Observation of extremely long electron-spin-relaxation times in *p*-type δ-doped GaAs/$Al_xGa_{1-x}As$ double heterostructures

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Spin-relaxation dynamics of electrons have been studied in *p*-type δ-doped GaAs:Be/$Al_xGa_{1-x}As$ double heterostructures by time-resolved photoluminescence polarization measurements. A relaxation time of $\simeq 20$ ns has been observed which is two orders of magnitude longer than that reported for corresponding acceptor concentrations in homogeneously doped GaAs. This enhancement, which arises from a drastic reduction of the electron-hole wave-function overlap, demonstrates that electron-hole scattering with a simultaneous exchange interaction dominates spin relaxation at low temperatures.

Polarization-dependent measurements of optical interband transitions have been proven to be a powerful method to study the symmetry of electron and hole wave functions as well as the dynamics of carrier and spin relaxation in semiconductors.[1–5] In homogeneously *p*-type doped semiconductors and in *p*-modulation-doped quantum wells, it is possible to study spin-polarized conduction electrons and their spin-relaxation kinetics by luminescence polarization spectroscopy.[1–8] Low-temperature spin-relaxation times in *p*-type bulk-doped GaAs are ≤ 200 ps for acceptor concentrations $\geq 10^{19}$ cm^{-3} (Ref. 8). Based on measured hole density and temperature dependences it was suggested that electron-hole scattering with a simultaneous exchange interaction [the so-called Bir-Aronov-Pikus (BAP) mechanism[9]] dominates electron-spin relaxation at low temperatures and high doping levels. In contrast, the Elliott-Yafet[6,7] and D'yakonov-Perel' (DP) (Ref. 10) mechanisms should determine the relaxation rates at high temperatures and/or low hole concentrations.[6–8] An enhancement of the spin-relaxation rate by a factor of 3 to 4, as compared to bulk material with comparable hole densities, has been observed in *p*-type quantum wells. This has been attributed to the enhanced electron-hole overlap and has been taken as additional evidence for the BAP mechanism being the dominant one at low temperatures.[4] An unambiguous proof of the BAP mechanism, however, would result from experiments on a sample structure where the electron-hole overlap is drastically reduced. Such structures should show an enhancement of the spin-relaxation time by at least an order of magnitude.

In this paper we present time-resolved circularly polarized photoluminescence (PL) experiments on δ-doped GaAs:Be/$Al_xGa_{1-x}As$ double heterostructures, in which the photogenerated electrons are spatially separated from the holes, while still maintaining sufficient wave-function overlap for efficient radiative recombination to probe the spin polarization of the electrons. This separation and the resulting reduction in electron-hole interaction strength lead to electron-spin-relaxation times of about 20 ns in the present sample. This value is two orders of magnitude larger than that reported for comparable acceptor concentrations in bulk-doped GaAs.[8] The drastic enhancement in the spin-relaxation time proves directly that electron-hole scattering (the BAP mechanism) is the dominant process for electron-spin relaxation at low temperatures. Thus the present heterostructure allows us to tune the spin-relaxation time to values which are impossible in homogeneously doped material or in *p*-type modulation-doped quantum wells, and therefore provides a new model system for future studies of electron-spin dynamics in semiconductors.

The samples used for the present study consist of a 60-nm wide GaAs layer δ doped in its center with Be. The data shown in the following were taken from a particular sample with a Be concentration of 8×10^{12} cm^{-2} but similar results were obtained for other dopant concentrations. This GaAs layer is sandwiched between a 20-nm thick $Al_{0.33}Ga_{0.67}As$ barrier on the substrate side and a corresponding 10-nm thick barrier on the surface side. Figure 1 displays the self-consistently calculated valence and conduction-band potential profile together with electron and hole subbands and the corresponding probability densities.[11] A spread of the dopant atoms along the growth direction of 2 nm was taken in accord with secondary ion mass spectroscopic measurements,[11] which results in an effective volume *p*-type dopant concentration of 4×10^{19} cm^{-3}. For the given dopant density of 8×10^{12} cm^{-2} the first heavy-hole (hh1) and the first light-hole (lh1) subband are populated. Excitation for time-resolved measurements was performed with a mode-locked Ti:sapphire laser producing 1.0 ps pulses. For detection a synchroscan streak camera coupled to a single monochromator was used with an overall time resolution of ≤ 20 ps. Polarized (I^+) and depolarized (I^-) luminescence spectra were recorded with the detected light having the same and the opposite circular polarization of the pump light, respectively. The pump polarization was kept fixed.

