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Spin-Dependent Exciton–Exciton Interaction in Quantum Wells under an Electric Field

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We give conclusive experimental evidence that spin-dependent exciton–exciton interaction processes can be altered by directly modifying the spatial exciton structure applying an electric field. The previously observed energy splitting in spin-polarized, dense exciton gases can be varied by the electric field from a maximum of 4 meV to 0 meV. On the basis of an earlier developed theoretical model it is seen that inter-exciton scattering processes dominating at zero field are quenched and others are enhanced on increasing the field strength.

1. Introduction

In time resolved photoluminescence (TRPL) experiments on two-dimensional (2D) semiconductor systems where a spin-polarized exciton gas was excited, a lifting of the spin-degeneracy of the one-exciton energies has been observed [1 to 3]. In these experiments the system is excited at (or near) the heavy hole (hh) energy with circularly polarized light pulses at an intensity I_{ex} creating excitons with angular momentum $J = +1(-1)$, for $\sigma^+(\sigma^-)$ polarized light, at a density $n_{\mathrm{X}} = n_+(n_-)$. If a +1 exciton gas is excited initially, spin-flip processes will depopulate the +1 component creating a −1 component[2]) which after a few tens of picoseconds still has a density n_- much lower than that of the +1 population, i.e. $n_- \ll n_+$ and $n_{\mathrm{X}} = n_+ + n_-$. The obviously time-dependent polarization of the system is given by $P = (n_+ - n_-)/(n_+ + n_-)$. Recombination of the polarized excitons leads to a polarized photoluminescence (PL) signal and thus $P = (I_+ - I_-)/(I_+ + I_-)$, where $I_\pm$ are the PL intensities for the two polarizations σ^+ and σ^-. The experiments showed an energy splitting, $\delta\epsilon = E_+ - E_-$, between the two spin-polarized exciton components of several meV which decreased with decreasing I_{ex} and P. $E_\pm$ is the PL peak position of each component.

A theoretical model which takes into account the spin-dependent inter-exciton interactions has quantified this splitting [4, 5]. Using the spin-exciton wave function to calculate the above-mentioned exciton–exciton scattering processes and assuming a spin-polarized exciton gas (i.e. $P > 0$) the model yields the dependence of the splitting on P

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[2]) It is neglected here that spin-flip processes can also populate the optically inactive ± states [10, 11].

and n_X (in the approximation that the ±2 exciton population is zero and interaction between +1 and −1 excitons is neglected):

$$\delta\epsilon \propto n_X P(I_{VC} - I_{EC})\,. \qquad (1)$$

Here I_{VC} is a vertex correction (VC) term and I_{EC} an inter-excitonic exchange (EC) term. These two corrections to the excitonic binding energy E_b, have opposite sign since the VC can be understood as an effective repulsive and the EC as an attractive interaction. From Eq. (1) we see that the relative magnitudes of I_{VC} and I_{EC} determine the sign of the splitting $\delta\epsilon$. So far only positive splittings have been reported from experiments. Thus, the majority +1 excitons always have higher energies than the minority −1 excitons and the interaction is dominated by the VC.

Fernández-Rossier et al. expanded their model from Ref. [4] to a system where the electrons and holes forming an exciton can be separated locally [5]. It is assumed that

