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Electrical spin injection from ferromagnetic MnAs metal layers into GaAs

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The spin injection into GaAs has been studied for the ferromagnetic metal MnAs. Evidence for preferential minority-spin injection is obtained from the circular polarization of the electroluminescence in GaAs/(In,Ga)As light-emitting diodes (LED). The spin-injection efficiency of 6% at the MnAs/GaAs interface is estimated on the basis of spin-relaxation times extracted from time-resolved photoluminescence measurements. This efficiency, as well as the preferential spin orientation, resembles very much the injection behavior found for epitaxial Fe layers. The results do not depend on the azimuthal orientation of the epitaxial MnAs injection layer.

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The realization of spintronic devices relies on the ability to inject a spin-polarized current into a semiconductor,[1] which still remains to be a challenge. Electrical spin injection from ferromagnetic metals into semiconductors has been demonstrated only recently using Fe as the spin-injector material.[2,3] The ferromagnetic metal MnAs is a further promising choice since it has a high Curie temperature ($T_C \approx 40\,°\mathrm{C}$) and a relatively small coercive field (50 Oe).[4,5] However, spin injection from (Ga,Mn)As has so far been demonstrated only for diluted material with low Mn content, which is semiconducting (mostly *p*-type) and becomes ferromagnetic only at low temperature ($T_C < 110$ K).[6,7] The hybrid system MnAs/GaAs can be prepared by molecular beam epitaxy (MBE) with interfaces of extremely high quality and different azimuthal orientations.[8–10] These different orientations can be utilized to study the possible influence of the symmetry matching between the conduction-band wave functions in MnAs and GaAs.

We present here experimental evidence for spin injection from MnAs into the semiconductor GaAs obtained by analyzing the polarization degree of the electroluminescence (EL) spectra from *n-i-p* (In,Ga)As/GaAs light-emitting diodes (LED). The actual spin-injection efficiency at the MnAs/GaAs interface is estimated by taking into account the spin-relaxation times measured by time-resolved photoluminescence (TRPL) spectroscopy. We show that this efficiency as well as the preferential spin orientation resemble the results obtained with Fe injection layers.

The LED device structures were grown by molecular beam epitaxy (MBE) on *p*-GaAs(001) substrates with 500-nm-thick *p*-GaAs buffer layers.[2] The active region consists of two 4-nm-thick (In,Ga)As QWs separated by a 10-nm-thick GaAs barrier sandwiched between two 50-nm-thick undoped GaAs spacer layers. On top of this intrinsic region, a 70-nm-thick *n*-GaAs layer was grown. The MnAs injection layers were deposited on the *n*-GaAs layer in a separate MBE chamber. After sample transfer through air, the GaAs surface was first treated by thermal annealing (cleaning) followed by the regrowth of 5-nm-thick GaAs. The subsequent growth of the 50-nm-thick MnAs layer was carried out at a temperature of about 250 °C and a growth rate of 20 nm/h.[9] By varying the As coverage of the GaAs surface, two epitaxial orientations of MnAs with respect to the substrate have been realized: Type A with MnAs$[0001] \parallel$GaAs$[1\bar{1}0]$ and type B with MnAs$[11\bar{2}0] \parallel$GaAs$[1\bar{1}0]$. In both cases, the surface orientation is given by MnAs$(\bar{1}100) \parallel$GaAs(001). In the case of the B orientation, a reduced As coverage leads to an inclined MnAs c-axis: MnAs$(\bar{1}101) \parallel$GaAs(001). For reference purposes, one part of each wafer was not capped with a MnAs layer, but subsequently covered with a nonmagnetic AuGe alloy layer. After metal electrode deposition, the epitaxial wafers were processed into 50-μm-wide mesa stripes defined by dry chemical etching and cleaved into pieces of 240 to 670 μm length. For TRPL experiments, reference QW samples without p^+- and n^+-GaAs layers were grown under the same conditions.

For the EL measurements, the LED was placed into a superconducting magnet system with the temperature controlled in a continuous flow cryostat. The experiments were done in the Faraday geometry, i.e., with the magnetic field parallel to the light-propagation direction.[2] The EL signal was collected from the wafer backside, dispersed in a single spectrograph, and detected by a charge-coupled device (CCD) array. The circular polarization was analyzed by passing the EL light through a photoelastic modulator[11] (PEM) and a linear polarizer with its optical axis rotated by 45° with respect to the optical axis of the PEM. The LED was operated with current pulses (0.4 μsec pulse width at a frequency of 42 kHz) locked to the maximum or minimum phase shifts of the PEM. The degree of circular polarization is determined by $P=(I_+ - I_-)/(I_+ + I_-)$, where the right (left) circularly polarized component I_+ (I_-) is obtained for EL generation pulses locked to $+\lambda/4$ ($-\lambda/4$) phase shifts of the PEM.