Figure 2 shows low-temperature cw PL spectra excited at 1.65 eV. In the total intensity spectrum ($I^+ + I^-$) plotted in Fig. 2(a) the emission from the two-

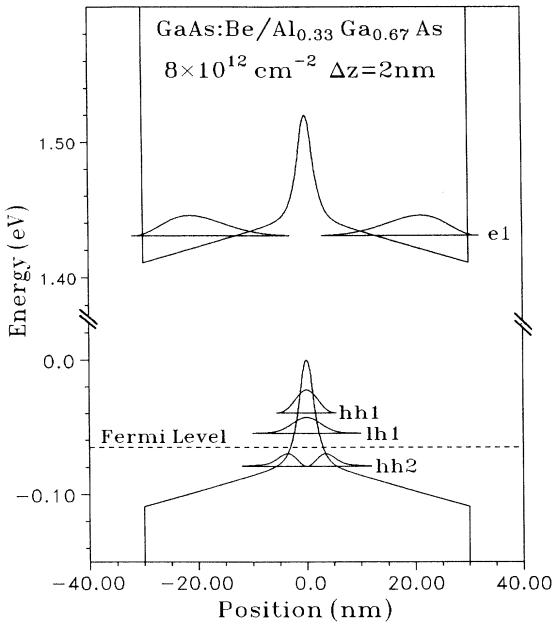

FIG. 1. Self-consistent potential profile for the quasi-two-dimensional hole gas in a δ-doped $Al_{0.33}Ga_{0.67}As/GaAs:Be/Al_{0.33}Ga_{0.67}As$ double heterostructure. An 8×10^{12} cm^{-2} Be concentration is taken with a 2-nm dopant spread. Subband energies and probability densities at $k_{\parallel}=0$ are also shown. hh and lh denote heavy-hole and light-hole subbands, respectively; el denotes the first electron subband.

dimensional hole gas (2DHG) appears as a broadband centered around 1.47 eV. The peaks marked by asterisks at 1.513 and 1.493 eV arise from, respectively, bound-exciton and donor-to-acceptor pair recombination in the GaAs buffer layer. The difference spectrum (I^+-I^-) [Fig. 2(b)] shows a maximum at 1.46 eV and a minimum at 1.48 eV. Based on the polarization selection rules[1,6–9] these extrema are assigned to recombination involving the hh1 and the lh1 subband, respectively. The spacing of the calculated subband energies, marked in Fig. 2(b), is in good agreement with the difference between the experimental transition energies. Figure 2(c) displays the degree of circular polarization $P=(I^+-I^-)/(I^++I^-)$ plotted versus the emitted photon energy. For the hh1 transition the maximum value of P is 0.26.

As the incident light is absorbed essentially in the almost bulklike intrinsic regions of the 60-nm wide GaAs layer above and below the doping spike, which is evident from PL excitation spectroscopy,[11,12] excitation with, e.g., right circularly polarized light generates heavy holes with spin $-\frac{3}{2}$ and spin $-\frac{1}{2}$ electrons as well as spin $-\frac{1}{2}$ light holes and spin $+\frac{1}{2}$ electrons. The probabilities for these transitions are 3:1, leading to a majority of spin $-\frac{1}{2}$ electrons with an initial degree of polarization of 0.5.[1,10] Since the number of photogenerated holes is small compared to the doping level ($\simeq 1\%$), the spin polarization of these holes is irrelevant and therefore neglected. The radiative recombination of the spin-polarized electrons involves quantized hole states with the heavy-hole–light-hole degeneracy removed. Therefore the maximum polarization of the emitted light is also 0.5. Recombination with holes in the hh1 subband appears as a maximum in the difference spectrum I^+-I^-. Recombination with light holes results in the emission of photons with circular polarization opposite to that from heavy-hole recombination and appears therefore as a minimum in the difference spectrum [Fig. 2(b)].

