# Subpicosecond Spin Relaxation Dynamics of Excitons and Free Carriers in GaAs Quantum Wells

T. C. Damen, Luis Viña, J. E. Cunningham, and Jagdeep Shah
*AT&T Bell Laboratories, Holmdel, New Jersey 07733*

L. J. Sham
*Department of Physics, University of California at San Diego, La Jolla, California 92093*

(Received 21 June 1991)

We have obtained a coherent understanding of spin relaxation processes of electrons, holes, and excitons in quantum wells by investigating subpicosecond dynamics of luminescence polarization. We show that the spin behavior for electrons and holes in quasi-two-dimensional systems is distinct from that in bulk semiconductors and that many-body effects and formation processes play an important role in exciton spin relaxation.

PACS numbers: 78.47.+p, 71.35.+z, 71.70.Ej, 71.70.Gm

Investigations of polarization of interband optical transitions provide considerable information about the symmetry of electron, hole, and excitonic wave functions in semiconductors. Such studies have led to the identification of different spin relaxation processes [1,2] in bulk semiconductors. In quantum wells, cw measurements of linear [3] and circular polarization [4,5] of near-band-gap luminescence have been reported and various theories have been proposed to explain these results [6–12]. Time-resolved spectroscopy of luminescence polarization has also been reported recently for very high ($10^{19}$ cm$^{-3}$) carrier densities [13], for different samples at intermediate carrier densities with 150-fs time resolution [14,15], and for low densities [16,17].

In spite of this intense activity, spin relaxation in quantum wells is poorly understood. The quality of samples is important since spin dynamics can be strongly influenced by localization or defects. On a more fundamental level, the differences in the behavior of electrons, holes, and excitons must be recognized and carefully investigated. The prediction of slower hole spin relaxation in quantum wells compared to bulk semiconductors [10] has not been investigated. Finally, the influence of the formation dynamics of excitons and of many-body effects must be considered.

In this Letter, we discuss new insights into many of these unresolved issues obtained through our investigations of spin dynamics of electrons, holes, and excitons in quantum wells. Using *n*-modulation-doped quantum wells, we obtain the first measurement of hole spin relaxation time in a semiconductor. The measured $\approx 4$ ps demonstrates that the usual assumption of instantaneous hole spin relaxation is incorrect. We also show that the electron Fermi sea can be spin polarized under certain excitation conditions. In *p*-modulation-doped quantum wells, we measure electron spin relaxation time of $\approx 150$ ps, approximately a factor of 4 shorter than that in comparably *p*-doped bulk GaAs. We will show that an electron-hole exchange is responsible for this reduction. For intrinsic quantum wells, we show that polarized and unpolarized spectra exhibit an unusual splitting that depends on excitation density and time delay. This splitting results from many-body exchange interactions between excitons and contributes to an increase in the exciton spin relaxation time at higher density. Finally, we explain why spin relaxation in nonresonantly created excitons is faster than in resonantly created excitons. We believe that our quantitative measurements provide a standard of comparison for theoretical calculations.

The intrinsic sample used in the present studies is the same very-high-quality [18] GaAs/AlGaAs multiple-quantum-well (MQW) sample (80-Å wells, 150-Å barriers with $x = 0.3$, and 50 periods) that was used for earlier studies [17,19]. Intrinsic band-to-band luminescence was investigated in a high-quality *p* (*n*) sample, a GaAs/Al- GaAs MQW sample with 60-Å (50-Å) wells, 280-Å barriers with $x = 0.3$, with central 80 Å doped with Be (Si) and 60 periods. The estimated doping densities are $p_0 = 4 \times 10^{11}$ cm$^{-2}$ and $n_0 = 3 \times 10^{11}$ cm$^{-2}$. The samples were mounted in a cold finger cryostat (10–300 K) and weakly excited with 0.5-ps pulses [20] from a synchronously pumped Styryl 8 dye laser, tunable from 720 to 810 nm. The circularly polarized exciting beam was incident normal to the surface of the sample and luminescence was collected at about 10° to the normal.

