Inhibited recombination of charged magnetoexcitons

H. Okamura
Physics Department, Kobe University, Kobe 657-8501, Japan;
Physics Department, Northeastern University, Boston, Massachusetts 02115;
and Francis Bitter Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

D. Heiman
Physics Department, Northeastern University, Boston, Massachusetts 02115
and Francis Bitter Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

M. Sundaram* and A. C. Gossard
Materials Department, University of California, Santa Barbara, California 93106
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Time-resolved photoluminescence measurements show that the decay time for charged excitons in a GaAs two-dimensional electron gas increases by an order of magnitude at high magnetic fields. Unlike neutral excitons, the charged exciton center-of-mass is spatially confined in a ''magnetically adjustable quantum dot'' by the cyclotron orbit and the quantum well. The inhibited recombination is explained by a reduced phase coherence volume of the magnetically confined charged excitons.
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Charged excitons or ''trions'' were first identified in optical-absorption experiments on electron-doped CdTe quantum-well (QW) structures through their polarization properties in a magnetic field.[1] The negatively charged exciton (X^-) transition in a narrow QW was manifest in the spectra as a peak lying several meV below the uncharged exciton (X^0) peak. Although both X^0 and X^- transitions had probably been observed in an earlier photoluminescence (PL) spectra of GaAs QW's, the high electron density precluded their identification.[2] In hindsight, it is not surprising that the recently identified X^- is often the most common exciton found in a system with excess electrons, similar to the X^0 in a system without excess electrons. An X^0 in the presence of excess electrons becomes polarized by a nearby electron and binds the electron by a dipolar attraction. The properties of X^- transitions in GaAs QW's have been explored in several recent experimental[3–7] and theoretical[8–12] studies.

An interesting facet of the charged exciton that has yet to be explored is the *confinement* produced by the cyclotron motion in a magnetic field. The X^- complex (two electrons plus one hole) is singly charged and a magnetic field confines the X^- center-of-mass motion to a cyclotron orbit, unlike the neutral exciton (X^0) which is free to move in a magnetic field. This will be referred to as the *magnetically confined charged exciton* (MCX). A magnetic field applied perpendicular to a two-dimensional (2D) QW effectively confines the exciton to a quantum dot (QD) whose size is adjustable with magnetic field. The 3D MCX volume, defined roughly by the QW width and the area of the cyclotron orbit in the plane of QW, is inversely proportional to the perpendicular magnetic field, $V_{MCX} \propto 1/B_\perp$. At high magnetic fields this volume is typically smaller than QD's currently available via patterned nanostructures.

The purpose of the present study is to examine the radiative recombination of excitons confined in these MCX QD's. Exciton recombination times were determined by measuring the PL decay times in low-density GaAs/Al_xGa_{1-x}As electron gases in magnetic fields to 18 T, at temperatures 0.5–7 K. At low temperatures, the X^- decay time was found to increase by an order of magnitude for increasing perpendicular magnetic field. In contrast, the recombination was rapid for both the X^- in fields applied in the plane of the QW and for the uncharged X^0. In the latter two cases the exciton is not confined to a QD. The linear dependence of exciton decay time with magnetic field is explained by a model in which the transition strength for optical recombination is inversely proportional to the MCX QD volume or phase coherence volume.

Experiments were performed on a symmetrically modulation-doped electron gas contained in wide parabolic GaAs/$Al_{0.3}Ga_{0.7}$As QW's.[13] In these QW's the electrons are distributed uniformly over a thick layer $\sim$250 nm wide, with electron densities of 5 and 7×10^{15} cm^{-3}, and mobility of 1.2 and 1.9×10^5 cm^2/V sec. The photogenerated holes were confined within a layer $\sim$25 nm wide at the center of the much wider electron layer. Thus although the electrons are spread over $\sim$250 nm, the excitons are confined to a narrow 2D plane in the center of the QW. Samples were mounted on a fiber optic probe inserted into a ^{3}He cryostat, which was placed in the bore of an 18 T Bitter or a 30 T hybrid magnet.[14] Time-resolved PL measurements employed standard time-correlated single-photon counting electronics and a multichannel plate. A pulsed diode laser operating at 1.58 eV (200 ps pulse length at 17 MHz) and a 0.85 m double spectrometer provided a system response of 300 ps full width at half maximum.[15] Deconvolution of the system response resulted in a time resolution of $\sim20-100$ ps for PL decay times.

