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# Thermal dissociation of free and acceptor-bound positive trions from magneto-photoluminescence studies of high quality GaAs/Al$_x$Ga$_{1-x}$As quantum wells

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Thermal dissociation of free and acceptor-bound quasi-two-dimensional positive trions is investigated by measuring the temperature dependence of the integrated emission intensity in magnetic fields up to 17 T in high-quality GaAs/Al$_x$Ga$_{1-x}$As quantum wells. Three distinct dissociation processes are observed for the well-resolved hole cyclotron replicas („shake-up") of positive trions bound to neutral acceptors in the spin-doublet state (SU–$A^0X_d^+$). To demonstrate that the hole involved in the shake-up process is not bound by the Coulomb interaction to the charged $A^0X^+$ complex, we calculate the valence Landau levels using the Luttinger model beyond the axial approximation. The calculated value of the hole cyclotron energy agrees well with the experimental data for the energy separation of the $A^0X^+$ and SU–$A^0X^+$ lines, determined from the emission spectra. At low temperatures, below 6 K, the dominant dissociation results in a free hole and an exciton bound to the neutral acceptor in the spin-singlet or -triplet state, ($A^0X_d^+ \to A^0X_s + h$ or $A^0X_t + h$). At higher temperatures, above 9 K, the dissociation into the free positive trion and the neutral acceptor ($A^0X_d^+ \to A^0 + X^+$) predominates. From the temperature evolution of the integrated emission of the free trion lines ($X^+$) we evaluate the transition energy between the two triplet trion states, the dark one ($X_{td}^+$) and the bright one ($X_{tb}^+$). The ionization energies of all detected dissociation processes are compared with the spectral positions of the relevant radiative recombination lines from which excellent quantitative agreement is achieved.

## I. INTRODUCTION

Trions (or „charged excitons") predicted by Lampert in 1958 [1] have not been unambiguously identified in bulk materials, mainly because of the small binding energy of the additional electron or hole to the neutral exciton [2, 3]. Stébé and Ainane pointed out [4] that in two-dimensional (2D) structures the trion binding energy significantly increases due to the strong geometric confinement of the three-body complex. The first observation of trions was reported by Kheng et al. [5] in photoluminescence (PL) spectra of modulation doped CdTe/Cd$_x$Zn$_{1-x}$Te multiple quantum wells. This observation was rapidly followed by experimental evidence of trions in other 2D semiconductor structures based on III-V (GaAs/Al$_x$Ga$_{1-x}$As) [6, 7] and II-VI (CdTe/Cd$_{1-x}$Mg$_x$Te, Cd$_{1-x}$Mn$_x$Te/Cd$_{1-y-z}$Mg$_y$Zn$_z$Te) [8, 9] compounds.

Three-particle complexes of fermions bound by Coulomb interaction are a fundamental problem in nuclear, atomic and solid-state physics. Negative and positive trions ($X^- = 2e + h$ or $X^+ = 2h + e$) are the analogs of the negative hydrogen ion ($H^-$) and the positive hydrogen molecule ($H_2^+$) [10]. However, comparable electron and hole masses, confinement in one dimension, nonparabolic and anisotropic hole dispersion, coupling to the crystal lattice and free carriers cause a complex behaviour of trions with multiple facets which give a quite unique possibility to study various fundamental aspects of a three particle system.

Most efforts have been devoted to the investigation of trion states in high magnetic fields [11–14]. According to theory, in the limit of zero magnetic field the only bound trion state in a 2D system is a spin singlet, whereas in the limit of extremely high magnetic fields the only bound trion state is an optically inactive („dark") spin triplet [15–18]. Theoretical calculations [19] also predict formation of weakly bound optically active „bright" triplet states. Indeed these all trion states were observed experimentally and successfully identified in optical spectroscopy experiments of 2D structures with electrons and/or holes [11, 20–22]. Besides free trions, a variety of excitons bound on shallow impurities were indentified in magneto-PL spectra from 2D structures [22–24]. Investigations of trions are typically performed at low temperatures, mainly due to their small binding energy. Temperature dependent PL studies of trions, whether free (mobile) or bound on impurities, may provide additional information on trion properties. The ionization energies of different trions calculated from the temperature dependence of the integrated emission can be compared with the corresponding energies obtained by different experimental methods or calculated numerically. Due to the additional carrier, thermal dissociation processes of trions are expected to be considerably more complicated than those of neutral exciton complexes [25].