The collected luminescence passed through an analyzer quarter-wave plate and was time resolved using an up-converter spectrometer [21] with 0.5-ps time resolution and 10-meV spectral resolution, determined primarily by the laser spectral width. Only vertically polarized light was up-converted. Polarized and depolarized luminescence intensities ($I_+$ and $I_-$ with the same and opposite circular polarization from the pump laser, respectively) were measured by rotating the quarter-wave plate in the exciting laser beam and keeping the analyzer quarter-wave plate fixed. Identical results were obtained by keeping the polarization of the incident beam constant and rotating the analyzer quarter-wave plate.

Figure 1(a) shows the cw photoluminescence (PL), photoluminescence excitation (PLE), and luminescence polarization ($P$) spectra as a function of the excitation energy for the *p*-modulation-doped sample. The PL corresponds to *band-to-band recombination* of photoexcited electrons with majority holes. The polarization, measured near the peak of PL (carriers near $k_\parallel \approx 0$), is large

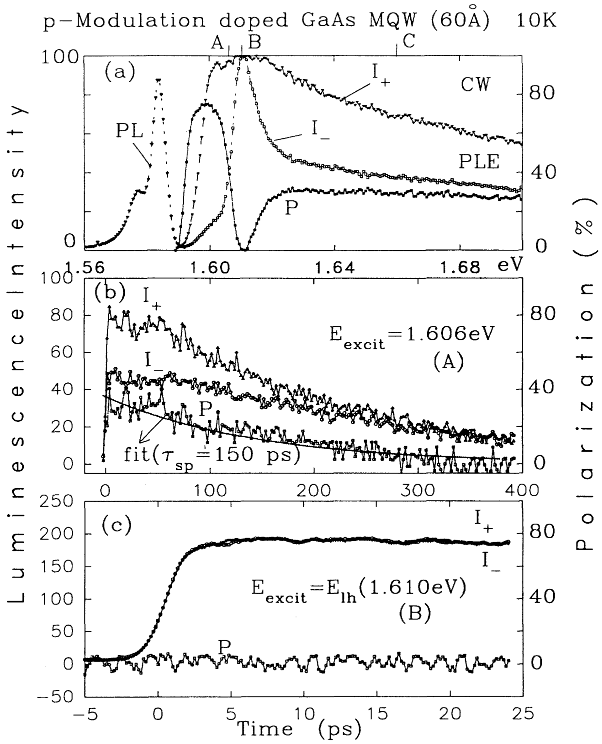

FIG. 1. $p$-modulation-doped sample. (a) cw PL, PLE, and polarization ($P$) spectra, (b) time evolution of $I_+$, $I_-$, and $P$ for excitation at $A$, and (c) same as (b) for excitation at $B$. Excitation density is $2\times10^{10}$ cm$^{-2}$ for (b) and (c).

for excitation close to the chemical potential ($A$), vanishes at light-hole (LH) exciton energy ($B$), and then recovers with further increase in excitation energy ($C$). The polarization is similar to that calculated earlier [10,11].

Figures 1(b) and 1(c) show the time evolutions of $I_+$, $I_-$, and $P=(I_+-I_-)/(I_++I_-)$ at the peak of the PL spectrum for excitations at $A$ and $B$, respectively; the time evolution at $C$ is similar to that at $A$. For excitations at $A$ and $C$, the polarization rises within our time resolution and then decays with a time constant of $150\pm25$ ps; for excitation at $B$, there is no polarization at any time.

We explain the results on the $p$-modulation-doped sample as follows. Assuming, without any loss of generality, that the incident light is right circularly polarized ($\sigma^+$), excitation of electrons from the heavy-hole (HH) band only ($A$) primarily produces spin $-\frac{3}{2}$ holes and spin $-\frac{1}{2}$ electrons [22]. Since the number of photoexcited holes is small compared to the doping level, the spin relaxation of holes is irrelevant and the measured decay of $P$ gives the electron-spin relaxation time of $\tau_{sp,e}=150\pm25$ ps for this sample.

