Optical detection of spin relaxation of 2D electrons during photoexcitation

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Direct optical detection of the relaxation of the spin orientation of 2D electrons in a quantizing magnetic field has been achieved under conditions such that the corrections for the exchange interaction of the spins of an electron spin and a photoexcited hole are reduced to zero. This is the first such detection. The time scale of the relaxation to a spin equilibrium has been found through time-resolved measurements to be on the order of 5 ns in a field of 2.6 T. The spin relaxation slows down as the index of the Landau level decreases and as the magnetic field simultaneously increases. The experimental results point to spin-flip mechanisms involving a spin–orbit interaction.

1. The spin relaxation of electrons in 2D systems has been the subject of numerous recent studies, both experimental[1–7] and theoretical.[8,9] A distinguishing feature of the 2D problem is a quantization of the spin in the direction in which the motion of the electrons is quantized. Another feature is the discrete nature of the energy spectrum in high magnetic fields, $\hbar\omega_c \gg kT$. Since a flip of an electron spin requires a magnetic interaction, one can distinguish four basic mechanisms for spin relaxation: a scattering by a paramagnetic impurity, a contact interaction with the host nuclei of the lattice, an exchange interaction with the spin of photoexcited holes, and a spin–orbit interaction, with the associated corrections to the Hamiltonian of the electron system which lead to a mixing of different spin states. In high-quality heterostructures grown by molecular beam epitaxy, the concentration of paramagnetic impurities may be so low that these impurities have no substantial effect on the relaxation of 2D electrons. The very long relaxation times (tens of minutes[5]) of the nonequilibrium magnetization of lattice nuclei caused by the contact interaction with 2D electrons means that the inverse scattering channel also has a low rate. Exciton effects which unavoidably arise during optical pumping in symmetric quantum wells should lead to a rapid exchange of the spins of an electron and a hole. The relaxation of a hole spin occurs rapidly because of the strong spin–orbit coupling; the holes in GaAs differ from the conduction electrons (the s-wave state) in having a p-wave wave function. As a result, the relaxation of a hole spin is on the order of the momentum relaxation time; the spin of an electron follows this relaxation because of exchange. Studies of the depolarization of the optical orientation of 2D carriers in a crossed magnetic field[3] (the Hanle effect) and time-resolved measurements during resonant optical orientation[1,2] yield values

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~ 150 ps for the spin relaxation time of an electron bound to a free hole in an exciton. Transport studies of spin-dependent processes carried out to date[4-7] yield indirect estimates of the spin relaxation time; these estimates are much longer; ~ 5–10 ns. There is every reason to believe that the last of the four mechanisms listed above—the spin–orbit coupling—can be manifested under these conditions. In this case a mixing of spin states occurs because of terms associated with the absence of an inversion center, both at the heterojunction itself (including the electric field that forms the well) and in the interior of the GaAs itself. The solution of the Schrödinger equation in a quantizing magnetic field,

$$E_{n\downarrow}=(n+1/2)\hbar\omega_c-1/2g\mu_bB, \quad E_{n\uparrow}=(n+1/2)\hbar\omega_c+1/2g\mu_bB, \tag{1}$$

determines the energy of the Fermi level n, including the Zeeman splitting. When the spin–orbit terms are taken into account[9-11] the eigenenergies

$$E_{n\pm}=(E_{n+1\uparrow}+E_{n\downarrow})/2+[((E_{n+1\uparrow}-E_{n\downarrow})/2)^2+\beta^2\hbar\omega_c(n+1)]^{1/2} \tag{2}$$

differ only slightly from those of unperturbed states of the Landau levels, $E_{n+}\approx E_{n+1\uparrow}$ and $E_{n-}\approx E_{n\downarrow}$. However, the occurrence of a mixing of states with different spin directions, $\Psi_{n\pm}=a_\pm(n)\,|n,\downarrow\rangle+b_\pm(n)\,|n+1,\uparrow\rangle$, will determine spin-flip processes which involve a unit change in the index of the Landau level. The elastic scattering of electrons by large-scale fluctuations of the random potential (charged impurities which have moved beyond the spacer) and the emission of acoustic phonons may lead to transitions between different Landau levels and will be accompanied by spin flip to the extent that there is a spin–orbit interaction.

