Optically induced charge conversion of coexistent free and bound excitonic complexes in two-beam magnetophotoluminescence of two-dimensional quantum structures

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We report on extensive polarization-resolved photoluminescence (PL) studies of a variety of excitonic complexes formed in high-quality symmetric GaAs quantum wells containing a high-mobility two-dimensional (2D) hole gas in a broad range of magnetic fields from 0 to 23 T and under two-beam illumination, allowing for dynamical control of the hole concentration beyond the point of conversion from *p*- to *n*-type structures. We have demonstrated charge conversion between positive and negative complexes bound to acceptors in the well, differing from the charge conversion of free trions due to charge reflection symmetry breaking by a fixed impurity, leaving a qualitative trace (exchange splitting) in the PL spectrum. The effect of switching between the electron and hole gases (in the same well) on different emission lines has also allowed us to distinguish the (direct and cyclotron-satellite) emission lines from positive trions moving almost freely in the quantum well and bound to nearby ionized acceptors in the barrier, thus demonstrating their coexistence in high-quality structures.

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I. INTRODUCTION

Continuous improvement of the quality of semiconductor nanostructures and of the sensitivity of probing techniques reveals subtlety of their optoelectronic characteristics and invites deeper understanding of the underlying fundamental physics.[1] The structures relevant for this work are quasi-two-dimensional (2D) quantum gases with thicknesses not much greater that the effective Bohr radius and concentration corresponding to about one filled Landau level in accessible high magnetic fields. Their quality (synonymous with lateral uniformity on a short length scale and rarity of local scatterring or binding centers) is measured by the carrier mobility.[2–4]

Of the two kinds of carriers, it is the valence band holes which have recently received greatly increased attention. Their magneto-optical spectra are not equivalent to those of the electrons—predominantly because of the larger effective mass yielding different relations between the binding energies of neutral and charged complexes.[5–7] An important contrast is also found in the weaker coupling of the holes with the nuclei,[8,9] making the localized hole spins more attractive for storage of quantum information.[10,11] The hole mobilities have now reached $\mu = 10^6$ cm^2/Vs in the best *p*-type GaAs structures.[12,13]

Early photoluminescence (PL) studies of 2D hole gases in high magnetic fields were largely focused on the identification of positive trions,[14–19] in analogy to the previously established negative trions in electron gases.[20,21] However, the later, increasingly precise, measurements from higher-mobility structures have revealed great complication of the spectrum, with multiple emission lines emerging above a few tesla, varying in intensity, polarization, and energy and differently dependent on magnetic field, composition, and width of the structure and on illumination or simply varying from sample to sample.[22–24] Although this complexity is not unexpected but naturally attributed to variety of possible excitonic complexes interacting with the surrounding hole gas and the however sparse imperfections of the structure, understanding of the particular lines has been problematic. Numerical calculations certainly help in predicting binding energies and oscillator strengths and their trends, but their accuracy is limited, even for the simplest complexes like excitons or trions (not surprising, given the strong Coulomb mixing of high single-particle levels and significant coupling with the surrounding lattice or free carriers).[25–27]

The difficulty in unravelling the rich PL spectra available from present experiments can also be viewed from the opposite, conceptual perspective. It is predictable—and known from computations alone—that the considered physical systems may host various excitonic complexes (bound states of a small number of electrons and holes), such as excitons or trions.[28–31] These complexes may be laterally tied to localization centers of a different nature and placement, such as width fluctuations[32] or ionized impurities positioned differently with respect to the 2D layer.[33] And, certainly, they may have multiple recombination channels, due to energy splittings in the initial or final state (e.g., spin-singlet versus triplet[34]) or assisted decay processes involving excitation of the surrounding atomic lattice or gas of free carriers.[35] Again, for the lack of precise quantitative predictions, unambiguous identification of these transitions in real PL spectra is problematic. Hence, while demonstration of many anticipated transitions and the underlying effects has been reported (e.g., electron and hole shake-up[36,37]), some rather fundamental questions of relative stability of complexes or intensity of lines still remain controversial.

For example, based on a series of measurements Volkov *et al.*[22] questioned the previous assumption that recombining trions are essentially free particles. The claim of lateral confinement of optically active trions resonated well with some experiments[38,39] and a later realization[40] that the 2D translational symmetry must be broken for the (eventually observed[41]) direct recombination of the "dark" spin-triplet trion. But it contradicted a contemporary study of emission

temperature dependence by Ron *et al.*[42] And it was later spectacularly disproven by direct measurements of trion mobility.[43–45]

The aim of this work has been bridging the gap between the complexity of sensitively measured magneto-PL spectra and the multitude of predicted transitions. In particular, we have demonstrated coexistence and coemission of laterally nearly free and impurity bound trions. More generally, we have identified in our complex spectra essentially all of many lines originating in the 2D gas, thereby confirming various predicted cyclotron assisted recombination processes. This was possible by using superior-quality p-doped samples, recording PL with high spectral resolution, at low temperature, in high magnetic fields, and under combined two-beam illumination, enabling dynamical control of the carrier concentration (beyond the point of conversion to an n-type structure) and critical comparison with the tendencies found from own numerical simulations. Especially important was the possibility to immerse each considered radiative complex in the gas of either holes of electrons in the same sample, combined with sufficient sensitivity to detect several weak lines in addition to the dominant exciton and trion emission (e.g., cyclotron replicas). This allowed us to demonstrate charge conversion between positive and negative excitonic complexes bound to acceptors inside the well and to contrast it with the known[5,17] charge conversion of free trions.

