Manipulation of the Spin Memory of Electrons in n-GaAs

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We report on the optical manipulation of the electron spin relaxation time in a GaAs-based heterostructure. Experimental and theoretical study shows that the average electron spin relaxes through hyperfine interaction with the lattice nuclei, and that the rate can be controlled by electron-electron interactions. This time has been changed from 300 ns down to 5 ns by variation of the laser frequency. This modification originates in the optically induced depletion of an n-GaAs layer.

A long spin relaxation time for electrons, τ_s [1–3], could hold promise for magnetoelectronics and quantum information applications [4,5]. This time can be controlled by an external magnetic field [6], by choice of heterostructure geometry [7], by doping level [2,6,8], and by a gate voltage that changes the electron concentration in the two-dimensional channel [9].

This Letter is devoted to the *optical* manipulation of the spin memory of electrons. We show, using optical orientation methods, that the spin relaxation rate in a GaAs-based heterostructure can be modified by a factor of 100 by variation of the laser frequency. The comparison between experiment and theory shows that the observed effect is the result of turning on or off the hyperfine channel of spin relaxation of electrons localized on shallow donors. The giant changes of spin relaxation time allow one to extend the concept of spin dynamics engineering in quantum wells [9] to quasibulk semiconductors having much longer spin lifetimes.

The optical orientation experiments were carried out on a sample with a semi-insulating GaAs substrate, on which a 500 nm thick GaAs buffer and a 25 nm thick AlAs barrier separating the substrate from the main structure were grown by molecular beam epitaxy. Following this is a 100 nm thick GaAs layer capped by a 25 nm thick $Al_{0.3}Ga_{0.7}As$ barrier, above which a series of five quantum wells (QWs) of different thickness were prepared (Fig. 1b). The sample, though nominally undoped, has a background doping level of order 10^{14} cm^{-3} n-type from Hall measurements on test structures. The dominant photoluminescence (PL) at the wavelength close to the GaAs band edge is due to the 100 nm layer. The sample was placed in a liquid-helium cryostat and pumped by a tunable Ti-sapphire laser, with the circular polarization of light being alternated in sign at a frequency of 26.61 kHz with a photoelastic quartz modulator. The geomagnetic field was compensated to less than 0.1 G at the sample. The PL polarization was measured in the reflection geometry with a fixed circular-polarization analyzer by a double-grating spectrometer. The electronics provided measurement of

the *effective* degree of circular polarization $\rho = (I_+^+ - I_+^-)/(I_+^+ + I_+^-)$, where I_+^+, I_+^- are the intensities of the σ^+ PL component under σ^+ and σ^- pumping, respectively.

When the excitation energy, $h\nu$, is below the absorption edge of the quantum wells, the PL spectrum of the 100 nm GaAs layer (curve 1 in Fig. 1a at $h\nu = 1.520$ eV) is dominated by two lines, corresponding to the recombination of the free exciton (X) and of the exciton bound to a neutral donor (D^0X) [10]. The GaAs PL spectrum changes drastically when the laser frequency is in resonance with the heavy-hole exciton (HHX) transition of the quantum well (14 nm) nearest to the 100 nm layer
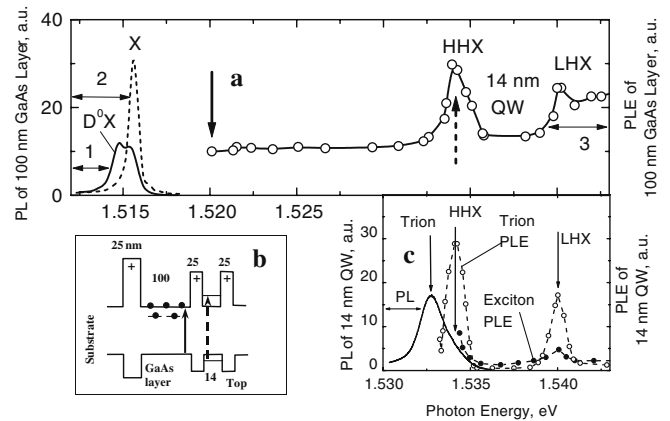