2. In the present experiments we studied the recombination spectra of 2D electrons in a single heterojunction with a hole bound at an acceptor in a δ-layer lying 300 Å from the electron channel. The electron density was set by charged impurities (there was a doping of the AlGaAs) beyond the spacer, which was 225 Å. The effect of free holes on the relaxation of 2D electrons was eliminated in our experiments, in contrast with Refs. 1–3; a hole at an acceptor is a neutral formation. That the overlap of the wave functions of the 2D electron and hole is small follows from the anomalously long radiative times for recombination with a photoexcited hole at an acceptor:[13] ~ 270 ns. Effects associated with relaxation to a spin equilibrium can be observed during either steady-state or pulsed excitation. The inset in Fig. 1 shows magnetooscillations of the rate of recombination with electrons from an excited quantum-well subband populated in a nonequilibrium way.[14] These oscillations are known to reflect oscillations in the time scale of the radiationless relaxation of nonequilibrium electrons induced by the optical pumping to the level of an excited subband.[15,16] The lifetime at the maxima of the oscillations amounts to several tens of nanoseconds, while that at the minima[15] is ~ 5 ns. The recombination spectra were measured in magnetic field corresponding to a filling factor of 4 in the ground subband, and also at the maximum and minimum of oscillations in the intensity of recombination from the excited subband. These spectra are shown in the σ^- and σ^+ polarizations in Fig. 1; the spectra in the σ^+ polarization have been normalized to the polarization of the holes. Also shown in this figure is the scheme of optical transitions between electron and hole levels. By taking account of the difference between the matrix elements ($\sim 1{:}3$) for transitions

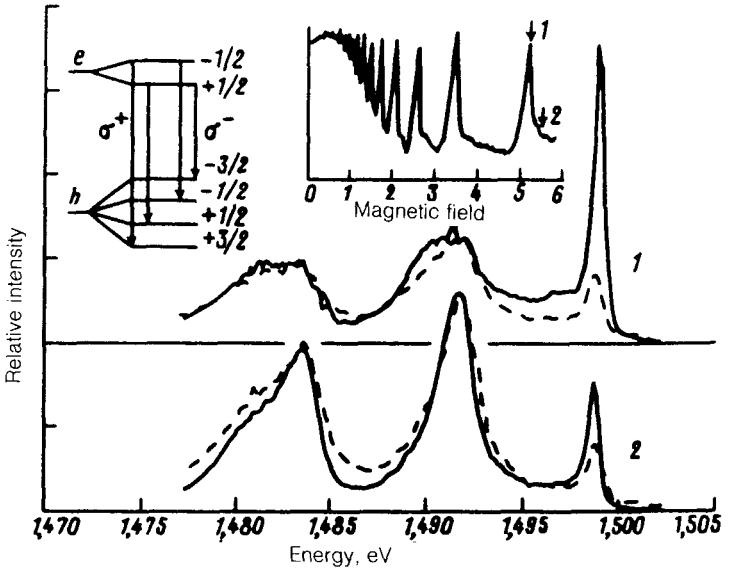

FIG. 1. Recombination spectra of 2D electrons measured at the maximum (1) and minimum (2) of magnetooscillations in the intensity of electron recombination from an excited quantum-well subband (as shown in the inset). The spectra are shown in pairs in the σ^- and σ^+ polarizations. The spectra in the σ^+ polarization are normalized to the spin polarization of the holes. The reason why the pairs of spectra coincide for the lower subband is that its zeroth and first Landau levels (those at the far left) are completely filled in these magnetic fields. The difference in the normalized spectra for recombination with electrons of an excited subband (the level at the far right) stems from electron spin polarization.

with light ($\pm 1/2$) and heavy ($\pm 3/2$) holes, one can show that the polarization of the luminescence will depend on the electron polarization even in strong magnetic fields. This dependence is approximately linear: $\rho_1 = \rho_0 + \alpha\rho_e$. At a filling factor of 4 the electrons of the lower subband cannot be spin-polarized; all the spin sublevels of the zeroth and first Landau levels are completely and thus identically filled. The polarization of the radiation from the recombination with electrons of the ground subband reflects only the polarization of the spins of the hole system, so the σ^+ and σ^- spectra have been multiplied by factors which bring them into coincidence. The difference between the normalized spectra in the region of recombination with nonequilibrium electrons of the excited subband is a manifestation of their spin polarization. We can clearly see a difference between the situations corresponding to the longer lifetime of the nonequilibrium electrons (a large spin polarization) and the shorter lifetime (a small spin polarization). Since the orientation of the spins of excited electrons caused by the quantizing magnetic field cannot be completed in the first few nanoseconds, the orientation is brought to completion in the next few tens of nanoseconds.

