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The Role of Spin in Interacting Excitonic Gases

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The spin-dependent exciton–exciton interactions in GaAs quantum well (QW) systems have been studied, under resonant and non-resonant excitation conditions at liquid helium temperatures. In the former case, a pure exciton gas is created, whereas in the latter the excitons are surrounded by a charged electron–hole plasma. In a double QW structure, the distance between electrons and holes comprising the excitons could be externally tuned by applying an electric field perpendicular to the well planes. The interactions were studied in terms of their dependence on excitation intensity, excess energy, spin polarization and electron–hole separation using time- and polarization-resolved photoluminescence up-conversion spectroscopy.

Introduction When an exciton gas is so dense that the exciton wave functions begin to overlap, i.e. the volume that one exciton occupies multiplied by the density approaches unity, the single-particle picture is no longer valid. In dense exciton gases, interactions or non-linear effects have to be taken into account. In this respect, it is also important to distinguish between the cases of excitation of a pure exciton gas and that where an additional electron–hole (e–h) plasma is excited in the material. In low-temperature experiments, a resonant excitation leads to a pure exciton gas. However, a non-resonant excitation also produces an e–h plasma at short times after excitation, which relaxes with time either into excitons or recombines directly into photons. At higher lattice temperatures, a resonant excitation initially creates a pure exciton gas, which is rapidly ionized by phonons (<300 fs in GaAs quantum wells (QWs) at room temperature [1]). When a dense e–h plasma is excited, the electronic states change with respect to the single-particle states, which results in a renormalization of the band gap [2, 3]. The binding energy, E_b^{2D}, of the exciton measured from the renormalized band gap decreases so that the size of the orbitals, a_{2D}, increases and the oscillator strength decreases. Finally, at a certain density, the exciton resonance disappears and is replaced by the shifted continuum. This effect of the band gap renormalization on the excitons is generally called 'screening' [1]. The underlying processes for this behaviour are long- and short-range Coulomb interactions. In the presence of a charged e–h plasma, the long-range Coulomb interaction effectively screens the e–h attraction in an exciton, which in turn reduces its binding energy. As a result the electron and hole are further apart and the orbital wave function is enlarged in space. In two-dimensional (2D) sys-

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tems this screening is much less effective than in bulk materials [4, 5]. If only a pure exciton gas is present, this long-range part of the Coulomb interaction is of the van der Waals type. A dipole–dipole attraction occurs which has been shown experimentally as very small [6] and is normally neglected.

The exchange interaction in fermions is defined as the short-range contribution of the Coulomb interaction and consists of two parts [7]. The first is the self-energy correction which comprises the electron and hole exchange, i.e. the energy that parallel spin particles gain by avoiding each other, and is commonly termed exchange correction (EX). The second describes the weakening of the attractive e–h interaction due the exclusion principle, which is generally called the vertex correction (VC). When only excitons are present, a small blue shift with increasing density, n_X, is expected in two dimensions [7], which has been seen experimentally [8, 9] (in three dimensions it is partially compensated by the screening red shift).

Until now the spin of the excitons has not been taken into account. Including this degree of freedom in the above-developed scheme for the interacting exciton gas leads to surprising new features appearing [10, 11]. The direct Coulomb interaction acts as charge screening and is therefore invariable on spin orientation. Only the exchange and vertex correction depend on the spins of the excitons. Fernández-Rossier et al. studied theoretically the spin effects on the exciton binding energy using a mean-field approach. They considered a pure exciton gas at $T = 0$ and focused on the ± 1 spin component, neglecting the ± 2 states (although this approximation is not strictly justified, the good agreement with experiment seems to validate it). The gas is spin polarized, which means that one spin component has a higher density, $n_X^{\pm 1}$, than the other, where the polarization is defined by $P = (n_X^{+1} - n_X^{-1})/(n_X^{+1} + n_X^{-1})$, assuming the $+1$ component as the majority spin population. At excitation densities well below the saturation limit, the interaction between excitons with parallel spin is in the meV range, while that between cross-polarized excitons is much smaller and will be neglected in the following. Since the gas is polarized, different effects on the binding energy of majority and minority excitons due to the exchange and vertex interaction are expected. As stated above, the exchange between two parallel spin particles reduces the energy and thus increases the binding energy of the exciton with increasing exciton density. The vertex interaction, which results from the Pauli exclusion principle for fermions, in contrast becomes more and more repulsive on increasing the density and decreases the exciton binding energy. For low densities, $n_X a_{2D}^2 < 0.1$, Fernández-Rossier et al. obtained a linear expression (neglecting cross-polarized interactions) for the change of binding energy in a spin-polarized 2D system:

$$\Delta E_b^{2D\pm} = n_X^{\pm}(I_{EX} - I_{VC}) ,$$

where I_{EX} and I_{VC} denote the expectation values of the interaction Hamiltonian for the exchange and vertex terms, respectively. The analytical calculations for low density, and the numerical simulation for higher densities, showed that I_{VC} is always greater than I_{EX} and $\Delta E_b{}^{2D}$ is negative, leading to a decrease of the binding energy. Since the exciton gas has two non-equal components, i.e. $n_X^{+1} > n_X^{-1}$ as assumed before, the majority component has a higher total energy (lower binding energy) than the minority component. If emission due to optical recombination of excitons is observed, an energy splitting, δ_{sp}, in the emission lines is expected, which could reach several meV at densities below saturation: $\delta_{sp} = n_X P(I_{VC} - I_{EX})$. Fernández-Rossier et al. then introduced an

additional degree of freedom in the system by considering the distance between the electrons and holes by means of a parameter d. Assuming the electrons and holes are each confined to parallel 2D layers, which are separated by a distance d, they found a significant dependence of the relative strength of I_{EX} and I_{VC} on d. The vertex correction is reduced when the electron plane is separated from the hole plane in space, while the exchange energy increases. So by increasing d from zero to a finite value could lead to a quantum phase transition between a paramagnetic phase (dominated by vertex correction) and a ferromagnetic phase (dominated by exchange). Experimentally, this would manifest itself in a reduction of the splitting, δ_{sp}, when electron and hole layers are separated. On increasing d, δ_{sp} reaches zero and eventually reverses its sign. The existence of a negative splitting indicates that a transition to a ferromagnetic phase has taken place. Such a phase would have important consequences for the spin-flip processes, since the majority excitons now would see an energy barrier that prevents them from flipping to the opposite spin and so the phase would be stable, or P could even increase to unity.

Before the calculations of Ref. [10] were made, the spin splitting had been observed in GaAs multiple QW (MQW) samples. Damen et al. [12] reported a splitting between the two spin components of up to 1.5 meV on increasing the excitation density under non-resonant excitation conditions. The polarization values were between $P = 0.1$ and 0.3. In a later experiment by Viña et al. [13] initial polarizations of $P = 0.8$ were achieved under non-resonant excitation conditions and a splitting up to 4.5 meV was observed, which is comparable to the exciton binding energy, E_b^{2D}. Subsequent experiments under resonant excitation conditions verified these results very clearly [14].

