Spin Conservation of Photocreated Carriers in Quantum Wells in High Magnetic Fields: A New Spectroscopic Tool

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The spin relaxation of photocreated carriers in GaAs/GaAlAs quantum wells in high magnetic fields is investigated by luminescence experiments under selective optical excitation. The complete quantization of the two-dimensional energy structure in high magnetic fields implies a simultaneous exchange of energy and momentum for spin relaxation and makes spin-conserving relaxation processes, in both thermalization and recombination, much faster than spin-flip processes. This results in a new spectroscopic tool to identify magnetoexcitonic states with the same electronic spin orientation.

The full quantization of the energy spectrum of a two-dimensional (2D) system, like a quantum well, in high magnetic fields makes inefficient all the elastic processes which are usually invoked to explain the rapid spin relaxation in bulk materials[1-3] or in 2D systems without magnetic field.[4] As the density of states between spin-split Landau levels is, in principle, zero, the relaxation of photocreated carriers between spin-split levels can only take place by a change in energy *and* magnetic moment. The inhibition of the elastic spin-flip processes may thus lead to a substantial increase of the spin-relaxation time.[5] We have observed, by interband photoluminescence and photoluminescence-excitation experiments on GaAs/GaAlAs quantum wells in high magnetic fields, that (i) the relaxation time of the magnetic moment of the electron is much longer than the recombination time and (ii) the magnetic moment is conserved in both recombination and during thermalization from the excited states. Although similar phenomena are well known for free atoms, where the states are localized and discrete, they have never been observed for band states in solids. In a 2D system in a magnetic field these band states are fully quantized (like in atoms), but they are at the same time delocalized (like in solids). Our experimental results show that this full quantization alone leads to a substantial increase in the spin-relaxation time.

A direct consequence of these long spin-relaxation times is the possibility to perform optical-pumping experiments, i.e., to populate electronic states with preferential spin orientation using selective optical excitation. This technique is a powerful spectroscopic tool in atomic

physics[6] and, in general, for localized centers which possess spin-relaxation times longer than the radiative lifetime. Optical-pumping experiments on band states in bulk semiconductors are instead less easy because of the rather rapid spin relaxation of photocreated carriers;[1] typical values of the spin-relaxation time at low temperatures are often shorter than the recombination time.[2,3]

We have performed such experiments on high-quality GaAs undoped quantum wells (4.5 to 20 nm thick) sandwiched between $Ga_{1-x}Al_xAs$ barriers ($x = 0.26$–0.43). The photoluminescence and photoluminescence-excitation spectra have been measured at 1.7 K and in magnetic fields up to 20 T in the Faraday configuration. Both exciting and emitted light were analyzed with different helicity (σ^+ and σ^-) of the circular polarization. Dye lasers (with LD700 or DCM as dyes) pumped by ion lasers (Kr^+ or Ar^+) are used as excitation sources.