Figure 1(a) shows the PL spectra at 0.5 K in magnetic fields applied perpendicular to the QW layers, and the inset

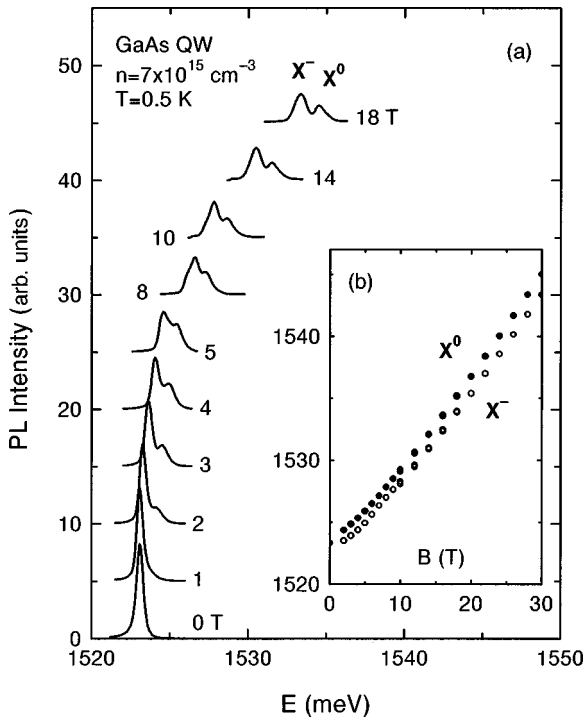

FIG. 1. (a) Photoluminescence spectra at $T=0.5$ K from a GaAs quantum well for various magnetic fields applied perpendicular to the well. The 250 nm wide electron layer had a density of 7×10^{15} cm^{-3}; holes were confined to a 25 nm wide well in the center of the electron layer. (b) Spectral energies of the X^- and X^0 peaks versus magnetic field.

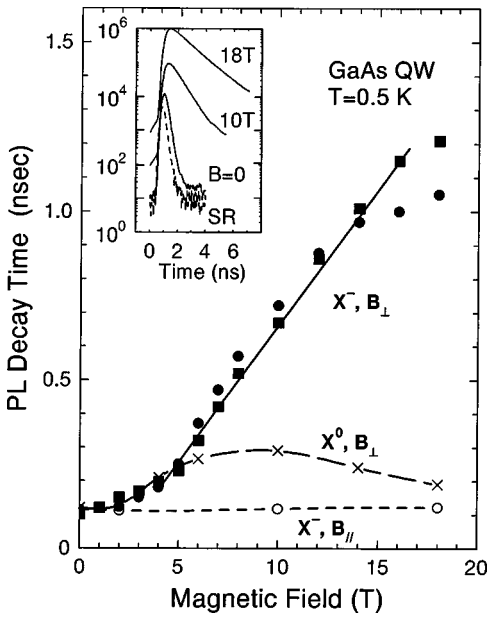

FIG. 2. Photoluminescence decay time of excitons in GaAs quantum wells at $T=0.5$ K versus magnetic field. The solid points are for the X^- charged exciton in perpendicular ($B_\perp$) magnetic field, for electron densities of 5 (squares) and 7 (circles) $\times10^{15}$ cm^{-3}. Open circles are for X^- in parallel ($B_\parallel$) field. The crosses are for the X^0 with $B_\perp$ in an undoped sample. The solid, straight line shows the nearly linear dependence of decay time on $B_\perp$. The inset shows PL decay at 0, 10, and 18 T, and system response (SR, dashed curve).

(b) plots the PL peak positions up to $B=30$ T. There are two prominent PL peaks, both showing a quadratic spectral shift at low fields and a nearly linear shift at high fields, which is typical of exciton emission. Excitonic character of these PL lines was further supported by the presence of a clear onset of absorption in the PL excitation spectrum, and also by strong resonant Rayleigh scattering.[16] In each spectrum the PL peak at higher energy is assigned to recombination emission from the X^0 neutral exciton and the peak at lower energy is assigned to the X^- charged exciton. Assignment of the lower energy peak to the X^- rather than to a trapped exciton is in agreement with many other optical studies of electron-doped GaAs QW's.[3–7] The singlet (antiparallel electron spins) and triplet (parallel electron spins) states are not resolved in this sample, however, another sample having smaller PL linewidths showed X^- peaks similar to those found in a previous study of triplet X^-.[5] The X^- and X^0 peaks here had strong opposite circular polarizations at high fields, consistent with their peak assignments. The energy separation between the two peaks is the binding energy of the second electron, $\sim$1meV at high fields. With increasing temperature the spectral intensity shifts from the X^- peak to the X^0 peak due to thermal ionization of the second electron. These spectral features for the two peaks are quite similar to previous reports of X^- transitions in GaAs QW's.[3–7]

