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Spin dynamics and spin-dependent interactions in semiconductor heterostructures

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Abstract

We present new aspects on the spin dynamics of carriers and excitons in semiconductor heterostructures. In doped wells, we show that the two spin components of an optically created two-dimensional electron gas are well described by Fermi-Dirac distributions with a common temperature but different chemical potentials. In intrinsic wells, where excitonic effects dominate the optical properties, we show that exciton–exciton interaction produces a breaking of the spin degeneracy in two-dimensional semiconductors, which can be tuned by the application of an external electric field. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Spin; Semiconductors; Nanostructures

1. Introduction

Useful electronic devices rely on the precise control of electronic charge, and in general the fact that the electrons also have a spin is ignored. However, the scattering processes for electrons depend on their spin state. Recently, interest in electronic spin polarization in solid-state systems has grown fuelled by the possibility of producing efficient photoemitters with a high degree of polarization of the electron beam, creating spin memory devices and spin transistors as well as exploiting the properties of spin coherence for quantum computation. A new field, known as "spintronics", deals with the possibility of manipulation of electronic spin to read and write information through magnetism [1,2]. One of the main difficulties in the development of these devices is the lack of an efficient mechanism to inject spin-polarized charge [3–5]. The determination of the spin-flip rates is extremely important for electronic applications, because if the spins relax too rapidly, the distances traveled by spin-polarized currents will be too short for practical applications.

There are various experiments to detect optical orientation, such as spin polarization of electron photoemission, electron paramagnetic resonance, nuclear magnetic resonance, spin-dependent transport, Faraday rotation, and spin-dependent "pump and probe" or recombination. The Hanle effect [6], i.e., the spin depolarization of carriers subject to a transverse magnetic field, has been traditionally used to obtain quantitative values for the spin relaxation times [7]. Optical pumping, with linearly polarized light, and orientation, with circularly polarized light, of carrier spins is a powerful method to investigate relaxation processes in

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semiconductors and has also found applications in spin-polarized electron sources. With the appearance of ultrafast lasers, the spin relaxation time can be also measured in a direct way. The emitted luminescence, after an excitation with a light pulse, reflects the temporal evolution of the carrier distribution and can be analyzed in polarization to study energy as well as orientation relaxation rates.

Experimental investigations on the spin dynamics in low-dimensional semiconductors have flourished in the last decade. Many works deal with the spin processes of excitons [8–13], including the study of the influence of external electric fields [14]. Fewer investigations deal with the spin-flip of individual electrons and holes in 2D systems [9,10,15,16]. Extensive theoretical studies have been also done on the spin-flip relaxation of excitons [17–20], and free carriers [21–24].

In this work, we will concentrate on the study of the time evolution of the light emission, using time-resolved photoluminescence (TR-PL) spectroscopy, in different semiconductor heterostructures. We will pay special attention to phenomena related with the spin of the carriers and the excitons. The rest of the manuscript is organized as follows: Section 2 gives the experimental details. The dynamics of a spin polarized, optically pumped, electron gas is presented in Section 3. The interaction between excitons in a spin-polarized exciton gas is shown in Section 4. Finally, we summarize in Section 5.

2. Experiments

The experiments are performed in a temperature variable, cold finger cryostat exciting the samples with light pulses from a Ti:Sapphire mode-locked laser pumped by an Ar^+-ion laser. The pulse width is 1.2 ps. The incident light is directed along the growth axis of the heterostructures and a back-scattering geometry is used. The PL is time-resolved in a standard up-conversion spectrometer. The time resolution, obtained by overlapping on a non-linear crystal, $LiIO_3$, the luminescence from the sample with a delayed pulse from the laser, is basically determined by the temporal pulse width. A double grating monochromator is used to disperse the up-converted signal. The exciting light is circularly polarized by means of a $\lambda/4$ plate, and the PL is analyzed into its σ^+ and σ^- components using a second $\lambda/4$ plate before the non-linear crystal. Time delays at a given emission energy or TR-PL spectra at different delays after the excitation pulse are obtained using this system.