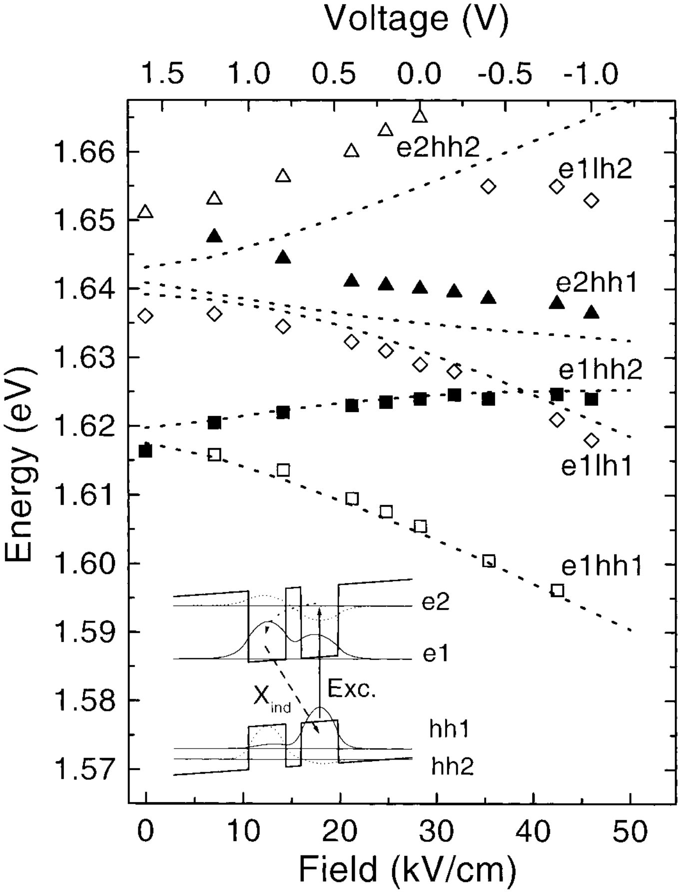

Fig. 1. Excitonic energy levels as a function of electric field. The data were obtained from PLE measurements at $T = 8$ K. The inset shows the schematic structure and the levels of the cDQW. The solid, dotted and dashed arrows indicate the excitation, relaxation and recombination processes, respectively. The solid (dotted) line is the symmetric (antisymmetric) wave function calculated for $\mathcal{E} = 14$ kV/cm. The light hole levels are not shown for simplicity

the electrons and holes are confined to separate planes, parallel to the quantum well (QW) plane, at a distance d. With increasing d, the overlap between holes and electrons decreases, which affects the inter-excitonic scattering mechanisms. The two parameters responsible for the splitting between the spin components I_{VC} and I_{EC} turn out to be functions of d; VC tends to decrease and EC to slightly increase with increasing d. Looking at Eq. (1) we see that this leads to a reduction of $\delta\epsilon$ (at unchanged n_{X} and P) and, eventually, it becomes negative at a certain d. A $\delta\epsilon < 0$ means that the minority excitons have higher energies than the majority ones, thus promoting a repolarization of the exciton gas or at least a stabilization of the polarization as the system can win energy going to the majority states. The exciton gas would be in a ferromagnetic phase. It is argued that a ferromagnetic phase could make the Bose-Einstein condensation of the exciton gas experimentally more accessible [6]. In practice, the local separation of electrons and holes in 2D systems can be achieved by the use of a type-II QW or by applying an electric field to a type-I QW, where the parameter d is proportional to the field $\mathcal{E}$.

In this paper, we present TRPL studies on the energy splitting between the two spin components of an exciton gas in a coupled double quantum well structure (cDQW) under an electric field perpendicular to the QW plane. The investigated sample consists of 10 periods of two 50 Å wide GaAs wells separated by a 20 Å $\mathrm{Al}_{0.3}\mathrm{Ga}_{0.7}\mathrm{As}$ barrier. The wells are embedded in a p–i–n structure. A schematic representation of the potential profiles of the cDQW structure at a field of 14 kV/cm is shown in the inset of Fig. 1. The electric field dependence of the different excitonic transitions, obtained from cw-photoluminescence excitation (PLE) measurements (spectra not shown here), is depicted in Fig. 1. For a detailed description of the characteristics of the different transitions, see Ref. [7].