Time-resolved PL spectra provide direct information on the electron recombination and spin dynamics. Figure 3(a) shows a series of total intensity spectra (I^++I^-). With increasing time delay the 2DHG emission shifts to lower energies. After the exciting laser pulse, photogenerated carriers reduce the spatial modulation of the band-edge energies. This causes an initial blue shift of the luminescence due to the spatial separation of the photocreated electrons and the 2DHG. The subsequent red shift results from the decrease of the photogenerated carrier concentration. The long-lived emission at 1.49 eV is due to donor-to-acceptor recombination in the GaAs buffer layer. Figure 3(b) displays the temporal evolution

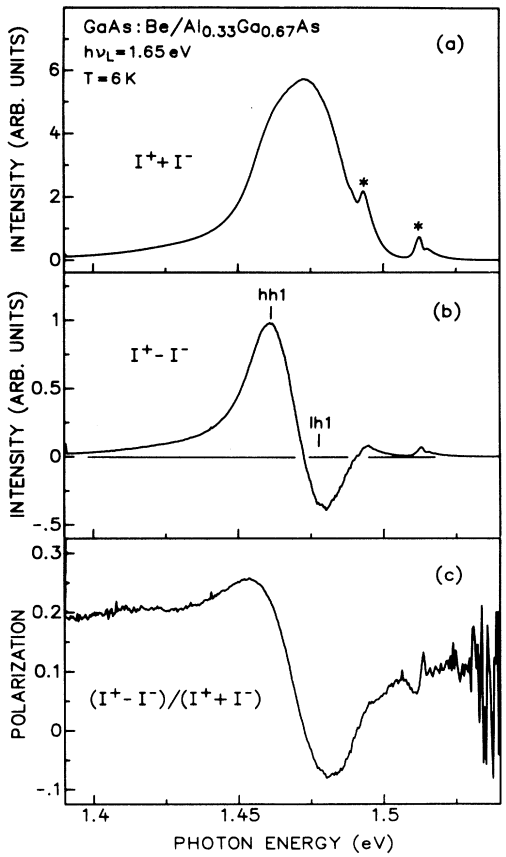

FIG. 2. Low-temperature PL spectra recorded at 6 K with excitation at 1.65 eV. (a) Total intensity spectrum I^++I^-, (b) difference spectrum I^+-I^-, and (c) polarization spectrum $(I^+-I^-)/(I^++I^-)$. Subband energies, calculated for a dopant spread of 2 nm, are marked in (b) with the position of the first heavy-hole level fixed at the peak of the hh1 emission band.

of the circular polarization P, showing again the low-energy shift of both the hh1 and lh1 transition with increasing time delay.

From the time-resolved total intensity spectra [Fig. 3(a)] and polarization spectra [Fig. 3(b)] the recombination time τ_{rec} and the electron-spin-relaxation time τ_{sp} can be extracted. At 6 K the recombination shows a biexponential behavior with an initial time constant $\tau_{rec}=2$ ns followed by a somewhat slower decay time of 3.5 ns. For the spin-relaxation time, defined by[2] $P=P_0\exp(-2t/\tau_{sp})$, a significantly larger value of $\tau_{sp}=20\pm4$ ns is obtained. The initial degree of polarization P_0 for the hh1 transition is 0.3 This value is lower than the maximum achievable polarization of 0.5, which indicates that some spin relaxation takes place while the photogenerated electrons relax in energy and get spatially separated from the photoexcited holes. Taking into account the reduction factor[1] $\tau_{sp}/(\tau_{rec}+\tau_{sp})$, which relates the initial polarization P_0 to the time-averaged one P_{av} observed in the cw measurement, one thus obtains an expected averaged value of $P_{av}=0.27$ in good agreement with the measured cw polarization of $P_{av}=0.26$ [see Fig. 2(c)].