Because of size quantization, the energy spectrum of GaAs quantum wells consists of several subbands and, at zero field, the optical spectra are characterized by a series of excitonic lines, one for each subband. The lowest state, i.e., the heavy-hole exciton ground state, is observed in low-temperature luminescence experiments. In excitation spectroscopy the intensity of this luminescence is measured as a function of the energy of the exciting light. Thus, if all excited carriers relax to the ground state and then recombine radiatively, this excitation spectrum reflects the absorption. In a magnetic field, the heavy-hole ground state splits into two components.[7,8] At sufficiently high magnetic fields, both
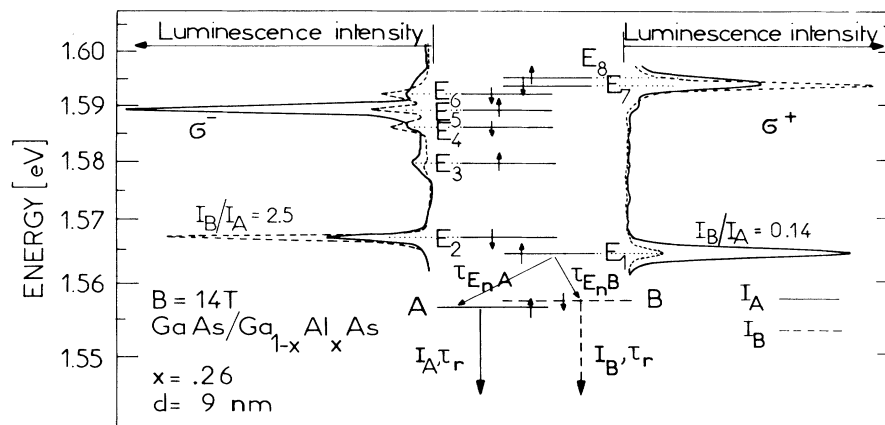
FIG. 1. Luminescence intensity as a function of the energy of the σ^- (left) and σ^+ (right) polarized exciting light at 14 T. A and B are the energies position of the two components of the heavy-hole spin-split ground state. The luminescence of A (B) is σ^- (σ^+) polarized. Solid lines: excitation spectra taken by measuring the intensity of A; dashed lines: intensity of B. The relevant relaxation processes are shown in the central part. The small arrows label the orientation of the conduction-band magnetic moment involved in the transition (see also Fig. 2). Note that in σ^-, the peaks corresponding to E_3 and E_5 are more pronounced in I_A, and E_2, E_4, and E_6 in I_B. Since the exciting light is absorbed in the same excited states (all peaks occur at the same energy in both I_A and I_B) this means that the states with electron spin-up orientation (E_3 and E_5) relax preferentially to A and those with spin-down (E_2, E_4, and E_6) to B. Similarly, in σ^+, E_1 and E_8 relax preferentially to A and E_7 to B.

these components, labeled A and B, are seen in luminescence spectra. Interestingly enough the higher-energy component B is also visible in emission, even though the thermal energy ($kT = 0.13$ meV) is much less than the splitting between A and B (e.g., 1.5 meV for a 9-nm quantum well at 18 T) and even if the excitation power is very low (less than 0.1 mW/cm^2), which excludes heating. Hence an appreciable number of carriers do not relax from the higher-lying state of the doublet to the lower one but recombine directly with emission of light. The intensity ratio between these two luminescence peaks depends strongly on the energy and helicity of the exciting light, i.e., on which excited state is actually pumped. This effect is illustrated in Fig. 1, where representative experimental results are shown. For a given polarization of the exciting light, say σ^+, the peak positions in the excitation spectra are obviously the same, independent of whether luminescence intensity (I_A) from the lower component (A) or luminescence intensity (I_B) from the higher component (B) of the ground state is measured. However, the relative intensity of the peaks in the two excitation spectra is drastically different. For σ^- polarization a similar behavior, but not involving other excited states, is observed. Since, for a fixed polarization, the exciting light is absorbed in the same states, the difference in the excitation spectra for A and B must be attributed to different relaxation rates. Therefore, we conclude that carriers excited in some higher magnetoexcitonic level, labeled E_n, relax preferentially to one of the two components (A or B) of the ground state. All the states among which the relaxation is efficient must have a common property: We will show that all these

states possess the same orientation of the conduction-band spin.