2. Experimental

Time resolved PL measurements were performed with a standard up-conversion set-up 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 exciting laser beam and the PL collecting optics. The sample was mounted on a cold-finger cryostat and held at $T = 8$ K for all measurements presented here. The excitation energy was always set to the e1hh2 exciton (see Fig. 1) and the excitation intensity was $I_{\mathrm{ex}} = 15$ mW which corresponds approximately to a density of $n_{\mathrm{X}} \approx 5 \times 10^{10}$ cm^{-2}.

3. Results

At flat band and low fields we clearly see a splitting between +1 and −1 components as is illustrated in Fig. 2a and d.^{3}) This is similar to what has been observed in the works discussed above. Increasing the field while maintaining the same n_{X} (i.e. I_{ex}), the splitting decreases approximately linearly (Fig. 2b and d) and vanishes at an electric field $\mathcal{E} \approx 35$ kV/cm. The PL spectra of the two spin components overlap (Fig. 2c). Note that

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^{3}) The spectra in Fig. 2a, b and c were taken at $t = 32$ ps after the excitation. At very short times ($t < 15$ ps) the high energy side of the spectra is broadened by contributions of the EHP since excitation is not resonant with the e1hh1 exciton. Between $t = 15$ and 30 ps the EHP mostly relaxes to bound exciton states.

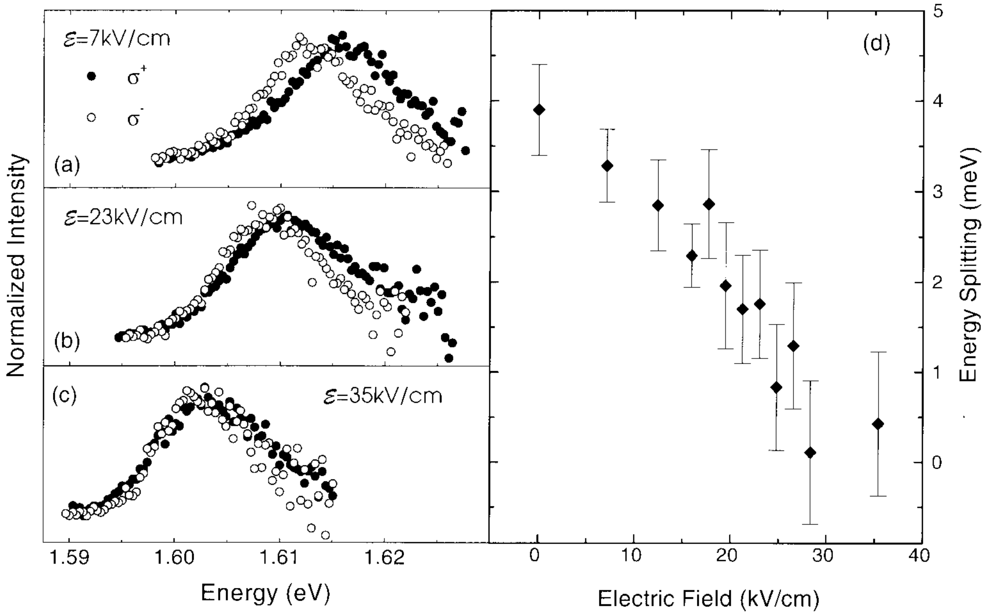

Fig. 2. a), b) and c) Normalized spectra for the two polarizations taken at $t = 32$ ps and $I_{ex} = 15$ mW for $\mathcal{E}$ = 7, 23 and 35 kV/cm, respectively. The excitation light was σ^+ polarized. d) Initial splitting as a function of electric field $\mathcal{E}$. All measurements were performed at T = 8 K

