Electric-Field Tuning of Spin-Dependent Exciton-Exciton Interactions in Coupled Quantum Wells

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We have shown experimentally that an electric field decreases the energy separation between the two components of a dense spin-polarized exciton gas in a coupled double quantum well, from a maximum splitting of ~4 meV to zero, at a field of ~35 kV/cm. This decrease, due to the field-induced deformation of the exciton wave function, is explained by an existing calculation of the change in the spin-dependent exciton-exciton interaction with the electron-hole separation. However, a new theory that considers the modification of screening with that separation is needed to account for the observed dependence on excitation power of the individual energies of the two exciton components.

Spin-dependent exciton-exciton interactions in low dimensionality semiconductors have been studied extensively during this decade [1–8]. The first observation of an energy splitting between the two spin-polarized components of an exciton gas without a magnetic field was reported by Damen *et al.* [1]. Since then, this effect has been considered both experimentally [5,8] and theoretically [6,7]. Fernández-Rossier *et al.* attributed that splitting to a density- and spin-dependent modification of the two-dimensional (2D) exciton binding energy (E_{2D}) by two competing processes [6,7]. One of these many-body processes is the exchange interaction (I_{EC}) between electrons (and holes) of different excitons, which always *increases* E_{2D} in proportion to the spin-polarized exciton density, so that the change, $\Delta E_{2D}^{\pm}$, is $\Delta E_{2D}^{\pm} \propto n_X^{\pm} I_{EC}$, where $n_X^{\pm}$ refers to the spin +1 and −1 exciton populations, respectively [9]. The second process is the vertex correction (I_{VC}) to the Coulomb interaction between electrons and holes of different excitons. This correction effectively reduces the electron-hole attraction due to the occupation of final states in the electron-hole scattering processes and *lowers* E_{2D} proportionally to $n_X^{\pm} I_{VC}$. Hence, neglecting the small coupling between +1 and −1 states due to valence band mixing, the total change of the exciton binding energy is $\Delta E_{2D}^{\pm} \propto n_X^{\pm}(I_{EC} - I_{VC})$.

When the overlap between the electron and hole wave functions is large the vertex correction outweighs exchange effects [that is, $(I_{EC} - I_{VC}) < 0$] and the exciton binding energy is reduced (with respect to that of a single exciton). Moreover, when $n_X^+ > n_X^-$, the reduction is such that $|\Delta E_{2D}^+| > |\Delta E_{2D}^-|$, leading to a smaller binding energy of the +1 excitons than that of the −1. As a result, an energy splitting $\delta\epsilon$ between the components of a dense spin-polarized quasi-2D exciton gas is expected. Such a splitting has been observed in time-resolved photoluminescence (PL) experiments in quantum wells, in which a circu-

larly polarized light pulse (σ^+) creates excitons with spin +1 that gradually relax into the spin −1 state, until both populations are equal. The PL spectrum corresponding to the spin +1 exciton has been found to be at higher energy than that of the −1 exciton, with the splitting between the two, $\delta\epsilon$, decreasing exponentially with time.

However, until now, no experiment has tested the theoretical prediction of a decrease or even a sign reversal in $\delta\epsilon$ caused by a modification of the vertex correction and the exchange term when the overlap of electron and hole wave function is reduced [7]. We report here the first evidence of such a change, determined from measurements of $\delta\epsilon$ in coupled double quantum wells (CDQW) subjected to a longitudinal electric field that separates spatially the electrons and holes. We not only find good agreement between the observed energy splitting and the predicted one but our results go beyond available models. Specifically, we have found that screening of these many-body corrections is strongly reduced by the field, an effect not contemplated by theory so far.

The material structure used for the experiments was an n^+-i-p^+ GaAs-AlGaAs heterojunction. The active region consisted of ten CDQW periods separated from each other by a 200 Å $Al_{0.3}Ga_{0.7}As$ layer, and each formed by two 50 Å GaAs wells with a 20 Å $Al_{0.3}Ga_{0.7}As$ barrier in between. An undoped $Al_{0.3}Ga_{0.7}As$ layer on each side of the CDQW stack (100 and 80 nm thick, respectively) completed the intrinsic (i) region of the structure, whose p^+ and n^+ electrodes were a 500 nm p^+-doped GaAs:Be layer and a 1 μm thick n^+-doped GaAs:Si layer on a [100] GaAs substrate, respectively.