II. SAMPLES AND EXPERIMENTS

The samples used in our experiments were high-quality $w = 15$ nm wide GaAs/$Ga_{0.66}Al_{0.34}As$ quantum wells with energy gaps of 1.519 eV in the well and 2.128 eV in the barrier, symmetrically δ doped with carbon in both barriers. They were grown by molecular beam epitaxy on the (001)-oriented semi-insulating GaAs substrate, with the following growth sequence: 50-nm GaAs, a superlattice consisting of 10 repetitions of 5 nm of GaAs and 5 nm of AlAs; a 200-nm GaAs, 150-nm $Ga_{0.66}Al_{0.34}As$ superlattice consisting of 33 repetitions of 2 nm of GaAs and 1 nm of AlAs; a 20-nm $Ga_{0.66}Al_{0.34}As$, carbon δ doping, 57-nm $Ga_{0.66}Al_{0.34}As$ superlattice consisting of seven repetitions of 2 nm of GaAs and 1 nm of AlAs; a 15-nm GaAs quantum well superlattice consisting of 7 repetitions of 2 nm of GaAs and 1 nm of AlAs; 59 nm of $Ga_{0.66}Al_{0.34}As$, carbon δ doping; 78 nm of $Ga_{0.66}Al_{0.34}As$; and a 5-nm carbon δ-doped GaAs cap.

In all investigated samples, the hole mobility measured at $T = 4.2$ K was nearly equal, $\mu = 10^5$ cm^2/Vs, while the hole concentration measured in the dark varied in the range of $p = (1.2 - 1.9) \times 10^{11}$ cm^{-2}. Photoluminescence was excited by a combination of two beams: a "red" 720-nm line of a titanium sapphire tunable laser with the photon energy $E = 1.72$ eV (below the band gap of the barrier) and a "green" 514-nm line of an ion argon laser with the photon energy $E = 2.41$ eV (above the band gap of the barrier). As shown previously,[5,17] by selective photoexcitation inside and outside of the quantum well, thanks to different electron and hole kinetics in the induced electric field, this allows control of the dynamical population and even of the sign of majority carriers inside the well.

The measurements were performed in a bath liquid helium cryostat at temperatures varying from $T = 1.8$ to 4.2 K. Fiber optics was used, with a linear polarizer and a wave-quarter plate placed close to the sample. The spectra were analyzed with a 0.7-m-long monochromator and a nitrogen-cooled CCD camera with resolution exceeding 0.02 nm. The measurements conducted in a magnetic field employed a Faraday configuration, with the field increased with a small step $\Delta B = 0.05$ T, up to the maximum value, $B = 23$ T. The switch between the σ^- and σ^+ helicities was realized by reversing the field direction.

III. RESULTS AND DISCUSSION

Let us begin the discussion with the results obtained in the absence of the magnetic field and illustrated in Fig. 1 with the set of spectra collected in one representative sample using different combinations of red (720-nm) and green (514-nm) laser beams. When only the red laser was applied, a single slightly asymmetric emission line was observed in each sample. Its energy position and shape changed slightly between different samples, but they were essentially independent of the laser power density, varied in a broad range. This implies that in each given sample the hole concentration in the well remained constant regardless of the power density. Based on our previous study[23] we attribute this line to the singlet state of a positively charged exciton (X_s^+).

When a green laser is additionally applied the spectrum changes. In Fig. 1 we kept a constant laser power density of the red beam, $P_{red} = 25$ mW/cm^2, and used several power densities of the green beam, $P_{green} = 1.5 \times 10^{-3}$, 1.5×10^{-2}, 0.75, and 15 mW/cm^2. On addition and increase of the green excitation, the X_s^+ line initially shifts to higher energies. This is

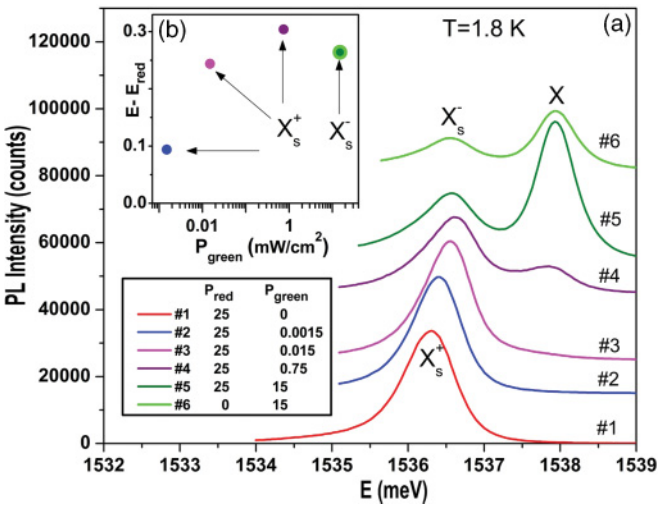

FIG. 1. (Color online) Comparison of photoluminescence spectra recorded in the absence of a magnetic field, under excitation with different combinations of red and green lasers, in the symmetric GaAs quantum well of width $w = 15$ nm and dark hole concentration $p = 1.51 \times 10^{11}$ cm^{-2}. (Inset) Trion emission energy E as a function of green laser power density P_{green}, relative to pure red illumination (#1). The change of assignment from positive to negative trions beyond the maximum in $E(P_{green})$ indicates dynamical conversion of the structure from p to n type.

