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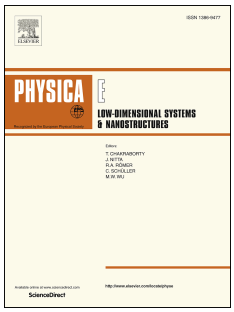

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Magnetic and power tuning of spin-asymmetric multiple excitons in a GaAs quantum well

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A comprehensive study of excitonic complexes in high-quality GaAs quantum wells has been performed. The polarization-resolved photoluminescence results show exciton, biexciton, and trion emissions tuned by temper- ature, excitation power, and by an external magnetic field. Excitons and biexcitons show important differences in their dependence on the external field. While the Zeeman splitting of biexcitons depends monotonically on the magnetic field, with a nearly constant g-factor, the behaviour of the exciton energy splitting is nonmonotonic and includes an inversion of sign as a function of the magnetic field. More importantly, we observe that a trion resonance emerges at a finite magnetic field, and only appears at low laser excitation power and for σ^+ polarization emission. The non-trivial dependence of the energy levels on the external magnetic field, together with the polarization selective trion emission, leads to an intricate exciton and trion dynamics, which can be reproduced by a set of coupled rate equations. Based on this theoretical approach, we show that a fast spin-flip process can lead to the spin-asymmetric emission, which reveals a contrasting spin-dynamics of excitonic complexes ruled by their charge.

I. INTRODUCTION

The optical properties of bound electron-hole complexes (excitonic quasiparticles) in high-quality GaAs/AlGaAs quantum wells have been the subject of extensive studies. The optical excitation and relaxation processes of these complexes grounded on the electronic structure and on the coupling between exciton states are the basis of a wide range of photo-electronic applications, such as light-emitting devices, laser technologies, and photovoltaics [1–6]. Excitons dominate the optical spectra of semiconductors close to the band gap. To obtain information about the spin-relaxation processes in this energy range, photoluminescence spectroscopy and coherent optical experiments in the time domain have been carried out with polarized light [3; 4]. For doped quantum wells, it has been established that the spin population of minority carriers at the band edge determines the luminescence polarization when the intensity of the exciting light is weak [7]. In turn, for undoped wells, the photoexcited populations of electrons and holes have the same density and the polarization of the observed luminescence is influenced by the relaxation processes that occur in both bands [4].

Magneto-optic studies are of interest to also explore magnetic g-factors and the effective Zeeman splitting of electrons, holes, and excitonic complexes in quantum wells [8; 9]. The radiative recombination efficiency of excitons can be improved by introducing a magnetic field, and this becomes relevant for phenomena, such as the quantum Hall effect, magneto-optical polarization measurements, electron-nuclear spin coupling, and optical detection of nuclear magnetic resonance [5; 8]. Although magnetoexcitons are beneficial for many emission-based devices, the short recombination lifetime of electron-hole pairs is detrimental to applications where the separation and transport of carriers is relevant [5; 8; 9]. In spite of many investigations on exciton complexes and their recombination in undoped quantum wells [10–13], the mechanisms driving these dynamics and the identification of the contribution of each complex are still controversial. For instance, the photoluminescence peak sometimes observed at the lower energy side of the neutral exciton emission (0.6 - 1 meV) has been ambiguously attributed to either biexciton or trion recombination [10–21]. In Ref. 12, a series of undoped GaAs/AlGaAs quantum wells with different widths showed a PL line around 0.7 meV below the excitonic emission with σ− polarization that was ascribed to positive trions (X^+), where the excess of charges would come from unavoidable background concentration in the barriers or in doped samples, due to the influence of possible ionized donors/acceptors [12; 13]. An analogous correlation between this emission and the X^+ contribution has been presented in Ref. 14. In this case, at moderated applied magnetic fields, a circular polarized sensitive spin triplet was also observed in an intentionally doped QW sample. In contrast, due to similarities of the binding and excitation energies and their behavior as a function of temperature, other works have also described this transition as resulting from biexcitons [16–21]. The present work addresses the duality of the interpretations of the nature of this excitonic emission by providing an analysis of the binding energies and the interplay of the spin relaxation process of excitons, biexcitons and trions determined experimentally by polarization- resolved photoluminescence and magnetospectroscopy (see inset of Fig. 1). These findings are explained in terms of an effective analysis of population evolution via rate

equations that include creation, recombination, and scattering between different spin-resolved exciton complexes.