FIG. 1. (a) PL and PLE spectra of the 100 nm thick GaAs layer. PL curve 1 was measured at excitation, $h\nu = 1.520$ eV (only GaAs layer was excited), and curve 2 was measured at $h\nu = 1.534$ eV (the heavy-hole exciton in the 14 nm QW was excited resonantly). PLE curve 3 was measured at the exciton (X) energy (1.5155 eV). Maxima in the PLE spectrum correspond to the resonant excitation of the heavy-hole (HHX) and light-hole (LHX) excitons in the 14 nm QW. (b) The part of the structure containing the 100 nm thick GaAs layer and the 14 nm QW. Without illumination, additional electrons from barriers and quantum wells are accumulated in the GaAs. (c) PL and PLE spectra of the 14 nm thick QW. PL (solid curve) was measured at excitation, $h\nu = 1.540$ eV (LH exciton). PLE of the exciton line (1.534 eV, full circles) and trion line (1.5326 eV, open circles).

(curve 2 at $h\nu = 1.534$ eV, Fig. 1a). The intensity of the exciton line becomes 3 times larger, whereas the intensity of the D^0X complex decreases. The same changes take place when one excites the light-hole exciton (LHX) ($h\nu = 1.540$ eV). Curve 3 shows the PL excitation (PLE) spectrum of the GaAs layer exciton line when the laser frequency is tuned through the exciton resonances in the 14 nm QW.

One can see sharp enhancements in the PL intensity of the 100 nm GaAs exciton peak under resonant excitation of heavy-hole and light-hole excitons in the QW. The PLE spectrum of the D^0X complex evolves with phase opposite to that of the exciton PLE. There is also a striking interplay between the exciton and trion lines in the spectrum of the QW PL. The solid line in Fig. 1c shows that the PL trion peak (1.5326 eV) dominates over the HHX PL (1.534 eV) when one excites the LHX transition in the 14 nm QW. The full (open) circles show the PLE spectrum of the QW exciton (trion) peak. One can see that resonant excitation of excitonic transitions favors the trion line, whereas the exciton line is stronger at excitation out of resonance. The quenching of D^0X in the PL of bulk GaAs and the simultaneous sharp enhancement of the trion in the PL spectrum of the QW indicate that the GaAs layer is depleted of electrons, whereas the 14 nm QW is enriched of them [11]. This phenomenon disappears when the pump density is below 0.1 mW/cm². In this case the PL of the GaAs layer consists of D^0X and X lines, and the QW PL reveals only the exciton line. These results can be understood qualitatively with the use of Fig. 1b. Without excitation of the QW, the GaAs layer is charged negatively: besides donor-bound electrons there are extra free electrons escaping the donors from the surrounding barriers and wells. The excitation of the QW induces their redistribution, depleting the GaAs layer and filling the QW. One possibility is the tunneling of the QW photoexcited holes into the 100 nm GaAs layer and recombination with electrons. Another possibility is the annihilation of one QW exciton with the simultaneous ionization of another one through the Auger process. In this case the hole goes into the GaAs by jumping over the barrier while the electron goes back due to repulsion from the negatively charged layer.

Overall, the experimental data clearly show that the density of electrons in the 100 nm layer is reduced and that in the QW is increased independent of the specific mechanism responsible for the recharging effect. In this paper we will *exploit* this phenomenon to manipulate the 100 nm layer electron concentration optically in order to trace the electron *spin* relaxation in the 100 nm GaAs layer. We show that the depletion of the 100 nm GaAs switches on a fast electron spin relaxation mechanism (5 ns) involving lattice nuclei, whereas the presence of extra electrons in the layer strongly suppresses it (up to 300 ns).

The electronic spin relaxation in GaAs can be probed accurately with the use of the Hanle effect under optical pumping conditions [6]. Circularly polarized light creates electronic spin polarization oriented along the exciting beam. The degree of circular polarization, ρ, is determined by the projection, S_z, of the mean electron spin on the pump beam direction: $\rho = S_z$. An external magnetic field in Voigt geometry (i.e., a transverse field) decreases the steady state PL polarization due to Larmor precession with frequency $\omega = \mu_B g_e B/\hbar$ (μ_B is the Bohr magneton, $g_e = -0.44$ [12] is the electron g factor). The field dependence of ρ is described by a Lorentzian curve with half-width $B_{1/2} = \hbar/\mu_B g_e T_s$ (Hanle effect), where the electron spin lifetime $T_s = (1/\tau + 1/\tau_s)^{-1}$ is determined by recombination $1/\tau$ and spin relaxation $1/\tau_s$ rates. The electron lifetime in n-type semiconductors depends strongly on the pumping intensity (photohole concentration), and in the low pumping limit is very long ($\tau \gg \tau_s$). In this limit the spin lifetime coincides with the spin relaxation time, e.g., $T_s(W = 0) = \tau_s$, and determines directly the linewidth of the Hanle curve.