For the measurements of the electron spin-relaxation, we have studied p-type modulation doped GaAs/GaAlAs QW's with hole sheet concentrations of $\sim 3\times10^{11}\,cm^{-2}$, mobilities of $\sim 4000\,m^2\,V^{-1}s^{-1}$ and well widths from 30 to 80 Å. Here, we will show the results obtained in a 30 Å-thick QW modulation-doped with Beryllium. We have measured PL spectra at fixed times, as well as the PL decay at fixed emission energies, both as a function of temperature ($T = 10$–50 K) and as a function of the laser power, giving concentrations of excess carriers in the range between $\sim 10^{10}$ and $\sim 10^{11}\,cm^{-2}$ [25].

For the study of the dependence of the exciton interaction on initial carrier concentration we concentrate on the results of a GaAs/AlAs multiquantum well (MQW), consisting of 50 periods of nominally 77 Å-wide GaAs wells and 72 Å-wide AlAs barriers. The experiments were performed under quasi-resonant excitation at the free heavy-hole exciton detecting at the weakly-bound exciton seen in PL. The influence of the exciton localization has been reported in the literature [12]. The electric field dependence of the splitting has been investigated using a coupled double quantum well (cDQW). This sample consisted of 10 periods of two 50 Å wide GaAs wells separated by a 20 Å $Al_{0.3}Ga_{0.7}As$ barrier. Each cDQW period is separated by a 200 Å $Al_{0.3}Ga_{0.7}As$ layer. More details about the sample can be found in Ref. [26].

3. Spin polarization of an optically pumped electron gas

Exciting a p-type GaAs quantum well below the light-hole resonance, electrons with almost purely one spin component are photocreated. Filling of the conduction band is clearly different for both electron-spin components, leading to an appreciable shift between σ^+ and σ^- emission spectra.

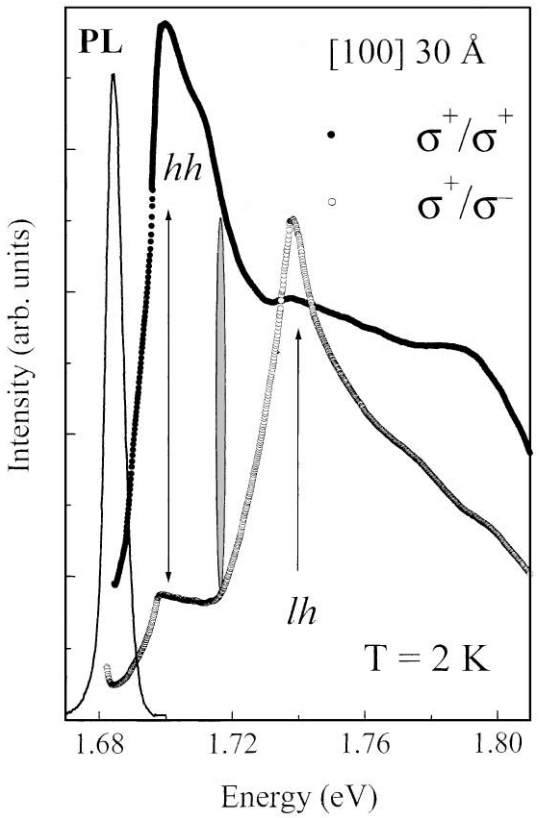

Fig. 1. PL (solid line) and excitation spectra of a 30 Å thick p-type modulation doped ($p = 3 \times 10^{11}\,\mathrm{cm}^{-2}$) QW measured for aligned (solid points) and crossed (open symbols) polarization of the exciting/emitted light. The shaded area indicates the energy used for excitation on the time-resolved experiments.