3. The spin relaxation time was determined accurately by a time-correlated photon counting method during optical pumping by picosecond laser pulses at $\lambda = 6050$ Å. The intense laser pulse was defocused to a spot ~ 3 mm in size at the sample (the average power density was ~ 50 mW/cm^2). To eliminate effects due to irregularities

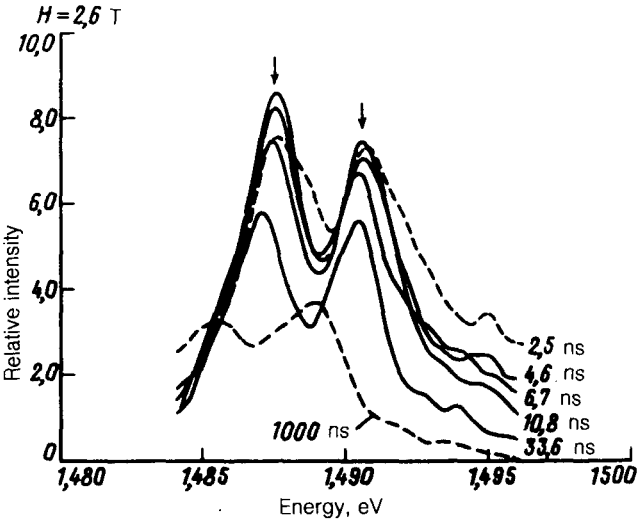

FIG. 2. Recombination spectra measured at various times after the arrival of a laser pulse. The changes in the positions of the Landau levels in the spectrum are due to a change in the density of 2D electrons.

in the illuminated system, we measured the luminescence from a central part of the laser spot about 0.1 mm in size. Further measurements were based on the decrease in the density of 2D electrons during photoexcitation by photons with an energy greater than the bandgap in AlGaAs (Ref. 17). As was mentioned in Ref. 13, the following steady-state time dependence of the electron density can be observed if the pump pulses are sufficiently intense: an abrupt change in the density immediately after the arrival of the laser pulse and a slow subsequent relaxation which continues until the next pulse arrives, in 1.2 μs. Figure 2 shows recombination luminescence spectra recorded at various times after the arrival of the laser pulse. There is clearly a difference in the positions of the Landau levels in the spectra before the arrival of the laser pulse (at a delay of 1 μs) and immediately after the arrival (2.5 ns). The changes in the positions of the levels over the first 40 ns are extremely small, so that we were able to study relatively brief spin-relaxation processes after an abrupt change in the density of 2D electrons. The filling factor in the 2D gas before and after the arrival of the laser pulse can be determined accurately (within $\sim$5%) from the complete extinction of the line corresponding to recombination with electrons of the first Landau level. The magnetic field corresponds to a filling factor $\nu=2$ for the 2D electrons in this case. The filling factor in an arbitrary field can be calculated easily: $\nu=2H/H_0$. We selected a laser power level and a magnetic field ($H=2.6$ T) such that there was an abrupt change from a filling factor of 4 to a filling factor of 3 immediately after the arrival of the laser pulse (the corresponding spectra are also shown in Fig. 2). The 2D electrons, which were initially not spin-polarized (at a filling factor of 4, all the spin sublevels are completely filled), abruptly found themselves in a situation in which a polarization should arise in the upper Landau level because of Zeeman splitting. We intend to determine the rise time of this polarization in future measurements.

