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Cyclotron-resonant exciton transfer between the nearly free and strongly localized radiative states of a two-dimensional hole gas in a high magnetic field

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Avoided crossing of the emission lines of a nearly free positive trion and a cyclotron replica of an exciton bound to an interface acceptor has been observed in the magnetophotoluminescence spectra of p-doped GaAs quantum wells. Identification of the localized state depended on the precise mapping of the anticrossing pattern. The underlying coupling is caused by an exciton transfer combined with a resonant cyclotron excitation of an additional hole. The emission spectrum of the resulting magnetically tunable coherent state probes weak localization in the quantum well.

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I. INTRODUCTION

Carrier localization is attracting strong current interest fueled, to some extent, by anticipated applications in quantum-information technology. For example, charged quantum dots may be used for storage purposes,[1] whereas, localized spins are considered as promising quantum bit candidates.[2] In that respect, it seems promising to also study carriers bound to defect atoms,[3] as a smaller extension of their wave function (compared to quantum dots) enhances protection from relaxation. Because of their weaker coupling with the nuclei,[4] valence-band holes appear especially attractive for information storage by localized spins. The holes are heavier than electrons; in optics, especially in high magnetic fields where the spectra split into multiple lines, this asymmetry yields a different hierarchy of binding and localization energies of excitonic complexes.[5]

On the other hand, small spatial extent also aggravates external manipulation. For example, matrix elements for optical excitation are strongly reduced. In addition, long-range coupling between confined excitations is expected to be rather weak. A possible solution may be coupling of the localized excitations to delocalized ones. This can be obtained by placing the defects close to a quantum well, serving as an interface channel. The quantum well may also be used for efficient optical excitation, after which the photoinjected (and possibly spin-polarized) carriers are exchanged with the localization centers. For deterministic coupling between localized and delocalized excitations, they must be brought in resonance where they show a pronounced avoided level crossing—very similar to the avoided crossing observed for tunnel-coupled electronic states in quantum dot molecules.[6]

In this paper, we report on the coupling between the nearly free and the strongly localized states of different excitonic complexes formed in a two-dimensional hole gas in a high magnetic field and study optical emission from the resulting magnetically tunable coherent state. (By "nearly free" we mean either mobile or, more likely, weakly localized on remote charges or width fluctuations, in contrast to the "strong localization" on nearby charges.) The interacting states are the positive trion[7] X^+ and the AX complex whose binding center A^- is a bare acceptor at an interface of the well. In a high magnetic field, the difference in their binding energy is compensated by a cyclotron excitation of the hole gas, enabling their coupling through resonant exchange of an electron-hole pair. This is observed in photoluminescence as an avoided crossing of the emission lines attributed to the X^+ and a cyclotron replica (CR) of the AX as shown in Fig. 1 (discussed further in detail).

In contrast to the previous studies of the shake-up (SU) effect,[8] we used a gas of holes instead of electrons. This was motivated by the higher hole mass, placing the replica CR-AX in the suitable energy range for achieving resonance with the X^+. On the other hand, in contrast to the previous studies of the hole shake-up of nearly free trions,[9,10] we detect an opposite replica (with cyclotron gap *increasing* emission energy) of a *localized* complex.

II. SAMPLES AND EXPERIMENTS

We have examined a selection of superior quality GaAs quantum wells, ranging in width between $w = 15$ and 40 nm. They were grown by molecular-beam epitaxy on a (001)-oriented semi-insulating GaAs substrate, employing the following growth sequence: 100-nm GaAs, 5-nm AlAs, 200-nm GaAs, and 150-nm $Ga_{0.65}Al_{0.35}As$, the superlattice consisting of 33 repetitions of 2-nm GaAs and 1-nm AlAs, 57-nm $Ga_{0.65}Al_{0.35}As$, w-wide GaAs quantum well, 40-nm $Ga_{0.65}Al_{0.35}As$, carbon δ doping, 80-nm $Ga_{0.65}Al_{0.35}As$, and