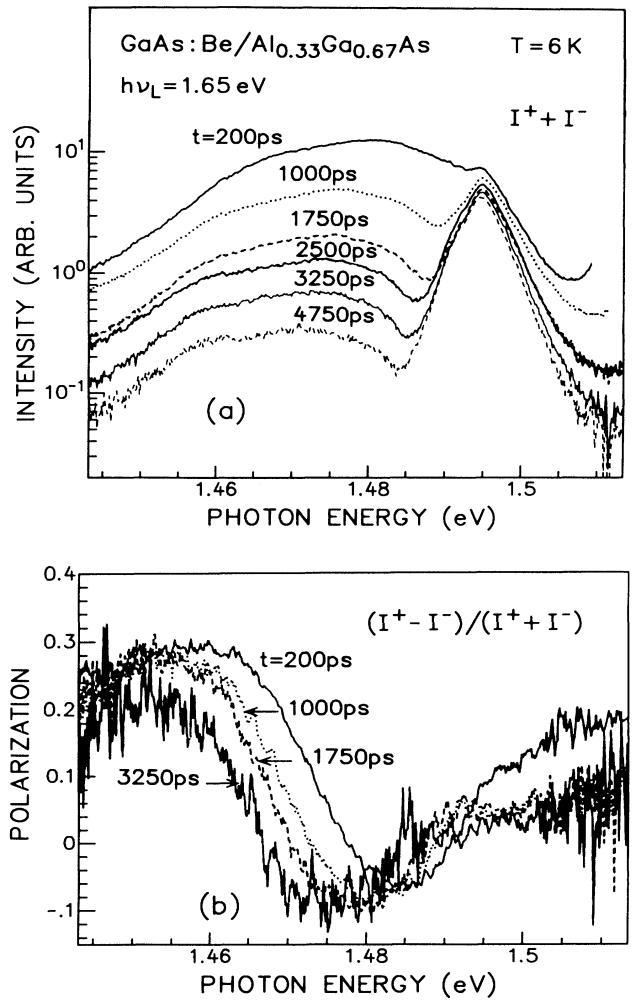

FIG. 3. Time-resolved PL spectra for various time delays t recorded at 6 K with excitation at 1.65 eV. (a) Total intensity spectra $I^+ + I^-$ and (b) polarization spectra $(I^+ - I^-)/(I^+ + I^-)$.

The present spin-relaxation time $\tau_{sp}=20$ ns is two orders of magnitude longer than the relaxation time of $\simeq 200$ ps found in homogeneously p-type doped GaAs with a comparable acceptor concentration of 2.8×10^{19} cm^{-3} (Ref. 8). From work on homogeneously doped samples, it has been concluded that the BAP mechanism is relevant for electron-spin relaxation in degenerate p-type GaAs at low temperatures.[7,8] For this mechanism the spin-relaxation rate $1/\tau_{sp}$ in a degenerate semiconductor is proportional to $(\Delta_{exch})^2$ (Refs. 7–9). Δ_{exch} is the exchange splitting of the exciton ground state and is given by[13]

$$\Delta_{exch}=\Delta_{exch}(3D)|\Phi^{2D}(0)|^2/|\Phi^{3D}(0)|^2\int[X_e(z)X_h(z)]^2dz\ , \tag{1}$$

where $\Delta_{exch}(3D)$ is the exchange splitting of the 3D exciton, $\Phi^{2D}(0)$ and $\Phi^{3D}(0)$ are, respectively, the 2D and 3D exciton wave function amplitudes at zero electron-hole relative coordinate, and $X_e(z)$ and $X_h(z)$ are the electron and hole subband envelope wave functions, respectively. Ignoring quantum size effects, we can write $|\Phi^{2D}(0)|^2/|\Phi^{3D}(0)|^2=8a_0$, where a_0 is the 3D exciton radius (15 nm for GaAs). Based on the scheme presented in Fig. 1 we have calculated the electron-hole wave function overlap in Eq. (1). From these calculations we expect a two orders of magnitude decrease in $1/\tau_{sp}$, in agreement with the experiment. This confirms that the drastic increase in electron-spin-relaxation time upon reduction of the electron-hole overlap can be taken as an unambiguous proof that electron-hole scattering with a simultaneous exchange interaction (the BAP mechanism) is the dominant electron-spin-relaxation process in p-type doped GaAs at low temperatures. Although the electrons are confined in the nearly intrinsic regions of the present sample structure, the interaction with the hole gas is still important as both the electrons and the holes are part of a quantum confined system.