In this paper we report on detailed studies of both the magnetic field and temperature dependence of the PL spectra of superior quality GaAs/$Al_xGa_{1-x}$As 15 nm wide quantum wells confining a 2D hole gas. In high magnetic fields our PL spectra reveal all theoretically predicted states of the positive trion: the spin singlet ($X_s^+$), and the dark and bright spin triplets ($X_{td}^+$ and $X_{tb}^+$). At lower photon energies we also record the radiative recombination of excitons and trions bound to carbon acceptors. Specifically, three complexes of different charge have been detected: the positive trion bound on a neutral acceptor ($A^0X^+$), the exciton bound on a neutral acceptor ($A^0X$), and the exciton bound on an ionized acceptor ($A^-X$). The latter, negatively charged complexes are related to ionized barrier acceptors ($A^-$), which serve as suppliers of holes to the quantum well. Additionally, we observe in the PL spectra the hole cyclotron ($\hbar\omega_{hc}$) resonance processes leading to replicas, decreasing or increasing the emission energy of various exciton complexes. Emission at lower energies, so called shake-up processes, are observed for the free and acceptor bound trions (SU–$X^+$, SU–$A^0X^+$). In a shake-up process, the radiative recombination of the electron-hole pair is accompanied by the excitation of an additional electron or hole to a higher Landau level (LL), which lowers the emission energy by a quantum of the cyclotron energy [22–26]. Emission with higher energy, so called combined cyclotron-exciton resonance [27, 28], is observed for excitons bound to ionized barrier acceptors (CR–$A^-X$). In the combined cyclotron-exciton resonance process, the radiative recombination of the electron-hole pair is accompanied, as in the shake-up process, by transition of an additional electron or hole between $LL$s but in the opposite direction from a high to a lower LL, which results in increasing the emission energy.

We study the dissociation of trions, both free and bound on neutral acceptors, by measuring the temperature dependence of the integrated emission. The ionization energies evaluated from the temperature evolution of the integrated emission are compared with the spectral positions of the relevant radiative recombination lines. Excellent quantitative agreement is obtained in this comparison.

## II. SAMPLES AND EXPERIMENTS

The studied samples are two modulation - doped 15 nm thick GaAs/$Al_{0.33}Ga_{0.67}$As quantum wells, fabricated by molecular beam epitaxy on (001) semi-insulating GaAs substrates. Holes are delivered to the well by carbon doping of both $Al_{0.33}Ga_{0.67}$As barriers but with different concentrations: in the bottom barrier - $5\times10^{16}$ cm$^{-3}$ (in both samples) and in the top barriers - $3\times10^{17}$ cm$^{-3}$ and $2.3\times10^{17}$ cm$^{-3}$ in samples 1 and 2, respectively. The 200 nm wide doping layer was set back from the quantum well by undoped 60 nm wide spacers. The hole concentrations measured in dark at temperature $T = 4.2$ K are $p_1 = 2.22\times10^{11}$ cm$^{-2}$ and $p_2 = 1.45\times10^{11}$ cm$^{-2}$, with mobilities of $\mu_1 = 7.67\times10^5$ cm$^2$/Vs and $\mu_2 = 8.67\times10^5$ cm$^2$/Vs.

The photoluminescence is excited by a red diode laser line emitting at 680 nm wavelength, corresponding to a photon energy below the band gap of the barriers. The measurements are performed as a function of the temperature from $T = 2$ K to 30 K. The magnetic field is applied in the Faraday configuration perpendicular to the quantum well plane and changed with a small step $\Delta B = 0.05$ T up to $B = 17$ T. A fiber optics was used for excitation and detection with a linear polarizer and a quarter-wave plate placed close to the sample in helium gas. The $\sigma^-$ and $\sigma^+$ polarizations were switched by reversing the field direction. The spectra were analyzed using a high-resolution monochromator equipped with a liquid-nitrogen-cooled charge coupled Si device.

## III. RESULTS AND DISCUSSION

### A. Evolution of PL spectra in magnetic fields.

In Fig. 1 the magnetic field evolutions of the PL spectrum of sample 1 recorded at two temperatures, $T = 2.3$ K and 10 K, in both light polarizations are presented. Examples of zero and high-field spectra in $\sigma^-$ polarization are shown in Fig. 2. The results for sample 2 are similar. Due to the significant difference in emission intensities of 2D related lines the spectra are presented using a logarithmic scale. The magnetic field evolution of the PL spectra is presented in Fig. 1 for the two different temperatures in order to reveal more details. At low temperatures the emission lines in the higher energy sector are well resolved, whereas the lines in the lower energy sector are extremely weak and difficult to detect. At higher temperatures the PL lines in the high energy sector decrease in intensity and merge, which makes it impossible to distinguish between individual lines. In contrast, the lines in the low energy sector increase strongly in intensity with temperature rise and are well resolved at temperatures above 10 K.

The opposite temperature behavior of the lines detected in the high and low energy sector of the PL spectra is consistent with our previous division of all 2D related lines into two groups [22–29]. The main feature of the lines from the first group, positioned in the high energy sector, is the strong dependence of their energies on the well width, which reflects their origin from electron-hole complexes, whose wave functions are spread over the whole quantum well. In contrast, the lines recorded at lower energies are insensitive to the well width, which points to the strong localization of the related electron-hole complexes by the deep Coulomb potentials of ionized acceptors located in the barrier.