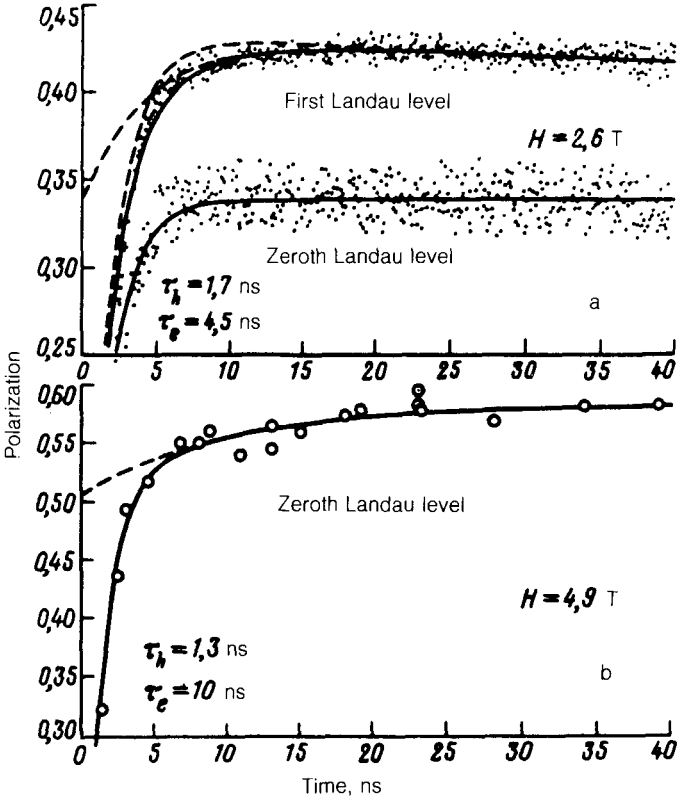

FIG. 3. Time evolution of the degree of circular polarization of the radiative recombination of 2D electrons after the arrival of a laser pulse. Two processes can be observed: a relaxation, over a time τ_h, of a spin polarization of photoexcited holes and a relaxation, over a time τ_e, of a spin polarization of the electron system. a: An abrupt change from a filling factor of 4 to a filling factor of 3 has occurred; the polarization was determined at fixed spectral positions (Fig. 2). b: An abrupt change from a filling factor of 2 to 1 has occurred, and the polarization was determined from the spectra. The relaxation of the difference between the polarizations of the luminescence from fully and partially filled levels results from a relaxation of the electron spin polarization.

4. Figure 3a shows the polarization $\rho=(\sigma_- -\sigma_+)/(\sigma_- +\sigma_+)$ of the zeroth and first Landau levels in the situation described above. As we mentioned earlier, the difference between the curves stems from a difference in the spin polarizations of the recombining electrons. The rise time of the spin polarization of a hole at an acceptor, $t_h \sim 1.7$ ns, has been studied previously.[18] It was found from the behavior of the degree of polarization $\rho=\rho_0(1-e^{-t/t_h})$ of the zeroth Landau level, in which there is absolutely no electron polarization. It can be seen from Fig. 3a that the behavior of the polarization of the first Landau level cannot be described by a single exponential function, in contrast with the behavior of the zeroth level. The difference $\Delta\rho=\rho_1-\rho_0=\gamma(1-e^{-t/t_e})$, associated with the spin polarization of the electrons of the first Landau level, is shown by the dashed line in Fig. 3 for a time constant

t_e=4.5 ns. Here $\gamma(t)$ is a slow decay reflecting the increase in the density of 2D electrons. By the time the next pulse arrives, after 1.2 μs, the quantity $\gamma(t)$ has dropped to zero, since the filling factor becomes equal to 4, and both spin sublevels of the first Landau level are completely and identically filled. A corresponding procedure was carried out for a higher laser power and in a magnetic field H=4.9 T. In this case the filling factor changed abruptly from 2 to 1. Since the zeroth Landau level (all the electrons are in this level under these conditions) changes spectral position too rapidly at such laser power levels, we determined the degree of polarization from the spectra. The set of points found in this manner is shown in Fig. 3b. The spin relaxation time turns out to be 10 ± 3 ns.

5. The rise times found here for the spin polarization of the excited system of 2D electrons, $\sim$5 ns in a field of 2.6 T, are very different from the results of previously published optical studies.[1-3] In those other studies, the leading role was played by exchange interactions of the spins of a photoexcited electron and a photoexcited hole. Those times are the same as the times extracted indirectly from transport measurements.[6,7] The slowing of the relaxation in strong magnetic fields for lower-index Landau levels may correspond to a mechanism of spin relaxation through a spin-orbit interaction, as can be seen from Eq. (2), in which the spin–orbit corrections appear with a weight of $(n+1)/\hbar\omega_c$.

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