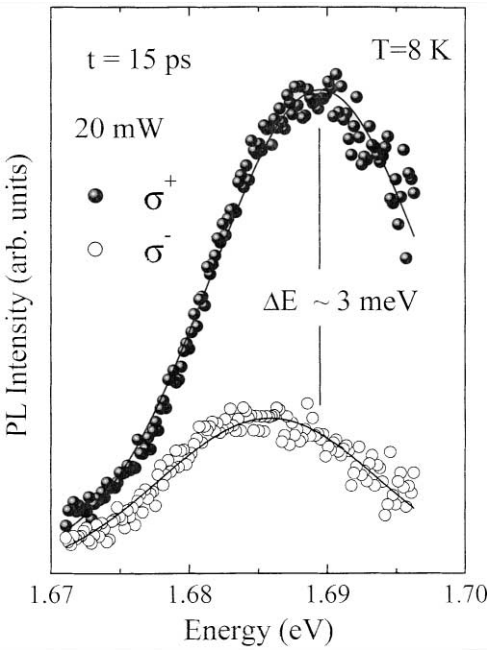

Fig. 2. Co-polarized (solid points) and counter-polarized (open points) components of the PL spectra, measured at 15 ps after a σ^+-polarized pulsed excitation with a mean laser power of 20 mW and excitation energy of 1.717 eV. Bath temperature 8 K.

Fig. 1 depicts the PLE spectra of a 30 Å, [1 0 0]-oriented QW at 2 K, exciting with σ^+-polarized light, recorded at the tail of the PL. The onset of the absorption due to the hh transition is clearly seen as a peak in the $\sigma^+\sigma^+$ spectrum at 1.7 eV, while the one corresponding to the lh transition is dominant in the $\sigma^+\sigma^-$ spectrum at 1.738 eV. The degree of polarization of the emission, which for a given helicity of the exciting light, i.e., σ^+ is defined as the fractional difference of the PL intensities of the two circular polarizations, σ^+ and σ^-, $\wp = (I^+ - I^-)/(I^+ + I^-)$, gives information about the spin relaxation. As can be deduced from this figure, the sample shows a high degree of optical alignment, indicating a long spin-flip relaxation time for the photoexcited electrons. For excitation energies above the threshold of lh states, the polarization decreases rapidly, since electrons with opposite spin direction are produced from the light-hole subband. Large values of $\wp$ have been already reported in p-doped strained films [27], and superlattices [28].

Using p-type samples, we can easily investigate spin alignment effects in the conduction band: electrons with an unbalanced population of the two spin components, created under circularly polarized excitation, recombine with non polarized holes which mostly originate from doping. The spin relaxation of the electrons can be obtained from the difference of the time evolution of the two orthogonally polarized emissions. Fig. 2 represents the σ^+ and σ^- components of the luminescence spectra excited with σ^+ pulses at 1.717 eV. These spectra clearly show the difference in the occupation of electronic states with opposite spins. This difference, which vanishes at longer delay times, leads to