The complementary measurements of the magnetic

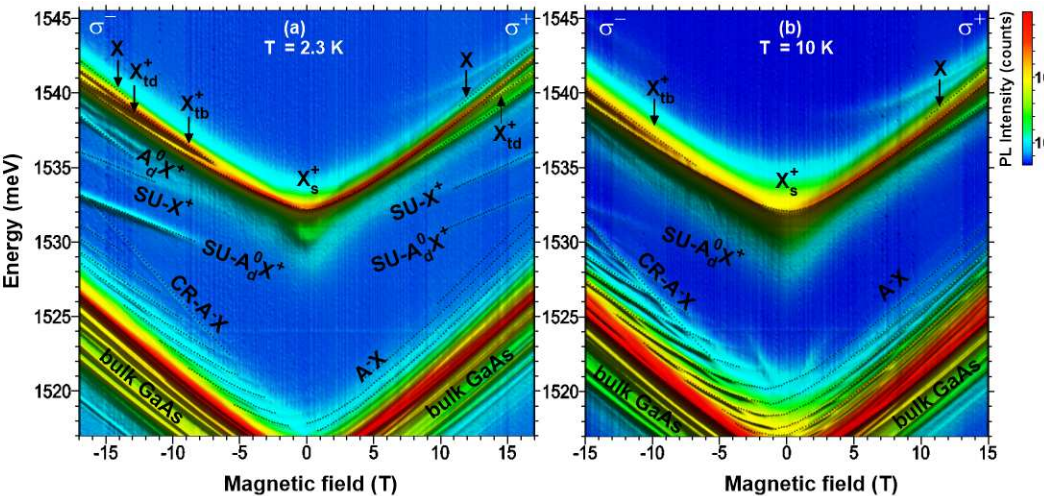


FIG. 1. (color online). Magnetic field evolution of the PL spectrum of sample 1 in $\sigma^-$ and $\sigma^+$ polarizations at temperatures (a) $T = 2.3$ K and (b) $T = 10$ K.

field and temperature dependencies of the photoluminescence, as well as the comparison with numerical calculations of the studied structures enable us to interpret all 2D related emission lines, including those, which cannot be well resolved as individual peaks. At zero magnetic field only one line is observed (see Fig. 2). We attribute it to radiative recombination of the positive trion in the hole spin singlet state ($X_s^+$) [29]. When the magnetic field is applied, new lines emerge successively in the PL spectra with field rise. At the energies above the $X_s^+$ emission, the exciton ($X$) and the trion in the triplet bright ($X_{bt}^+$) and dark ($X_{dt}^+$) states are observed. On the low energy wing of $X_s^+$ two not well resolved lines are observed as well (see e.g. the spectrum at $B = 13$ T in Fig. 2). Examination of the temperature evolution of these lines allows us to attribute them to emission of the exciton bound to the neutral acceptor in the hole spin triplet ($A^0X_t$) and singlet ($A^0X_s$) states (see also text below). Next to $A^0X_t$ and $A^0X_s$, at slightly lower energy we observe a line which we interpret as radiative recombination of the positive trion bound to the neutral acceptor in the hole spin doublet state ($A^0X_d^+ = A^- + 3h + e$). Note that as shown in our previous calculations [22], within the quantum well hosting the hole gas, acceptors can be either neutral ($A^0$) or positively charged ($A^+$). Thus, the positively charged complex of $A^0X^+$ can be created in two ways: (1) as positive trion ($X^+$) bound on the neutral acceptor ($A^0$), or (2) as exciton ($X$) bound on the positively charged acceptor ($A^+$). The two different ways of formation of this complex lead to two different notations: $A^0X^+$ or $A^+$X. In the whole text we use the first notation. Lower in energy relative to $A^0X_d^+$, two shake-up replicas of the positive trion in the hole spin triplet (SU–$X_t^+$) and singlet (SU–$AX_s^+$) states are observed (see e.g. the spectrum at $T = 2$ K in Fig. 3). The shake-up replica of the positive trion bound to the neutral acceptor in the hole spin doublet state (SU–$A^0X_d^+$)

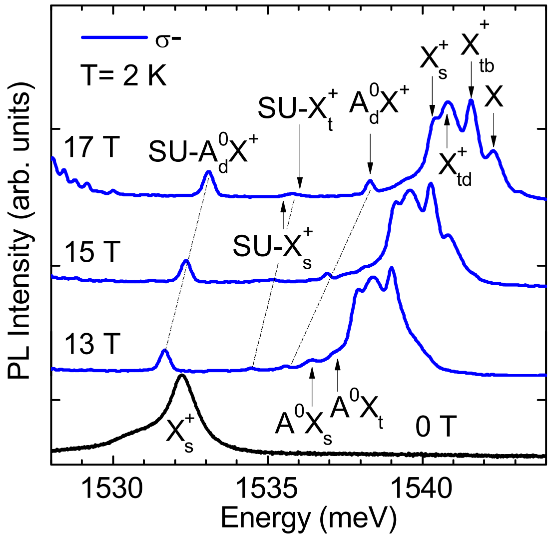


FIG. 2. (color online). Examples of zero and high-field PL spectra of sample 1 in $\sigma^-$ polarization at $T = 2$ K.

complete the lines from the first group. The omparison of the energy vs. magnetic field slope of the shake-up lines with those of $X^+$ and $A^0X^+$ yields a difference of 0.3 meV/T.