Magnetoexcitons in quantum wells have been studied theoretically[8,9] and experimentally.[7,10] Although the resulting optical spectra (see, e.g., Figs. 1 and 2) are rather complicated, it has been shown that, at high fields, the main features of experimental data can be well described with calculated transitions between the free electron and hole Landau levels.[11] The energy-level structure of the electrons in a magnetic field can be seen as a simple Landau-level ladder, with each Landau level split into two well-defined spin components. The hole Landau levels are instead more complicated[11] and, due to spin-orbit coupling, do not have a simple spin-up or spin-down character. In the left panel of Fig. 2 we show the field dependence of the same magnetoexcitonic states which, for $B = 14$ T, are shown in Fig. 1. In the right panel of Fig. 2 the calculated free-electron-hole interband transition energies at high fields[11] are shown. The theoretical curves are not identical to the experimental ones because excitonic effects are not included. The Coulomb interaction lowers the heavy- and light-hole ground-state energies and changes the field dependence of the transitions; it affects the lower states more than the higher ones.[8,9] However, the qualitative agreement between theory and experiment is good enough to identify each transition. Thus, we can attribute to each level E_n an electron-spin orientation, corresponding to the conduction-band Landau level involved (see Fig. 2). In particular, the peaks A and B, observed in luminescence, are seen to possess the opposite spin orientation. In Fig. 1 we have labeled all the observed states with their electron-spin orienta-
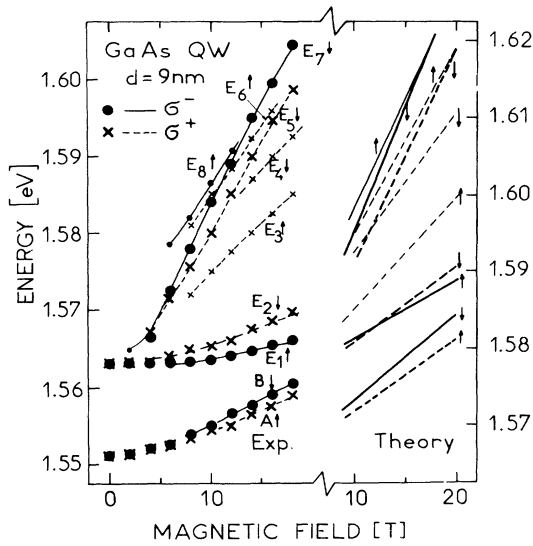
FIG. 2. Magnetic field dependence of the peaks in the excitation spectra in the two circular polarizations; the size of the symbols indicates the strength of the transition (left panel). Calculated field dependence of interband transitions in both polarizations; the thickness of the lines is proportional to the calculated intensity of the transitions. Notice the shift of the energy scale (right panel). The spin magnetic moment of the conduction band involved in the transitions (indicated by arrows) is assigned by comparing theory and experiments.

tion, and it can be seen that, in a given excitation spectrum, those states are enhanced which possess the same conduction-band spin orientation in both absorption and emission. Therefore, the experimental results, together with the theoretical assignment, show that the inhibition of spin-flip processes is responsible both for the non-thermalization between the spin-split ground states (peaks A and B) as well as for the preferential relaxation from a higher excited state to that ground state which has the same conduction band.

The previous analysis, based on a comparison of experiments with theory, shows that the inhibition of the spin flip of conduction electrons only dominates the thermalization processes. The hole thermalization apparently does not show such a bottleneck. This observation can be understood in terms of the very different nature of the conduction- and valence-band states. Electronic states in a field, in wide-gap materials like GaAs, are characterized by their magnetic moment (electron-spin quantum number) and their orbital moment (Landau-level quantum number). Conversely, the valence-band states are characterized by the magnetic sublevels of the total angular momentum J, as a consequence of spin-orbit coupling. Therefore, any nonmagnetic interaction like that with phonons, impurities, and defects may induce transitions between hole levels. Instead, only magnetic processes, like hyperfine interaction, exchange interaction,

lack of inversion symmetry, and the nonpure character of the conduction-band states (none of them being very efficient[3,12]), can be invoked for the relaxation of conduction-band spin. In a 2D system in high magnetic fields such processes will be even less efficient since, contrary to the 3D case, no elastic processes can occur because there are no states available between spin-split levels due to the absence of energy versus momentum dispersion in the direction of the field.

To estimate the relevant relaxation times we analyze the intensity ratio $R = I_B/I_A$ between the luminescence peaks A and B using rate equations. A simplified scheme of the processes involved is shown in Fig. 1. The relevant processes can be described as follows. Carriers are created optically in some state E_n. From this state they relax toward the states A and B, with characteristic relaxation times τ_{E_nA} and τ_{E_nB}. The deviation from unity of the ratio between these two relaxation times indicates preferential relaxation, i.e., spin-conserving processes. When the carriers have relaxed down to the lowest-energy state A, they can only recombine radiatively, giving rise to the luminescence I_A. Carriers which relax down to state B can instead either relax further to A, which implies an electron spin flip, or recombine directly, giving rise to the luminescence I_B. The intensity ratio $R = I_B/I_A$ will, therefore, also depend on the ratio of the recombination lifetime τ_r (assumed to be the same for A and B) to the relaxation time between A and B, τ_{BA}. With this model we can write

$$R = \frac{I_B}{I_A} = \frac{\tau_{BA}/\tau_r}{(\tau_{E_nB}/\tau_{E_nA})(1 + \tau_{BA}/\tau_r) + 1}. \qquad (1)$$