consistent with the gradual decrease of the hole concentration in the well[5,17] through the band-gap renormalization effect. Further increase of the green excitation also causes emergence of a new peak at higher energy, associated with the neutral exciton (X). Distance of 1.35 meV between the X^+ and X lines is well consistent with the expected trion binding energy in our structures, and the transfer of intensity from trionic to excitonic emission is a convincing signature of decreasing charge accumulation in the well. We have also checked that for comparably intense green excitation ($P_{\text{green}} = 15$ mW/cm^2 in Fig. 1) removing the red laser leaves the spectrum nearly unchanged (apart from the trivial overall reduction in proportion to the total excitation power, $P_{\text{red}} + P_{\text{green}}$).

Since it is already well established[5,17] that sufficiently intense above-barrier illumination converts the structure from the p into the n type, we have not aimed at locating the precise point at which this conversion occurs. However, as it will become evident from the spectra collected in high magnetic fields (discussed further), it certainly does occur in the range of applied power densities of the green laser, with the critical value of P_{green} (for the sample of Fig. 1) of the order of 1 mW/cm^2. At lower values of P_{green} the majority carriers in the well remain holes and the charged excitonic complex is the positive trion X^+, while above this value of P_{green} the well contains predominantly electrons and the trion observed in photoluminescence is negative, X^-. What makes detection of the p- to n-type conversion in Fig. 1 relatively more difficult is, most of all, the previously established[5,17] near degeneracy of the binding energies of positive and negative trions in the absence of magnetic field, keeping the trion emission energy nearly continuous through the point of conversion between X^+ and X^-.

Furthermore, P_{green} from the range used in Fig. 1 is unable to induce sufficient electron concentration to quench the neutral exciton emission and, in consequence, the behavior of the exciton-to-trion peak ratio near the point of conversion is altered by different positive and negative trion kinetics (instead of simply measuring the charge concentration). However, a cleaner signature of the carrier concentration passing through zero can be found in the trion emission energy, thanks to the already-mentioned effect of band-gap renormalization: While the initial increase of the trion emission energy as a function of P_{green} is attributed to the decreasing hole concentration (through the weakening exchange of the recombining hole with the surrounding gas), its subsequent decrease beyond $P_{\text{green}} = 0.75$ mW/cm^2 points to the concentration rising again (only, this time, of electrons instead of holes). Let us stress, however, that the interpretation of our limited zero field data of Fig. 1 in terms of the p- to n-type conversion at P_{green} of about 1 mW/cm^2 is based more firmly on the high field data discussed further.

Let us now turn to the magnetic field measurements, in which we applied the above-described method of tuning concentration of the 2D hole gas to investigate the major issue of trion localization. Indeed, most of the following measurements were repeated for different combinations of red and green laser beams, whose effect on the carrier type and concentration was earlier tested in the absence of the field (cf. Fig. 1). Figure 2 presents (in all its complexity) the magnetic field evolution of the photoluminescence spectrum excited by