The second group of lines observed in the low energy sector is very weak in intensity at low temperatures but their intensities grow rapidly with temperature increase (see also Fig. 3). These multiple, almost equidistant lines shift in magnetic field with the same energy vs. magnetic field ratio as the $X^+$ and $A^0X^+$ lines. In this group of features we detect an additional line with higher energy vs. magnetic field slope. We attribute the multiple lines to excitons bound on ionized acceptors positioned on subsequent crystallographic planes in the barrier ($A^-X$) and the line of higher energy vs. magnetic

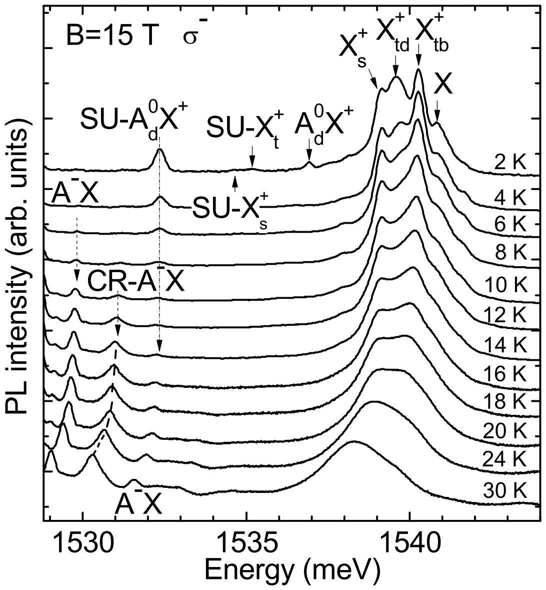


FIG. 3. (color online). Evolution of PL spectra of sample 1 in $\sigma^-$ polarization as function of temperature in magnetic field $B = 15$ T.

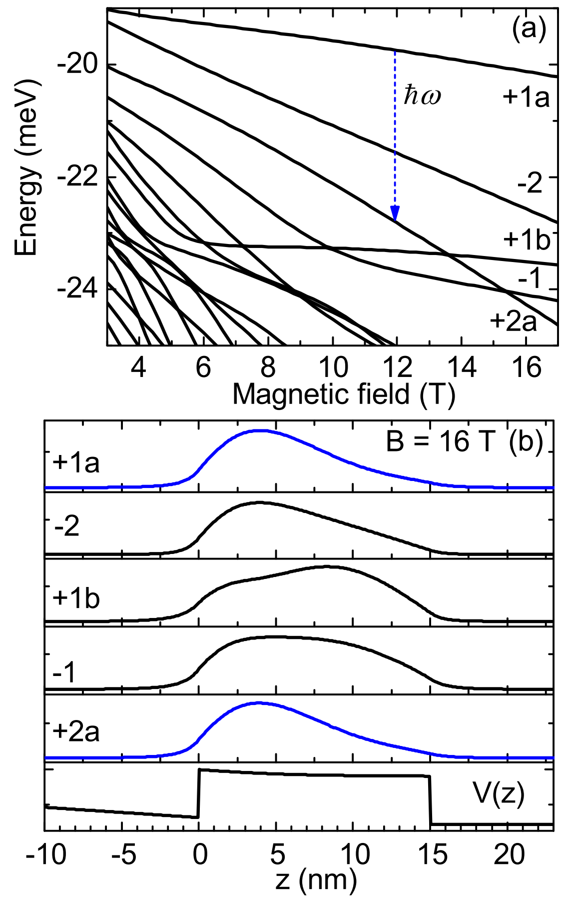


FIG. 4. (color online). Results of theoretical calculations: (a) energies and (b) envelope wave functions of the topmost hole Landau levels and the profile of the potential of sample 1.

field slope to the hole cyclotron replica of the nearest energy line $A^-X$, relating to it as the combined exciton-cyclotron resonance of an exciton bound to an ionized acceptor (CR–$A^-X$) [28, 29]. The comparison of the energy vs. magnetic field slope of the $A^-X$ and CR–$A^-X$ lines gives the same difference equal to 0.3 meV/T as for the SU–$A^0X_d^+$ and $A^0X^+$ lines, only with opposite sign.

### B. Evolution of PL spectra with temperature increase.

In Fig. 3 the evolution of the PL spectra of sample 1 in $\sigma^-$ polarization as function of temperature is presented at a magnetic field of $B = 15$ T. A strong decrease of the intensities of the lines located at higher energies (SU–$AX_d^+$ and above) accompanied by a simultaneous increase of the intensities of the lines located at lower energies (CR–$AX^-$ and below) with increasing temperature is observed.

#### Dissociation processes of $A^0X_d^+$.

Let us begin the discussion of the temperature evolution of the PL spectra with the results obtained for the shake-up replica of the positive trion bound to the neutral acceptor in the hole spin doublet state (SU–$A^0X_d^+$). We choose the hole cyclotron replica instead of the ground state since SU–$A^0X_d^+$ can be much better resolved in the spectra than $A^0X_d^+$. In order to evidence that the hole involved in the shake-up process is not one of the holes of $A^0X^+$, i.e., it is not bound with the complex by Coulomb interaction, but originates from the surrounding 2D hole gas, we evaluate the energies and wave functions of the carriers confined in the investigated structures. In the first step the potential distribution V(z) and energies of the hole subbands at $B = 0$ T are determined by self-consistent solution of the Schrödinger and Poisson equations. The Hartree approximation is used to include the contribution of mobile holes to $V(z)$. The exact 2D hole eigenfunctions are represented in the Luttinger Hamiltonian. In the next step, the potential $V(z)$ is used to find the energies and wave functions of the hole LLs in a magnetic field normal to the quantum well plane. The calculations are extended beyond the axial approximation by inclusion of the cubic term. The details of the numerical method are described in Refs. [29] and [30].