Figure 2 shows the Hanle depolarization curves measured at different laser energies with fixed power density, $W = 40$ mW/cm². Curve 1 shows the data measured under the quasiresonant creation of the exciton of the 100 nm layer. The half-width value, $B_{1/2} = 2.9$ G, corresponds to $T_s = 90$ ns. The spin lifetime becomes shorter when one excites above the GaAs energy gap (curve 2): $T_s = 40$ ns ($B_{1/2} = 6.8$ G). The inset of Fig. 2 shows the dependence of $1/T_s$ on W under quasiresonant excitation of the GaAs exciton. The low power limit of this dependence gives $\tau_s = (290 \pm 30)$ ns.

Increasing the laser frequency, with power fixed, up to the heavy-hole exciton energy in the 14 nm QW brings no
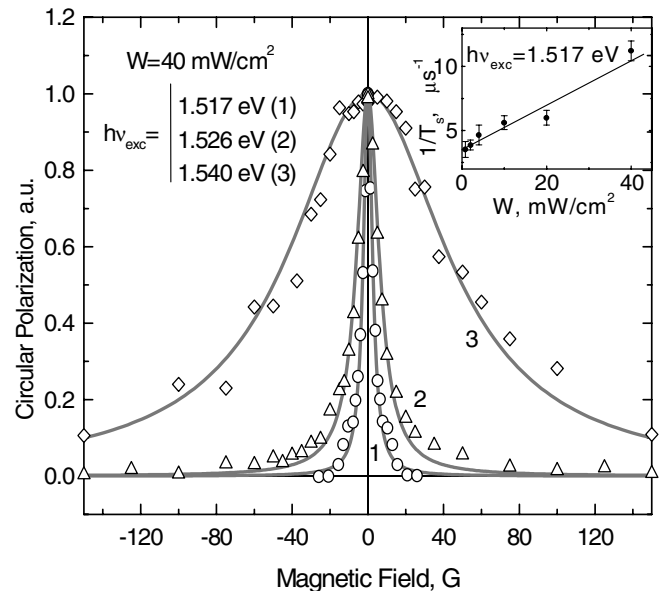

FIG. 2. The Hanle effect of the 100 nm GaAs electrons at three excitation conditions: (1) with quasiresonant excitation of GaAs excitons; (2) with excitation above the GaAs energy gap but in the transparency region of QWs; (3) with resonant excitation of the LH-exciton in the 14 nm QW. Inset: power dependence of the reciprocal spin lifetime.

drastic changes to the half-width of the Hanle curve, $B_{1/2}$. The situation changes dramatically when either the QW heavy-hole or light-hole exciton is excited: $B_{1/2}$ broadens to 50 G. As an example we present the data obtained under resonant LHX excitation (curve 3 in Fig. 2). When the exciting frequency is between the HHX and LHX, the QW is not excited and the polarization behavior of the 100 nm layer luminescence is similar to curve 2. Further increasing the laser frequency above the QW resonances increases the QW excitation and $B_{1/2}$ again approaches to 50 G. We conclude that there is a large quantitative difference in $B_{1/2}$ of the 100 nm layer when the QW is excited (the layer is depleted of electrons) or not (extra free electrons fill up the layer). The change of $B_{1/2}$ cannot be connected with spin transfer from the QW across the 25 nm AlGaAs barrier during the recharging process. Direct evidence against this is that both the zero-field polarization values (+2%) and the Hanle effects in the GaAs layer are identical under LHX (Fig. 2, curve 3) and HHX (Fig. 3b) excitation of the 14 nm QW. However, the QW exciton polarization has opposite signs in these two cases due to selection rules (experimental values are −10% for LHX and +20% for HHX excitations for positive laser polarization).