TRPL measurements were performed using a synchro-scan streak camera system in conjunction with a Ti:sapphire laser emitting 150 fs pulses at photon energies between 1.56 and 1.70 eV (repetition rate 76 MHz). The average excitation power density was about 10 Wcm^{-2}. The luminescence was dispersed by a single monochromator and focused onto the photocathode of the streak tube. The streak images were recorded by a cooled CCD array. The nominal temporal resolution of the synchro-scan system is 2 ps. The samples were mounted on the cold finger of a He flow cryostat. An initial spin polarization of photo-excited carriers was created by pump pulses, which were circularly polarized by means of a quarter-wave plate.[12,13] The emitted PL light was analyzed

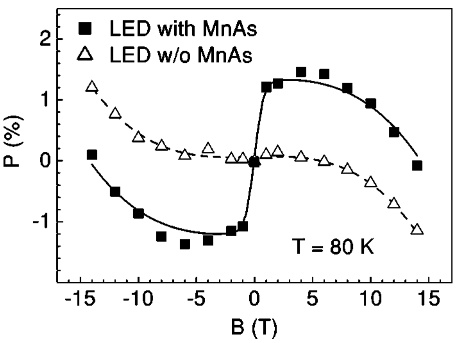

FIG. 1. Circular polarization degree P as a function of external magnetic field measured at 80 K from LEDs with (full squares) and without (open triangles) a MnAs injection layer. The solid and dashed lines are guides to the eye.

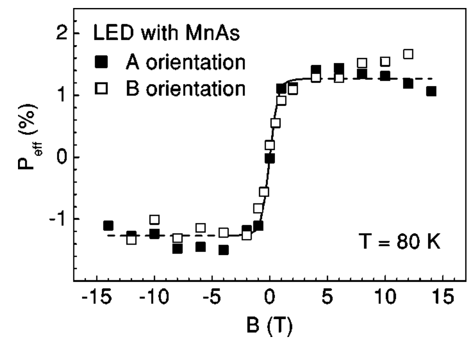

FIG. 2. Effective polarization degree P_{eff} as a function of external magnetic field measured at 80 K from LEDs with MnAs injection layers of A orientation (full squares) and B orientation (open squares). The contribution due to the Zeeman splitting in the LED layers has been subtracted. The magnetization curve of a thin MnAs layer is shown for comparison in arbitrary units (solid line; dashed line is the continuation as a guide to the eye).

into its right (I_+) and left (I_-) circularly polarized components using a second quarter-wave plate. The total carrier lifetime τ_R (which at low temperatures corresponds to the radiative recombination time) and the spin relaxation time τ_S have been obtained by fitting single exponential decay curves to the total PL intensity ($I_+ + I_-$) and $(I_+ - I_-)/(I_+ + I_-)$, respectively.

The EL spectra of the LEDs with MnAs injection layer reveal one peak at 1.44 eV ($T = 80$ K) in accordance with the design of the active region. Since the EL peak width of 22 meV is smaller than the heavy-hole/light-hole splitting, only heavy-hole transitions contribute to the emission. For a given polarization degree of injected polarized electrons and unpolarized heavy holes, the absolute value of P is identical to the spin polarization of the radiatively recombining electrons.[14] As a proof for spin injection, we require that the polarization degree P as a function of an external magnetic field B follows the out-of-plane magnetization curve of the MnAs injection layer obtained independently using a superconducting quantum interference device (SQUID). As shown in Fig. 1, the measured polarization degree P (full squares) fulfills this requirement for magnetic fields $|B| < 2$ T indicating spin injection.

However, for $|B| > 2$ T the polarization curve does not exhibit the saturation observed for the out-of-plane magnetization (solid line in Fig. 2). To explain this deviation, we have to consider that the effect of spin injection is expected to be superimposed on the influence of electron and hole thermalization in the semiconductor layers of the LED structure due to Zeeman splitting of spin-up and spin-down states in large external magnetic fields. For the present LED, the (In,Ga)As QWs in the active region were grown with an In content of 0.1, whereas for previous experiments with Fe injection layers an In content of 0.2 was chosen.[2] It turns out that the influence of spin alignment due to the Zeeman splitting is more pronounced in the LED structure with the lower In content. This finding becomes evident from the magnetic-field dependence of P for the reference LED without a MnAs injection layer (open triangles in Fig. 1). This behavior is probably due to the specific spin-relaxation times and g factors in the QWs with lower In content, i.e., less strain in the (In,Ga)As layers.[15,16] The polarization of the EL light is influenced by the thermalization of both electrons and heavy holes into the energetically lowest Zeeman states. The nonlinear dependence on the magnetic field can then be explained by assuming Fermi-Dirac distributions for electrons and holes in quasiequilibrium and relatively large g factors in the (In,Ga)As QWs.