Figure 4 presents results of calculations of the high magnetic field evolution of the topmost valence-band levels in sample 1. All shown states belong to the ground heavy-hole subband. They are harmonic oscillator functions labeled by the index $n = -2, -1, 0, 1, \ldots$ of the largest axial component of the wave function. For $n \geq 1$, there are two axial levels with the same number $n$ in each subband, distinguished by the letters a and b. Only the ground states $+1a$ and $-2$ have opposite spins (up and down) whereas all excited states, due to the strong mixture of heavy and light hole subbands contain both spin orientations.

The comparison of the energy separation of the $A^0X_d^+$ and SU–$A^0X_d^+$ lines with the numerical LL calculations indicates that the shake-up process corresponds to the excitation of holes from the ground state $+1a$ to the excited state $+2a$. In the regime of high magnetic fields the hole cyclotron energy is a linear function of the magnetic field ($\hbar\omega = \alpha \cdot B$), with energy/magnetic field coefficients determined experimentally and numerically equal to $\alpha_{ex} = 0.3$ meV/T and $\alpha_{th} = 0.27$ meV/T. The excitation of the hole in the shake-up process from the ground state $+1a$ to the excited state $+2a$ even in fields above 14 T when the states $+1b$ and $-1$ are lower in energy than the state $+2a$ can be understand by comparing the wave functions of the LLs involved in the process. The inspection of the shape of the envelope functions for the topmost valence-band levels, presented in Fig. 4(b), shows that the ground states $+1a$ and the excited state $+2a$ are localized near the doped barrier, whereas the excited states $+1b$ and $-1$ are spread toward the center of the well. Thus, the ground state $+1a$ has a larger overlap with the excited state $+2a$ than with the $+1b$ and $-1$ states. This increased overlap results in a larger transition probability of the hole from the state $+1a$ to the state $+2a$ in the shake-up process. This result allows us to regard the SU–$A^0X_d^+$ line as the hole cyclotron replica of the $A^0X_d^+$ line.

Figure 5 presents, in more detail than Fig. 3, the temperature evolution of the radiative recombination of the hole replica SU–$A^0X_d^+$ and the CR–$A^-X$ line at a magnetic field $B = 15$ T. The SU–$A^0X_d^+$ line can be well resolved at the lowest temperatures. Its emission intensity decreases rapidly with temperature growth and disappears from the PL spectra for temperatures above $T = 10$ K. The line detected at the same energy for higher temperatures ($T > 10$ K) is one of the multiple lines $A^-X$, which PL intensity increases strongly with temperature (see also Figs 1 and 3).

In contrast, the emission intensity of the CR–$A^-X$ line is very weak at the lowest temperatures (see also Fig. 1) but its intensity grows rapidly with temperature. The observed temperature-induced exchange of emission intensities of the two lines indicates close spatial proximity of the excitonic complexes involved in the radiative recombination in the quantum well. At low temperatures photo-excited electron-hole pairs are bound (and recombine) on positively charged acceptors, whereas at higher temperatures recombination channels through ionized acceptors are more efficient. The higher emission intensity of $A^0X_d^+$ as compared to $A^-X$ at the lowest temperatures, in spite of the lower energy of the latter complex, is related to the much large number of radiative recombination centers of $A^0$ and $A^+$ than $A^-$. The neutral and positively charged acceptors are spread over the whole quantum well, whereas the negatively charged acceptors,

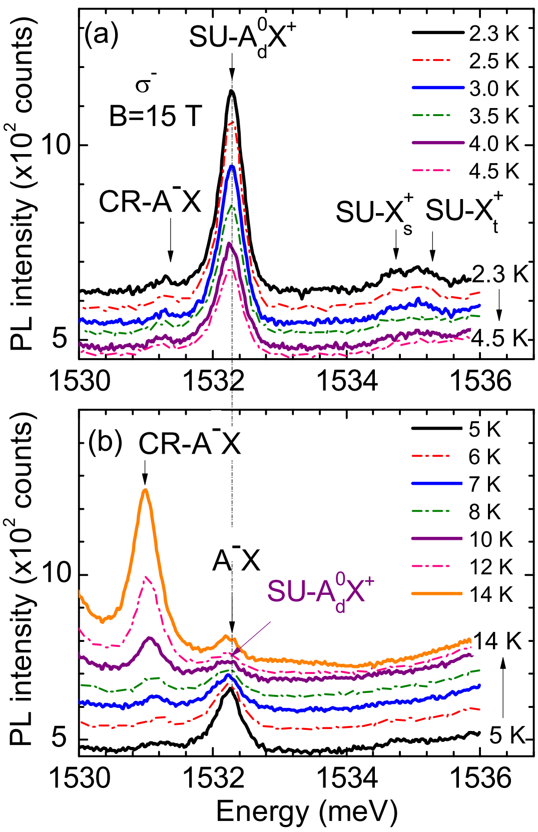


FIG. 5. (color online). Evolution of the radiative recombination of the hole replicas in sample 1: SU–$A^0X_d^+$ and CR–$A^-X$ at a magnetic field of $B = 15$ T for different temperatures, in (a) in the range $T = 2.3 - 4.5$ K, and in (b) in the range $T = 5 - 14$ K.

which can bind an exciton, are located only on a few crystallographic planes in the barrier [29].

