Shallow Quantum Well Excitons: 2D or 3D?

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(Received 17 September 1992)

A new regime is obtained in semiconductor heterostructures with constituents of nearly identical band gaps. Previously, it has been shown that even extremely shallow quantum wells (SHQWs) exhibit excitonic and electroabsorption properties typical of a 2D system, but 3D transport features. We show that, surprisingly, even when carriers are two-dimensionally confined in SHQWs, the hole spin relaxation is extremely fast (~400 fsec) as in the bulk (3D) limit and that a 2D-3D transition in the hole spin dynamics in GaAs/$Al_xGa_{1-x}As$ SHQWs takes place at $x \sim 5\%$.

PACS numbers: 71.35.+z, 42.65.Re, 73.20.Dx

The fabrication of deep semiconductor quantum wells (QWs) in which carriers are confined to one or more thin layers has allowed the study of new and interesting properties of quasi-two-dimensional carriers. When the QWs are made extremely shallow, the well depth becomes comparable to the quantum confinement energy, and a new intermediate dimensionality regime occurs. Excitonic absorption and electroabsorption measurements in the GaAs/$Al_xGa_{1-x}As$ system [1] have shown that excitons and carriers maintain a two-dimensional (2D) nature even down to $x \sim 0.82\%$. It is expected that for such very shallow wells, the band structure will be almost bulklike, and it has been recently shown that transport in shallow quantum wells (SHQWs) is essentially three dimensional (3D) [2]. Even small band structure changes should have a significant influence on carrier spin dynamics. In the 2D limit, considerable changes in carrier spin relaxation have been reported [3–5] as compared to bulk materials [6]. The purpose of this Letter is to investigate the carrier and exciton spin dynamics in this interesting intermediate dimensionality regime. The most dramatic changes are expected in the hole spin relaxation as it is strongly dependent on the valence-band structure [7,8]. We present the first direct evidence of a 2D-3D transition in the hole spin relaxation rate in QWs taking place at $x \sim 5\%$. Thus, in the regime of SHQWs, two-dimensional carrier confinement is clearly attained but hole spin relaxation is still extremely fast (~400 fsec), as in bulk GaAs.

The SHQW samples studied consist of fifty GaAs wells 10 nm wide, separated by 10 nm barriers of $Al_xGa_{1-x}As$ with $0 < x \leq 6\%$, grown by molecular beam epitaxy. We also use "deep" QWs with $x = 30\%$ and various well widths as control samples in the 2D limit. All the samples had the substrates etched and were double side antireflection coated. Our experimental setup consists of a mode-locked Ti-sapphire laser which generates tunable 60 fsec pulses which are amplified and converted to white light at 8 kHz repetition rate [9]. The probe beam (< 1% of the white continuum beam) is passed through the sample and detected with a spectrometer-optical multichannel analyzer.

Nonlinear circular dichroism (NCD) measurements [10] are done by chopping the pump beam on and off, while synchronously changing the helicity of the pump polarization. Thus for every time delay, four differential spectra are collected corresponding to the different combinations of pump on or off and right or left pump circular polarization. We define NCD as the difference in the nonlinear spectrum $-(\Delta\alpha l)$ measured for copolarized and for counterpolarized pump and probe beams. In order to understand the information obtained from the time evolution of the NCD we analyze the different contributions to the nonlinear spectra for the different combinations of pump and probe polarizations [10]. The pump-induced changes in the absorption are caused by the interaction between the photocreated pump and probe excitons. Excitons and carriers with low kinetic energy photogenerated by circularly polarized light have a well-defined angular momentum and their interactions can be classified into spin-dependent and spin-independent interactions. Phase-space filling (PSF) and molecular repulsion or attraction are the dominant spin-dependent exciton-exciton and exciton-carrier interactions and they manifest themselves differently in copolarized or counterpolarized nonlinear spectra. On the other hand, Coulomb interactions like dielectric screening are naturally spin independent. The latter will be present when pump and probe excitons are generated with parallel or opposite angular momentum. Therefore, by subtracting the nonlinear spectra measured with counterpolarized pump and probe from the copolarized spectra we leave only the contributions originating from spin-dependent interactions. The temporal evolution of the NCD spectra contains information on the spin dynamics of the photocreated excitons and, therefore, of the electrons and holes bound as an exciton.