Spin polarization for excitation at $B$ is zero at all times, because increased absorption at LH excitons and increased spin admixture in the HH wave function at large $k$ creates a nearly equal population of spin $+\frac{1}{2}$ and $-\frac{1}{2}$ electrons. For this reason, zero or small (positive or negative) spin polarization for excitation at LH exciton should be a general feature in all $p$ samples. For excitation at $C$, the HH absorption dominates because one has moved away from LH exciton resonance. This reduces the creation of spin $+\frac{1}{2}$ electrons compared to excitation at $B$ and leads to the observation of a positive polarization at $C$. We note that the cw polarization data are consistent with the reduction factor $\tau_{sp,e}/(\tau_{rec}+\tau_{sp,e})$ averaged over the spectral bandwidth, where the spin relaxation time $\tau_{sp,e}$ and the recombination time $\tau_{rec}$ are from the time-resolved measurements. These features of the polarization spectrum are in complete agreement with the theory of Ref. [11]. The description above of the causes of these features is based on the actual calculation.

For comparable hole density ($6\times10^{17}$ cm$^{-3}$) and temperature (10 K), our measured spin relaxation time of $\approx150$ ps in quantum wells is about a third to a quarter of that in $p$-doped bulk GaAs [2]. In Ref. [2] it was inferred from the hole density and temperature dependence that electron-hole scattering with simultaneous exchange [Bir-Aronov-Pikus (BAP) mechanism [23]] dominates, rather than the D'yakonov-Perel (DP) mechanism [7]. The enhancement of the spin relaxation rate in the quantum well compared to the bulk due to the exchange interaction [24] is $3\pi/2k_FL$, where $k_F$ is the Fermi vector for the 3D density and $L$ the well width, whereas the enhancement due to the DP mechanism [7] is $4/(k_FL)^4$. For our sample, we calculate that the BAP enhancement is 3 and the DP mechanism is less than 1. Therefore, the BAP is likely to be the dominant mechanism for electrons in $p$-quantum wells.

Luminescence from the *n-modulation-doped sample* results from band-to-band recombination of photoexcited holes with electrons. Figure 2(a) shows the time evolution of $I_+$, $I_-$, and $P$ for excitation at 1.645 eV that creates electrons close to the Fermi energy and holes at $\approx3$ meV. The peak polarization of about 30% decays with a time constant of $\approx4$ ps. Data for excitation at 1.7 eV [Figs. 2(b) and 2(c)] that excites electrons from both HH and LH bands show the presence of an additional slow component with a decay time constant of $\approx200$ ps.

Using arguments similar to the $p$ sample, we identify the fast decay with the spin relaxation of holes ($\tau_{sp,h}=4$ ps). This provides the first measurement of the *hole* spin relaxation rate in a semiconductor. Since hole states are in general an admixture of various spin states, any energy or momentum relaxation process (e.g., with acoustic phonon or defects or other carriers) lead to spin relaxation [10–12].

Our measurements show that the usual assumption of instantaneous hole spin relaxation is not valid in quasi-2D systems. The increased hole spin relaxation time is a result of the lifting of valence-band degeneracy at $k_\parallel=0$. This is consistent with the increase in hole spin relaxation