Spin-Dependent Interaction in Double QWs under an Electric Field In this section we show the effects of varying the parameter d on the exciton interactions. In a double QW (DQW) structure, the electrons and holes of the excitons can be confined to opposite wells by applying an electric field, E, to the structure [15, 16]. Thus the emission energies of the spin-polarized exciton gas can be measured as a function of the interplane separation d, which will be defined as the distance between the z-expectation values, using the electron and hole wave functions. The structure used in this study consists of ten 50 Å GaAs DQWs separated by 20 Å $Al_{0.3}Ga_{0.7}As$ barriers. A 200 Å wide $Al_{0.3}Ga_{0.7}As$ layer separates each of the 10 periods. The intrinsic (i) DQW structure is embedded in an n^+–i–p^+ heterojunction, which allows the application of a voltage perpendicular to the DQWs. The p^+ and n^+ electrodes consist of a 500 nm GaAs:Be layer and a 1000 nm thick GaAs:Si layer on a [100] GaAs substrate, respectively. The doped layers are separated from the DQW stack on both sides by a 100 and 80 nm wide intrinsic $Al_{0.3}Ga_{0.7}As$ layer. The confined conduction (e) and heavy- (light-) hole valence band states h (l) of both wells couple and form symmetric and anti-symmetric states e1, e2, h1, h2, l1 and l2, which can form eight excitonic transitions. The voltage to field correspondence, depicted in the upper and lower x-axes in Fig. 1a, was obtained by calculating the voltage drop over the intrinsic region of the structure. The points in Fig. 1a represent the energy positions of the peaks measured by photoluminescence excitation (PLE) at $T = 8$ K for several voltages. The dotted lines in Fig. 1a represent the calculated transition energies using our sample parameters. The effective distance between electron and hole can be defined as the sum of the z-position expectation value of the respective wave functions, $z_{e/h} = |\langle \Psi_{e/h} \, |z| \, \Psi_{e/h} \rangle|$. When elec-

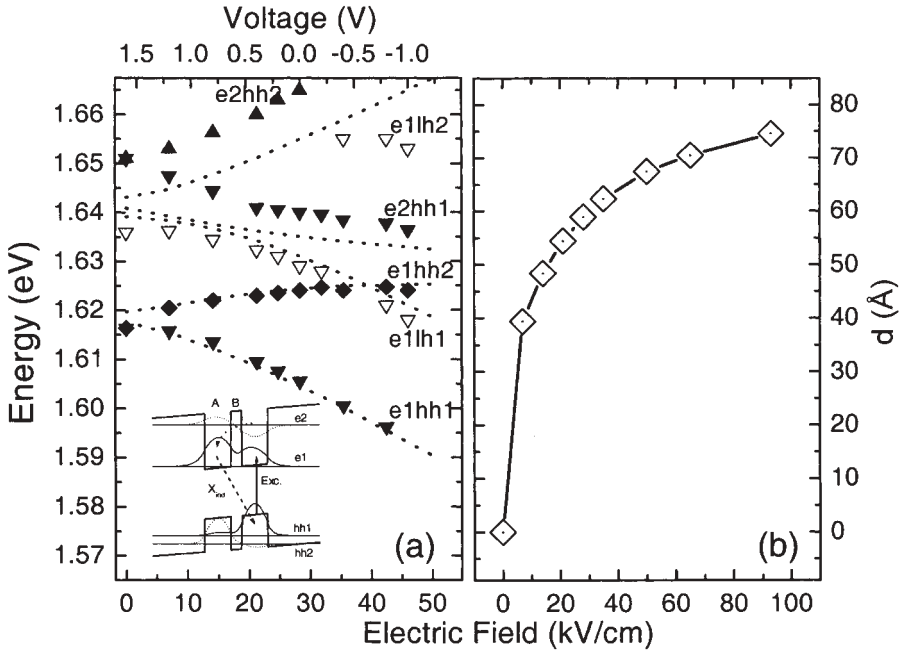

Fig. 1. a) Excitonic transition energies obtained by PLE measurements (symbols). The transitions are labelled according to the text. Dotted lines represent the results of a calculation. The inset shows the level scheme in the DQW. b) Correspondence between inter-plane separation d and electric field E, obtained from the z-expectation values using electron and hole wave functions

tron and hole are confined to opposite wells, we define the inter-plane separation, d, as $d = |z_e| + |z_h|$. Figure 1b shows the relation between the electric field and d as calculated from the single-particle wave functions. The wave functions become localized already at small fields and therefore d increases quickly until it begins to saturate for $E \sim 40$ kV/cm. The maximum separation is finally limited by the geometry of the structure. The time-resolved PL measurements were done using a standard one-colour (1 ps resolution) and two-colour (150 fs resolution) up-conversion set-up for non-resonant and resonant excitation experiments, respectively.