Figure 1 shows the nonlinear absorption spectra for counter- and copolarized beams and for two samples representative of the two different regimes achieved in this study: (I) a GaAs/$Al_{0.02}Ga_{0.98}As$ SHQW: Here the photocreated excitons exhibit a quasi-2D nature, but the hh-lh splitting is extremely small (~2 meV); (II) a deep 75 Å GaAs/$Al_{0.3}Ga_{0.7}As$ QW. All the spectra were mea-

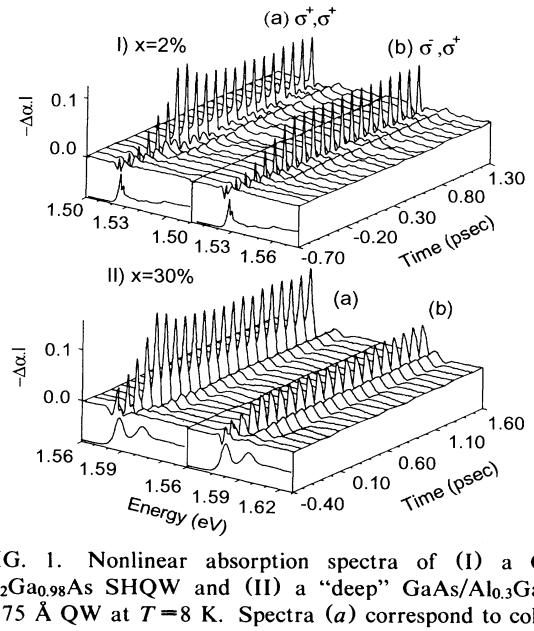

FIG. 1. Nonlinear absorption spectra of (I) a GaAs/$Al_{0.02}Ga_{0.98}As$ SHQW and (II) a "deep" GaAs/$Al_{0.3}Ga_{0.7}As$, $L_z = 75$ Å QW at $T = 8$ K. Spectra (a) correspond to collinear circular pump and probe polarizations and (b) to counterpolarized beams. The traces at the base projection are the linear absorption spectra of each sample.

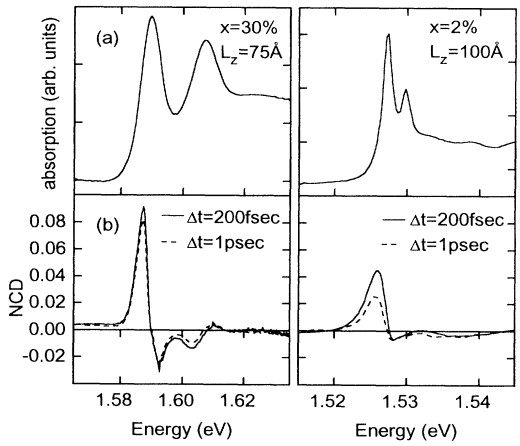

FIG. 2. (a) Linear absorption spectra; (b) nonlinear circular dichroism at $\Delta t = 200$ fsec and $\Delta t = 1$ psec, for the samples shown in Fig. 1.