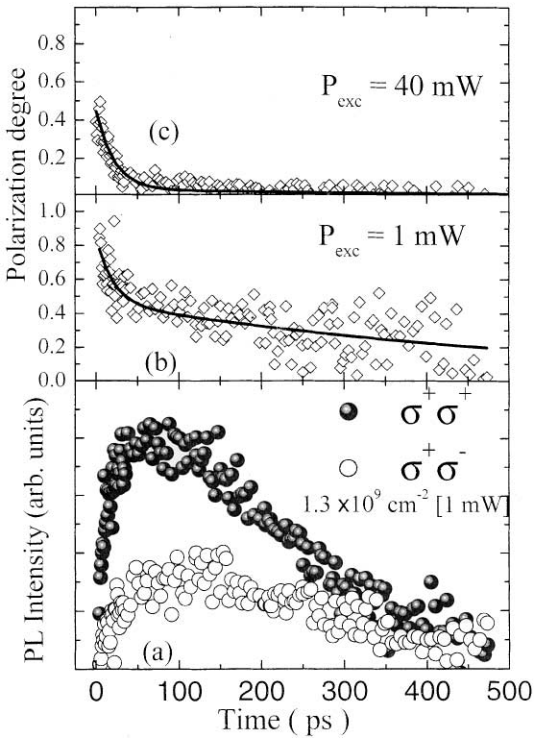

Fig. 3. (a) Decay of σ^+ (solid points) and σ^- (open points) PL under pulsed σ^+ excitation with a mean laser power of 1 mW. (b) Time evolution of the polarization degree for 1 mW. (c) Same as (b) for a laser power of 40 mW. The lines represent the best fits according to the sum of two exponential decays: $A \times \exp(-t/\tau_1) + B \times \exp(-t/\tau_2)$.

the energy difference between the positions of the maxima in the σ^+ and σ^- luminescence spectra.

The most striking finding is illustrated in Fig. 3: (a) shows the evolution of the two components of the polarized PL, and (b) and (c) depict the time evolution of the polarization degree, $\wp$, for two different powers of the exciting light. The decay of spin polarization of the electron gas depends very much on the intensity of the laser excitation. The decay of $\wp$ measured at $T = 8$ K can be well reproduced by a sum of two exponential decays with two distinct characteristic times of $\tau_1 = 20$ ps and $\tau_2 = 550$ ps. The amplitude of the fast component vanishes at low excitation power, whereas this fast process almost completely determines the electron spin depolarization at the highest level of laser excitation. It is well known that for any mechanism of electron spin relaxation [29], the probability of spin flip transitions increases as a function of the electron k-vector. Therefore, the increase in the rate of spin relaxation as a function of the laser power may result partially from a larger electron kinetic energy caused either by an increase of the effective electron temperature or of the initial electron concentration.

However, this simple reasoning hardly explains our data. From an analysis of the time evolution of the luminescence spectra, we have estimated an increase in the mean kinetic energy of the electron gas of only 4 meV at the highest laser power [25]. This amount is not sufficient to reduce the spin relaxation time down to 20 ps, since at low excitation powers but high lattice temperatures ($kT \sim 3.44$ meV) we still observe a relatively long spin relaxation time (~ 80 ps).

We have simulated measured spectra at different times after the excitation by the broadened convolution of Fermi-Dirac statistics for non-polarized gas of holes and two spin components of the 2DEG, assuming the conservation of k-selection rules.

An analysis of pairs of σ^+ and σ^- PL spectra leads us first to conclude that each component is well described assuming a common temperature for the two electron spin components (and for holes), but different values of the chemical potential. Excluding very short delay times after excitation, i.e., already after a few picoseconds, each spin component of the electron gas can be qualitatively characterized by its own Fermi distribution, each one with different chemical potential but both with very similar temperatures. Under our experimental conditions, the exchange interaction between electrons is unable to stabilize a common chemical potential of the electron gas for the two spin components. This latter situation might be expected under equilibrium conditions and would imply a difference in the renormalization of the conduction band edges for the two spin components.

The obtained time evolution of carrier temperature, is shown in Fig. 4 (diamonds). T rises up to ~ 100 K just after the laser pulse. This fact, in conjunction with other measurements as a function of lattice temperature [30], accounts for the fast