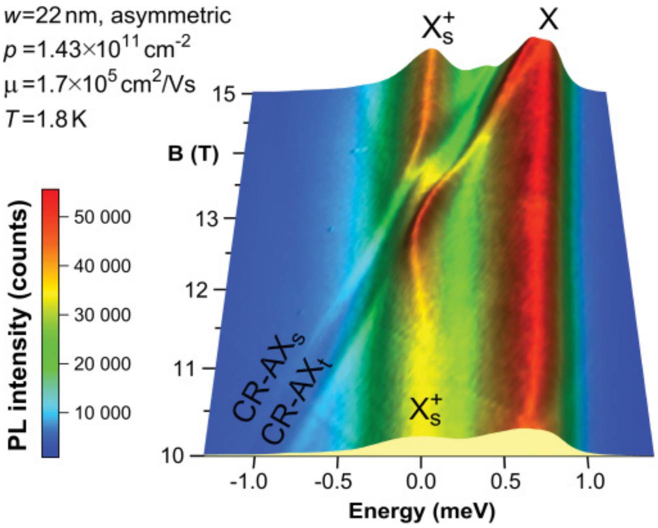

FIG. 1. (Color online) Evolution of the photoluminescence spectrum in the σ^- polarization as a function of magnetic field B. In fields around $B_c = 13.2$ T, avoided crossings between the pair of exchange-split CR-AX lines and the X^+ line are clearly observed, demonstrating the coupling via cyclotron-resonant exchange of an electron-hole pair. For better visibility, at each magnetic field, the energy is linearly shifted to the X^+ resonance (slope 0.7 meV/T, constant 1520.8 meV).

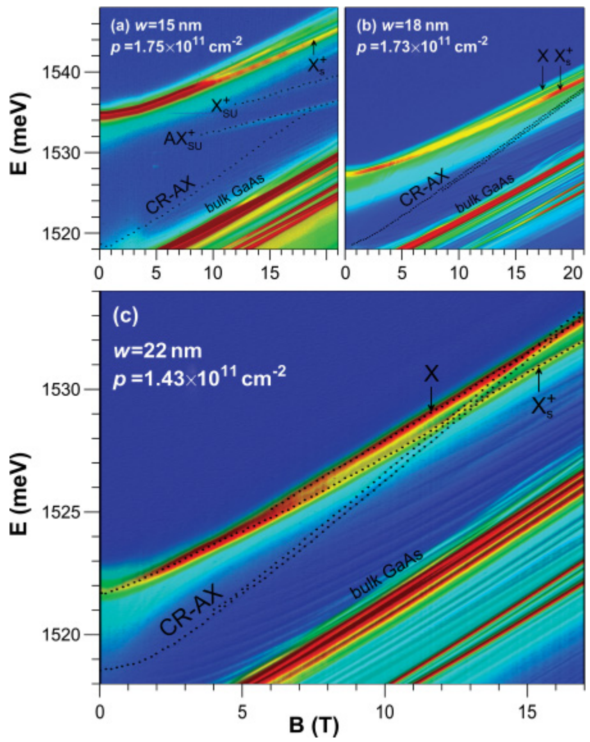

FIG. 2. (Color online) Evolution of the photoluminescence spectra of the narrow symmetric quantum well of width (a) $w = 15$ nm and two wider asymmetric wells of widths (b) $w = 18$ nm and (c) 22 nm in magnetic field B, collected in polarization σ^- (higher intensity), at low temperature $T = 1.8$ K, and under laser power density $P = 20\ \mathrm{mW/cm^2}$. The identified lines are as follows: exciton (X), positive trion family (X_s^+, X_{tb}^+, and X_{td}^+), acceptor-bound complexes (AX and AX^+), SU lines, and CRs. The color scheme is the same as in Fig. 1.

the 5-nm carbon δ-doped GaAs cap. An exception is the $w = 15$-nm well, doped symmetrically on both sides.