sured at 8 K and with a carrier density of $\sim(2\text{-}4)\times10^{10}$ cm^{-2} as estimated independently from pulse energy and spot size, and from the magnitude of the density-dependent exciton bleaching. The pump spectrum was selected with interference filters (bandwidth 12–15 meV) and was kept resonant with the hh exciton minimizing free-carrier generation by careful spectral control [11]. The nonlinear response for copolarized pump and probe beams [Figs. 1 I(a) and 1 II(a)] arises mainly from three different mechanisms: (i) a bleaching contribution caused by PSF, (ii) a blueshift originating from the molecular repulsion between excitons of parallel angular momentum, and (iii) dielectric screening. The small rise in the nonlinear absorption around $\Delta t = 0$ is due to the coherent coupling between the pump and probe fields and it is only present during the temporal overlap of both optical fields. The spin-dependent contributions to the nonlinear spectra are greatly reduced in the case of the counterpolarized pump and probe beams [Figs. 1 I(b) and 1 II(b)] and the main contribution comes from dielectric screening. The main differences in the nonlinear spectra between the shallow and the deep QWs are visible in the first picosecond of the delay time. After a few hundred femtoseconds both copolarized and counterpolarized nonlinear spectra are almost identical in the SHQW, whereas clearly this is not the case for the deep well sample. This is reflected in the magnitude and the time evolution of the NCD, which is shown in Fig. 2 for $\Delta t = 200$ fsec and $\Delta t = 1$ psec (after the first 200 fsec there are no effects of the coherent coupling between pump and probe fields).

In this figure it is clear that (a) the magnitude of the NCD for a given delay time is stronger for the deeper wells, and (b) the NCD signal for the deep wells shows a very slow decay behavior. For the SHQW sample, however, there is a considerable decay of the NCD on a subpicosecond time scale. From a detailed line shape analysis of the spectra in Fig. 1, we determine that in the case of the SHQW, the blueshift component in the copolarized nonlinear spectra is the first one to decay, followed then by the bleaching part which is mostly due to PSF.

Figure 3 shows the time decay of the NCD *spectrally integrated* across the hh exciton bands for all the samples studied. Clearly, all the samples show a biexponential decay behavior with the solid lines representing the experimental fit. For SHQWs, the most dramatic changes

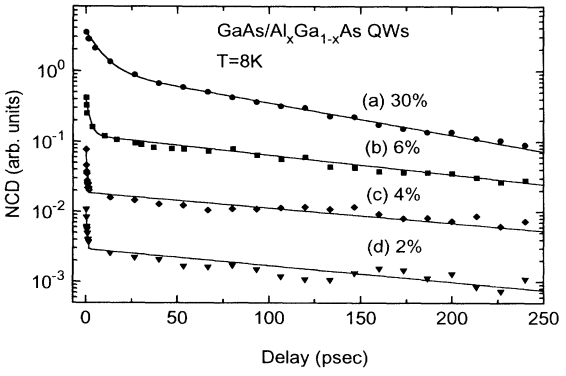

FIG. 3. Spectrally integrated NCD across the hh band excluding the first 200 fsec. (a) Deep $x = 30\%$, $L_z = 75$ Å QW; (b)–(d) SHQWs with $L_z = 100$ Å and $x = 6\%$, 4%, and 2%, respectively. The solid lines are biexponential fits to the experimental data.

occur on a subpicosecond time scale, and basically for $x < 6\%$, most of the spin memory of the exciton system is lost in the first picosecond. Figure 4(b) shows the short time constant (τ_s) for the different samples studied and as a function of the hh-lh energy splitting $\Delta E_{\text{hh-lh}}$. A bulk GaAs sample was also included in the measurements. The long time constant changes from 200 ± 30 psec in the $x = 2\%$ SHQW to 94 ± 5 psec in the $x = 30\%$, $L_z = 75$ Å sample. This long time constant has been previously assigned to the electron spin relaxation [5,12,13].

We now turn to the analysis of the short decay time constant τ_s. The exciton spin relaxation reflects the spin dynamics of its constituents. If the *e-h* exchange interaction J is strong enough, then the spin relaxation of the exciton will reflect that of the most rapidly relaxing particle, which for bulk GaAs is the hole. Theory [14] and experiment [15] have shown that the strong exchange regime, defined as $J/\hbar \gg (\tau)^{-1}$, with τ the shortest of the spin relaxation times, occurs only for strong 2D confinement. As the well width is increased, or conversely, as the barrier height is decreased, both J decreases and τ for the holes becomes shorter, thus entering the weak or intermediate exchange regime. When this is the case, the exciton spin relaxation as measured by femtosecond NCD should reflect the spin dynamics of *both* particles. This supports the existence of more than one time constant in the time evolution of the NCD, in agreement with our experiments.