A schematic representation of the potential profiles and energy levels of the CDQW structure at a field of 14 kV/cm is shown in Fig. 1a, where $e1$ and $e2$ represent the coupled-well states in the conduction band, and $hh1$

and $hh2$ the corresponding states for heavy holes in the valence band. (For simplicity, the light hole states have not been shown.) The field-induced symmetry breaking of the wave functions makes all the transitions optically allowed and the ground-state exciton (X_{e1hh1}) becomes spatially indirect due to the localization of electrons and holes in wells A and B, respectively [10–12].

The experimental electric-field dependence of the various excitonic transitions, obtained from photoluminescence excitation (PLE) measurements on the above heterostructure, is summarized in Fig. 1b. The lower lying indirect transitions X_{e1hh1} and X_{e1lh1} undergo a redshift with increasing field, primarily because of the tilting of the potential, although at high fields the Stark effect contribution becomes appreciable [13]. The two direct transitions X_{e1hh2} and X_{e2hh1} have a less marked field dependence and at high fields they approach each other in energy. The identification of the transitions and determination of the effective field in the intrinsic region were done through a comparison with calculated values obtained by solving numerically the Schrödinger equation within the envelope function approximation [14]. (See dotted lines in Fig. 1b.)

For the study of the spin-dependent exciton-exciton interaction, time-resolved PL measurements were performed with a standard up-conversion setup using a Ti:sapphire laser with 2 ps pulse width. For polarization-selective excitation and PL detection, $\lambda/4$ wave plates were placed in the paths of the laser beam and the PL collecting optics. The sample was mounted in a cold-finger cryostat and held at $T = 8$ K.
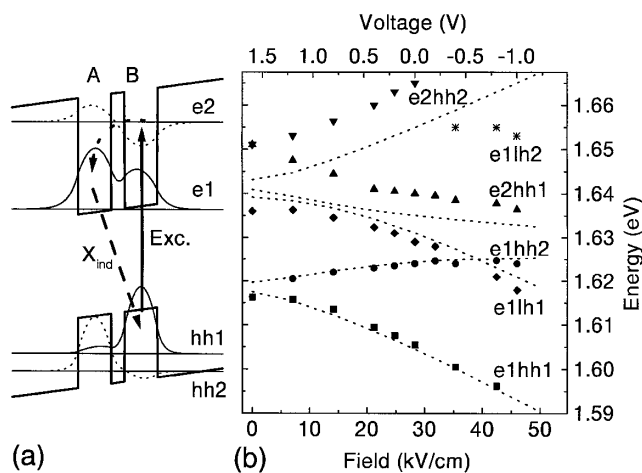

FIG. 1. (a) Schematic structure and levels of the coupled double quantum well. The solid, dotted, and dashed arrows indicate the excitation, relaxation, and recombination processes, respectively. The solid (dotted) line is the symmetric (anti-symmetric) wave function calculated for $\mathcal{E} = 14$ kV/cm. The light hole levels are not shown, for simplicity. (b) Excitonic energies measured by PLE spectroscopy at $T = 8$ K as a function of electric field (points). The upper scale shows the applied voltage corresponding to the internal field. The lines represent the calculated energies (see text for details).

Our focus is on the two spin components of ground-state excitons, X_{e1hh1}, created after optically induced electrons relax from the $e2$ to the $e1$ states (Fig. 1a). An exciton population $n_X \approx 5 \times 10^{10}$ cm^{-2} was created with 15 mW pulses of circularly polarized light, whose photon energy was tuned to the X_{e2hh1} direct exciton. Initially, a $+1$ exciton population was created; however, spin-flipping processes gave rise to a nonequilibrium mixture of $+1$ and -1 excitons with populations n_X^+ (majority) and n_X^- (minority), respectively. The degree of polarization, defined as $P = \frac{n_X^+ - n_X^-}{n_X^+ + n_X^-}$, was found to be initially $P_0 = 0.5 \pm 0.05$, independent of the electric field.

Also almost independent of the field was the excitonic PL rise time, which amounted to 110 ± 10 ps for the X_{e1hh1}^+ and to 140 ± 10 ps for X_{e1hh1}^-. In contrast, the PL decay time, τ_d, increased linearly (almost independently of spin polarization) with increasing field, from 400 ps at $\mathcal{E} = 0$ to 1000 ps at $\mathcal{E} = 35$ kV/cm. For even higher fields the decay time decreased again, and at $\mathcal{E} = 40$ kV/cm it dropped to 250 ps. These two regimes have been explained in terms of a reduction of wave-function overlap and of tunneling of the carriers out of the wells, respectively [15,16].