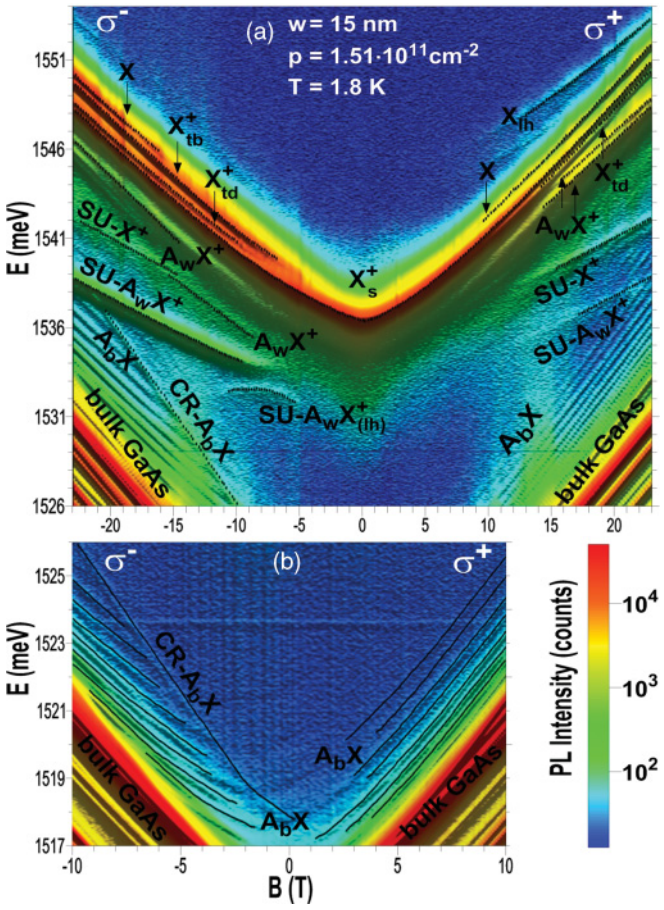

FIG. 2. (Color online) Evolution of the photoluminescence spectrum of the symmetric GaAs quantum well of width $w = 15$ nm, with a high-mobility hole gas of (dark) concentration $p = 1.51 \times 10^{11}$ cm^{-2}, in magnetic field B varied from 0 to 23 T, collected in polarizations σ^+ and σ^-, at low temperature $T = 1.8$ K, and under power density $P = 25$ mW/cm^2 of the red laser. Emission intensity is color coded in logarithmic scale. A great number of recombination lines have been indicated; their assignment is explained in the text (e.g., A_b and A_w are acceptors in the barrier and in the well, "SU-" denotes shake-up, and "CR-" denotes cyclotron resonance). A pair of frames, (a) and (b), show adjacent areas of the map, recorded separately in two different spectral windows.

the red laser only, with the power density $P = 25$ mW/cm^2 (corresponding to the red curve in Fig. 1 for $B = 0$). It is shown in fairly standard form of color maps of emission intensity as a function of emission energy and magnetic field. The two light polarizations are shown separately, with σ^- (strong intensity) and σ^+ (weak intensity) being indicated at the negative and positive values of B, respectively.

The complementary Fig. 3 presents variations of the spectrum as a function of the power density of an additional green laser, in a particular high magnetic field $B = 23$ T (in which the greatest variety of emission lines related to the 2D gas recombination are observed). Clearly, in addition to revealing the dependence on carrier concentration, by using intensity curves rather than color maps, this graph also provides much greater detail in terms of the shapes, widths, and heights of the individual peaks than Fig. 2.

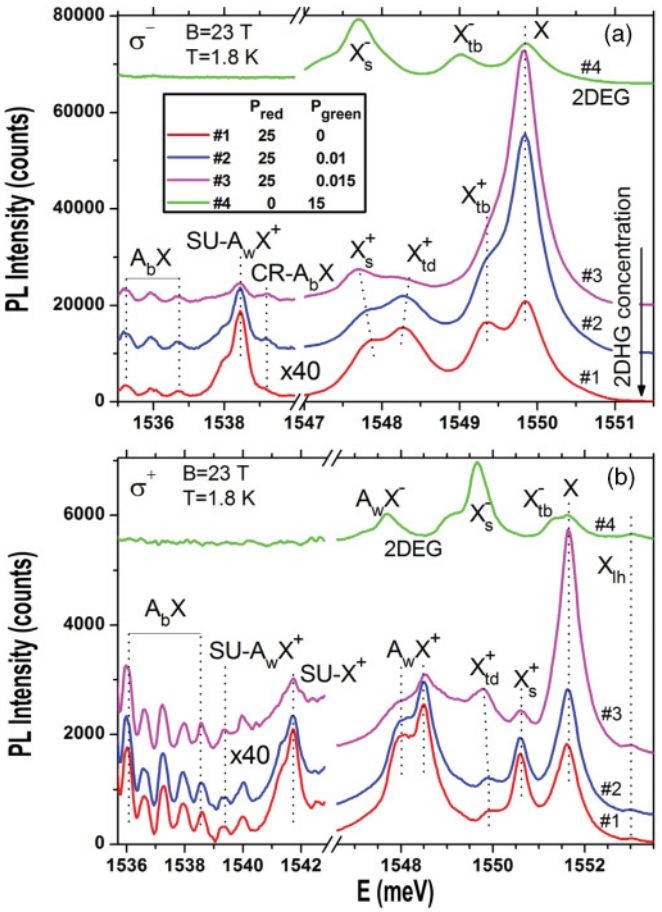

FIG. 3. (Color online) Similar to Fig. 1 but for the spectra recorded at magnetic field $B = 23$ T, resolved into the two polarization channels, σ^- (a) and σ^+ (b). The horizontal (energy) axis is broken between $E = 1543$ and 1547 meV, and the energy scale differs in the two disconnected ranges. The assignment of lines is explained in the text, notation (X, A, SU, and CR) is the same as in Fig. 2, and "2DEG" and "2DHG" denote 2D electron and hole gas.

Careful analysis of the field evolution of the emission spectra recorded for different concentrations or holes or electrons, aided with the realistic numerical calculations[23,27] (exact diagonalization of the relevant few-electron-hole systems) allowed us to convincingly identify and interpret all detected emission lines related to the 2D gas. They can be divided into the following two main groups.

The common feature of the first group of lines is their strong dependence of their energy position on the quantum well width, namely all these lines shift to higher energies in the narrower wells, as has been observed in numerous studies, including ours.[24] Furthermore, they generally occur in the higher energy sector of the spectrum. This shared behavior originates from radiative recombination of the electron-hole complexes whose wave functions are located inside the quantum well and spread through its whole width. While for all optically active complexes some form of lateral localization (on various ubiquitous imperfections of the well, e.g., width fluctuations or shallow impurities) is generally expected, this lateral confinement may be strong or weak.[46] This gives rise to further classification of lines. By weakly localized or "nearly free" states we shall mean those for which localization has an insignificant effect on the emission energy, even though it may be essential for breaking the symmetry-related optical selection rules. The opposite is true of the "strongly localized" states.