Figure 6a shows the integrated PL intensity of the SU–$A^0X_d^+$ and CR–$A^-X$ lines as function of $1/T$ at $B = 15$ T. From this representation the opposing trends of the intensities become particularly clear: the intensity of SU–$A^0X_d^+$ decreases, that of CR–$A^-X$ increases with temperature growth.

From the temperature decrease of the integrated emission intensity of the SU–$A^0X_d^+$ line we estimate the related activation energies of the positive trion bound to the neutral acceptor. We use a formula similar to the one proposed by D. Bimberg and co-workers in studies of the dissociation of excitons bound to neutral acceptors in GaAs [25]:

$$I\left(T\right) = \frac{I_o}{\left(1 + \sum_{i=1}^{N} \frac{\alpha_i}{\alpha_o} \cdot e^{-E_i/k_B T}\right)}, \quad (1)$$

where $I_o$ is the total number of photo-excited electron-hole pairs, $\alpha_i$ and $\alpha_o$ are the degeneracies of the excited levels $E_i$ and the ground level $E_0$, respectively. The accurate evaluation of the experimental data shows that

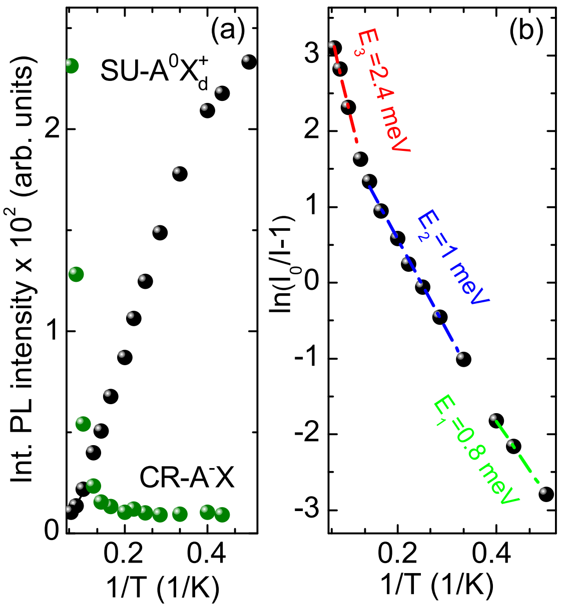


FIG. 6. (color online). (a) Integrated PL intensities of SU–$A^0X_d^+$ and CRA-X lines in sample 1 as function of $1/T$ in a magnetic field of $B = 15$ T. (b) Integrated PL intensity rate $\ln(1 + I_o/I)$ of the SU–$A^0X_d^+$ line as function of $1/T$ (symbols). The lines are linear fits used to deduce the activation energies (see Eq. 1).

three activation energies $E_1 = 0.8$ meV, $E_2 = 1$ meV, and $E_3 = 2.4$ meV are necessary for a satisfactory fit to the data (see Fig. 6b).

Let us identify the dissociation processes leading to the three activation energies $E_1$, $E_2$ and $E_3$. There are thirteen different processes for the dissociation of $A^0X_d^+$. From these processes different dissociation energies $E_d$, which we will compare below with the measured ionization energies, result. (a) Two dissociations result in a free exciton and a positively charged acceptor in the hole spin-singlet or triplet state:

$$A^0X_d^+ \to A_s^+ + X, \qquad E_{d1} = E(A_s^+, X) \qquad (2)$$

$$A^0X_d^+ \to A_t^+ + X, \qquad E_{d2} = E(A_t^+, X) \qquad (3)$$

(b) Two dissociations result in a free electron, a free hole and a positively charged acceptor in the hole spin-singlet or triplet state:

$$A^0X_d^+ \to A_s^+ + e + h, E_{d3} = E(A_s^+, e, h) = E_{d1} + E(X) \qquad (4)$$

$$A^0X_d^+ \to A_t^+ + e + h, E_{d4} = E(A_t^+, e, h) = E_{d2} + E(X) \qquad (5)$$

(c) Two dissociations result in a free hole and an exciton bound to a neutral acceptor in the hole spin-singlet or triplet state:

$$A^0X_d^+ \to A^0X_s + h, \qquad E_{d5} = E(A^0X_s, h) \qquad (6)$$

$$A^0X_d^+ \to A^0X_t + h, \qquad E_{d6} = E(A^0X_t, h) \qquad (7)$$

(d) Two dissociations result in a neutral acceptor and a free positive trion in the hole spin-singlet or triplet state:

$$A^0X_d^+ \to A^0 + X_s^+, \qquad E_{d7} = E(A^0, X_s^+) \qquad (8)$$

$$A^0X_d^+ \to A^0 + X_t^+, \qquad E_{d8} = E(A^0, X_t^+) \qquad (9)$$

(e) Dissociation results in a neutral acceptor, a free exciton and a free hole:

$$A^0X_d^+ \to A^0 + X + h,$$
$$E_{d9} = E(A^0, X, h) = E_{d7} + E(X, h) \qquad (10)$$