The possible contribution of spin injection to the measured magnetic-field dependence of P might therefore be masked by the superposition with the effect of the spin alignment due to the Zeeman splitting. Consequently, we subtract the polarization curve obtained for the reference LED (P_{ref}) from that of the LED with a MnAs injection layer (P_{tot}). Indeed, this effective net polarization $P_{eff} = (P_{tot} - P_{ref})$ displayed in Fig. 2 follows the MnAs out-of-plane magnetization curve obtained by SQUID measurements. This result provides evidence for successful spin injection from the ferromagnetic metal MnAs with a circular polarization degree of about 1.5% in the saturation range.

If the symmetry matching between the conduction-band wave functions in MnAs and GaAs plays an important role, the spin injection into GaAs might depend on the orientation of the MnAs injection layer.[17] However, no significant difference has been found between MnAs layers with orientation of type A (full squares in Fig. 2) and type B (open squares in Fig. 2). A comparison with our previous results for Fe injection layers reveals that electrons are injected with a polarization degree of the same sign. In accordance with the results of Ref. 3, the preferential polarization is identified to be spin-down, i.e., electrons are injected with the minority spin direction from both Fe and MnAs injection layers.[18]

The observed spin injection from a ferromagnetic metal into a semiconductor can be explained by tunneling through the Schottky barrier at the interface. Such a tunneling process can lead to an enhanced spin-injection efficiency, since it is not affected by the resistance mismatch.[19] Recent theoretical work on the basis of a ballistic picture includes the possibility of minority spin injection.[20] This model demonstrates that from the quantum mechanic point of view, there is an intrinsic contact resistance due to momentum mismatch, which determines the spin-injection rate across a hybrid junction. Such a spin-dependent intrinsic contact resis-

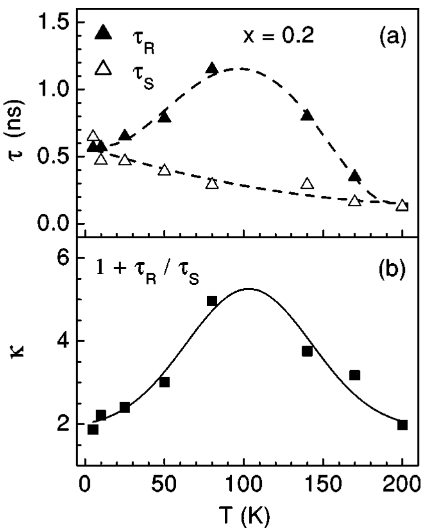

FIG. 3. (a) Measured carrier lifetime τ_R (full triangles) as well as spin-relaxation time τ_S (open triangles) and (b) the correction factor $\kappa=1+\tau_R/\tau_S$ (full squares) as a function of sample temperature for an (In,Ga)As QW structure with an In content of 0.2. The solid and dashed lines are guides to the eye.

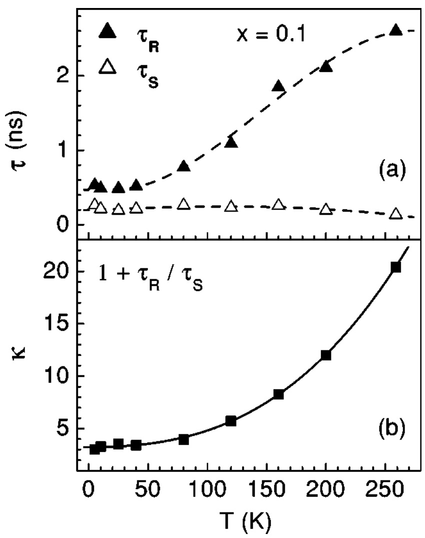

FIG. 4. (a) Measured carrier lifetime τ_R (full triangles) as well as spin-relaxation time τ_S (open triangles) and (b) the correction factor $\kappa=1+\tau_R/\tau_S$ (full squares) as a function of sample temperature for an (In,Ga)As QW structure with an In content of 0.1. The solid and dashed lines are guides to the eye.

tance overcomes the resistance mismatch obstacle and results in a negative spin-injection rate at the percentage level for a typical hybrid junction. The observed preferential injection of minority carriers from both MnAs and Fe into GaAs with comparable efficiency might be understood in such a picture. However, according to that theory, the sign of the spin-injection rate depends not only on the interface scattering potential (tunneling barrier and the bias), but also on the Fermi energy of GaAs. Concerning the contribution of tunneling processes, we did not succeed in extracting related information from I-V curves of our LED structures, since they consist of two diodes in series. During the EL measurements, the (In,Ga)As/GaAs *n-i-p* diode is operated in forward direction, while at the same time the Schottky diode at the metal/GaAs interface is reversely biased.