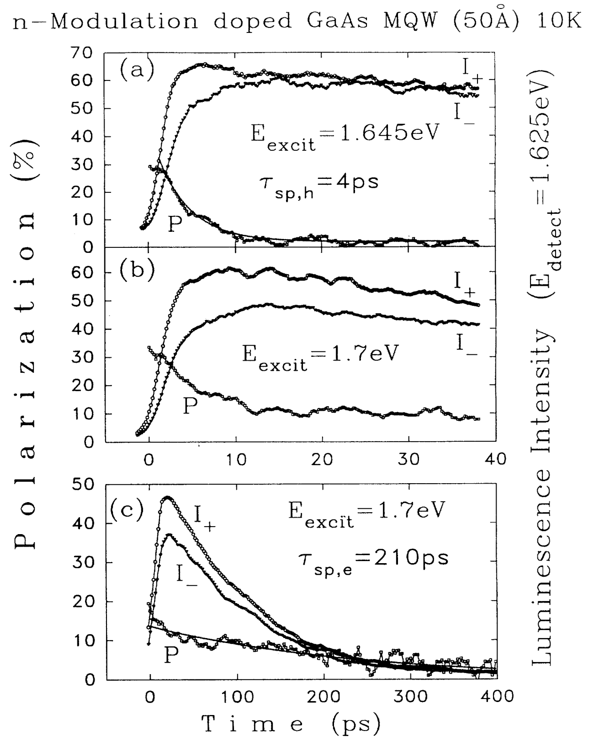

FIG. 2. Luminescence intensities and polarization for the $n$-modulation-doped sample at $2\times10^{10}$ cm$^{-2}$ for excitation at (a) 1.645 eV and (b),(c) 1.7 eV.

time in bulk GaAs under strain [25] and also with recent theoretical considerations [10–12]. However, a quantitative comparison with theory is not possible at present because different scattering processes contribute to hole relaxation and because we measure the spin relaxation of a thermalized, hot-hole distribution (photoexcited holes achieve this distribution in $<1$ ps).

For high-energy excitations, the electrons are created with energies comparable to or larger than the electron Fermi energy ($\approx 10$ meV). Electron-electron collisions mix the electrons and give the Fermi sea a net spin polarization that decays with $\tau_{\mathrm{sp},e}$. We find that the ratio of the slow to the fast component increases with intensity as expected. Since the slow component is present only if there is a net polarization within the spectral width of detection, it is absent for low-energy excitations for which the photoexcited electrons mix primarily with electrons near the Fermi energy. A small, net polarization of the Fermi sea in the $p$-doped sample also should be present at very short times ($<4$ ps); however, its effects are probably masked by the much larger polarization of the photoexcited electrons.

For the *intrinsic sample* we monitor the HH *exciton luminescence.* Earlier results [17] on this very-high-quality sample [18] showed that the exciton spin relaxation time is about 50 ps for resonant excitation and less than the streak-camera resolution ($\leq 20$ ps) for nonresonant (band-to-band) excitation. The present up-

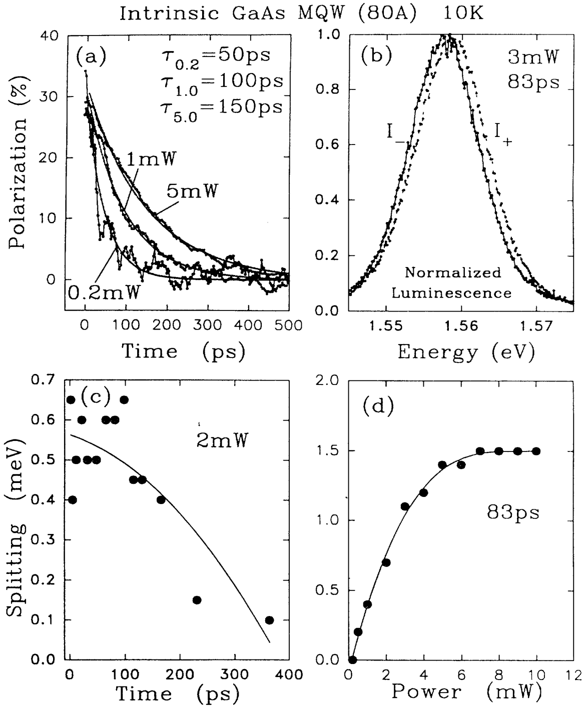

FIG. 3. Intrinsic sample. (a) Time evolutions of $P$, (b) polarized and depolarized spectra at delay = 83 ps, (c) dependence of splitting on time at the power indicated, and (d) density at the delay indicated. 10 mW corresponds to a density of $\approx 1\times10^{11}$ cm$^{-2}$. Note that the spectral width in (c) is determined primarily by the laser spectral width and the spectrometer slits; the cw spectral width is 0.65 meV [18].