The group of width-sensitive lines can also be further divided into those associated with the recombination of neutral and charged complexes. Of the weakly localized states, this is a distinction between the excitons and trions. They can be reliably distinguished in our spectra excited with varied two-beam illumination, due to the clear correlation of their relative intensity with the carrier type and concentration (the emission from neutral complexes grows stronger at the expense of the related charged ones when the carrier concentration is decreased).

The second group of lines which generally occur in the lower energy sector of the photoluminescence spectrum are distinctly insensitive to the width of the quantum well (at least above certain minimum width; after all, they, too, originate from inside the quantum well). These lines are attributed to the radiative recombination of electron-hole complexes which are strongly localized by deep Coulomb potentials produced by the residual ionized acceptors (all investigated samples being nominally p type) located in the barriers, at some distance away from the interface at the edge of the quantum well. These complexes also can be characterized by the total electric charge drawn in the well around the remote acceptor. In addition to strong lateral confinement, Coulomb attraction to the external charged impurities also causes clinging of the captured electron-hole complexes to one side of the quantum well, which is responsible for the distinctive width insensitivity of the corresponding lines in the photoluminescence spectrum.

Let us now focus on a few particular, most interesting, lines identified in Fig. 2 and discuss them in more detail. Beginning with the first group corresponding to the width sensitive transitions of weakly laterally localized neutral and charged excitonic bound states, the highest energy lines belong to the pair of radiative states of the neutral exciton ($X = e + h$), distinguished by the valence subband occupied by the hole. Neglecting (for the sole purpose of commonly accepted labeling) the intersubband mixing of light and heavy hole states ("lh" and "hh" respectively) induced by the Coulomb interaction with the electron, the upper and lower excitonic lines are denoted in Fig. 2 as X_{lh} and $X \equiv X_{\mathrm{hh}}$. Of these two transitions X is considerably stronger than X_{lh}. However, the intensity of the X_{lh} line can be significantly enhanced relative to X by an increase of the green laser power density (as a straightforward consequence of an increased relative occupation of the higher hole energy level caused by optical excitation with higher-energy photons).

Next to the exciton lines, at the slightly lower energies, we find a series of transitions attributed to recombination of the whole family of bound states of the positive trion complex ($X^+ = 2h + e$), including the spin-singlet state (X_s^+), observed in all magnetic fields, as well as two spin-triplet states, bright (X_{tb}^+) and dark (X_{td}^+), both emerging only at sufficiently high fields. With the decrease of the hole concentration (conveniently realized by additional green laser illumination) the emission intensities from all trion states decrease, thereby confirming the expected dependence of the

exciton to trion population ratio on the total charge density in the quantum well.

Remarkably, when the addition of sufficiently intense green laser illumination results in increasing the average number of confined electrons above the number of holes, i.e., conversion of the structure from the p- to the n-type (judging from Fig. 1, this occurs at the green laser power densities exceeding about 15 mW/cm^2), the positive trion lines (X^+) are rather abruptly replaced in the emission spectra by a differently positioned set of lines attributed to the recombination of the (analogous) family of the negative trion (X^-). This sudden switch in the positions of trion lines in the spectrum is accompanied by a discontinuous drop in the ratio of exciton to trion emission intensity.

Our most convincing evidence for the conversion of the structure from the p to the n type comes from the comparison the of Coulomb binding energy of the trion ground state before and after the switch, fitting well the understanding as a switch from the positive to the negative spin-singlet trion. In the magnetic field, the raw energy separation of exciton and trion emission lines is not precisely equal to the Coulomb binding energy, because it also includes the Zeeman contribution (while the electron Pauli g factor is constant to a good approximation, for the valence holes it depends on the wave vector; this causes a difference in the average Zeeman contribution in different bound states, for example, in the exciton and in various positive and negative trions). However, averaging the peak separation over both polarizations of light, equivalent to averaging over both spin states of the hole annihilated during the recombination, nullifies the Zeeman effect and so allows extraction of the pure Coulomb binding energy from the emission spectrum—also in the high magnetic fields.[17,19]

The results of such $\sigma^{\pm}$ polarization averaging of the exciton-to-trion peak distance extracted from our field-dependent photoluminescence spectra is shown in Fig. 4. The color coding is the same as in the other figures, so the two sets of data points correspond to the illumination with only red and only green lasers. Clearly, despite their near degeneracy at $B = 0$, the field evolution of the two compared binding energies differs markedly.