Since R depends on two ratios of characteristic times, it cannot be derived from a single measurement involving only one excited state. However, the same equation is valid for different excited states E_n, with different values of τ_{E_nB}/τ_{E_nA}, but with the same value for τ_{BA}/τ_r since this latter ratio has no relation with the excited state. We note that, as follows from Eq. (1), for any E_n,

$$\tau_{BA}/\tau_r \geq R, \qquad (2)$$

the equal sign occurring when τ_{E_nB}/τ_{E_nA} is zero, i.e., when all carriers excited in E_n relax to B only.

In the data shown in Fig. 1, we observe $R = 2.5$ for the excited state E_2; we thus conclude that $\tau_{BA}/\tau_r \geq 2.5$, implying that the spin-relaxation time between the two components of the spin-split heavy-hole ground states is about 3 times longer than the recombination lifetime. Taking $\tau_{BA}/\tau_r = 2.5$, also when $R = 0.14$—which corresponds to the excitation to the E_1 state (see Fig. 1)—we obtain $\tau_{E_1B}/\tau_{E_1A} = 5$, which means that the spin-conserving relaxation for this state is at least 5 times more efficient than the spin-flip process. In the rate-equation model presented here, it is assumed that the degeneracy of the levels is large compared to the number of excited

2411

particles (low excitation power). Upon increasing the power, the values of R are changed, as we have indeed observed. The results we presented here were, therefore, all taken at low excitation power (< 0.1 W/cm^2).

The inhibition of spin-flip processes is strong enough to create a population inversion between spin-split levels using selective optical excitation (optical pumping) as shown in Fig. 1. More intense luminescence from the higher-energy component of the ground state is observed when one of the E_2, E_4, E_6, or E_7 excited states is pumped.

The possibility to distinguish between transitions allowed in either left- or right-circularly polarized light is an important tool for the identification of transitions in magneto-optical experiments. Our results yield an additional spectroscopical possibility, namely, to distinguish between states with different electron-spin orientation. This is indeed a complementary tool to polarization, since there are cases where light is absorbed in one polarization and gives rise to a strong luminescence of opposite polarization; for instance, as shown in Fig. 1, absorption in the state E_2 of σ^- radiation gives rise to a strong luminescence of B in σ^+ radiation, and similarly for E_1 and A.

From a study of this effect as a function of magnetic field and exciting energy and for different samples, it is found that inhibition of spin flip is more pronounced (i) for higher fields (for the sample of Fig. 1, but at 18 T, we found $\tau_{AB}/\tau_r \geq 4$ and $\tau_{E_2B}/\tau_{E_2A} = 8$, while no spin-conserving processes are observed at vanishing magnetic field), (ii) for lower-lying excited states, and (iii) for better samples (i.e., with sharper peaks). The common denominator of these trends is the reduced overlap in the density of states between spin-split levels. We then conclude that the conservation of magnetic moment is the result of the discrete nature of the levels in 2D systems in a magnetic field.

In conclusion, we have shown experimentally that energy relaxation of the photocreated carriers in quantum wells in a quantizing magnetic field takes place preferentially with conservation of the electron magnetic moment. We believe that these long spin-relaxation times are a consequence of the discrete nature of the spin-split Landau levels (zero density of states between the levels), which makes these states analogous to energy levels in atoms where similar effects are observed. In combination with excitation spectroscopy, these long spin-relaxation lifetimes can be used as a tool to distinguish between states which have the same and states which have the opposite electron-spin magnetic moment with respect to each of the two components of the spin-split ground state. Our results show that excitation spectra at high fields may not necessarily reflect the absorption intensity (oscillator strengths) of a transition. The proper

oscillator strength can be obtained from the sum of the luminescence intensity from both spin-split components of the ground state. The strong tendency to the conservation of the electron magnetic moment is shown to be sufficient to create a population inversion between the two spin-split components of the ground state. We hope that our results may contribute to a better understanding of spin-dependent properties of 2D systems.

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