conversion system provides much better time resolution but does not allow measurement for resonant excitation [21] and requires higher excitation intensity. For these conditions, the spin relaxation time decreases from 150 to 50 ps as the excitation intensity is decreased [Fig. 3(a)] [26]. A striking aspect of the data is that the polarized and depolarized spectra exhibit a splitting in energy [Fig. 3(b)]. The depolarized spectrum is always at lower energy, independent of the helicity of the exciting laser. The splitting decreases with delay and increases with intensity [Figs. 3(c) and 3(d)]. These results show that spin dynamics in intrinsic samples is extremely complicated.

We first consider why the spin relaxation time at the lowest intensity is shorter for nonresonant excitation than for the resonant excitation. We believe that a combination of the exchange interaction between the electron and the hole and the spin-orbit interaction on the hole is needed to flip the spin of the exciton from $|+1\rangle$ to $|-1\rangle$ or vice versa because exchange which conserves total angular momentum cannot flip the total spin by itself. Experimental results for the resonant excitation show that $\tau_{\mathrm{sp,exc}} \approx 50$ ps.

For nonresonant excitation, the electron-hole pair relaxes in two stages [19]: In less than 20 ps, it forms an exciton with a large center-of-mass momentum ($K$), and then in over 300 ps, the exciton loses its total momentum. During the first stage, the electron-hole pair forms an exciton by scattering with acoustic phonons. The hole, being the heavier particle, acquires the major share of the total momentum. This increases the spin-orbit mixing and thus enhances the spin flip with the aid of the exchange compared to that in the exciton ground state. This may explain why the nonresonant spin relaxation time is shorter than the resonant one.

We next explore the reason for the relative shift between the polarized and depolarized spectra. Consider excitation by a $\sigma^+$ laser which primarily creates $|+1\rangle$ excitons. For a sufficient density, the excitons will have an energy distribution because of the exclusion principle acting separately on the electrons and on the holes. The effect is opposite on $|+1\rangle$ and $|-1\rangle$ excitons because the former experiences a repulsion, while the latter an attraction. (The idea is similar to the antibonding and bonding states of a hydrogen molecule.) The energy difference depends on the density of excitons, and approaches zero as the densities of the two spin populations approach each other. Both of these expectations are confirmed by experiments [Figs. 3(c) and 3(d)]. For a larger intensity of excitation, the closer approach of the excitons also enlarges the electron-hole relative distance and thus lowers the exchange interaction. These many-body effects (the energy shift and the weakened exchange) lead to a longer relaxation time at higher excitation densities [Fig. 3(a)].

In conclusion, we have quantitatively determined electron, hole, and exciton spin relaxation times in quantum wells and find significant differences between quasi-2D and bulk semiconductors. The spin relaxation for holes is considerably slower in quantum wells than in bulk and just the opposite is true for electrons. We have elucidated the reasons for this seemingly contradictory behavior. The exciton spin relaxation time is between that for electrons and holes, as one might expect. We have shown that the exciton spin relaxation is influenced by how it is formed as well as by many-body effects. Although the parameter space in quantum wells is particularly large (different well widths, doping densities, etc.), these results should stimulate experimental investigations of this parameter space, and should provide a basis for developing a quantitative theory of spin relaxation in quasi-2D systems.

We thank K. Leo and G. 't Hooft for discussions. Research by L.J.S. was supported by the Center for Materials Science, Los Alamos National Laboratory and by NSF Grant No. DMR 88-15068, and research by L.V. was partially supported by DGICYT of the Spanish Ministry of Education.

---

[1] *Optical Orientation*, edited by F. Meier and B. P. Zachachrenya (North-Holland, Amsterdam, 1984).