In all investigated samples, the hole mobility measured at $T = 4.2$ K was nearly the same, $\mu \approx 10^5\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$. The hole concentration was accurately estimated from the quantum Hall-effect measurements (performed in the van der Pauw configuration, in parallel with photoluminescence). In the dark, it varied slightly in the range of $p = (1.2\text{–}1.9) \times 10^{11}\ \mathrm{cm^{-2}}$; under illumination, it decreased linearly with the excitation power density (by less than 10% in the actual experiments.)

Photoluminescence was excited by the 632.8-nm line of a helium-neon laser, with the photon energy below the band gap of the barrier. The applied laser power density varied from $P = 5$ to $20\ \mathrm{mW/cm^2}$. The measurements were performed in a bath liquid-helium cryostat at temperatures varying from $T = 1.8$ to 4.2 K. A magnetic field was applied in the Faraday configuration; it was changed with a small step $\Delta B = 0.05$ T up to the maximum value $B = 22$ T. The fiber optics was used, with a linear polarizer and a quarter-wave plate placed close to the sample. The σ^- and σ^+ helicities were switched by reversing the field direction. The spectra were analyzed using 0.5-m- and 1.0-m-long monochromators and a liquid-nitrogen-cooled CCD camera.

III. RESULTS AND DISCUSSION

In Fig. 2, the field evolution of the photoluminescence spectrum in polarization σ^- (corresponding to the higher emission intensity) is presented for one symmetric ($w = 15$ nm) and two asymmetric wells ($w = 18$ and 22 nm). Examples of high-field spectra are shown in Fig. 3. In each spectrum, we have identified, in the high-energy sector, several strong emission lines, attributed to the nearly free exciton (X) and spin-singlet trion (X_s^+). The complete trion family,[11,12] singlet (X_s^+), bright triplet (X_{tb}^+), and dark triplet (X_{td}^+), has been resolved exclusively in the symmetric $w = 15$-nm well.

We use the term nearly free to signify that, although efficient emission from an excitonic complex generally requires some lateral localization[13] (by remote-charged impurities or well-width fluctuations), for a nearly free state, its effect on recombination energy is not substantial. Also distinctive is the effect of localization on the dynamics in the growth direction: The emission energy is sensitive to the well width for the nearly

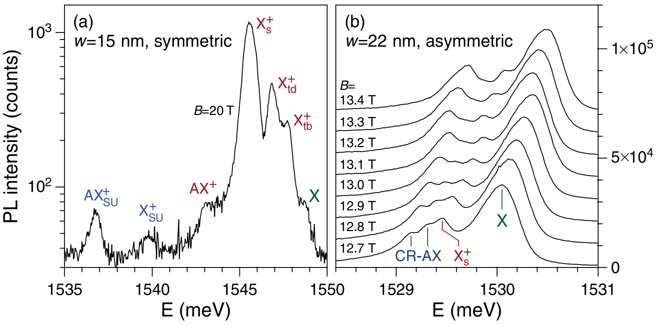

FIG. 3. (Color online) Examples of the σ^--polarized high-field photoluminescence spectra of two wells from Fig. 2: (a) $w = 15$ nm, symmetric; (b) $w = 22$ nm, asymmetric.

free states in contrast to the "strongly bound" ones, clung to one side of a well as the result of attraction to an interface acceptor.