A measure of the exciton-exciton interaction is the energy difference between the PL spectra of the X_{e1hh1}^+ and X_{e1hh1}^- excitons, plotted in the insets of Fig. 2 for three representative electric fields. These spectra were taken at a time, $t_d = 32$ ps, at which the short-lived peak broadening of the high energy side of the PL spectra due to electron-hole (e-h) plasma luminescence was already negligible [17]. The main body of Fig. 2 shows the time evolution of the peak energies of both spectra. At $\mathcal{E} = 7$ kV/cm (Fig. 2a), the $+1$ (-1) exciton peak shifted to lower (higher) energies and their energy difference decreased exponentially with a time constant $\tau_{sd} = 180$ ps. At higher fields, aside from a field-induced "redshift" of both spectra, the time dependence of the $+1$ exciton peak was not affected by the field, while the -1 exciton time dependence was quite different: at $\mathcal{E} = 23$ kV/cm its energy remained constant with time, while at $\mathcal{E} = 35$ kV/cm it evolved as the $+1$ exciton.

The electric-field dependence of $\delta\epsilon$, defined as the average splitting taken at delays $t_d = 7$, 15, and 32 ps, is summarized in Fig. 3. The maximum splitting of ~4 meV is reached at zero field, and then it decreases linearly until it becomes zero at $\mathcal{E} \simeq 35$ kV/cm. The parameter d in the upper scale of Fig. 3 represents the average separation (along the growth direction) between electron and hole as calculated from the field-dependent expectation values. The zero splitting is reached at an e-h separation of ~60 Å, which is a factor of 2.5 larger than the value predicted by the model [7]. This discrepancy is not surprising in view of the simplifications of the model, such as zero temperature and a strictly 2D confinement.

Since the splitting $[\delta\epsilon = E_X^+ - E_X^- \propto n_X P(I_{VC} - I_{EC})]$ is proportional to the total exciton population, the

polarization, and the many-body corrections, and since P is not much affected by the field, at a given excitation power any change of $\delta\epsilon$ with field has to be related to a modification of the exciton-exciton interaction. Furthermore, $\delta\epsilon(t)$ and $P(t)$ decay with the same time constant, which verifies the expected correlation between $n_X^+ - n_X^-$ and $\delta\epsilon$, assuming a constant n_X (i.e., that the exciton lifetime is much longer than the other decay times).

Analyzing the power dependence of the energies of ± 1 excitons at a short delay time, and hence at constant polarization, allows a direct comparison of the experiments with theory and a test of its underlying assumptions. The energy positions of the two spin polarizations are displayed in Fig. 4 as a function of excitation power. Let us consider first the energy splitting between the two components. At low fields (Fig. 4a), $\delta\epsilon$ increases considerably with power, in agreement with previous experimental results at zero field [5] and the predictions of theory [7]. At intermediate fields ($\mathcal{E} = 23$ kV/cm, Fig. 4b), the splitting still grows with increasing power; however, the values

of $\delta\epsilon$ are reduced with respect to the low field case, as a consequence of the enhanced e-h separation. At a field of $\mathcal{E} = 35$ kV/cm, when the splitting vanishes, it remains zero at all excitation powers, in agreement with the theoretical model, which predicts that at a certain d the I_{VC} and I_{EC} corrections become equal, independent of the total exciton population.

Turning our attention to the individual energies, we have observed that the power dependence of the individual energies of the two spin components has a strong field dependence, as seen in Fig. 4. At low fields, the energy of the $+1$ excitons does not change appreciably with power, while that of the -1 excitons decreases markedly. A similar behavior observed in zero field measurements [5] could be explained only after including screening corrections to the VC and EC effects. Without these corrections, the theory predicted positive slopes in the power dependence of both E_X^+ and E_X^-, in contradiction with the experiments. (Including the effects of screening was not necessary to explain the splitting since screening depends only on n_X and not on the individual $n_X^\pm$.) The same situation is observed in the present experiment at 7 kV/cm: the predicted blueshift of $E_X^\pm$, due to the negative correction to the binding energies $\Delta E_{2D}^\pm \propto n_X^\pm (I_{\mathrm{EC}} - I_{\mathrm{VC}})$, is canceled and overcompensated by the screening for E_X^+ and E_X^-, respectively.