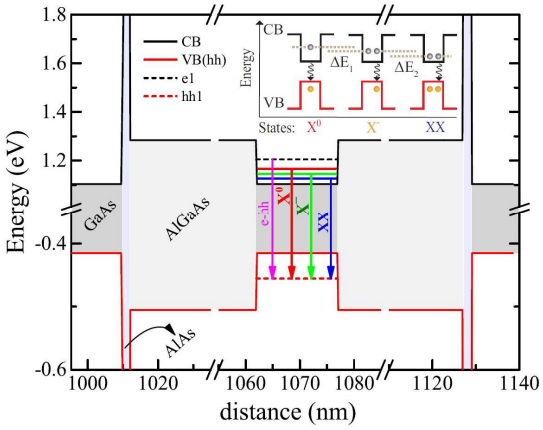

FIG. 1: Conduction (CB) and valence bands (VB) profiles of the studied quantum well and the observed optical transitions involving exciton (X^0), biexciton (XX) and negative trion (X^-); the latter only appears for the (σ^+) luminescence component at B > 3 T. The inset shows a schematic band structure of carriers confined in a quantum well and the different excitonic complexes forming when electrons (grey circles) and holes (orange circles) recombine.

II. METHODS

The undoped sample was grown by molecular beam epitaxy on top of a GaAs(001) substrate. The active layer consisted of a 15 nm wide GaAs quantum well with 50 nm-thick $Al_{0.18}Ga_{0.82}As$ barriers and an extra 2 nm-thick AlAs confinement layer on each side. The whole structure was grown at 580 □ and capped with a 5 nm-thick GaAs layer [22]. The band structure profile is presented in Fig.1. The temperature and magnetic field dependent microphotoluminescence measurements were performed using a cryogenic confocal microscope coupled to a cryostat (Attocube - Attodry 1000) allowing to range the temperature from 3.5 to 20 K. The applied magnetic field was oriented along the growth direction, and a linearly polarized 660 nm wavelength laser (Toptica-Ibeam) was used as an excitation source focused on a 1-μW diameter spot with four different laser-power values. The luminescence was analyzed in terms of the circular σ^+ and σ^- components, using a linear polarizer and a quarter-wave plate. The signal was detected by a 75 cm spectrometer (1200 lines/mm grating) and a Silicon CCD detector (Andor - Shamrock-Idus).

To interpret the experimental measurements, we considered a theoretical framework in which exciton, biexciton, and trion populations are intertwined through a set of rate equations. The exciton dynamics can be described by

$$\frac{dn_{X_0}^{\pm}}{dt} = P_{X_0}^{\pm} - \frac{n_{X_0}^{\pm}}{\tau_x} - F_{XX}^{\pm} - F_{X^-}^{\pm} \mp Z(X_0), \qquad (1)$$

corresponding to an exciton generation rate with a given spin, $P_{X^0}^{\pm}$, determined by the incident-light polarization. The exciton decay rate

$$\frac{1}{\tau_x} = \frac{1}{\tau_{r,X_0}} + \frac{D(E_{X0})}{\tau_{r,X_0}}, \qquad (2)$$

combines the optical recombination, characterized by τ_{r,X_0} and the nonradiative activation time, τ_{r,X_0} weighted by the thermal factor, $D(E_{X0}) = \exp(-E_{X0}/k_BT)$, with $E_{X_0} > 0$ being the exciton activation energy. The term $F_{XX}^{\pm} = 2\frac{\left(n_{X_0}^{\pm}\right)^2}{\tau_\beta} - \frac{n_{XX}^{\pm}}{\tau_{r,XX}}$ correlates the exciton and biexciton densities, $n_{XX}^{\pm}$, by assuming that the biexciton population is proportional to the square of the exciton concentration, as expected at thermal equilibrium. Here $\tau_{r,XX}$ is the biexciton recombination time, and $1/\tau_\beta$ the biexciton formation rate. The penultimate term in Eq. (1), $F_{X^-}^{\pm} = \frac{n_{X_0}^{\pm}}{\tau_t} - \frac{D(E_t)}{\tau_t}$, sets the link with the trion dynamics by introducing a formation rate, $1/\tau_t$. According to this model, the biexciton density evolution can be characterized by the expression

