$  and  $X^-$

system in more details. Figure 3(a) displays the measured decay times of the  $X$  and  $X^-$  as a function of temperature. The two-exponential decay of the  $X(X)$  signal gives rise to two decay times. It is seen that the fast decay time,  $X(X)_{fast}$ , is relatively temperature independent, while the slow component,  $X(X)_{slow}$ , increases with temperature. The  $X^-$  decay as a function of temperature is also shown for the two excitation conditions. For excitation at the  $X$  energy,  $X^-(X)$ , we observe a decay time which is identical to the long decay time of the exciton at all temperatures. The radiative decay time of the  $X^-$ , determined from the  $X^-(X^-)$  signal, is much shorter and is almost temperature independent.

A two-exponential decay of the exciton line after resonant excitation has already been observed and interpreted as a transition from a cold exciton distribution created by the laser pulse, to that thermalized with the lattice.<sup>13</sup> It is well established by now that in a QW only excitons with kinetic energies less than  $E_0 \sim 0.1$  meV are able to recombine (excitons with higher kinetic energies do not satisfy the in-plane momentum conservation with the emitted photon). For the case of thermal distribution of excitons, only a small fraction of the population  $\sim E_0/(k_B T)$  has kinetic energies less than  $E_0$ . As a result, the radiative decay rate of a thermalized exciton population is  $\sim E_0 \Gamma_{rad}^X/(k_B T)$ , where the radiative rate of excitons with energies less than  $E_0$  is denoted as  $\Gamma_{rad}^X$ .<sup>11</sup> The linear increase with temperature of the  $X(X)$  slow component [Fig. 3(a)] is therefore consistent with a decay of a thermalized excitons distribution.

The  $X-X^-$  system can be modeled in a way similar to that used by Vinattieri *et al.*<sup>13</sup> for excitons [see the inset of Fig. 3(b)]. The exciton continuum is divided into two discrete levels, corresponding to wave vectors within (cold) and outside (hot) the radiative zone. Since the times associated with polarization relaxation are shorter than our time resolution, the exciton spin is not revealed. We also introduce a third level for charged excitons. As was explained above, at the lowest temperatures we may safely neglect the  $X^-$  ionization into  $X$ , which further simplifies the model.

In solving the dynamics of the system we use the fact that the process, in which an exciton is trapped by an electron to form  $X^-$ , has a higher cross section for cold excitons. This is because the interaction cross-section for any long range potential (such as the Coulomb interaction between the electron and the exciton dipole) decreases with energy. A hot exciton, with a large kinetic energy, has therefore a lower probability to be captured. Furthermore, if one assumes that the excess energy is given to an acoustical phonon, then this process favors a low-energy transfer. If the excess energy is large the  $X^-$  acquires most of it, and can breakup to an exciton and an electron, through the emission of another acoustic phonon. Thus, one can assume that there is only one  $X^-$  formation rate,  $\Gamma_{form}$  [inset of Fig. 3(b)]. With this assumption the dynamics of the system can be easily solved. We fit a two exponential decay curve to the  $X(X)$  signal [Fig. 2(a), dashed line] and get the slow and fast  $X(X)$  decay rates. We then obtain from the model the rates for exciton heating and cooling, as well as the sum of the exciton radiative decay rate  $\Gamma_{rad}^X$  and  $X^-$  formation rate,  $\Gamma_{form}$  [inset Fig. 3(b)]. The escape rate from the cold exciton level,  $\Gamma_{rad}^X + \Gamma_{form}$ , is depicted by open squares in Fig. 3(b). We now recall our as-

sumption that only cold excitons can form  $X^-$ . Then the ratio of the number of excitons that bind to form  $X^-$  to the number of excitons that recombine radiatively is constant at any instant of time and equals to  $\Gamma_{\text{form}}/\Gamma_{\text{rad}}^X$ . We may therefore estimate it from the experiment as the ratio of the time-integrated  $X^-(X)$  and  $X(X)$  signals and find separately  $\Gamma_{\text{form}}$  and  $\Gamma_{\text{rad}}^X$  [Fig. 3(b)].

We find that the capture rate of excitons by electrons  $\Gamma_{\text{form}}$  decreases with temperature. This trend is similar to that found for the rate of electron capture by impurities in bulk semiconductors.<sup>21</sup> The cold exciton radiative rate  $\Gamma_{\text{rad}}^X$  is found to be temperature independent in the range of 2–6 K, as is expected. The value obtained,  $\sim 100$  ps, is larger than the theoretical limit of 50 ps, probably due to localization and the built-in electric field. It is interesting to study the dependence of this radiative lifetime on magnetic field, and compare it to that of  $X^-$ . The exciton radiative lifetime as a function of magnetic field is shown on the inset of Fig. 2(c). It can be seen that the lifetime exhibits an opposite trend (relative to that of  $X^-$ ) and slightly *decreases* with magnetic field. This is because the magnetic field increases the electron-hole overlap within exciton, and probably does not affect the localization length of an exciton as a neutral object.

Finally, let us analyze the  $X^-(X)$  signal [Fig. 2(b)]. The assumption that only radiative excitons can form  $X^-$  enables us to express this signal in terms of *measured quantities only* as

$$I_{X^-(X)} = \frac{\Gamma_{\text{form}}}{\Gamma_{\text{rad}}^X} \Gamma_{\text{rad}}^{X^-} \int_{-\infty}^0 \exp[-\Gamma_{\text{rad}}^{X^-}(t-t')] I_{X(X)}(t') dt', \quad (1)$$

where  $\Gamma_{\text{rad}}^{X^-}$  is the measured  $X^-$  radiative decay rate, obtained from the  $X^-(X^-)$  signal. The prefactor  $\Gamma_{\text{form}}/\Gamma_{\text{rad}}^X$  is given from the ratio of the time integrated  $X^-(X)$  and  $X(X)$  signals [Eq. (1)], and the  $X(X)$  intensity,  $I_{X(X)}$  is taken from Fig. 2(a). The dotted line in Fig. 2(b) is the curve resulting from Eq. (1). Note that it is not a fit—all of the parameters, including the amplitude, are taken from the experiment. The quality of the agreement between the measurement and the calculation is a good indication that our underlying assumptions are reasonable.

In conclusion, we have studied the dynamics of a neutral and charged exciton system in a GaAs QW at different temperatures and magnetic fields. We have measured the charged exciton radiative lifetime and its dependence on magnetic field. This time turns out to be short, suggesting a large coherence volume of  $X^-$ . We propose that  $X^-$  is created by sharing an exciton between several localized electrons. Finally, we have measured the  $X^-$  formation time ( $X + e \rightarrow X^-$ ). The  $X^-$  ionization ( $X^- \rightarrow X + e$ ) is found to be negligible at low temperatures ( $T \leq 6$  K).

*Note added in proof.* Recently, we have learned about another work, which investigates charged exciton dynamics in a magnetic field.<sup>22</sup>

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