[2] K. Zerrouati, F. Fabre, G. Bacquet, J. Bandet, J. Frendon, G. Lampel, and D. Paget, Phys. Rev. B **37**, 1334 (1988).

[3] R. Sooryakumar, D. S. Chemla, A. Pinczuk, A. C. Gossard, W. Wiegmann, and L. J. Sham, Solid State Commun. **54**, 859 (1985).

[4] R. C. Miller and D. A. Kleinman, J. Lumin. **30**, 520 (1985).

[5] M. J. Snelling, A. S. Plaut, G. P. Flinn, A. G. Trooper, R. T. Harley, and T. M. Kerr, J. Lumin. **45**, 208 (1990).

[6] A. E. Ruckenstein, S. Schmitt-Rink, and R. C. Miller, Phys. Rev. Lett. **56**, 504–508 (1986).

[7] M. I. D'yakonov and V. Yu. Kachorovskii, Fiz. Tekh. Poluprovodn. **20**, 178–182 (1986) [Sov. Phys. Semicond. **20**, 110–112 (1986)].

[8] L. J. Sham, J. Phys. (Paris), Colloq. **48**, C5-381 (1987).

[9] A. Twardowski and C. Hermann, Phys. Rev. B **35**, 8144 (1987).

[10] T. Uenoyama and L. J. Sham, Phys. Rev. Lett. **64**, 3070–3073 (1990).

[11] T. Uenoyama and L. J. Sham, Phys. Rev. B **42**, 7114–7123 (1990).

[12] R. Ferreira and G. Bastard, Phys. Rev. B **43**, 9687 (1991).

[13] H-S. Chao, K. S. Wong, R. R. Alfano, H. Unlu, and H. Morkoc, J. Soc. Photo-Opt. Instrum. Eng. **942**, 215–222 (1988).

[14] M. R. Freeman, D. D. Awschalom, J. M. Hong, L. L. Chang, and K. Ploog, in *Proceedings of the Twentieth International Conference on the Physics of Semiconductors*, edited by E. M. Anastassakis and J. M. Joannopoulos (World Scientific, Singapore, 1990).

[15] M. Kohl, M. R. Freeman, D. D. Awschalom, and J. M. Hong (unpublished).

[16] H. Stolz, D. Schwarze, W. von der Osten, and G. Weimann, Superlattices Microstruct. **6**, 271–282 (1989).

[17] T. C. Damen, K. Leo, J. Shah, and J. E. Cunningham, Appl. Phys. Lett. **58**, 1902–1904 (1991).

[18] The PL linewidth for this sample is 0.65 meV at 10 K and coincidence of PL and PLE peaks shows that there are no localization effects.

[19] T. C. Damen, Jagdeep Shah, D. Y. Oberli, D. S. Chemla, J. E. Cunningham, and J. M. Kuo, Phys. Rev. B **42**, 7434–7438 (1990).

[20] For measurements $< 20$ meV from excitation laser, the pulse width was increased to 1 ps.

[21] Jagdeep Shah, IEEE J. Quantum Electron. **24**, 276 (1988).

[22] We denote the hole spin $m$ by the valence-electron spin and the electron spin $s$ by the conduction-electron spin so that the exciton spin is $s-m$.

[23] G. L. Bir, A. G. Aronov, and G. E. Pikus, Zh. Eksp. Teor. Fiz. **69**, 1382 (1975) [Sov. Phys. JETP **42**, 705 (1976)].

[24] See, for example, U. Rossler, S. Jorda, and D. Broido, Solid State Commun. **73**, 209 (1990).

[25] G. Lampel, A. N. Titkov, and V. I. Safarov, in *Proceedings of the Fourteenth International Conference on the Physics of Semiconductors*, edited by B. L. H. Wilson (Institute of Physics, Bristol, 1978).

[26] This trend in the spin relaxation times shows that the two sets of data, taken over a nonoverlapping intensity range, are consistent.