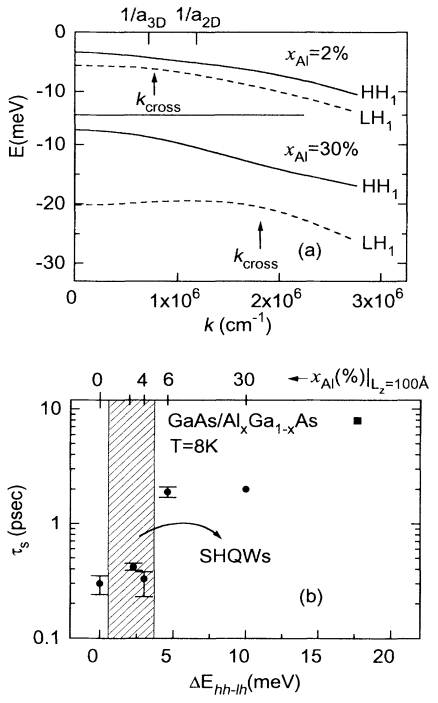

FIG. 4. (a) HH_1 and LH_1 bands calculated within the eight-band $\mathbf{k}\cdot\mathbf{p}$ formalism for a $x = 2\%$ SHQW and a deep $x = 30\%$ QW with $L_z = 10$ nm. k_{cross} is the anticrossing point. (b) Hole spin relaxation times τ_s as deduced from the biexponential decay (e.g., Fig. 3). The dashed area represents the regime of SHQWs in which carriers and excitons exhibit 2D optical properties but hole spin relaxation is 3D-like. ●, $L_z = 100$ Å; ■, $L_z = 75$ Å.

The short spin relaxation time τ_s is up to 2 orders of magnitude smaller than the measured electron spin relaxation times for bulk GaAs or $GaAs/Al_xGa_{1-x}As$ QWs [3,6]. Also, the trend in the electron spin relaxation time, as the dimensionality is reduced from 3D to 2D [3], is opposite to the one observed for τ_s and no abrupt changes are expected in this transition. We can therefore conclude that the short decay time cannot be caused by the electron spin relaxation. On the other hand, the picture is markedly different for the case of holes. In bulk semiconductors like GaAs, any scattering event of a hole (even for $\mathbf{k} \to 0$) will change its angular momentum (usually called "spin") due to the strong valence-band mixing for any $\mathbf{k} \neq 0$. As has been shown theoretically [7], the lifting of the hh-lh degeneracy at the Γ point by the quantum confinement makes the angular momentum of the holes (light or heavy) a good quantum number over a considerable range of $\mathbf{k}$ vectors, and there "spin flip" transitions will be greatly suppressed. Thus, very significant changes are expected in the case of the hole spin relaxation as the valence-band structure is changed from that of a bulk semiconductor to a QW, and, as we will discuss later, a power-law dependence of τ_s on $\Delta E_{\text{hh-lh}}$ is expected. Based on the previous arguments and the experimental observations, we assign τ_s to the hole spin relaxation time. This assumption is also supported by the observation that the fastest decaying component in the nonlinear spectra is due to the blueshift for copolarized pump and probe beams: This shift is a manifestation of the repulsion between two excitons with identical angular momentum. A spin relaxation of the hole implies a change in the exciton angular momentum which will immediately affect the molecular repulsion that pump and probe excitons were experiencing.

The dependence of the hole "spin-flip" time on the valence-band structure has been theoretically calculated [7]. For a free hole it can be expressed as

$$\tau_{s,k}^{-1} \propto \sum_{k'} |\langle \varphi_{\text{hh},k\uparrow} | V_{\text{scat}} | \varphi_{\text{hh},k'\downarrow} \rangle|^2 \delta[\varepsilon_{\text{hh},k\uparrow} - \varepsilon_{\text{hh},k'\downarrow}] ,$$