$$\frac{dn_{XX}^{\pm}}{dt} = P_{XX}^{\pm} + F_{XX}^{\pm} - \frac{n_{XX}^{\pm} X}{\tau_{nr,XX}} D(E_{XX}) \mp Z(XX), \qquad (3)$$

that contemplates a direct biexciton generation, $P_{XX}^{\pm}$, and a nonradiative dissociation rate, $\tau_{nr,XX}$, with a nonradiative activation energy, E_{XX}. In turn, the trion equation is given by

$$\frac{dn_{X^-}^{\pm}}{dt} = F_{X^-}^{\pm} - \frac{n_{X^-}^{\pm}}{\tau_{r,X^-}} \mp Z(X^-). \qquad (4)$$

The terms *Z(Y)* that appear in Eqs. (3-4) account for the spin thermalization between Zeeman split states with energies E_Y^+ and E_Y^- at finite magnetic fields, and are defined as

$$Z(Y) = \frac{n_Y^+}{\tau_Y^s} F(E_Y^- - E_Y^+) - \frac{n_Y^-}{\tau_Y^s} F(E_Y^+ - E_Y^-), \qquad (5)$$

where τ_Y^s is the spin-flip time of the corresponding excitonic complex, and

$$F(x) = \begin{cases} D(x); & x \geq 0 \\ 1; & x < 0 \end{cases} \qquad (6)$$

This definition allows intralevel Zeeman states to change

their relative positions by varying the magnetic field. The stationary condition can be simulated by setting the left-hand side of the rate equations equal to zero.

III. RESULTS AND DISCUSSIONS

A. Temperature and power dependence

The temperature dependence of the luminescence spectra corresponding to the GaAs/AlGaAs quantum well emissions is displayed in Fig. 2(a) for a fixed laser power of 1-μW, without magnetic field. We observe two peaks ascribed to the single exciton and the biexciton emissions, separated by the biexciton binding energy $E_b^{XX} \approx 1.5$ meV. This value is in excellent agreement with previous results about biexciton states in a 160 □ wide GaAs quantum well [17].

Figure 2(b) shows the integrated area from panel (a) as a function of $1/(k_B T)$. Both exciton and biexciton intensities decrease with rising temperature. To further investigate the nature of the low-energy peak, we fitted our experimental data according to the theoretical description of the exciton dynamics, (where we neglect the spin index due to the spin degeneracy of the excitonic states in the absence of magnetic field). Solving the coupled Eqs. (2) and (3) for the steady-state, we obtained the PL intensities of the exciton and biexciton emissions as $I_{X0} = n_{X0}/\tau_{r,X_0}$ and $I_{XX} = n_{XX}/\tau_{r,XX}$, respectively. We have used $P^{+}_{XX(X_0)} = P^{-}_{XX(X_0)}$ to describe a linearly polarized laser light without considering the trion contribution, since this state only appears in the presence of a magnetic field (see Figs. 3 and 4 and the related discussion). For the sake of simplicity, we set $\tau_{r,X_0} = \tau_{r,XX} = 1$ ps. The other parameters were obtained by fitting the experimental data (dotted lines in Fig 2(b)). The coupling of exciton and biexciton dynamics makes the simultaneous fitting of both X^0 and XX data an intricate task. However, we found a good qualitative agreement with the following values: $\tau_{nr,X_0} = 1.09363$ps, $\tau_{nr,XX} = 0.104407$ ps, $E_{XX} = 2.4$ meV, $E_{X_0} = 8.5$ meV, $P_{X_0} = 2.8\ \mathrm{kW/cm^2}$, $P_{XX} = 1.3\ \mathrm{kW/cm^2}$, and $1/\tau_\beta = 0.00007$ ps^{-1}. Note that the transition rate of non-radiative processes for biexcitons is an order of magnitude larger than for excitons. Furthermore, the formation rate of biexcitons from exciton states is negligibly small, i.e., the biexciton formation is almost instantaneous and doesn't result from a two steps process. The formation of secondary biexcitons has also been disregarded in Ref. 18, since the quasi-equilibrium between the exciton and biexciton populations is not established. The large value of τ_β indicates that the biexciton dynamics may be decoupled from the other excitonic complexes.