Non-Resonant Excitation Conditions As outlined above, the energy splitting in a spin-polarized exciton gas is expected to change when the electrons and holes comprising the excitons are localized in separate 2D layers. The contributions of the vertex and exchange corrections change by increasing the distance between these two confinement layers.

The excitation energy in the non-resonant experiment was tuned to the e2hh1 transition (see Fig. 1a) and the excess energy was $\sim$30 meV. A constant initial polarization at $t_d = 15$ ps of $P = 0.5 \pm 0.05$ was obtained over the whole range of excitation densities and electric fields used in the experiments (see Fig. 2e). Also, the density created in the indirect e1hh1 transition was independent of the field strength because of the excitation at the direct e2hh1 transition, the absorption of which does not change significantly with the field. These two facts are important for the interpretation of the results since

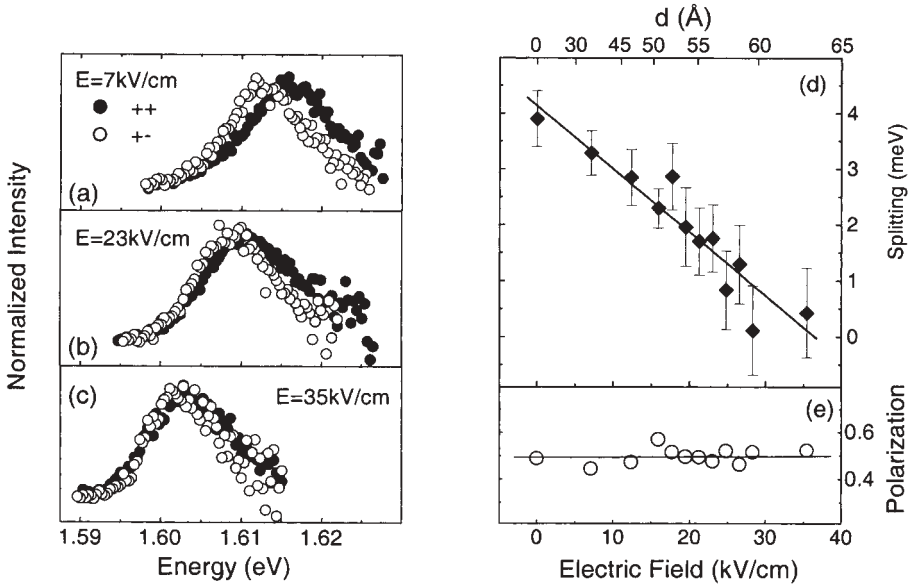

Fig. 2. PL spectra of the two polarizations corresponding to the +1 and −1 exciton energies for I_{exc} = 200 W/cm^2 under non-resonant excitation conditions and at three different electric fields: a) 7; b) 23; c) 35 kV/cm. d) Energy splitting as a function of electric field and distance, d, between the electron and hole layer. The correspondence between E and d is taken from Fig. 1 b. e) Initial polarization as a function of electric field

the energy splitting between the ±1 components is proportional to P and n_X. In Figs. 2a–c, a series of spectra taken at t_d = 32 ps and I_{exc} = 200 W/cm^2 for three different electric fields are shown for the two polarizations, exciting with σ^+-polarized light. In the low-field case a splitting similar to the one obtained in Ref. [13] is observed. The minority emission peak lies below the majority one. As the field is increased under constant n_X, t_d and P, the splitting becomes smaller and finally at a field of E = 35 kV/cm the spectra, corresponding to the two polarizations, overlap in energy. Apart from the splitting between +1 and −1 energies, a spin-independent red shift is induced by the field, which is due to the Stark effect. Figure 2d shows the splitting, δ_{sp}, as a function of electric field. A linear correlation between δ_{sp} and E is found. The splitting reaches zero just at the onset of ionization of the excitons in this sample, which hindered further exploration.