where $\varphi_{\text{hh},k\uparrow(\downarrow)}$ denotes the heavy-hole wave function for a specific $\mathbf{k}$ vector and "spin" up (down) (meaningful only at $k = 0$), and V_{scat} is the scattering potential. For any finite QW, there is always a $\mathbf{k}$ value for which the hh-lh mixing becomes significant (usually close to the hh-lh anticrossing, k_{cross}). This causes a strong deviation of the angular momentum of this eigenstate from $\pm \frac{3}{2}$, and a decrease in $\tau_{s,k}$. The anticrossing points for two 10 nm QWs with $x = 2$ and 30% are shown in Fig. 4(a) where we plot the HH_1 and LH_1 bands calculated within the eight-band $\mathbf{k}\cdot\mathbf{p}$ formalism. As in reality all the holes are bound into excitonic states, their wave vector will be

spread in $\mathbf{k}$ space up to $\sim 1/a_0$, with a_0 the exciton Bohr radius. Therefore a proper summing over the different $\mathbf{k}$ components needs to be done. As seen in Fig. 4(a), k_{cross} is comparable to $1/a_0$ (3D or 2D) for SHQWs but much larger for deep QWs. Since $k_{\text{cross}} \propto \Delta E_{\text{hh-lh}}^{1/2}$ (in the diagonal representation [7]) and the mixing coefficients in $\varphi_{\text{hh},k}$ are inversely proportional to $\Delta E_{\text{hh-lh}}$, a relationship of the form $\tau_s \propto \Delta E_{\text{hh-lh}}^{\beta}|_{k=0}$ with $\beta > 2$ is expected. A sudden increase of τ_s with $\Delta E_{\text{hh-lh}}$ is indeed observed in Fig. 4(b) for a value of $\Delta E_{\text{hh-lh}}$ corresponding to $x \sim 5\%$. For $x > 5\%$, τ_s continues to rise slightly as the deep well regime (and hence almost perfect 2D confinement) is approached. A similar scaling behavior was found when studying hole spin relaxation with cw techniques in bulk GaAs under stress [16], although their absolute values are somewhat higher than ours. Also, the highest valence band for stressed GaAs is the Γ_7 (light-hole) band, and some difference should be expected. These theoretical estimates of τ_s are very dependent on the scattering mechanism considered and published values are several orders of magnitude longer than our measured times. Whether the source of this disagreement lies in the character of the scattering process or in the excitonic nature of the problem, this will certainly provide a challenge for further theoretical investigations. Our work provides values of hole spin relaxation times for various well depths to which future calculations could be compared.

In conclusion, we report measurements of exciton and carrier spin dynamics in the intermediate dimensionality regime of shallow quantum wells of $GaAs/Al_xGa_{1-x}As$ with $0 < x \leq 6\%$. We use the technique of femtosecond nonlinear circular dichroism from which we extract the temporal decay of the spin-dependent interactions. An initial subpicosecond decay occurs in the SHQW samples which we assign to the hole spin relaxation. The same process takes several picoseconds as soon as the regime of SHQWs is abandoned, i.e., $x > 6\%$. The source for the measured changes of over 1 order of magnitude in the hole spin relaxation rate can be related to the amount of valence-band mixing which can be changed continuously as the well depth is decreased. Excitons, however, lose their two-dimensional nature when the Al concentration in the barriers is $< 0.82\%$. Therefore, for $0.82\% < x < 6\%$, a regime occurs in which carriers and excitons behave as two-dimensional confined entities, but the hole spin dynamics is essentially the same as the one observed in a bulk semiconductor. The existence of these two regimes is a mere consequence of the different source for these two dimensionality transitions. The 2D nature of the exciton in SHQWs is mainly determined by quantum confinement and possibly by a higher than expected effective confining potential, caused either by Coulomb correlations between adjacent wells or by failure of the "average band-gap" assumption in heterostructures with very dilute alloys. The spin relaxation behavior is mainly determined by the band structure and most significantly, by the amount of valence-band admixture for each SHQW structure.

We acknowledge the expert technical assistance of G. E. Doran and F. A. Feisser and useful discussions with T. C. Damen, J. Shah, D.-S. Kim, J. B. Stark, S.-L. Chuang, and R. T. Harley.

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