Specifically, the red symbols picture a binding energy E_b which at $B = 0$ begins slightly above 1.6 meV, then decreases to about 1.35 meV, roughly linearly with the field growing up to $B = 5$ T, and then remains essentially unchanged all the way up to the highest applied magnetic field $B = 23$ T. In contrast, the green symbols describe a binding energy E_b which is initially enhanced by the field, from just under 1.6 meV at $B = 0$ to about 2.0 meV at $B = 9$ T, and then nearly stops growing in higher fields. This contrasting behavior of the green and red data points, together with the earlier comparative experiments[5,17] and calculations[27] on positive and negative trions, decisively reinforce our earlier assignment of the X_s^+ and X_s^- lines at $B = 0$ in Fig. 1(b) and, thus, also confirm the conversion of the nominally p-type structure to the n type by means of green laser illumination.

In addition to establishing the charge conversion of our structure, Fig. 4 provides an unprecedented comparison of the positive and negative trions in the same quantum well of superior quality and in the magnetic field ranging from zero up

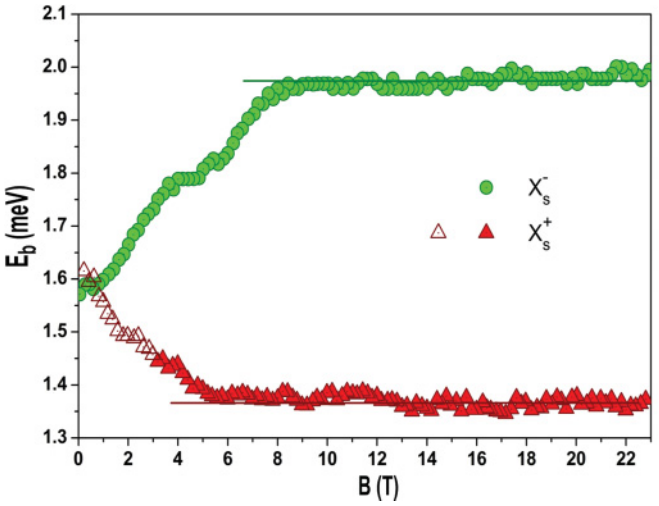

FIG. 4. (Color online) Comparison of the Coulomb binding energies of the positive and negative spin-singlet trions in the same quantum well (symmetric, GaAs, $w = 15$ nm), dynamically converted between the p and n types. For the X_s^+, a binding energy at $B \leqslant 3$ T was obtained by extrapolating the position of the exciton line (too weak to be seen in the spectrum).

to extremely high $B = 23$ T. We find (i) near degeneracy of the X_s^+ and X_s^- binding energy at $B = 0$ previously reported[5,17] in two undoped quantum wells made of different materials; (ii) approximately linear dependence of both binding energies on the weak field, with the opposite slopes ± 0.05 meV/T for X_s^+ and X_s^-, which differ strikingly from the behavior previously reported[17] for an slightly wider and undoped well of the same material; (iii) near-field independence of both binding energies in high fields (we are not aware of a similar comparison in such high fields). Our result also defines three distinct field ranges in our sample: (a) weak fields, symmetric X_s^+ and X_s^- binding; (b) intermediate fields, a strong and opposite effect of magnetic field on binding energy; (c) strong fields, a highly asymmetric and almost field-insensitive X_s^+ and X_s^- binding. The values (and, especially, their field dependence) obtained in the high-field range agree well with the previous numerical calculation[27] quoting for $B =$ 15 (25) T: $E_b = 1.14$ (1.11) meV for the X^+ and $E_b = 2.09$ (2.22) meV for the X^-.

The "recharging" of trions immersed in and coupled to the surrounding gas of free carriers—from the positive X^+ to the negative X^-, in reaction to the charge reversal of those carriers—from positive holes to negative electrons, is quite understandable but still a spectacular effect. Here, it has been demonstrated in a real structure by means of controlled two-beam illumination. The "recharging" idea is simple—an optically active quasiparticle must involve at least one electron and one hole, and in the presence of free carriers such a minimal complex (exciton, $X = e + h$) is unstable against binding one of them to become a trion, either $X^+ = X + h$ or $X^- = X + e$, depending on the type of available carrier.

An analogous effect may be anticipated for any charged quasiparticle coupled to an open reservoir of free carriers whenever these carriers switch the sign. Indeed, we have identified further examples in our spectra, including one from the first group of width-sensitive lines, with emission energies

falling slightly below the nearly free excitons and trions. We attribute these lines to radiative recombination of the charged excitonic complexes bound to the (very scarce) acceptors located inside the quantum well: A_wX^+ and A_wX^-.

A bare ionized acceptor A_w^- is a negative point charge, acting as a strong negative Coulomb center. In a p-type structure it binds two of the surrounding holes to become an $A_w^+ = A_w^- + 2h$. This stable but optically inactive object can further capture a photo-excited electron-hole pair to become a radiative bound state $A_wX^+ \equiv (A_w^+)X = A_w^- + 3h + e$ (with the spin-doublet three-hole configuration in the ground state). When recombining, an A_wX^+ leaves behind an A_w^+, which includes a pair of holes and, thus, has two available spin states, singlet and triplet. Hence, the A_wX^+ emission line is split by the two-hole exchange energy.[24]

Importantly, the spin-doublet A_wX^+ is the only stable and radiative state involving an acceptor inside a p-type well. Any complex which is smaller and still includes an electron is neutral or negative and, thus, unstable against capturing another hole from the surrounding gas (in fact, concentration of the acceptor's charge makes the binding energies of $A_wX = A_w^- + 2h + e$ and $A_wX^+ = A_w^- + 3h + e$ much higher than those of an exciton or trion, respectively). On the other hand, anything larger and energetically stable should include another (photoexcited) electron, which is improbable due to the A_wX^+'s short optical lifetime.