Turning our attention to the individual energies of the ±1 excitons as a function of excitation density, we have observed that these energies show a strong field dependence, as depicted in Fig. 3. At low fields, the energy of the +1 excitons does not change appreciably with power, while that of the −1 excitons decreases markedly. Again this is a similar behaviour to that observed at the zero field measurements [13], where it could be explained by adding screening corrections to the VC and EX effects. The same situation is observed in the present experiment at 7 kV/cm: the predicted blue shift of $E_X^{\pm}$, due to the negative correction to the binding energies, $\Delta E_b^{2D\pm} = n_X^{\pm}(I_{EX} - I_{VC})$, is cancelled and overcompensated by the screening for E_X^{+} and E_X^{-}, respectively. At intermediate fields (Fig. 3b) both PL components shift to higher

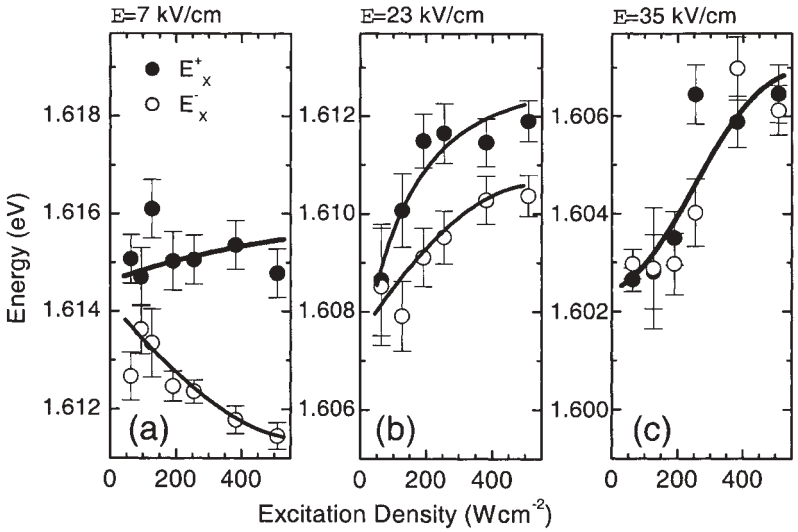

Fig. 3. Density dependence of the emission energies at $t_d = 15$ ps and: a) $E = 7$; b) $E = 23$; c) $E = 35$ kV/cm under non-resonant excitation conditions. The changes in the energy scales are due to the spin-independent Stark shift

energies with increasing excitation power, although at a different rate. Finally, at the highest fields (Fig. 3c), the rate of blue shift is the same for both components, and the splitting becomes zero, within experimental accuracy, at all powers.

Resonant Excitation Conditions Resonant excitation measurements were done on the same DQW structure. By exciting the indirect e1hh1 exciton directly, no e–h plasma is created and the excitons have a cold distribution around $K_{\|} = 0$. Due to the field-dependent absorption in e1hh1, the photoinduced exciton density, n_X, does not depend linearly on the excitation density, I_{exc}, and an effective density, I_{exc}^{eff}, has to be defined. In the following all excitation densities will be given as effective densities $I_{exc}^{eff} = I_{exc}\alpha(E)$.