Results of time-resolved measurements at $T=0.5$ K are shown in Fig. 2. The inset displays the PL intensity of the X^- on a log scale as a function of time. At $B=0$ the X^- decay is rapid and closely follows the system response (dashed curve). Deconvolution of the system response from the PL decay curve at $B=0$ yields a decay time of $t\sim100$ ps, which is close to that observed for high-mobility 2D electron gas.[14–16] For fields applied perpendicular to the QW, the decay becomes increasingly longer at higher fields. At $B=18$ T it reaches $t=1.2$ ns, an order of magnitude longer than at $B=0$. (Note that the field-induced increase in the PL decay time is not due to changes in nonradiative decay channels, as indicated by the nearly constant total intensity of the two PL peaks in magnetic fields.) Figure 2 plots PL decay times at $T=0.5$ K for fields up to B=18 T. The solid circles and solid squares represent X^- data measured for two samples with electron densities differing by a factor of 1.4. These data are nearly identical. They demonstrate that the MCX *decay time is linearly proportional to the magnetic field*. In contrast, for fields applied parallel to the QW, the X^- decay time is independent of magnetic field, shown by the open circles. Furthermore, the decay time for the X^0 peak (not shown) does not show appreciable lifetime increase, and $t\leqslant150$ ps for all fields.[17] Rapid decay of X^0 is also found in a similar but undoped sample (crosses in Fig. 2).[16]

Combining these observations, it is apparent that the inhibited recombination found at high fields and low temperatures is only observed for the X^-, and only for perpendicular magnetic field. The four cases of X^0 and X^- with magnetic fields perpendicular and parallel to the QW layer are illustrated in Fig. 3(a). For the X^0, the exciton is confined only to the 2D layer since it is free to move in the QW plane independent of the magnetic-field direction. On the other hand, for the X^- the exciton complex is effectively 1D for B parallel since it is free to move along the field direction, yet is 0D for magnetic fields perpendicular to the QW layer. Thus

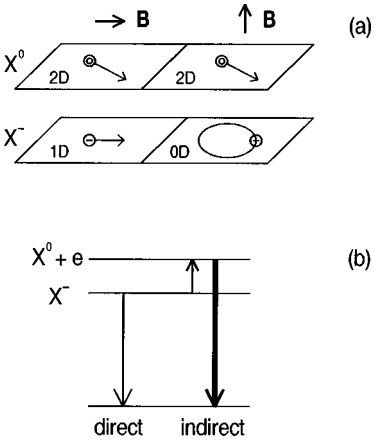

FIG. 3. (a) Schematic description for the center-of-mass motion of neutral X^0 and charged X^- excitons confined in a 2D plane for parallel and perpendicular magnetic fields. (b) Diagram of the two radiative processes for X^- recombination: the direct process of X^- emission which leaves an electron behind; and the indirect process in which the excess electron is first freed by ionization before the X^0 recombines.

the only configuration giving complete confinement in three dimensions is the latter case of X^- with B perpendicular, in agreement with the observed lifetime increase.

Concerning the free-exciton recombination in QW's, Feldmann *et al.*[18] first pointed out the importance of phase coherence volume for the center-of-mass motion of excitons. The coherence volume has a spatial extent which is usually much larger than the Bohr-orbit size. As a result, recombination radiation from free excitons emanates from a volume in which the radiating exciton is phase coherent. This phase coherence volume contains many unit cells radiating coherently, producing a macroscopic polarization. Thus the transition dipole strength and resulting radiative decay rate is a linear function of the coherence volume.[18–22] Feldmann *et al.* demonstrated this linear relationship experimentally through the dependence of exciton lifetime on the temperature-dependent homogeneous spectral linewidth.[18] Later, a similar relationship was demonstrated between exciton radiative lifetime and QD size for CuCl microcrystals.[23] Below, we model the present situation of MCX based on this coherence volume concept for excitons.

The characteristic volume for the magnetic confinement of X^- is given by

$$V_{MCX} = L\pi l_B^2 \propto 1/B_\perp , \tag{1}$$

where L is the well width (hole layer width in the present case), and $l_B = (c\hbar/eB_\perp)^{1/2}$ is the cyclotron radius. At small fields, V_{MCX} is larger than the intrinsic coherence volume V_0 at zero field, and the oscillator strength is determined by V_0 rather than V_{MCX}. At large fields, V_{MCX} becomes smaller than V_0, limiting the spatial coherence of X^-. The radiative decay time τ_r is then given by

$$\tau_r \propto \begin{cases} V_0^{-1} & \sim \quad \text{const} \quad (\text{at small } B_\perp) \\ V_{MCX}^{-1} & \propto \quad B_\perp \quad (\text{at large } B_\perp). \end{cases} \tag{2}$$

Although this simple model neglects effects of quantum confinement and magnetic field on the internal electron-hole (*e-h*) wave function of the exciton, the results in (2) are in remarkably good qualitative agreement with the data in Fig. 2, where t shows large, nearly linear increases at large $B_\perp$ fields, but it shows only small variation at lower fields.