Emergence of multiple emission lines from all of these excitonic complexes generally requires sufficient power density and/or magnetic field. Hence, all spectra in Fig. 2 begin at low fields with only one strong line (X or X_s^+) at high energy. In a sufficiently high field, it splits into two lines, gradually separating in energy with a further increase in the field. Their interpretation was aided by realistic numerics,[12] which included effects of finite well width, anisotropic hole mass, Landau level and quantum well sub-band mixing, and carrier correlations but excluded light-heavy-hole sub-band mixing[14] and disorder.[13]

On the other hand, the lower-energy part of each shown spectrum also contains multiple weaker lines even for the lowest magnetic fields and weakest excitations. Most of them shift with the magnetic field in parallel to neutral and charged excitons with the same slope of energy-field dependence, saturating at 0.7 meV/T at higher fields. However, in the spectra collected for higher power densities, we also distinguish two other kinds of lines, with either lower or higher slopes (i.e., either departing from the main lines or approaching them) and interpreted as hole cyclotron replicas of different excitonic complexes.

In particular, in the symmetric $w = 15$-nm well, we have observed shake-up lines of the nearly free and acceptor-bound positive trions (the latter with the A^- in the well). Their energy-field slope in high fields is 0.4 meV/T. The difference from the main lines matches the hole cyclotron energy determined from the cyclotron resonance of low-density GaAs wells (interpolation of data from Ref. 15 to $w = 22$ nm gives the slope 0.28 meV/T yielding an effective mass $m_h = 0.38$). Remarkably, these shake-up lines cannot be resolved in the spectra of our wider wells where they fall inside the emission range of bulk GaAs.

However, it is the other group of lines whose behavior we have found most intriguing. Two of them, denoted as CR-AX_s and CR-AX_t (as we justify further) in Fig. 2, are detected in the spectrum of each well, and remarkably, in each one, they follow a virtually identical energy-field dependence. In particular, regardless of the well width w, at $B = 0$, they begin at the same energy of 1518 meV and with the same splitting of 0.3 meV. When the magnetic field is switched on, they linearly approach the high intensity lines, with the energy-field slope saturating at 1.0 meV/T in high fields. Comparison of this slope with those of X and X^+ yields a field-independent difference of $\gamma = 0.3$ meV/T, i.e., exactly opposite that characterizing the hole shake-up lines.

At this point, let us express a few obvious conclusions: (i) The fact that the CR-AX lines repeat unchanged in wells of different widths ($w = 15$–40 nm) precludes their origin in the interior of the well. (ii) The low energy at $B = 0$ precludes the origin inside the barrier. Furthermore, the energy position several meV below the free X and X^+ states inside the (even fairly wide) well suggests strong localization, most likely on an acceptor. (iii) The energy-field slope (compared to X and X^+) reveals recombination accompanied by a hole cyclotron relaxation. The CR-AX transitions are, hence, related to the previously observed "combined exciton-cyclotron resonance,"[16] which is an *electron* cyclotron replica of the *nearly free* exciton.

The most intriguing effect is observed when the pair of CR-AX lines approach the trion line: Both X^+/CR-AX_s and X^+/CR-AX_t crossings are clearly avoided. We have only been able to observe this effect in the $w = 22$-nm well where it occurs at an accessible magnetic field $B_c = 13.2$ T and the lines are sufficiently sharp (0.4-meV full width at half maximum, compared to 0.2-meV thermal broadening). In a narrower $w = 18$-nm well, extrapolation of the available data points predicts the prohibitively high anticrossing field $B_c \approx 28$ T. In a wider well of $w = 25$ nm, the resonance occurs at $B_c < 4$ T, and the anticrossing is not resolved due to a larger linewidth in this field range.

The relevant sector of Fig. 2(c) has been magnified in Fig. 1, which clearly displays both avoided crossings—a convincing signature of the mixing of the corresponding states[17] CR-AX and X^+. Figure 1 also makes it clear that the intensities of the CR-AX lines become greatly enhanced in the anticrossing region—as a simple consequence of the mixing with the more radiative nearly free trion state. In higher magnetic fields, above the anticrossing region, the CR-AX and X^+ lines resume their initial energy-field dependences and intensities.