Figures 4a and b show the emission spectra for the two spin polarizations at zero field and at $E = 20$ kV/cm, respectively. At zero field, a splitting of $\sim$4.5 eV is obtained when $I_{exc}^{eff} = 200$ W/cm^2 and $P = 0.9$. The splitting is already reduced to zero at $E > 20$ kV/cm. The dependence of the splitting on the applied field for $I_{exc}^{eff} = 100$ W/cm^2 and $P = 0.9$ is shown in Fig. 4c. The energy splitting, δ_{sp}, decays rapidly at low fields and saturates at zero for higher fields. The dependence on E is different from that under non-resonant excitation and the exact balance between exchange and vertex correction is reached already at lower fields. However, it seems that δ_{sp} saturates at zero and does not become negative, i.e. the exciton system does not undergo a para-/ferromagnetic phase transition at this e–h separation of $d > 50$ Å. In Fig. 4c the experimental splittings for the resonant and the non-resonant cases are compared with the calculated values from Ref. [10]. Since the splittings are density dependent [13], the data for the resonant case correspond to an effective excitation density half of that used to obtain those for the non-resonant situation; under these conditions the splittings become comparable at $E = 0$, which facilitates the comparison with theory. According to theory, the splitting decreases very quickly with the e–h distance, d, and becomes negative at $d \sim 20$ Å.

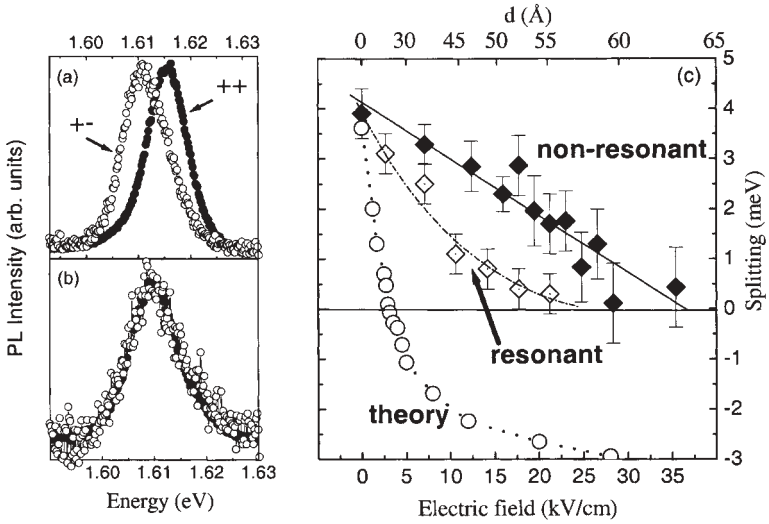

Fig. 4. a) PL of the two spin components at zero field, $I_{\mathrm{exc}}^{\mathrm{eff}} = 200$ W/cm and $P = 0.9$ under resonant excitation conditions. Spectra are normalized to the spectrally integrated intensities. b) PL at a field of 20 kV/cm and the same effective excitation intensity. c) Splitting as a function of E and d under resonant (open diamonds, $I_{\mathrm{exc}}^{\mathrm{eff}} = 100$ W/cm^2, $P = 0.9$) and non-resonant excitation conditions (filled diamonds, $I_{\mathrm{exc}}^{\mathrm{eff}} = 200$ W/cm^2, $P = 0.5$), and the theoretical values from Ref. [10]. The correspondence between E and d is taken from Fig. 1b

Although in the experiment, where only cold ($K \sim 0$) excitons are present, the reduction of the splitting is faster than in the non-resonant case (coexistence of e–h plasma and excitons), it still does not reverse its sign and becomes negative. To investigate whether the phase transition really exists, it would be desirable to use new sample structures, such as triple QWs or superlattices, to achieve a higher e–h separation while maintaining the exciton character. The theoretical model has to be improved since it assumes a delta-function-like confinement of electron and hole to strictly 2D layers, which are a distance d apart. Thus the excitonic interactions might be overestimated by these assumptions and d in reality has to be still larger in order to get into the ferromagnetic regime. The dynamics of the energy positions are similar to those observed under non-resonant conditions. However, since the spectra are not broadened by plasma contributions under resonant excitation conditions, these positions are accessible at very short delay times.