What should happen to the A_wX^+ when the structure is converted to the n-type? While it will certainly "recharge" to a negative complex, it cannot be a simple e-h reflection (like for the trions) because of the fixed charge of an ionized acceptor. Instead, this happens: The point charge A_w^- remains stable, surrounded by a gas of electrons. On photo-excitation it can capture an electron-hole pair to become an $A_wX^- = A_w^- + e + h$. This rather small complex recombines to the A_w^- which has no spin spectrum, thus yielding a single A_wX^- emission line. Compared to the A_wX^+, in addition to not being split the A_wX^- emission energy should also be lower for the smaller size of the complex.

Again, it is important to realize that the A_wX^- is the only stable and radiative state involving an acceptor inside an n-type quantum well. It is clearly the smallest such complex, and any larger one would have to involve another hole—which is improbable in an n-type structure, given the short A_wX^-'s optical lifetime.

The above prediction matches precisely the behavior observed in our spectra, and the corresponding lines have been marked in Figs. 2 and 3. The exchange splitting of the A_wX^+ and its energy position relative to X/X^+ have been observed and understood earlier,[24] but the switch to the A_wX^- (single line at a lower energy) is a new result. Like the excitons and trions, the $A_wX^\pm$ lines also exhibit a diamagnetic shift in low fields. In higher fields their emission energies shift linearly with the field, with roughly the same slope as X/X^+, of about 0.7 meV/T.

The list of transitions detected in the high energy sector of the spectrum is, finally, exhausted by a pair of parallel lines departing linearly (as a function of magnetic field) downward from the X and X^+, with an energy-field slope of about 0.4 meV/T. These lines labeled SU-X^+ and SU-A_wX^+ have already been understood as shake-up replicas of free and acceptor-bound trions.[23,36,37] Let us note that a shake-up process couples recombination of a given complex with a cyclotron excitation of an additional free carrier in the surrounding gas, which in effect lowers the emission energy (compared to the "unshaken" transition) by a cyclotron energy. In our spectra, comparison of the energy-field dependence of the main and shake-up lines yield the slope difference of 0.3 meV/T corresponding to the hole effective mass $m_h^* = 0.38\,m_e$, matching the value known from the cyclotron resonance experiments.[47]

Let us add a few other observations consistent with the shake-up assignment: (i) when extrapolated to $B = 0$, the SU-X^+ and SU-A_wX^+ emission energies merge with those of X^+ and A_wX^+, respectively; (ii) the SU-A_wX^+ line is stronger and better resolved in polarization σ^-; the SU-X^+ line is better resolved in σ^+, but only because in σ^- it coincides in energy with the A_wX^+; (iii) the intensity of the SU-A_wX^+ line is over an order of magnitude higher than of the SU-X^+; and (iv) both shake-up lines weaken with a decrease of hole concentration.

Not surprisingly, the SU-X^+ and SU-A_wX^+ shake-up lines disappear from the spectrum together with their X^+ and A_wX^+ parents when the well is converted to the n type. Their negative analogs are not observed together with the emergence of the X^- and A_wX^- lines, because the shake-up effect in an electron gas involves a larger cyclotron energy, thus pushing the respective emission lines outside of the recorded spectra.

It is well known[23,40] that conservation of angular momentum makes the emergence of the trion shake-up emission dependent on (symmetry-breaking) localization. Also, the relative intensity of the SU-X^+ and SU-A_wX^+ lines reflects relative strength of the lateral confinement of both complexes and so reveals the contrast between the "nearly free" and "strongly bound" trion states.

Finally, let us turn to the intriguing series of multiple, parallel, weak, narrow, equidistant, and closely spaced emission lines, positioned even below the shake-up lines in the photoluminescence spectrum. They are observed in both σ^+ and σ^- polarizations, their energy shifting linearly in high magnetic field with the slope of about 0.7 meV/T, similar to that of X or X^+.

In the σ^- spectra in Figs. 2(a) and 2(b) these multiple lines seem bounded by another line with a higher energy-field slope of about 1 meV/T in high fields. Comparison with the 0.7 meV/T slope of excitons and trions (X and X^+) yields a difference of 0.3 meV/T, which is the same as for shake-up replicas except for an opposite sign, suggesting a similar origin. Indeed, in our previous study[24] this line was shown to follow a virtually identical magnetic field dependence in several asymmetric GaAs quantum wells of different width 15 to 30 nm, which led us to its understanding as a hole cyclotron replica of a positive singlet trion confined inside the quantum well (X^+) bound to an ionized acceptor placed at the interface separating the well from the barrier (A_b^-). By analogy with a related effect observed earlier in the magnetoabsorption spectra of electrons[48] we called this transition CR-A_bX.