As anticrossing of the CR-AX lines with the X^+ line implies coupling between the corresponding radiative states (initial states for recombination), it also indicates spatial proximity of the CR-AX recombination to the quantum well (hosting the X^+). Combination of this fact with the earlier conclusions (i)–(iii) allows us to attribute the CR-AX lines to the cyclotron replica of an excitonic complex bound to an acceptor at the interface separating the GaAs well from its GaAlAs barrier. Emission from such a bound AX complex and its shake-up AX_{SU} have been observed at lower fields in an undoped quantum well,[18] but the detection of the $CR - AX$ line (requiring the hole gas) is a significant result of this work.

That the relevant bound excitonic state is neutral rather than charged follows from the exact course of emission lines near the anticrossing. The hypothetical charged complex AX^+ would consist of a bare interface acceptor (negative point charge), one electron, and three holes. Previous calculations[10] hint that its ground state would have a doublet spin configuration, corresponding to the unpolarized holes (total three-hole spin of 1/2; we adopt a convention assigning spin projections $\pm 1/2$ to the "up" and "down" heavy-hole states) with the quadruplet configuration (polarized holes) excited by several meV. Thus, only the doublet state AX_d^+ would be populated at low temperatures. In the relevant polarization σ^-, it would have two recombination channels, ending in a singlet or triplet state of the charged acceptor ($AX_d^+ \rightarrow A_s^+$ or A_t^+, where A^+ has two holes bound to a bare acceptor). Thus, its emission would be split by a small exchange energy gap in the final state (A^+; in the optically active angular momentum channel[10] $M = -1$). At first sight, this agrees with the observed 0.3-meV splitting of the CR-AX lines, but the exact course of the anticrossing lines turns out incorrect.

The following alternative scenario involves a neutral complex AX, which has an electron and two holes (all inside the well) bound to a bare interface acceptor A^-. Its two spin configurations of the pair of holes, singlet (AX_s) and triplet (AX_t), are separated by an exchange gap of a fraction of meV. Both configurations should be populated in the experiment,

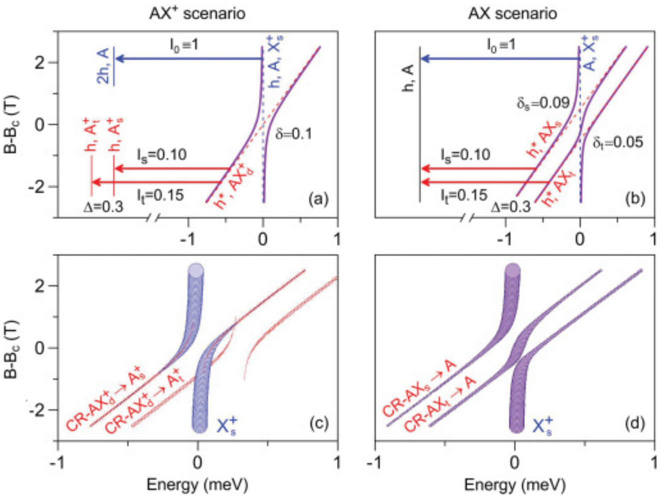

FIG. 4. (Color online) Top: Energy-field dependence for the initial and final states of two candidate scenarios for the observed anticrossing pattern of Fig. 1 (left/right: cyclotron replica of a charged/neutral acceptor-bound excitonic complex mixed with a free trion with the two-hole exchange splitting in the final/initial configuration). Magnetic field B is counted from the resonance field B_c and the energy—from the trion position at resonance. Exchange and coupling energies (Δ and δ) are quoted in meV. Oscillator strengths away from resonance (I) are defined relative to the free trion. Bottom: Corresponding field dependences of the emission energy and intensity (indicated by dot radii). Comparison with Fig. 1 allows positive identification of the CR-AX transition.

and each would have a single σ^--polarized recombination channel, ending in the neutral acceptor A (one hole bound to a bare acceptor). Thus, emission from a neutral complex is split by a singlet-triplet exchange in the initial state, naturally explaining the observed 0.3-meV splitting of the CR-AX lines. As we discuss below, it also correctly predicts details of the anticrossing pattern.