The dependence of the emission energies of the two spin components on excitation densities, shown in Fig. 5, is quite different to that obtained under non-resonant conditions (see Fig. 3). Although at zero field the +1 energy is almost independent of field and the −1 component shows a red shift, as in the non-resonant case, at larger fields the behaviour departs considerably from that of the non-resonant case. The +1 emission now also remains invariant with field and does not shift; for the minority −1 excitons the emission energy maintains the red shift that becomes smaller on increasing the field. Finally at high fields, the +1 and −1 energies overlap. This difference at finite fields from the non-resonant case is very interesting: it suggests that the e–h plasma effectively screens the attraction between electron and hole forming an exciton only

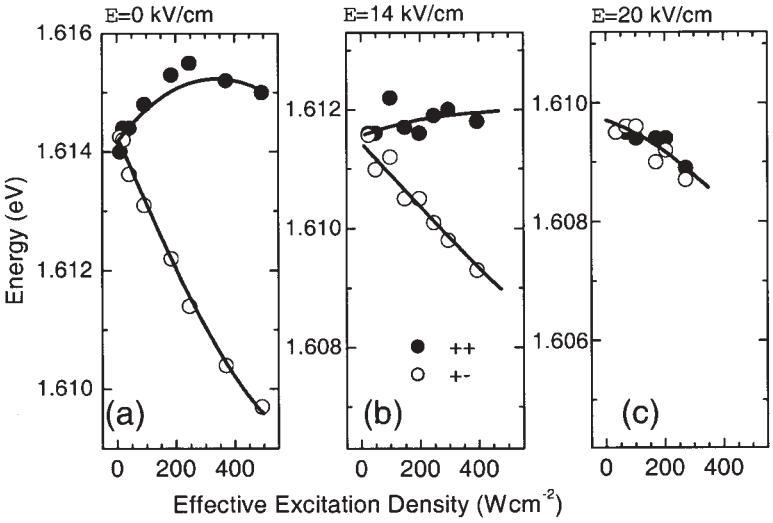

Fig. 5. Density dependence of the emission energies for ± 1 components under resonant excitation conditions ($P = 0.9$) and for three different electric fields: a) 0; b) 14; c) 20 kV/cm. Energy scales are adjusted for the spin-independent Stark shift

when they are confined to separate 2D layers, i.e. when the system becomes a mixture between 3D and 2D geometries. As the screening is much more effective in 3D than in 2D, at $d > 0$ the plasma reduces the exciton binding energy and leads to a blue shift of the emission (see Figs. 3b and c). To date there has been no theoretical work reported in the literature that treats the screening corrections for a DQW structure where electrons and holes are confined to separate layers. However, recently de Leon et al. [17] obtained from their theoretical approach that the screening due to the direct Coulomb interaction is negligible at zero field, but that it increases at separating electron and hole layers.

Conclusions In a DQW system it is possible to alter the spin-dependent exciton–exciton interactions by applying an electric field perpendicular to the DQW plane. Under both non-resonant and resonant excitation conditions, the energy splitting, δ_{sp}, between the $+1$ and -1 spin components in a polarized exciton gas can be tuned between $\sim$5 meV and zero by the applied field. An increasing field reduces the e–h overlap, which, in turn, reduces the vertex correction and increases the exchange correction, leading to a reduction of the splitting. Whereas under non-resonant excitation δ_{sp} decays linearly with increasing field, under resonant excitation this decay has a positive curvature and reaches $\delta_{sp} = 0$ at lower fields than in the former case. A theoretical model can reproduce this reduction of the splitting qualitatively; however, it overestimates the effect of the e–h separation on the vertex and exchange corrections, yielding $\delta_{sp} = 0$ already at lower fields than those observed in the experiment under resonant excitation conditions. The presence of an e–h plasma, coexisting with the polarized exciton gas, gives rise to a screening of the exciton–exciton interactions, which is only important when the system becomes a mixture between 3D and 2D geometry.

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