Remarkably, *qualitative* differences between the two regimes appear when the magnetic field is applied in the Faraday geometry, i.e., under a longitudinal field. In Fig. 3, polarization data taken in the Faraday and Voigt geometry are compared for the two regimes: when the QW is not excited and extra electrons remain in the 100 nm layer (Fig. 3a); and when the QW is excited and the electron population is reduced (Fig. 3b). With the extra electron population, there is practically no magnetic field dependence in the Faraday geometry (upper part of Fig. 3a). In contrast, when the electron population is reduced, application of the magnetic field in the Faraday geometry increases the polarization by a factor of 2.5 (upper part of Fig. 3b). Moreover, although there is no correlation in the change in polarization in the two geometries in Fig. 3a, the characteristic transverse and longitudinal magnetic field values that decrease and increase, respectively, the electron polarization are surprisingly similar in Fig. 3b. This difference in behavior reflects a qualitative difference in the spin relaxation process in the two experimental regimes, i.e., with high (Fig. 3a) and low (Fig. 3b) electron concentrations in the GaAs layer.

The experimental data shown in Fig. 3 correspond to the two limiting cases of the electron spin relaxation in a rapidly fluctuating or static random magnetic field. The spin relaxation can be viewed as the result of the action of random local magnetic fields on electron spin [13] and depends both on the amplitude of the random field and on its fluctuation rate. In the static limit the characteristic time (τ_c) of the local field fluctuation is much longer than the period of electron spin precession in this field (ω_f^{-1}), and the electron spins undergo precession in the static fields distributed randomly. The average spin of the ensemble relaxes to 1/3 of its initial value with a characteristic time
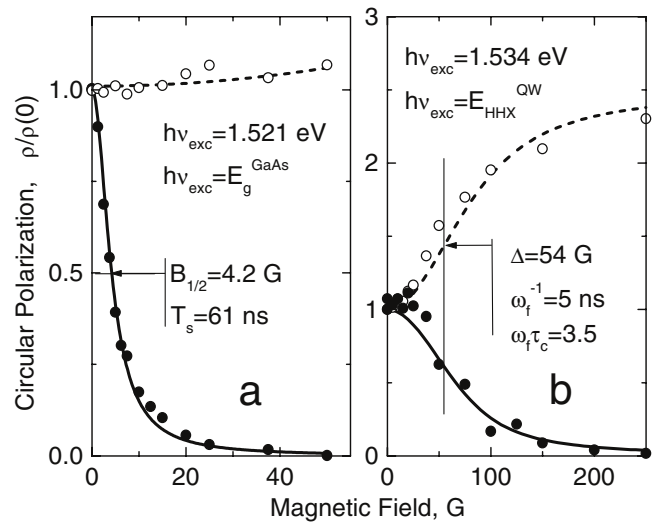

FIG. 3. Magnetic field dependence of the circular polarization in Voigt (full circles) and Faraday (open circles) geometries under two excitation conditions at $T = 4.2$ K. Polarization degree is normalized to the zero-field values, $\rho(0)$, of the exciton PL (1.5155 eV) in GaAs. (a) GaAs layer is excited only [$\rho(0) = 15\%$]. The solid curve is a Lorentzian fit with $B_{1/2} = 4.2$ G. The dashed line is a guide to the eye. (b) Resonant excitation of the HH exciton in the 14 nm QW [$\rho(0) = 2\%$]. The dashed and solid lines are the theoretical dependences of the electron spin calculated from Eq. (1) with the only fitting parameter, $\tau_c = 17$ ns.

$\tau_s \approx \omega_f^{-1}$ [14]. The longitudinal external field eliminates spin relaxation, and the transverse field depolarizes electrons when it overwhelms the local field, i.e., $\omega \gtrsim \omega_f$. In the opposite limit, $\tau_c \ll \omega_f^{-1}$ (the so-called motional narrowing case [15]), dynamic averaging takes place and the spin relaxation slows down: $\tau_s^{-1} \approx \omega_f^2 \tau_c$. In this case, the transverse magnetic field that depolarizes the electron spin (Hanle effect) satisfies the condition $\omega \gtrsim \omega_t \equiv \tau_s^{-1} \approx \omega_f^2 \tau_c$. The longitudinal field required to suppress spin relaxation satisfies the condition: $\omega \gtrsim \omega l \equiv \tau_c^{-1}$ (see Ref. [16]). It happens in much greater magnetic fields because $\omega_t = \tau_s^{-1} \approx \omega_f^2 \tau_c = (\omega_f \tau_c)^2 \omega_l \ll \omega_l$. Therefore, the experimental result of Fig. 3a, with additional electrons in the layer, corresponds to the motional narrowing case ($\omega_l \gg \omega_t$), and the result of Fig. 3b, with electrons removed, corresponds to the static limit ($\omega_l \approx \omega_t$).