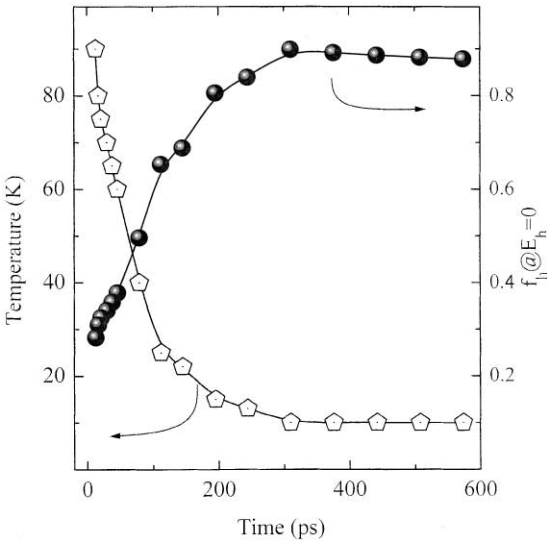

Fig. 4. Time evolution of the carrier temperature (diamonds) for a mean laser power of 10 mW The solid symbols depict the time evolution of the number of occupied hole states at the top of the valence band.

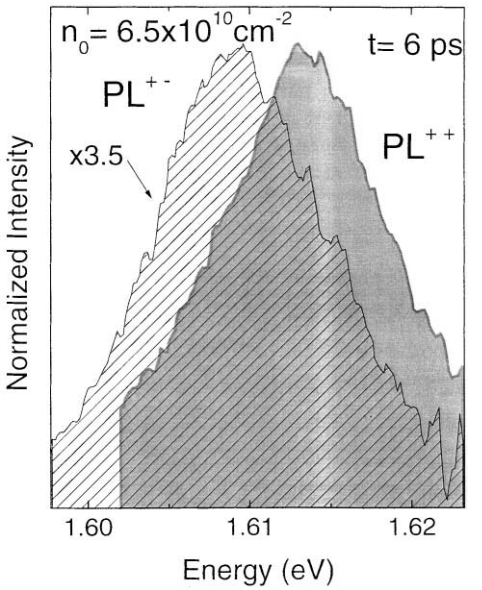

Fig. 5. Low T, 8 K, time-resolved PL spectra of the GaAs MQW taken 6 ps after excitation with σ^+-light at 1.625 eV. The gray (dashed) area depict the σ^+ (σ^-) emission. The initial carrier density is $6.5\times10^{10}\,\mathrm{cm}^{-2}$. The σ^- emission has been enlarged by a factor of 3.5.

depolarization of electronic spins, induced by the laser power. Our results confirm the high efficiency of carrier–carrier interaction in establishing a common temperature for electrons and holes. Cold before excitation, the gas of holes becomes nondegenerate almost immediately after the laser pulse. This nondegenerate character of the hole gas is illustrated in Fig. 4 (circles), where the number of occupied hole states at the top of the valence band is plotted as a function of time.

We can also conclude that fast spin depolarization in our structures is driven by the nondegenerate character of the carrier distribution and not exclusively by the increase of the electron kinetic energy. The nondegenerate carrier distributions favor the efficiency of spin-flip electron scattering via the exchange interaction with holes, in contrast, the available number of scattering configurations is appreciable reduced for the degenerate systems.

4. Spin splitting in a polarized exciton gas

Fig. 5 depicts TR-PL spectra of the GaAs MQW taken at 8 K, 6 ps after excitation with σ^+-pulses for a density of $6.5\times10^{10}\,\mathrm{cm}^{-2}$, exciting at 1.625 eV. The gray area corresponds to the polarized (σ^+, spin $+1$) emission while the dashed one shows the unpolarized (σ^-, spin -1) PL. A clear energy splitting of ~4.5 meV is seen between the two peaks. Increasing the excitation density, both a broadening of the lines and a strong enhancement of the splitting is obtained.