Let us now compare, in more detail, these two scenarios, involving either a charged or a neutral excitonic complex. In each case, the recombination event is accompanied by a cyclotron relaxation of a nearby hole, thermally excited to a higher Landau level. In a particular field, when the hole cyclotron energy equals the binding energy of the AX complex, the cyclotron transition brings to resonance the free and localized states, thus, allowing their efficient coupling (by an exciton transfer). Figure 4 presents the energy-field dependence for the relevant initial and final states and the connecting transitions, obtained in a simple phenomenological model.[19] The exchange gap Δ and the relative intensities I of the unmixed transitions were read from the experimental spectra (away from the crossings). For the "charged" scenario (left), degeneracy of the two final states $h + A_s^+$ and $2h + A$ (i.e., zero A_s^+ binding energy in the relevant $M = -1$ channel) is needed (and, actually, justified by the previous calculation[10]) to maximally reduce the predicted anticrossing pattern. Yet, regardless of the coupling constant δ, the experimental behavior cannot be reproduced as shown in Fig. 4(c) for $\delta = 0.1$ meV. For the "neutral" scenario (right), a single common final state $h + A$ removes the need for additional degeneracy. Moreover, assuming two coupling constants$\delta = 0.05$ and 0.09 meV (distinguished by spin configuration of the AX) produces a convincing match with the experiment, which is the basis for our interpretation (and notation) of the CR-AX lines.

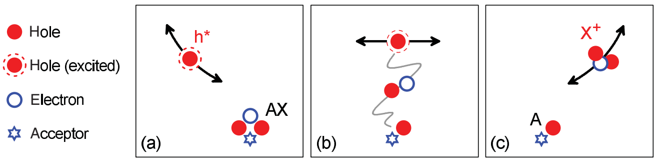

FIG. 5. (Color online) Schematic of the coupling between the acceptor-bound exciton AX and the nearly free trion X^+ through the exchange of an electron-hole pair combined with a hole cyclotron transition.

Having determined the relevant radiative states $h^* + AX$ and $A + X^+$, let us now discuss their coupling mechanism. The responsible process is the transfer of an electron-hole pair as illustrated in Fig. 5: A hole thermally excited to a higher Landau level (h^*) approaches an AX (in the singlet or triplet spin state). When their constituents engage in the Coulomb interaction, the hole relaxes to the lowest Landau level, releasing a quantum of cyclotron energy ($\hbar\omega_c \approx 4$ meV at the resonant magnetic field $B_c = 13.2$ T), which is used to unbind an e-h pair from the acceptor (breaking up the AX) and to bind it to the hole (forming an X^+). Of course, the reverse process is equally possible, beginning with an X^+ approaching an A and ending with the h^* leaving the AX. Remarkably, different localizations of the transferred e-h pair in the mixed configurations will make the coupling strength δ sensitive to the actual extent of the nearly free state and/or to the average distance between the acceptors.

Finally, Fig. 2(c) also reveals anticrossing of the CR-AX lines with the free exciton. This is straightforward to explain, in light of the above, the only difference being that a larger cyclotron energy $\hbar\omega_c \approx$ meV at the higher field $B_c \approx 16$ T brings the AX in resonance with the exciton instead of a trion: $h^* + AX \leftrightarrow h + X + A$.

IV. CONCLUSION

In magnetophotoluminescence spectra of positively doped quantum wells, we have demonstrated coupling between the nearly free and strongly localized excitonic complexes X/X^+ and AX, brought in resonance at a particular high magnetic field by an additional hole cyclotron transition. Coupling occurs through the exchange of an electron-hole pair. The result is a complex magnetically tunable coherent state whose optical spectrum probes weak localization in the quantum well.

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