APPLIED PHYSICS LETTERS VOLUME 85, NUMBER 16 18 OCTOBER 2004

Spin injection from Fe_3Si into GaAs

A. Kawaharazuka,[a] M. Ramsteiner, J. Herfort, H.-P. Schönherr, H. Kostial, and K. H. Ploog
Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5–7, 10117 Berlin, Germany

(Received 25 February 2004; accepted 17 August 2004)

We demonstrate room-temperature spin injection from the epitaxially grown ferromagnetic metal Fe_3Si into the semiconductor GaAs. The injection efficiency is comparable to values previously obtained for the Fe/GaAs and MnAs/GaAs hybrid systems using the emission of similar (In,Ga)As/GaAs light-emitting diodes for the detection of spin polarization. The temperature dependence of the detected polarization is explained by taking into account spin relaxation inside the semiconductor device. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1807014]

Electrical spin injection into semiconductors at room temperature has become a crucial issue for the realization of spin-electronic devices.[1,2] Hybrid structures consisting of a ferromagnetic metal layer on a semiconductor (FM/SC) are suitable for room-temperature operation. Actually, spin injection from epitaxially grown FM layers into SC has been demonstrated for Fe on GaAs,[3] Fe on (Al,Ga) As,[4] and MnAs on GaAs.[5] The growth temperature for Fe on (Al,Ga) As has to be kept low (close to room temperature) to prevent the formation of interfacial compounds, which are unfavorable for spin injection.[6] The drawback of MnAs is the Curie temperature of 40 °C, which is rather close to room temperature.[7,8] Regarding processing aspects as well as the heat dissipation in semiconductor devices, it is desirable to establish a FM/SC hybrid system for spin injection, which exhibits a high thermal stability. In this respect, Fe_3Si,[9,10] which is ferromagnetic up to 840 K[11] and almost lattice matched to GaAs, is a promising material for spin injection. Very recently, epitaxial growth of Fe_3Si on GaAs(001) was achieved with high crystalline and interfacial perfection at a growth temperature of $T_G = 200\,°C$, which is much higher than the one for Fe growth.[12] Moreover, Fe_3Si can be regarded as a Heusler alloy Fe_2FeSi and is therefore a candidate for being half-metallic,[13–15] which should be advantageous concerning the spin-injection efficiency.

In this letter, we demonstrate room-temperature spin injection for the Fe_3Si/GaAs hybrid system. Spin-polarized electrons are electrically injected from the ferromagnetic Fe_3Si layer and detected via the circular polarization of the electroluminescence (EL) intensity emitted by an *n-i-p* light-emitting diode (LED) on the semiconductor side.

The LED device structure consisting of a Fe_3Si injection layer on a $In_{0.1}Ga_{0.9}As$/GaAs LED structure (see Ref. 3) was grown by molecular-beam epitaxy (MBE). From the bottom, a 500 nm-thick *p*-GaAs buffer layer, a 50-nm-thick undoped GaAs barrier, a 4-nm-thick undoped $In_{0.1}Ga_{0.9}As$ quantum well (QW), a 50-nm-thick undoped GaAs barrier, and a 70-nm-thick *n*-GaAs layer were successively grown on the *p*-type GaAs(001) substrate. The doping concentration is $2\times10^{18}\ cm^{-3}$ for both *n* and *p*-GaAs layers. After growing the LED structure, the substrate was transferred into the As-free metal deposition chamber through ultrahigh vacuum, and then the 35-nm-thick Fe_3Si layer was grown on the *n*-GaAs layer at $T_G = 200\,°C$ For details about the growth of the Fe_3Si layer, we refer to Ref. 12. The epitaxial wafer was processed into LED devices with mesas of 230 μm diameter patterned by dry etching. We also prepared a reference sample from the same wafer by removing the Fe_3Si layer by wet etching and depositing the nonferromagnetic metal Ti. We measured the EL intensity from the back side of the LED structures placed in a superconducting magnet system. The degree of circular polarization of the EL was analyzed by using a photoelastic modulator. Details of the EL measurements are given in Ref. 3.