(f) Dissociation results in a neutral acceptor, a free electron and a pair of free holes:

$$A^0X_d^+ \to A^0 + e + h + h,$$
$$E_{d10} = E(A^0, e, h, h) =$$
$$= E_{d7} + E(X, h) + E(X) = E_{d9} + E(X) \qquad (11)$$

(g) Dissociation results in an exciton bound to an ionized acceptor and a pair of free holes:

$$A^0X_d^+ \to A^-X + h + h, \qquad E_{d11} = E(A^-X, h, h) \quad (12)$$

(h) Dissociation results in an ionized acceptor, a free exciton and a pair of free holes:

$$A^0X_d^+ \to A^- + X + h + h,$$
$$E_{d12} = E(A^-X, h, h) = E_{d11} + E(A^-, X) \qquad (13)$$

(i) Dissociation results in an ionized acceptor, a free electron and three free holes:

$$A^0X_d^+ \to A^- + e + h + h + h,$$
$$E_{d13} = E(A^-X, h, h) = E_{d12} + E(X) \qquad (14)$$

At lowest energy two dissociation processes with very low activation energies $E_1 = 0.8$ meV and $E_2 = 1$ meV dominate. According to our numerical calculations [22] of the Coulomb binding energies of different excitonic complexes in a 15 nm wide (but symmetric) QW, the binding energies of a free exciton to a positively charged acceptor in the hole spin-singlet or triplet state (Eqs. 2 and 3) are too low ($E_{b1/2} \approx 0.4$ meV) to account for the measured $E_1$ and $E_2$ activation energies. On the other hand the dissociation energies of the third and fourth processes (Eqs. 4 and 5) are much too high since they contain additionally the free exciton binding energy equal to $E(X) \approx 7$ meV [31]. Most probably the activation energies $E_1$ and $E_2$ are related to dissociations resulting in a free hole and an exciton bound to a neutral acceptor in the hole spin-singlet or triplet state (Eqs. 6 and 7). The ionization energy of a hole from the $A^0X_d^+$ complex can be evaluated from the energy separation of the $A^0X_d^+$ and $A^0X_s$ ($A^0X_t$) lines in emission spectra, since the

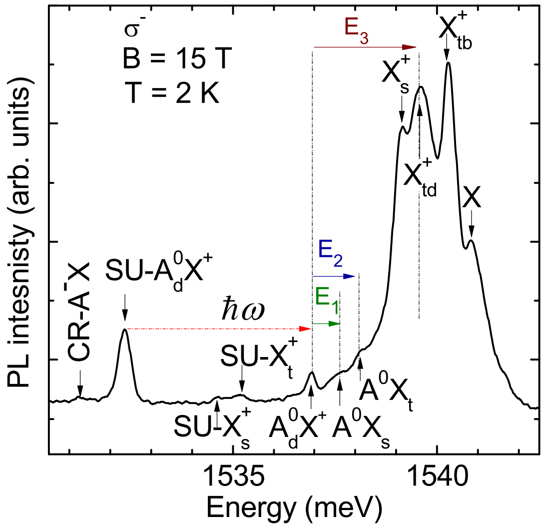


FIG. 7. (color online). Comparison of the activation energies of the dissociation processes with the low temperature ($T = 2$ K) PL spectra of sample 1 recorded at $B = 15$ T.

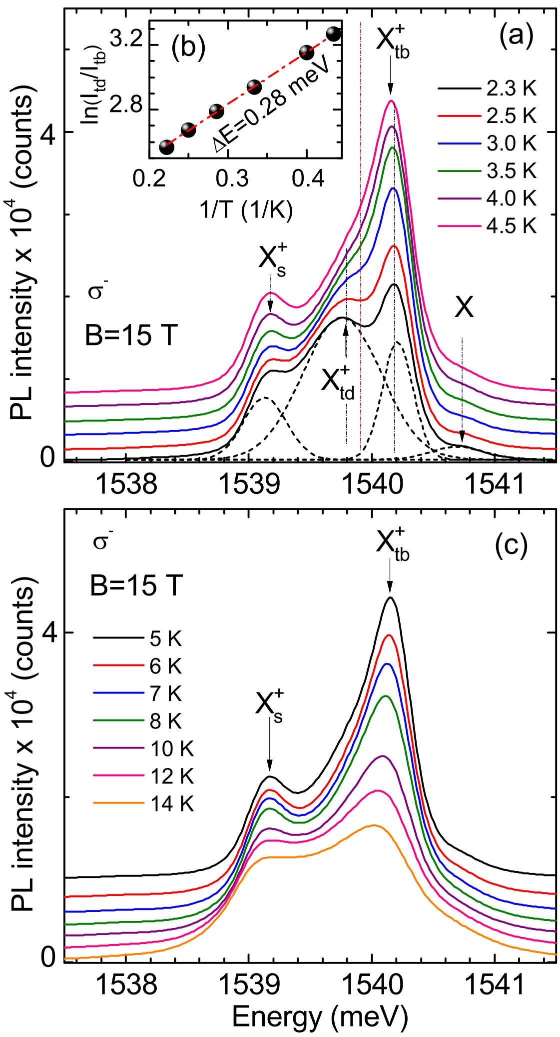


FIG. 8. (color online). Evolution of the PL spectra of the positive trion in the hole spin singlet ($X_s^+$) and triplet (dark–$X_{td}^+$ as well as bright–$X_{tb}^+$) states in sample 1 as function of the temperature in the ranges (a) 2.3 K $\leq T \leq$ 4.5 K and (c) 5 K $\leq T \leq$ 14 K. In (a) the dashed lines result from a numerical fit of the PL spectrum by three individual lines. In (b) the integrated PL intensity rate $ln(I_{tb}/I_{td})$ of the lines $X_{tb}^+$ and $X_{td}^+$ is presented as function of $1/T$ (the symbols). The dashed line is the linear fit from which the activation energy is deduced (see Eq. 2).