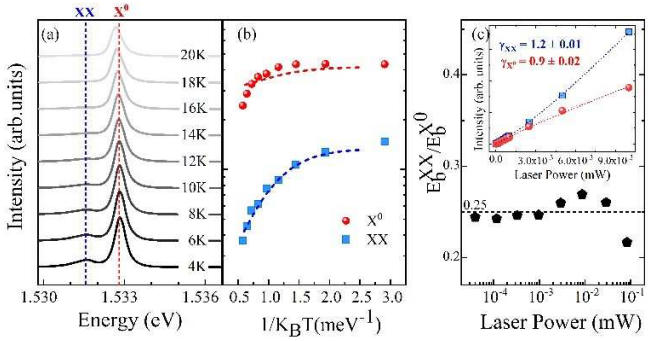

FIG. 2: (a) Photoluminescence spectra of the quantum well emissions at B = 0 T as a function of temperature and with a laser power of 1μW. Verticals blue and red lines represent biexciton (XX) and exciton (X^0) emissions, respectively. (b) Integral of the intensity of the biexciton (blue squares) and exciton (red circles) emissions. The dashed lines represent the best fit. (c) Ratio of the biexciton to exciton binding energy taken from the PL peak position as a function of power. The inset shows the integrated intensity behavior as a function of power at 10 K.

At a fixed temperature of 10 K, photoluminescence emissions were also collected for different laser powers ranging from ~10 nW to ~0.1mW. Again, two peaks are observed and the ratio of the exciton to biexciton binding energies is nearly constant (~0.25), irrespective of the laser power, as shown in Fig. 2(c). The emissions evolution with increasing power is opposite to that obtained with rising temperature. As shown in the inset of Fig. 2(c), the peak associated with biexciton emissions grows strongly with increasing power, while the one associated with exciton grows less intensely. The fit of the integrated PL intensity as a function of power was obtained using an expression of the type, $I \propto P^{\gamma}$. The range of the γ values helps interpreting the nature of the transitions. We observe that the exciton peak has a linear behavior with power (γ ~ 0.9), while the biexciton peak has a superlinear behavior (γ ~ 1.2). All these results are in quantitative agreement with previous reports of biexciton states [17].

B. Magnetic-field dependence of the excitonic emission

Now we focus on the magnetic-field dependence of the quasiparticle emissions. Figure 3 shows the PL spectra at a fixed temperature T = 3.5 K and four different laser powers, namely, 0.1, 0.8, 7.9 and 68 μW, in the presence of a magnetic field of 9 T for both circular σ^- (Fig. 3 (a)) and σ^+ (Fig. 3 (b)) polarization components. Polarization-dependent magneto-PL results revealed that the optical response of the GaAs/AlGaAs quantum well strongly depends on the excitation power of the laser. The exciton to biexciton intensity ratio Increases with increasing laser power. More interestingly, in Fig. 3(b) an additional transition emerges at a finite magnetic

field, ascribed to the trion spin triplet state X^-, which appears predominantly for the σ^+ polarization at low laser power and disappears at the highest power. In the case of σ^-, the corresponding transition could only be identified at very low intensity. Yet, due to the lack of experimental resolution, it was not possible to perform a reliable deconvolution with the fitting procedure.