The low-temperature (25 K) EL spectrum reveals one peak at 1.427 eV due to the recombination of electrons with heavy holes. The linewidth of 11 meV is clearly smaller than the estimated heavy-hole/light-hole splitting of about 45 meV. The degree of circular polarization is determined by $P = (I_+ - I_-)/(I_+ + I_-)$, where $I_+(I_-)$ denotes the intensity of right (left) circularly polarized light integrated over the width determined by the full width at half maximum of the spectrum. The absolute value of the polarization degree P is identical to the spin polarization of the radiatively recombining electrons, if the heavy holes are assumed to be unpolarized. The magnetic-field dependence of the circular polarization degree obtained at 25 K is shown in Fig. 1 (open squares). Successful spin injection must be accompanied by a circular polarization degree that follows the out-of-plane magnetization curve of Fe_3Si (solid line) independently obtained by using superconducting-quantum-interference-device (SQUID) magnetometry. Indeed, the steep increase of the polarization degree in the lower magnetic field region

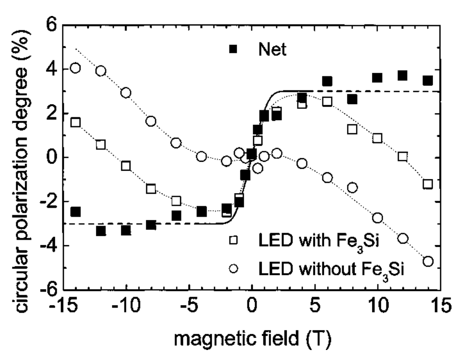

FIG. 1. Circular polarization degree as a function of external magnetic field at 25 K from LEDs with (open squares) and without (open circles) Fe_3Si. Dotted lines are guides to the eyes. The net polarization (closed squares) is plotted together with the magnetization curve measured by SQUID in arbitrary units (solid line, dashed line is a continuation as guide to the eyes).

[a]Electronic mail: kawaha@pdi-berlin.de

($|B|<2$ T) indicates the injection of spin-polarized electrons from Fe_3Si. However, the decrease at higher magnetic fields ($|B|>4$ T) does not correspond to the saturation observed for the out-of-plane magnetization. This discrepancy is due to the contribution of the polarization, which originates from carrier thermalization into the lowest of the Zeeman-split spin-up and spin-down states in the present semiconductor structure (see Ref. 5 for QWs with an In content of 0.1). The contribution of the Zeeman effect to the EL polarization is superimposed on the polarization induced by spin injection at the $Fe_3Si/GaAs$ interface and becomes more pronounced in the higher magnetic-field region. The bare Zeeman-induced polarization in the semiconductor structure can be determined from the EL of the reference LED without a ferromagnetic injection layer. The obtained degree of circular polarization P_{ref} (open circles) decreases monotonously as expected from the contribution due to the Zeeman effect. In order to eliminate the Zeeman contribution, the polarization curve obtained from the reference sample P_{ref} has been subtracted from the total polarization P_{tot} obtained from the sample with Fe_3Si injection layer. This subtraction eliminates the Zeeman contribution for small EL polarizations as can be seen by a first-order approximation of the theoretical expression given in Ref. 16 [Eq. 5]. The resulting net polarization $P_{net}=P_{tot}-P_{ref}$ (closed squares) indeed follows the Fe_3Si out-of-plane magnetization curve obtained from SQUID measurements. This result provides evidence for the successful spin injection from the ferromagnetic metal Fe_3Si into the semiconductor GaAs. The net polarization degree in the saturation region is about 3%, which is comparable to the values previously reported for the injection from Fe and MnAs into similar semiconductor LED structures.[3,5] The similar polarization degrees obtained for the three different FM/SC hybrid systems indicates that the spin polarization is strongly influenced by spin relaxation processes inside the semiconductor device. Therefore, in order to determine the injection mechanism and efficiency at the FM/SC interface, it is essential to understand the spin relaxation processes.

In order to study the influence of the spin relaxation in the semiconductor structure, we investigated the spin injection at higher temperatures. The total and net EL polarizations are shown in Figs. 2(a) and 2(b), respectively. The magnetic-field dependencies of the net polarization degree obtained at 150 and 295 K [Fig. 2(b)] clearly exhibit the signature of spin injection. However, the saturation value decreases with increasing temperature. This observation can be explained by taking into account the carrier lifetime τ_R of the recombining electrons and the spin-relaxation time τ_S in the QW. From a simple rate equation model,[16] the EL polarization degree P is obtained as

$$P=\frac{\eta+r(1-\nu)}{1+r(1+\nu)}, \quad (1)$$

where η denotes the injection efficiency at the $Fe_3Si/GaAs$ interface, $r=\tau_R/2\tau_S$ the ratio between carrier lifetime and spin-relaxation time, and ν the ratio of the electron population between upper and lower Zeeman levels given by Fermi–Dirac distribution functions. In the following we will discuss the dependence of P on the ratio r, which characterizes the semiconductor structure by spin relaxation time and carrier lifetime. Due to their very fast spin relaxation, holes are assumed to be in quasi-equilibrium, i.e., the spin polarization of holes is regarded to be independent of r. Since, consequently, only the spin relaxation time of electrons enters in the ratio r, we eliminated the Zeeman contribution of holes for the sake of simplicity ($\pi=1$ in Ref. 16). When the spin-relaxation time is much longer than the carrier lifetime ($r \ll 1$), the measured EL polarization represents the spin polarization due to electrical injection: $P \approx \eta$. In contrast, when the spin-relaxation time is much shorter than the carrier lifetime ($r \gg 1$), the spin polarization in the QW approaches the quasi-equilibrium state defined by Zeeman splitting. Consequently, the contribution of the Zeeman effect to the observed polarization is significant: $P\approx(1-\nu)/(1+\nu)$.