ground states of both radiative recombination processes ($A^+$ and $A^0$) have almost the same energy.

As we showed in our previous calculations [23] the additional hole is only weakly bound to $A^0$ in the hole spin triplet state $A_t^+$ and almost unbound in the singlet state $A_s^+$.

In Fig. 7 the activation energies are overlaid with emission spectra. As seen the $E_1$ and $E_2$ energies are in excellent agreement with the energy separation of the $A^0X_d^+$ and $A^0X_s/(A^0X_t$ emissions. We attribute the third activation energy $E_3$ to the dissociation into a free positive trion and a neutral acceptor ($A^0X_d^+ \rightarrow A^0 + X^+$) (Eqs. 8 and 9). Only one activation energy is observed for this dissociation process, even though there are two states of $X^+$ with hole spin singlet $X_s^+$ and triplet $X_t^+$ configurations. These results can be related to the small energy difference between $X_s^+$ and $X_t^+$ states in comparison to their energy distance from the $A^0X_d^+$ state, and hence we obtain only one energy, which is an average of two activation energies, in the numerical fit. The activation energies of the other dissociation processes (Eqs. 10–14) are much too high to explain the experiments.

### Dissociation processes of dark triplet $X_{td}^+$ into bright triplet $X_{tb}^+$.

Figure 8 presents the temperature evolution of the emission of the positive trion ($X^+$). The PL intensities of all trion states increase with the temperature increasing from $T = 2.3$ K up to 5 K and then rapidly decrease at higher temperatures. The increase of the trion emission intensity at the lowest temperatures is most probably related to the dynamics of the trion. The integrated PL intensity of the positive trion in the singlet and the bright triplet states increase at a higher rate than those of the dark triplet state. Since for temperatures $T > 2.5$ K the bright triplet emission cannot be well resolved in the PL spectra we perform a numerical fit to determine the exact shapes and the energy positions of all trion lines. An example of the fit at $T = 2.3$ K is presented in Fig. 1a by the dashed line. The main characteristics of the PL spectra from the trion emission is the almost two times larger width of the dark triplet line compared to the singlet and the bright triplet lines. This can be explained as follows. The dark triplet is optically inactive [22]. To become optically active some admixture of the nearby optically active singlet and bright triplet states is generally expected. The admixture with the lower and higher energy states results in a significant broadening of the dark triplet emission line. This interpretation is also consistent with the observed, much stronger temperature-induced energy shift of the $X^+_{td}$ line towards higher energies compared to the $X^+_s$ and $X^+_{tb}$ lines.

From the temperature evolution of the integrated emission intensities of the dark and the bright triplet lines we evaluate the transition energy between the $X^+_{td}$ and $X^+_{tb}$ states using the Boltzmann law:

$$\frac{I_{td}(T)}{I_{tb}(T)} = \frac{\alpha_{td}}{\alpha_{tb}} \cdot e^{-(E_{td}-E_{tb})/k_B T} \tag{15}$$

The energy obtained from Eq. (15), $\Delta E = 0.28$ meV, is in good agreement with the energy separation of the $X^+_{td}$ and $X^+_{tb}$ lines in the PL spectra which is equal to 0.3 meV at $T = 4.5$ K and 0.4 meV at $T = 2.3$ K.

## IV. CONCLUSION

In conclusion, we have studied the dissociation of positive trions, both free and bound to a neutral acceptor, by measurements of the temperature dependence of the integrated emission in high magnetic fields up to 17 T. Three dissociation processes are observed for the well resolved hole cyclotron replica (shake-up) of the positive trion bound to a neutral acceptor in the hole spin doublet state (SU–$A^0X^+_d$). To prove that the hole involved in the shake-up process is not bound to the $A^0X^+$ complex we have evaluated the valence band Landau levels using the Luttinger model and show that the calculated hole cyclotron energy is almost equal to the experimental value for the energy separation of the $A^0X^+$ and SU–$A^0X^+_d$ lines in the PL spectra. At low temperatures the dominant dissociation results in a free hole and an exciton bound to a neutral acceptor in the spin-singlet or triplet state, whereas at higher temperatures the dissociation into a free positive trion and a neutral acceptor predominates. From the temperature evolution of the integrated emission of the free trion lines ($X^+$) we have evaluated the transition energy between the two triplet trion states, the dark one ($X^+_{td}$) and the bright one ($X^+_{tb}$). The ionization energies of all detected dissociation processes have been compared with the spectral positions of the relevant radiative recombination lines from which excellent quantitative agreement has been achieved.

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