To explore the dependence of the exciton and biexciton states on the external magnetic field, and also investigate the new magnetically-induced peak, we show, in Fig. 4, the polarization-dependent magneto-PL results as a function of the magnetic field, for a low laser power of 0.8 μW. Fig. 4(a), illustrates the position of all the observed emissions as a function of the applied magnetic field. Panel (b) shows the corresponding PL intensity, calculated as the integrated area of each individual peak. Note that, in the presence of an external magnetic field, the exciton intensity increases, while the biexciton intensity decreases. Figure 4(c) displays the Zeeman splitting ΔE, obtained as the energy difference between the $\sigma+$ and σ^- emission peaks. Finally, in Fig. 4(d), we exhibit the degree of circular polarization (DCP) of the photoluminescence, determined as $DCP = (I_Y^+ - I_Y^-)/(I_Y^+ + I_Y^-)$.

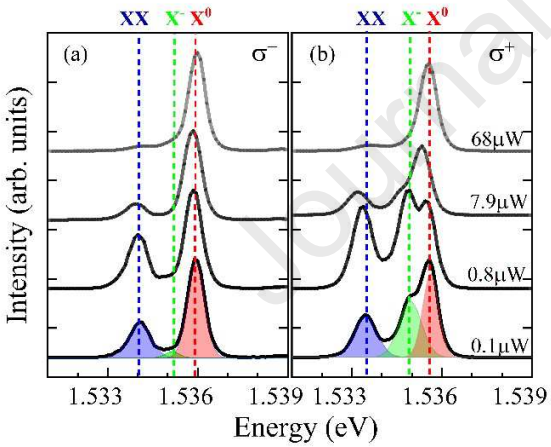

FIG. 3: Photoluminescence spectra of the quantum-well emissions at T = 3:5 K as a function of laser power for B = 9 T with (a) σ^- and (b) σ^+ luminescence components. The vertical green line indicates a new state (trion, X^-), which is clearly present for the (σ^+) luminescence component.

Let's first focus on the differences between the exciton and biexciton dependence on the magnetic field. In Fig. 4(c), we observe a nonlinear Zeeman splitting for the excitons, including a change of its sign. These features were reported before and are related to the spin-dependent field-induced admixture between the light- and heavy-hole valence subbands [23–25]. For the biexciton state, on the other hand, we observe that the Zeeman splitting is essentially linear with the field, especially for B > 3T, which indicates a nearly constant g-factor. Biexcitons with different Zeeman splittings from exciton states were reported before in quantum dots [26], and interpreted as a consequence of non-trivial spin configuration. These magnetically induced energy splitting lead to a population imbalance of the Zeeman sublevels that manifests itself as a finite degree of DCP, as shown in Fig. 4(d). The magnetic-field evolution of the DCP allows to corroborate the seeming decoupling of the biexciton dynamics from the rest of the excitonic complexes. According to Eqs. (3) and (5), the dynamics of the uncoupled biexciton states, in stationary conditions, can be reduced to,

$$0 = P_{XX}^{\pm} - \frac{n_{XX}^{\pm}}{\tau_{r,XX}} \mp Z(XX) \tag{7}$$

By defining $DCP_{XX} = (n_{XX}^+ - n_{XX}^-)/(n_{XX}^+ - n_{XX}^-)$, then,

$$DCP_{XX} = \text{sign}(\Delta E_{XX}) \frac{D(|\Delta E_{XX}|) - 1}{D(|\Delta E_{XX}|) + 1 + \frac{\tau_{XX}^s}{\tau_{r,XX}}} \tag{8}$$

With, $\Delta E_{XX} = E_{XX}^+ - E_{XX}^-$. In the limit $\tau_{XX}^s/\tau_{r,XX} \to 0$, this expression reduces to

$$DCP_{XX} = -\tanh\left(\frac{\Delta E_{XX}}{2k_B T}\right). \tag{9}$$

Note that the absolute value in the argument has disappeared. Indeed, the biexciton DCP follows the Zeeman Splitting behavior, ΔE_{XX}, of Fig. 4(c) and can be fitted according to Eq. 9, describing the thermal equilibrium population of Zeeman sublevels. The experimental data (square symbols) and the theoretical prediction (line) are shown in Fig. 4(d).