At intermediate fields (Fig. 4b) both PL components shift to higher energy with increasing excitation power,
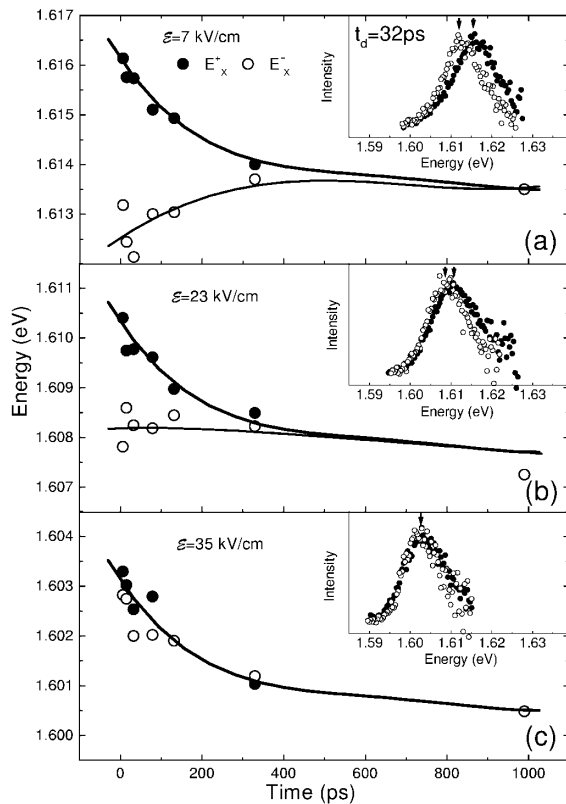

FIG. 2. Peak positions of the two spin-polarized exciton components as a function of time for three different electric fields, at 15 mW excitation power. The excitation is σ^+ polarized and set to the X_{e1hh2} energy. The solid lines are guides to the eye. The line for the $+1$ exciton is always the same, only shifted rigidly in energy by 6 meV (b) and 13 meV (c) with respect to the position in (a). The insets show the normalized spectra for σ^+ (solid dots) and σ^- (open dots) polarization of the emission, 32 ps after excitation. The arrows mark the peak positions. All measurements were taken at $T = 8$ K.
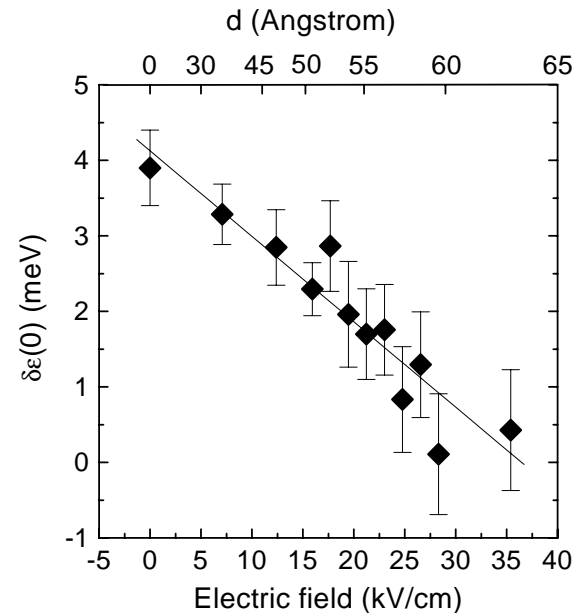

FIG. 3. Energy splitting between the $+1$ and -1 exciton components as a function of electric field. Each data point corresponds to an average over three measurements at delay times $t_d = 7$, 15, and 32 ps. The upper axis represents the electron-hole separation along the growth direction (z) defined as $d = \langle \psi_{\mathrm{electron}} | z | \psi_{\mathrm{electron}} \rangle - \langle \psi_{\mathrm{hole}} | z | \psi_{\mathrm{hole}} \rangle$. The excitation power was 15 mW and the sample was held at 8 K. The solid line is a linear fit to the data.
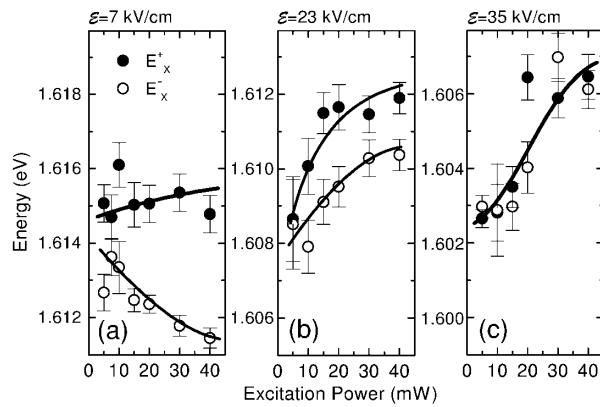
FIG. 4. Position of the E_X^+ and E_X^- PL peaks for three different electric fields as a function of excitation power. Excitation was set to the X_{e2hh1} energy and was σ^+ polarized. The solid lines are guides to the eye. All measurements were performed at $T = 8$ K and $t_d = 15$ ps.

although at a different rate. Finally, at the highest fields (Fig. 4c) the rate of "blueshift" is the same for both components, and the splitting remains zero at all powers. The latter behavior for E_X^+ and E_X^- is similar to that found by a model whose primary focus was on the energy splitting and therefore did not include screening [7]. At first sight, such an agreement at high fields may seem fortuitous, but since at intermediate fields the behavior found experimentally was intermediate between the two extreme fields, it is reasonable to suggest that screening decreases with increasing e-h separation and eventually vanishes. This reduction of screening is far from intuitive; its explanation will require a theoretical model that takes screening properly into account.