Fig. 6 depicts the dependence of the energy positions of the PL on the initial carrier density (open and solid points). Under the conditions presented in this figure, 12 ps after excitation at 1.631 eV, the σ^+ emission remains practically constant, while the σ^- red shifts with increasing carrier density up to $9\times10^{10}\,\mathrm{cm}^{-2}$. The lines correspond to a model, which takes into account interexcitonic exchange interaction and screening [31], that gives the *changes* in the energies of the interacting ±1 excitons as a function of the total and the ±1 populations of excitons as:

$$\Delta E^{\pm} = 2(n^{\pm} + fn^{\mp})(I_{\mathrm{VC}} - I_{\mathrm{SE}}) - \frac{0.82\pi e^2 na}{\varepsilon}, \qquad (1)$$

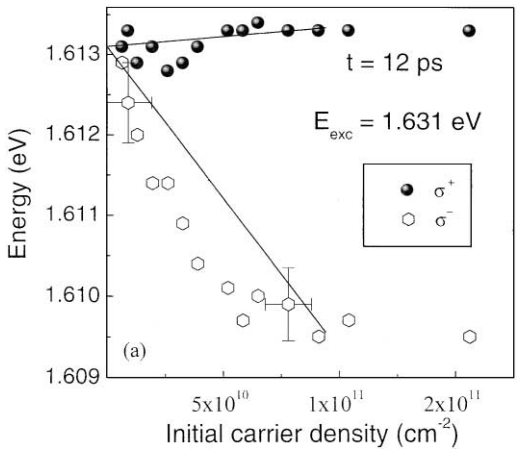

Fig. 6. Carrier density dependence of the emission energies of the GaAs MQW at 12 ps after excitation for both circular polarizations.

where n is the total density of excitons, $n^{\pm}$ are those of ± 1 excitons, ε the dielectric constant and a the three-dimensional Bohr radius. I_{SE} describes a "self energy" (SE) correction, also known in the literature as "exchange" correction, that weakens electron–electron and hole–hole repulsion. I_{VC} is a "vertex" correction (VC) that, due to the Pauli exclusion, reduces the inter-excitonic electron–hole attraction. The term involving f is a small coupling between ± 1 excitons, essentially due to valence band mixing. The last term is a screening correction using the random phase approximation. In spite of the strong approximations used in the theory, such as neglecting the presence of dark, ± 2, states and assuming that the excitons are all at $\boldsymbol{K} = 0$, which are not borne out by the experiments, the agreement with the experiments is satisfactory.

The energy splitting, $\delta = \Delta E^{+} - \Delta E^{-}$, neglecting the second term of Eq. (1), is given by:

$$\delta \propto n\wp(I_{VC} - I_{SE}). \tag{2}$$

The experiments corroborate the theoretical predictions concerning the relative strength of the electron–hole vertex (I_{VC}) and of the electron(hole)–electron(hole) (I_{SE}) "self-energy" corrections: the splitting grows with increasing initial carrier density.

Fernández Rossier et al. expanded their model from Ref. [31] to a system where the electrons and holes forming an exciton can be separated locally [32]. In their model, it is assumed that the electrons and holes are confined to separate planes at a distance d. The overlap between holes and electrons is tuned by changing d, therefore modifying the inter-excitonic scattering mechanisms: I_{VC} (I_{SE}) tends to decrease (slightly increase) with increasing d. According to Eq. (2), this should lead to a reduction of δ (at unchanged n and $\wp$) and, eventually, it could become negative at a certain $d = d_{cr}$. A $\delta < 0$ 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. Hence, by tuning d and n, one could obtain a quantum phase transition between a ferromagnetic phase (dominated by self-energy) and a paramagnetic phase (dominated by the vertex corrections). 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 $\mathscr{E}$.

Fig. 7 depicts the TR-PL spectra taken at 32 ps after excitation with 15 mW for different field strengths. At flat band (Fig. 7a) and low fields (Fig. 7b) we observe a splitting between $+1$ and -1 excitons. The splitting at zero field amounts to approximately half of the binding energy of the exciton. Increasing the field while maintaining the same initial exciton density, n, the splitting decreases and finally it vanishes.