In reality, however, the exciton internal wave function is strongly affected by quantum confinement and magnetic field. Effects of quantum confinement were studied theoretically by Takagahara,[24] taking into account the valence-band mixing and *e-h* exchange interaction. He showed that the exciton oscillator strength in a QD is a nearly linear function of the dot volume even in the intermediate confinement regime, primarily due to changing number of unit cells in QD's of different sizes. It has been known that the effective Bohr radius of an exciton decreases in a strong magnetic field,[25] known as ''magnetic shrinkage.'' This would result in a larger *e-h* wave-function overlap, and acting alone would produce a *shorter* decay time. Clearly, this prediction is contrary to the present observation of decay time *increase* in magnetic fields. Indeed, the observed X^0 decay time in an *undoped* QW shows only small changes at high fields (see Fig. 2). This demonstrates *experimentally* that even though a magnetic field modifies the internal wave function substantially, it does not lead to a large change in decay time in the present work. It is unclear why the magnetic shrinkage does not cause a large change in the X^0 decay time. However, for the confined X^-, the coherence volume effects could dominate over the internal wave-function effects, similarly to the case of quantum confinement for X^0.[24] The above considerations nevertheless explain the qualitatively good agreement between the observed behaviors of PL decay time and our model in spite of neglecting field- and confinement-induced effects on the internal wave function.

Inhibited exciton decay could also have a contribution arising from field-dependent changes of the mixing in the valence-band (*vb*) states. In the Luttinger model of QW *vb* levels, most levels contain several wave-function components having different angular momentum states.[26,27] For a given level, only certain components contribute to allowed optical transitions, the other components do not contribute. Thus if the magnetic field reduces the ratio of allowed to unallowed components of a hole level, the exciton will become ''dark'' and have a longer decay time.[28] Calculations indicate that these changes are small, typically much smaller than a factor of 2.[29] These changes in *vb* mixing do not appear to account for the order of magnitude increase observed in the decay time.

Figure 4 plots the measured PL decay times for both X^- and X^0 as a function of temperature. At $B=14$ T and $T=0.5$ K, there is a large difference between the decay times for X^- and X^0 excitons. This difference rapidly diminishes for increasing temperature, and for $T \simeq 3$ K the two decay times are nearly identical. Data for $B=6$ T show similar behavior but the effects are smaller. These behaviors can be explained by considering the thermal ionization of X^-. There are two competing channels for X^- decay, namely

$$X^- \rightarrow h\nu_- + e, \tag{3}$$

$$X^- \rightarrow X^0 + e \rightarrow h\nu_0 + e, \tag{4}$$

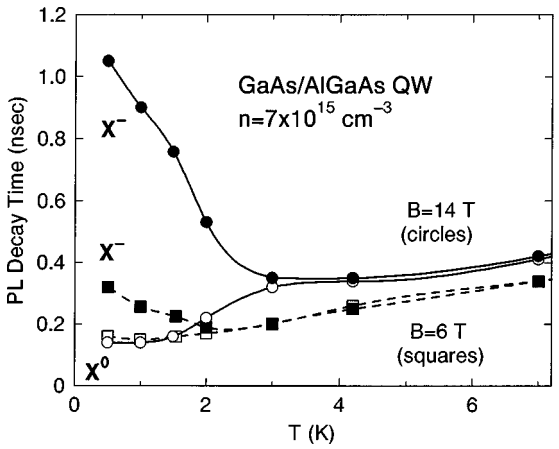

FIG. 4. Photoluminescence decay time of the X^- charged exciton versus temperature for perpendicular magnetic fields of $B=6$ and 14 T, for a GaAs quantum well.

where $h\nu_-$ and $h\nu_0$ are the corresponding photons energies. These decay channels are illustrated in Fig. 3(b). In the direct process (3), an electron recombines with the hole but leaves the remaining electron with excess energy. In the ionization process (4), X^- first loses the excess electron to the electron gas by thermal excitation, followed by a rapid X^0 recombination. At low temperatures, the direct process is not thermally activated, and most X^- recombine directly with a long decay time. At elevated temperatures, in contrast, the indirect decay channel (4) also becomes available for X^-, resulting in a rapid decrease of the X^- population.

The present work points out that charged excitons in a QW are confined by a perpendicular magnetic field to QD's having a size on the order of the cyclotron radius. These QD's are magnetically adjustable and their volume is inversely proportional to the magnetic field. The large increase in radiative lifetime of the magnetically confined charged exciton is attributed to a decrease in its coherence volume. Similar changes in radiative lifetimes are expected for other QW systems in high magnetic fields, and for nanofabricated QD structures.

Note added in proof. Recently, Finkelstein *et al.*[30] reported a time-resolved PL study of X^- in GaAs QW's.

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*Present address: Jet Propulsion Laboratory, Pasadena, CA 91109.

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