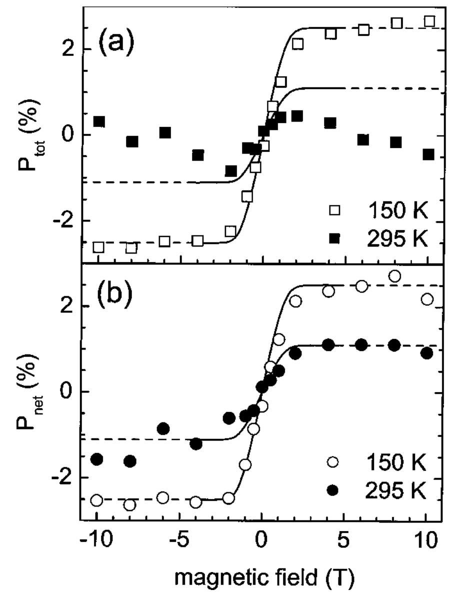

FIG. 2. (a) Total polarization P_{tot} obtained at 150 (open squares) and 295 K (closed squares). (b) Net polarization P_{net} obtained at 150 (open circles) and 295 K (closed circles). Solid lines are magnetization curves measured by SQUID normalized to the net polarization in the saturation region for each temperature. Dashed lines are continuations as guides for the eyes.

The total polarization shown in Fig. 2(a) reveals that the saturation polarization due to spin injection strongly decreases with increasing temperature, whereas the contribution of the Zeeman polarization exhibits a minimum around 150 K. Assuming a constant spin-injection efficiency η (Fe_3Si is ferromagnetic up to 840 K), the spin polarization due to electrical injection does not explicitly depend on temperature. This fact can be deduced from Eq. (1), since a vanishing Zeeman polarization ($\nu\approx 1$) results in

$$P\approx\frac{\eta}{1+2r}=\frac{\eta}{1+\tau_R/\tau_R}. \quad (2)$$

Therefore, the decreasing polarization due to spin injection can be explained only by a temperature-dependent ratio $r=\tau_R/2\tau_S$. Indeed, r has been found to strongly increase above 150 K for QWs with an In content of 0.1 (see Ref. 5). The Zeeman polarization, however, is assumed to decrease monotonously with increasing temperature, since the population ratio ν approaches unity. Without spin injection ($\eta\approx 0$), we obtain

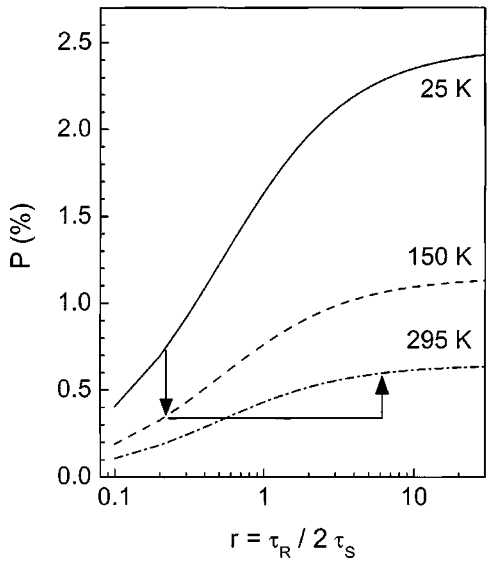

FIG. 3. Zeeman polarization calculated by Eq. (3) as a function of the ratio *r* for temperatures of 25 (solid line), 150 (dashed line), and 295 K (dash-dotted line). The changes from 25 to 150 K and 150 to 295 K are indicated by arrows. ν is calculated as described in Ref. 16.

$$P \approx \left(\frac{1}{r}\frac{1}{1+\nu}+1\right)^{-1}\frac{1-\nu}{1+\nu}. \quad (3)$$