Fig. 8 compiles the dependence of the initial splitting δ_0 on $\mathscr{E}$ and demonstrates that it decreases linearly with increasing $\mathscr{E}$. We have defined δ_0, for a given field, as the mean value of three measurements taken at delays $t_d = 7$, 15 and 32 ps. The maximum splitting of 4 meV is reached at zero field. At a field of $\mathscr{E} \sim 35\,\mathrm{kV\,cm^{-1}}$ the splitting becomes zero, this is in agreement with the predictions of the model of Ref. [32]. To ensure that the decrease of the splitting is actually due to a change in exciton–exciton interactions by the local separation of the electrons and holes, the field dependence of $\wp$, which is the only additional parameter in Eq. (2),

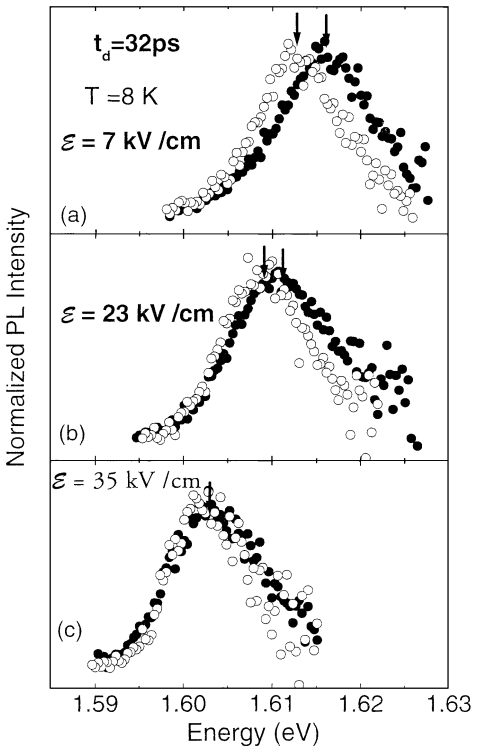

Fig. 7. TR-PL spectra for the cDQW at different field strengths, taken 32 ps after excitation. The arrows mark the positions of the PL maxima.

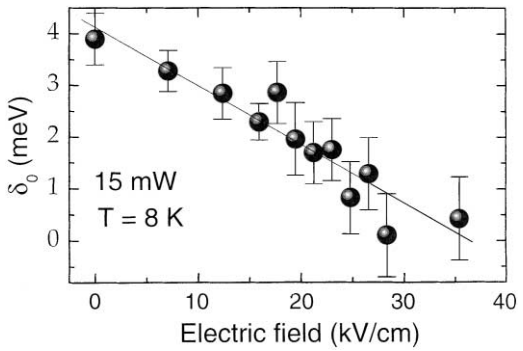

Fig. 8. Field dependence of the splitting in the cDQW at an excitation power of 15 mW, the magnitud of δ is a meanvalue of those at 7, 15 and 32 ps.

has to be checked. Exciting at the direct exciton transition, an initial ($t \sim 0$), field independent, polarization of the indirect, ground-state exciton, $\wp_0 = 0.5 \pm 0.05$, is obtained [26]. Since the spin splitting, δ, is proportional to the total exciton population n and to the polarization, and $\wp$ does not change significantly with $\mathscr{E}$, any change in δ, at a given excitation power, can be unambiguously related to a variation in the exciton–exciton interaction (I_{VC} and/or I_{SE}).

5. Summary

We have shown that for a 2DEG, which is created polarized by means of optical orientation, strong effects, nonlinear in the excitation power, are observed in the polarization of the emission, which originate from the nondegenerate character of the carrier distribution at short times after the laser excitation. The 2DEG can be well described by two separate Fermi-Dirac distribution functions, one for each spin component, with common temperature but different chemical potentials.

We have also described the existence of a splitting between the two circularly polarized components of a dense 2D exciton gas in the absence of any external magnetic field. Using an electric field and a cDQW heterostructures, to spatially separate the electrons and holes forming the excitons in different wells, we demonstrate that the splitting decreases with increasing separation, due to the reduction of the vertex correction to the inter-excitonic interaction with increasing electron–hole separation.

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