Experimental limitations prevented us from testing the prediction of a negative splitting (ferromagnetic phase) when the electron-hole separation is sufficiently large [7]. Field-induced tunneling of carriers out of the wells, and a concomitant exponential increase in photocurrent, limited the maximum applicable field to ~ 35 kV/cm, which probably falls short of the fields needed to explore the complete phase diagram [7].

In conclusion, we have demonstrated that it is possible to alter spin-dependent exciton-exciton interactions in a GaAs CDQW structure by applying an external electric field. This effect manifests itself as a change in the energy splitting between the two components of a dense spin-polarized exciton gas. The splitting can be tuned from a maximum value of 4 meV at zero field to 0 meV at $\mathcal{E} = 35$ kV/cm, which corresponds to an e-h separation of ~ 60 Å. The electric-field dependence of the two competing processes, interexcitonic exchange and vertex correction, reduces the splitting, due to a decrease (increase) of the vertex (exchange) correction with increasing electron-hole separation. Although the applicability of the model is limited, reasonable agreement is achieved

between experiment and theory. Going beyond existing models, our observations indicate that screening of the exciton-exciton interaction is also a function of the separation of electrons and holes, a finding we hope will stimulate further theoretical treatments.

[1] T. C. Damen, L. Viña, J. E. Cunningham, and J. Shah, Phys. Rev. Lett. **67**, 3432 (1991).

[2] J. B. Stark, W. H. Knox, and D. S. Chemla, Phys. Rev. B **46**, 7919 (1992).

[3] B. Dareys, X. Marie, T. Amand, J. Barrau, Y. Shekun, I. Razdobreev, and R. Planel, Superlattices Microstruct. **13**, 353 (1993).

[4] T. Amand, X. Marie, B. Baylac, B. Dareys, J. Barrau, M. Brosseau, R. Planel, and D. J. Dunstan, Phys. Lett. A **193**, 105 (1994).

[5] L. Viña, L. Muñoz, E. Pérez, J. Fernández-Rossier, C. Tejedor, and K. Ploog, Phys. Rev. B **54**, 8317 (1996).

[6] J. Fernández-Rossier, C. Tejedor, L. Muñoz, and L. Viña, Phys. Rev. B **54**, 11 582 (1996).

[7] J. Fernández-Rossier and C. Tejedor, Phys. Rev. Lett. **78**, 4809 (1997).

[8] P. LeJeune, X. Marie, T. Amand, F. Romstad, F. Perez, J. Barrau, and M. Brousseau, Phys. Rev. B **58**, 4853 (1998).

[9] Throughout this work only excitons with spin ± 1, which in first order are the only ones which couple to light, will be considered.

[10] Y. J. Chen, E. S. Kotelcs, B. S. Elman, and C. A. Armiento, Phys. Rev. B **36**, 4562 (1987).

[11] E. E. Mendez, F. Agulló-Rueda, and J. M. Hong, Phys. Rev. Lett. **60**, 2426 (1988).

[12] P. Voisin, J. Blouse, C. Bouchet, S. Gaillard, C. Alibert, and A. Regreny, Phys. Rev. Lett. **61**, 1639 (1988).

[13] L. Viña, E. E. Mendez, W. I. Wang, L. L. Chang, and L. Esaki, J. Phys. C **20**, 2803 (1987).

[14] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (Les Éditions de Physique, Les Ulis Cedex, 1988).

[15] K. Köhler, H.-J. Polland, L. Schultheis, and C. W. Tu, Phys. Rev. B **38**, 5496 (1988).

[16] A. Alexandrou, J. A. Kash, E. E. Mendez, M. Zachau, J. M. Hong, T. Fukuzawa, and Y. Hase, Phys. Rev. B **42**, 9225 (1990).

[17] This contribution to the emission was taken into account to fit the excitonic PL peaks at short times ($t_d \leq 15$ ps). It resulted from the simultaneous excitation of the X_{e2hh1}^{+1} and continuum states in other bands.