the σ^+ peak is only slightly broader due to $n_+ > n_-$. To assure that this behaviour is actually due to a change in exciton–exciton interactions by the local separation of the electrons and holes, the field dependence of the degree of polarization, which is the only additional parameter in Eq. (1), has to be checked. Fig. 3a demonstrates that the initial polarization, that is the average polarization at $t \leq 32$ ps, is approximately constant with $\mathcal{E}$ and $P = 0.5 \pm 0.05$. The decay of the polarization with time due to the spin-flip processes and the decay of the splitting, as both populations n_+ and n_- become similar, are strongly correlated. Both are practically independent of $\mathcal{E}$ as shown in Fig. 3b. The mean value of the decay times, as obtained from a fit to a monoexponential decay, amounts to $\bar{\tau} \approx 120$ ps. Note that the error in $\tau_{\delta\epsilon}$ becomes quite large at high fields due to the fact that the initial $\delta\epsilon$ becomes close to zero. Therefore, we have established that P and τ_P are field independent and that the proportionality between $\delta\epsilon$ and P (at constant n_X) predicted by Eq. (1) holds. So, according to the model, only I_{VC} and I_{EC} can be functions of $\mathcal{E}$.

The maximum field that can be applied to our structure, 35 kV/cm, yields a zero splitting between +1 and −1 excitons. Unfortunately, any further increase in $\mathcal{E}$ leads to a strong ionization of the excitons hindering the investigation of the possible existence of a negative splitting. The field ionization of the excitons gives rise to a strong increase of the tunneling current and reduces markedly the exciton lifetime. The PL decay time τ_d is shown in Fig. 3c as a function of the electric field strength for both polarizations of the emission. An initial linear increase of τ_d with field is followed by a sharp drop at $\mathcal{E} \approx 35$ kV/cm, which can be attributed to carrier tunneling out of the wells [8, 9].

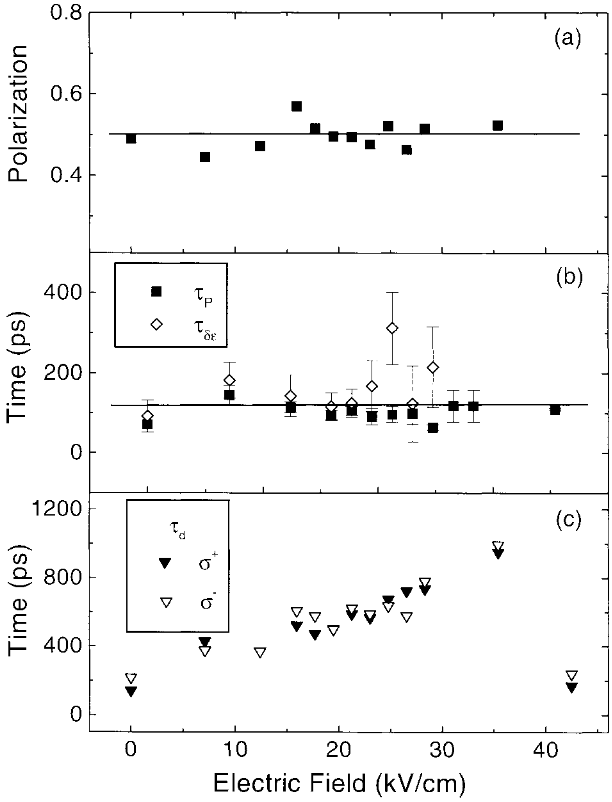

Fig. 3. a) Initial polarization of the exciton system as a function of field. The values are an average over $t = 7$, 15 and 32 ps. b) Decay times of the polarization (τ_P) and splitting ($\tau_{\delta\epsilon}$). These times were obtained from a single exponential fit of $P(t)$ and $\delta\epsilon(t)$. c) Decay times of the PL intensity (τ_d) for the two spin components as function of $\mathcal{E}$

4. Conclusions

We have shown that the spin-dependent splitting in an exciton gas is reduced by the changes in the exciton–exciton interaction effects, namely a reduction of the VC and an increase in EC, as predicted by the theory. The existence of a ferromagnetic phase, also predicted by theory, could not be demonstrated due to experimental limits in the maximum field strength. Resonant excitation of the e1hh1 exciton could help to overcome this difficulty.

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