As mentioned above, the polarization degree *P* is identical to the spin polarization of the recombining electrons in the (In,Ga)As QWs. However, if the spin relaxation time τ_S in the QWs[12,13] is considerably shorter than the carrier lifetime τ_R, the measured value of P_{eff} represents a lower limit for the spin polarization of the injected electrons. From a simple rate equation model, it follows that the spin-injection efficiency for our experiments would then be

$$\eta=(1+\tau_R/\tau_S)\times P_{eff}, \tag{1}$$

which defines a correction factor $\kappa=(1+\tau_R/\tau_S)$. Here, we neglect the possible spin relaxation during the transport from the Fe/GaAs or MnAs/GaAs interface to the (In,Ga)As QWs.

The crucial time constants τ_R and τ_S obtained by TRPL spectroscopy are displayed in Fig. 3(a) for a reference sample containing (In,Ga)As QWs with an In content of 0.2 as in the LED with an Fe injection layer studied in Ref. 2. Whereas τ_S (open triangles) shows a monotonic decrease with increasing temperature, τ_R (full triangles) exhibits a pronounced maximum at about 100 K. The decrease of τ_R for temperatures above 100 K is attributed to an increasing contribution of nonradiative recombination. For the correction factor κ displayed in Fig. 3(b), we find $2<\kappa<3$ at temperatures below 50 K and above 150 K. Consequently, with a correction factor $\kappa\approx2.5$, we deduce an effective spin injection efficiency of $\eta\approx5\%$ for our previous experiments with an Fe injection layer.[2]

For (In,Ga)As QWs with an In content of 0.1 like in the present LEDs with a MnAs injection layer, the contribution of nonradiative recombination is found to be weaker, which manifests itself in a monotonic increase of τ_R over the whole temperature range as shown in Fig. 4(a). At the same time, τ_S reveals a relatively weak temperature dependence. Consequently, the resulting correction factor κ, displayed in Fig. 4(b), reaches values as large as 20 at temperatures around 250 K. For our experiments with a MnAs injection layer at 80 K, we obtain $\kappa\approx4$, which leads to a spin-injection efficiency of $\eta\approx6\%$.

As shown above, our measurements reveal that the time constants τ_R and τ_S depend strongly on the In content in the (In,Ga)As QWs. Thus the accuracy of the chosen correction factors κ depends crucially on the uncertainty in the In content of the QWs in the LED structures. Nevertheless, the temperature dependence of κ should reflect itself in the measured spin injection efficiencies. However, the successful demonstration of spin injection was possible only for temperatures where κ is small. This condition was fulfilled for the Fe-LED (In content of 0.2)[2] at $T\leqslant30$ K and $T\geqslant200$ K, where κ has a similar value of about 2.5 (cf. Fig. 3). Larger values of κ lead to an EL polarization $P=\eta/\kappa$ which becomes too small to be measured. For the MnAs-LED (In content of 0.1), spin injection could only be measured for temperatures in the range $20\leqslant T\leqslant80$ K with similar EL polarization values. Assuming a constant spin-injection efficiency η, this finding would be in accordance with the small temperature dependence of κ below $T \approx 80$ K (cf. Fig. 4). Finally, it should be noted that the temperature dependencies of τ_S do not agree with the assumption that only one of the possible DP (D'Yakono-Perel), EY (Elliot-Yafet), or BAP (Bir-Aharonov-Pikus) mechanisms dominates.[13]

In conclusion, spin injection from the ferromagnetic metal MnAs into the semiconductor GaAs is demonstrated with an efficiency of about 6%, which makes MnAs another promising candidate for the use in spintronics devices. The circular polarization degree of the electroluminescence signal from (In,Ga)As/GaAs LEDs with ferromagnetic injection layers is found to be a lower limit for the spin-injection efficiency, since the spin-relaxation times in the active regions of the LEDs are shorter than the carrier recombination times. Different azimuthal orientations of the epitaxial MnAs layer lead to the same injection behavior. Concerning the spin injection efficiency as well as the preferential orientation of injected spins, the ferromagnetic metals MnAs and Fe exhibit a very similar behavior.

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