Consequently, the increasing Zeeman contribution for temperatures above 150 K has to be explained again by a temperature-dependent ratio $r=\tau_R/2\tau_S$. The Zeeman polarization according to Eq. (3) is shown in Fig. 3 as a function of *r* for different temperatures (at a fixed magnetic field of 14 T). As reasonable parameters, we used a Zeeman splitting of 0.5 meV (observed EL peak splitting at 14 T) and a Fermi energy of 10 meV. The changes from 25 to 150 K as well as from 150 to 295 K are indicated by arrows in Fig. 3 and explain the observed behavior at least qualitatively. We assume *r* to be almost constant between 25 and 150 K and to increase by about one order of magnitude between 150 to 295 K (cf. Ref. 5). Altogether, the complete temperature dependence of the EL polarization can be explained in a consistent way by an increasing carrier lifetime τ_R (or decreasing spin-relaxation time τ_S) at temperatures above 150 K. According to Eq. (2), the net polarization represents the lower limit of the spin-injection efficiency at the $Fe_3Si/GaAs$ interface. The estimated spin-injection efficiency is about 10% at 25 K by assuming the spin-relaxation time and carrier lifetime obtained from a similar semiconductor structure (Ref. 5). For a more quantitative consideration, carrier lifetimes and spin-relaxation processes in the actual semiconductor LED device have to be studied in more detail.

Note that the observed temperature dependence of the EL polarization cannot be explained by artifacts due to a possible magnetic circular dichroism (MCD), which in principle could influence the polarization of the EL light reflected at the $Fe_3Si/GaAs$ interface.[17] Such a MCD effect would indeed be proportional to the magnetization of the ferromagnetic layer. However, the magnetic properties of the Fe_3Si injection layer do not change below room temperature as shown in Ref. 18, since the Curie temperature of Fe_3Si is much higher.

In conclusion, injection of spin-polarized electrons up to room temperature has been demonstrated in a FM/SC hybrid structure $Fe_3Si/GaAs$. The spin-injection efficiency is comparable to values obtained for the Fe/GaAs and MnAs/GaAs hybrid systems, but Fe_3Si is, in particular, promising for device applications because of its high thermal stability. The temperature dependence of the detected polarization can be consistently explained by spin-relaxation processes in the semiconductor LED.

The authors would like to thank E. Wiebicke for sample processing. This work was partly supported by the German BMBF under Contract No. 13N8255.

[1] G. A. Prinz, Phys. Today **48**, 58 (1995).
[2] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
[3] H. J. Zhu, M. Ramsteiner, H. Kostial, M. Wassermeier, H.-P. Schönherr, and K. H. Ploog, Phys. Rev. Lett. **87**, 016601 (2002).
[4] A. T. Hanbicki, B. T. Jonker, G. Itskos, G. Kioseoglou, and A. Petrou, Appl. Phys. Lett. **80**, 1240 (2002).
[5] M. Ramsteiner, H. Y. Hao, A. Kawaharazuka, H. J. Zhu, M. Kästner, R. Hey, L. Däweritz, H. T. Grahn, and K. H. Ploog, Phys. Rev. B **66**, 081304 (2002).
[6] H.-P. Schönherr, R. Nötzel, W. Q. Ma, and K. H. Ploog, J. Appl. Phys. **89**, 169 (2001).
[7] M. Tanaka, J. P. Harbison, T. Sands, T. L. Cheeks, V. G. Keramidas, and G. M. Rothberg, J. Vac. Sci. Technol. B **12**, 1091 (1994).
[8] T. Plake, M. Ramsteiner, V. M. Kaganer, B. Jenichen, M. Kästner, L. Däweritz, and K. H. Ploog, Appl. Phys. Lett. **80**, 2523 (2002).
[9] M. Hong, H. S. Chen, J. Kwo, A. R. Kortan, J. P. Mannaerts, B. E. Weir, and L. C. Feldman, J. Cryst. Growth **111**, 984 (1991).
[10] S. H. Liou, S. S. Malhotra, J. X. Shen, M. Hong, J. Kwo, H. S. Chen, and J. P. Mannaerts, J. Appl. Phys. **73**, 6766 (1993).
[11] Y. Nakamura, *Landolt-Börnstein*, New Series III/19c (Springer, Berlin, 1988), p. 26.
[12] J. Herfort, H.-P. Schönherr, and K. H. Ploog, Appl. Phys. Lett. **83**, 3912 (2003).
[13] R. A. de Groot, F. M. Mueller, P. C. van Engen, and K. H. Buschow, Phys. Rev. Lett. **50**, 2024 (1983).
[14] M. J Otto, H. Feil, R. A. van Bruggen, and C. Haas, J. Magn. Magn. Mater. **70**, 33 (1987).
[15] S. Fujii, S. Sugimura, S. Ishida, and S. Asano, J. Phys.: Condens. Matter **43**, 8583 (1990).
[16] M. Ramsteiner, J. Supercond. **16**, 661 (2003).
[17] J. Zak, E. R. Moog, C. Liu, and S. D. Bader, Phys. Rev. B **43**, 6423 (1991).
[18] J. Herfort, H.-P. Schönherr, K.-J. Friedland, and K. H. Ploog, J. Vac. Sci. Technol. B **22**, 2073 (2004).