The experimental data obtained in the static case enable us to deduce that the main spin relaxation mechanism of electrons in this experiment is the hyperfine interaction of localized electrons with lattice nuclei. It follows from Fig. 3b that the characteristic precession frequency of the electron spins in the random local field is $\omega_f \approx 2 \times 10^8$ s^{-1}. Therefore $\tau_c > \tau_s \approx \omega_f^{-1} \approx 5$ ns. None of the known spin relaxation mechanisms of free carriers and excitons (Chap. 3 in Ref. [6]) can have such long correlation times. However, bound electrons at low electron concentration and low temperature may spend a long time at a given donor. Moreover, the ω_f value agrees well with the calculated precession frequency of spins of the

donor-bound electrons in the random hyperfine fields of surrounding nuclei as we now show.

To describe the average electron spin polarization in the low concentration limit we consider an evolution of the electron spin, S_n, localized at the nth donor. It is described by the Bloch equation [6] with the precession frequency, $\Omega_n = \omega + \omega_{fn}$:

$$\frac{dS_n}{dt} = \frac{S_i - S_n}{\tau_c} + \Omega_n \times S_n = 0, \qquad (1)$$

where S_i is the initial electron spin value in the moment of trapping. During the correlation time, τ_c, the electron spin dynamics is determined by the external and the local nuclear magnetic fields only. The static nuclear fields are described by a Gaussian distribution, $\Phi(B_N) = \pi^{-3/2}\Delta^{-3}\exp(-B_N^2/\Delta^2)$, with a dispersion, $\Delta = 54$ G, given by the root mean square of the random nuclear field [13]. The average nuclear polarization is zero because we are modulating the polarization of exciting light. Averaging the solution of Eq. (1) over the nuclear magnetic field distribution [14] results in a theoretical dependence of circular polarization on the longitudinal and transverse magnetic field that are presented in Fig. 3b by dashed and solid lines, respectively. The *only* fitting parameter for the curves in Fig. 3b is $\tau_c = 17$ ns, which satisfies the static condition, $\tau_c > \omega_f^{-1}$. This value is determined by the processes of donor electron ionization, recombination, exchange scattering by free electrons, etc. The good agreement between experiment and theory confirms that the interaction with nuclei is the main mechanism of the electron spin relaxation for the localized carriers.

The much longer spin relaxation time in the motional narrowing case ($\tau_s \approx 300$ ns) arises from the increase in electron concentration. Recall that the presence of extra free electrons, in addition to the "intrinsic" donor-bound ones, comes from donors in the barriers and quantum wells surrounding the GaAs layer. The suppression of spin relaxation is caused by the decrease of the correlation time (from $\tau_c > 5$ ns to $\tau_c \approx 0.1$ ns [17]) and can be explained by scattering processes between the free and bound electrons discussed in Ref. [18]. These processes average out the inhomogeneous nuclear field and suppress spin relaxation by 2 orders of magnitude. The exciton PL serves as a detector of spin polarization of donor-bound electrons [19].

In conclusion, we have shown that the spin relaxation time of donor-bound electrons in a thick GaAs layer depends strongly on the concentration of extra electrons. This enabled us to manipulate the spin relaxation rate by 2 orders of magnitude. The presence of extra free electrons in the conduction band suppresses strongly the spin relaxation of the donor-bound electrons because the inhomogeneous nuclear field is averaged out, leading to an extremely long spin relaxation time of 0.3 μs. Decrease of the electron concentration breaks up the electron spin system into an ensemble of individual donor-bound electrons,

and the electronic spin relaxation is the result of dephasing by the randomly distributed hyperfine fields. In our experiments, the average of the nuclear spin polarization is zero but its dispersion is approximately 54 G, corresponding to an inhomogeneous dephasing rate of 5 ns. The effect of inhomogeneous dephasing can be eliminated with the use of the spin echo technique Ref. [15]. This case may be important in efforts to implement quantum computing using localized electronic spins, which not only require long coherence times but also must be localized in order to be individually addressed.

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