A good agreement between both can be achieved by using the nominal value of the temperature, i.e., 3.5 K. This corroborates the biexciton decoupling that was already observed in the previous discussion on the temperature dependence. The polarization of the exciton state (circular symbols), on the other hand, does not simply follow the Zeeman splitting; instead, we observe a negative polarization even for negative ΔE, which indicates a weaker emission from the lower energy spin component.

To understand the intricate and contrasting magneto-optical emission of various excitonic states, one has to take account of the magnetically-induced state, which we attribute to negative trions. Such a state is observed for σ^+ polarization and finite magnetic field (B > 3 T), as shown in Fig. 4(a). The inset displays its binding energy,

i.e., the energy difference between the $X^0_{\sigma+}$ and the $X^-_{\sigma+}$ states. We observe that it depends linearly on the magnetic field, as previously observed for negative trions [10]. Furthermore, our results show that the emission intensity of the $X^0_{\sigma+}$ state is strongly affected by the emergence of the $X^-_{\sigma+}$ state [Fig. 4(b)]. Interestingly, it can be seen that the magnetic field at which the new peak appears coincides with the crossing between the two excitonic states with different polarization. That is, the X^- peak becomes visible when the energy of the σ^+ exciton state is lower than the energy of the σ^- state.

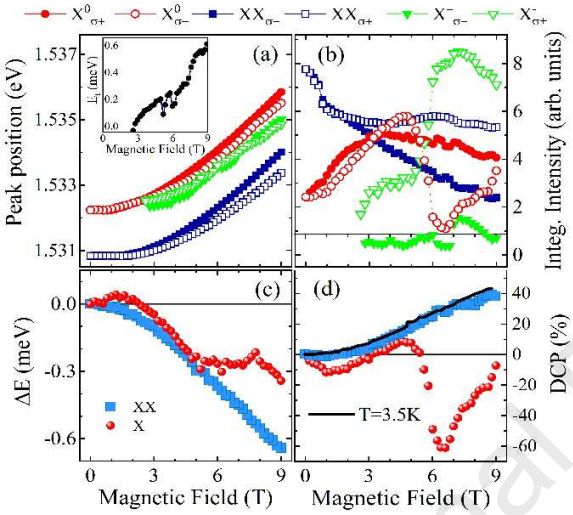

FIG 4: Results of the photoluminescence measurements of the quantum-well emissions at T = 3:5 K and laser power of 0.8 μW as a function of magnetic field. (a) Peak positions and (b) integrated PL intensities as a function of magnetic field for X^0 (red circles), XX (blue squares), and X^- (green triangles). Filled and empty symbols represent σ^- and σ^+ circular polarization, respectively. (c) Energy variation ΔE of circularly polarized emission of the exciton and biexciton. (d) Degree of circular polarization of the exciton and biexciton. The black line indicates the polarization of the biexciton obtained according to the equation of the thermal equilibrium population of Zeeman sublevels at T=3.5 K. The inset of (a) shows the trion binding energy, E_t, as a function of the applied magnetic field.

The spin asymmetry of the exciton and trion emissions can be further explored with our theoretical framework. To proceed, we solved the coupled Eqs. (2) and (4) by setting $F^{\pm}_{XX} = 0$ since the biexciton dynamics can be decoupled, as proven above (see Figs. 2(b) and 4(d) and related discussions). As presented along the temperature-dependence analysis, we kept a minimum number of free parameters to allow an accurate reproduction of the experimental results. Namely, we fixed $\tau_{r,X_0} = \tau_{r,X_-} = \tau_t = \tau^s_{X_0} = 1$ ps, and left only $\tau^s_{X_-}$ as a free parameter (see Eq. 5). In Fig. 5(a)-(c) we show the calculated PL intensity as a function of magnetic field for different values of $\tau^s_{X^-}/\tau^s_{X_0}$. We observe that the exciton emission is strongly influenced by this time ratio. Since the spin-down states (σ^-) have a higher energy than the correspondent spin-up states (σ^+), a faster spin-flip process suppresses the $X^-_{\sigma-}$ emission, in agreement with the experimental results displayed in Fig. 4(b). We also note that, while the $X^-_{\sigma+}$ and $X^-_{\sigma-}$ populations monotonically grow with increasing magnetic field, the $X^0_{\sigma+}$ intensity decreases, while $X^0_{\sigma-}$ remains almost constant. The physical reason for that is the thermally activated trion dissociation ($D(E_t)$) that decreases as the magnetic field increases, following the enhancement of the trion binding energy. This mechanism leads to the decrease of the $X^0_{\sigma+}$ emission and the simultaneous increase of $X^-_{\sigma+}$ emission, which is also corroborated in our experimental results. Indeed, our model can accurately describe the coupled exciton-trion spin-dependent magneto-optical emission, as shown in Fig. 5(d) by the color map of the PL intensity as a function of the magnetic field, obtained with $\tau^s_{X^-}/\tau^s_{X_0} = 0.1$.