The presence of a combined CR-A_bX transition in the emission spectrum predicts also the corresponding main recombination channel (in addition to the replica) of the same complex $A_bX = A_b^- + X^+ = A_b^- + 2h + e$. We are, indeed,

confident that the A_bX emission shows in our spectra, and that it is responsible for the (newly detected) multiple parallel lines apparently stemming from the CR-A_bX. More precisely, the parallel lines correspond to the A_bX complexes with their acceptors placed in the barrier at different distances from the quantum well (quantized into subsequent crystalline layers), and the lowest one has its acceptor right at the interface (indeed, it originates at $B = 0$ from the same energy as CR-A_bX). Note that assuming for the high-quality structure the residual bulk acceptor density in the barrier on the order of 10^{15} cm^{-3} yields the average sheet acceptor density per crystalline layer of 0.56×10^8 cm^{-2}. This is over three orders of magnitude below the nominal hole concentration in the well, consistent with the observable but relatively weak intensity of each single A_bX line (compared to X or X^+).

The arguments for our assignment of the A_bX lines are as follows:

(i) given the CR-A_bX line, the A_bX should also show in the spectrum;

(ii) the slope difference from the CR-A_bX line is consistent with that extracted from the shake-up lines;

(iii) the lowest A_bX line and the CR-A_bX line meet at $B = 0$ (see further for the explanation of higher lines);

(iv) the energy range of about 15 meV below the X^+ is consistent with rough estimates [note that this much larger value than for a donor-bound negative trion D_bX[46] stems from electron-hole mass asymmetry, allowing for a relatively much stronger electric polarization of an X^+ in the Coulomb field of an interface A_b^-; furthermore, as expected for the A_bX state divided by an interface, its absolute emission energy at $B = 0$ falls several meV above the well-known value 1512 meV of the AX state in bulk GaAs];

(v) given spatial distribution of acceptors in the barrier, sensitivity of the recombination energy to the acceptor distance from the well, and quantization of this distance in a crystal, the splitting of the A_bX into a series of parallel lines must be expected;

(vi) there are no other candidate transitions with the above features, in particular in this energy range and with this energy-field slope, any emission from the barriers would have much higher energy, emission from the bulk substrate would not have a cyclotron replica which anticross trion lines from the well[23] and would not disappear under green light (see further), and the well width variations would need an improbable amplitude of more than 10 lattice constants to affect emission energy by as much as 15 meV; and

(vii) the A_bX complex should exist, stabilized against capturing additional holes by the charged trionic component inside the quantum well.

On the other hand, there is no contradiction in marking only one CR-A_bX replica in Fig. 2, as it is considerably fainter than the A_bX main line. Actually, traces of higher CR-A_bX lines (corresponding to acceptors placed further away from the well) are seen more clearly in some other, unshown, spectra. There is also nothing special about the bounding of the series of A_bX lines by the CR-A_bX line in polarization σ^-; in fact, this behavior is seen only in some samples and at some temperatures.

Let us now repeat the question asked earlier about the other transitions: What happens to the A_bX and CR-A_bX lines when the sample is converted to the n-type by green illumination? Figure 3 shows that they simply disappear. This is completely consistent with their understanding as positive trions in the well bound to negative acceptors in the barrier; binding of negative trions formed in the n-type well would require positive donors, absent in our (nominally p-doped) structures. The vanishing of lines in the green light also points to the origin of recombination inside the well rather than in the bulk substrate (the lines denoted as "bulk GaAs" in Fig. 2 are not affected by the conversion from the p to the n type).

IV. CONCLUSION

In conclusion, we have carried out detailed studies of photoluminescence from symmetric GaAs quantum wells containing a high-mobility two-dimensional hole gas, in a broad range of magnetic fields, from $B = 0$ to 23 T. By comparing the spectra from wells of different widths, collected at different temperatures and in two different polarizations, and, especially, by varying concentration of the holes by means of two-beam illumination beyond the point of conversion from p to n type (thus, by dynamical switching between the gas of holes and electrons), we have been able to identify and understand many optical transitions, including the nearly free excitons and trions, excitonic complexes bound to charged impurities placed in the well and in the barriers, and also their cyclotron replicas. In particular, we have studied the effect of the p- to the n-type dynamical conversion on those transitions and on the stability of the corresponding excitonic complexes.

Arguably the most spectacular result is charge conversion between the positive and negative excitonic complexes bound to acceptors inside the well, $A_wX^+ = A_w^- + 3h + e$ and $A_wX^- = A_w^- + h + e$, differing from the previously demonstrated charge conversion of free trions (stripping of a pair of holes versus electron-hole substitution) due to the breaking of charge reflection symmetry by a fixed impurity. The optical signature of the $A_wX^\pm$ conversion is the emergence of a finite-state two-hole exchange splitting—exclusively for the A_wX^+ recombination.

Also insightful are the comparative results for the positive trions moving almost freely in the quantum well and bound to nearby ionized acceptors in the barrier (X^+ versus A_bX). We have shown strong electric polarization of the X^+ induced by the A_b^-, in contrast to the previous studies in the n-type structures where the energy splitting of the X^- and D_bX lines was much smaller. Our results distinguish the X^+ and A_bX transitions and prove coexistence of the two complexes in high-quality structures.

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