In Fig. 4 the temperature dependences of τ_{rec} and τ_{sp} are shown. The temperature dependence of τ_{sp} fits to a power law with an exponent of -0.6. This is apparently in agreement with various data obtained on homogeneously doped GaAs with hole concentrations $\geq 4\times10^{18}$ cm^{-3} (Refs. 6–8) for $T<100$ K. For a degenerate hole gas a $T^{-3/2}$ or possibly a T^{-2} (if the hole Fermi velocity exceeds that of the electrons) dependence is expected for the BAP mechanism[9] and has indeed been confirmed for $T>100$ K (Refs. 7,8). Aronov, Pikus, and Titkov[7] have shown that if the energy relaxation time of electrons is much longer than the electron lifetime, and hence there is an incomplete thermalization of the electrons in the conduction band, a $T^{-1/2}$ dependence can be expected. Zerrouati *et al.*[8] have also explained this deviation from a $T^{-3/2}$ dependence by a discrepancy between the lattice temperature and the higher electron temperature. For the present case with an electron lifetime of 2 ns and

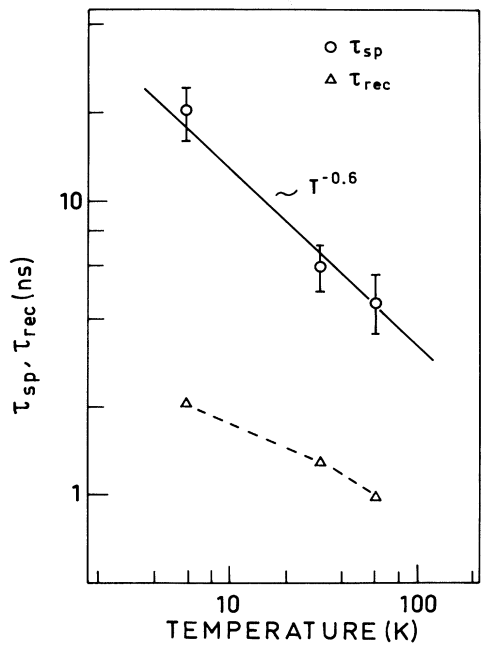

FIG. 4. Temperature dependence of the electron-spin-relaxation time τ_{sp} and luminescence recombination time τ_{rec}. The full line indicates a $T^{-0.6}$ power law, the dashed lines are drawn as a guide to the eye.

time-resolved measurements with an average excitation density of about 10 W/cm^2 an electron temperature significantly higher than the lattice temperature is not expected. But certainly an adequate formulation of the various electron-spin-relaxation mechanisms for the 2D case would be required to come to a final conclusion on this point.

When neglecting excitonic effects, electron-spin-relaxation times > 10 ns are expected only for hole concentrations below 10^{16} cm^{-3} (Refs. 8 and 9). However, it has been shown that exciton formation leads to a drastic reduction of the spin-relaxation time due to the enhanced electron-hole overlap.[3,4,14] Taking a hole concentration of about 2×10^{16} cm^{-3} as a limit where exciton formation becomes important, it follows that spin-relaxation times are always ≤ 4ns in homogeneously doped GaAs or in p-type doped quantum wells. In the present sample, in contrast, where exciton formation is suppressed by the built-in electric fields of the structure (Fig. 1), at least one order of magnitude longer relaxation times can be realized. It is therefore ideally suited for the investigation of electron-spin kinetics in semiconductors, even in the limit of weak electron-hole interaction with the 2DHG necessary for probing the electron-spin distribution.

In conclusion, we have investigated electron-spin relaxation in δ-doped GaAs:Be/$Al_xGa_{1-x}As$ double heterostructures by time-resolved photoluminescence spectroscopy. An extremely long electron-spin-relaxation time of $\simeq 20$ ns has been obtained which is two orders of magnitude longer than that found in homogeneously doped GaAs for comparable acceptor concentrations. This increase in relaxation time is due to the drastically reduced electron-hole interaction because of the spatial separation of electrons and holes in the double heterostructure. The present experiment provides an unambiguous proof that electron-hole scattering (the BAP mechanism) governs electron-spin relaxation in GaAs at low temperatures. It also demonstrates the potential of the present sample structure as a model system for future experiments on electron-spin dynamics in semiconductors due to its unique possibility for the variation of the electron-hole interaction strength.

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[12]This picture is further confirmed by the measured dependence of the cw circular polarization P_{av} on the excitation photon energy. For photon energies ranging from 1.55 to 1.83 eV only a modest decrease from $P_{av}=0.36$ to 0.23 is observed, whereas for photon energies exceeding the $E_0+\Delta_0$ band-gap energy P_{av} decreases rapidly and becomes zero in agreement with experiments on homogeneously doped p-type GaAs (Refs. 6 and 10).
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