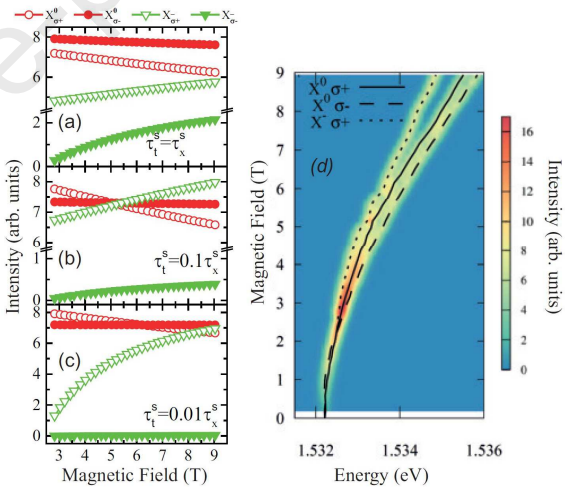

FIG. 5: (a)-(c) Theoretical simulation of the quasi-particle emission intensity for different values of τ^s_t/τ^s_x. (d) Color map of the evolution of the PL spectral intensity as a function of the external magneticfield. The four-level model with a minimum number of free parameters can accurately describe the coupled exciton and trion dynamics. The black dots, dashed and solid line are the corresponding experimental data used as eye guides.

IV. CONCLUSIONS

In conclusion, we experimentally and theoretically investigated magneto-optical and thermal properties of excitonic complexes in a GaAs quantum well. Via a systematic study of polarization-resolved photoluminescence measurements, we determined the Zeeman splitting and the quasi-particle binding energies. Through a combination of experimental results and theoretical framework, we shed light on an old controversy about the nature of a low energy excitonic

peak, and we show that it can be attributed to a biexciton emission. With our non-resonant excitation, the biexcitons are mainly photocreated and their dynamics is nearly independent of that of the excitons and trions. The biexcitons PL emission shows a supralinear dependence on laser power, strongly decreasing with increasing temperature and shows a monotonic Zeeman splitting, a constant g-factor, and a polarization that is defined by the thermal population of the Zeeman sublevels. The magnetically-induced state, which we showed to be consistent with a negative trion. This state appears predominantly for σ^+ polarization under a finite magnetic field and has a strong influence on the exciton emission. The intricate dynamics of excitons and trions, including their spin asymmetry, could be satisfactorily explained by our theoretical framework.

V. ACKNOWLEDGMENTS

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Highlights

- Excitonic complexes play a central role in the physics of semiconductor nanostructures as their quantum behavior is of high interest for fundamental research.
- Particularly in GaAs/AlGaAs quantum wells, the appearance of the photoluminescence emission at higher magnetic fields (> 3 T), for only a specific circular polarization degree is still controversial.
- We have identified and resolved all lines under different dependences such as temperature, laser power and magnetic fields, all in agreement with our proposed model.
- Thus, our work sheds light on a recurrent ambiguity in the identification of the dynamics of excitonic complexes recombination in a GaAs/AlGaAs quantum well.

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Dear Editors,

We declare that there are not conflict of interest in our manuscript ***"Magnetic and power range tuning of spin-asymmetric multiple exciton in a GaAs quantum well"***.

Sincerely,

Marcio Daldin Teodoro on behalf of the authors