Spin injection and relaxation in ferromagnet-semiconductor heterostructures

C. Adelmann,[1] X. Lou,[2] J. Strand,[2] C. J. Palmstrøm,[1] and P. A. Crowell[2,*]

[1]*Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, Minnesota 55455, USA*
[2]*School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455, USA*
(Received 20 December 2004; published 7 March 2005)

We present a detailed description of spin injection and detection in $Fe/Al_xGa_{1-x}As/GaAs$ heterostructures for temperatures from 2 to 295 K. Measurements of the steady-state spin polarization in the semiconductor indicate three temperature regimes for spin transport and relaxation. At temperatures below 70 K, spin-polarized electrons injected into quantum well structures form excitons, and the spin polarization in the quantum well depends strongly on the electrical bias conditions. At intermediate temperatures, the spin polarization is determined primarily by the spin-relaxation rate for free electrons in the quantum well. This process is slow relative to the excitonic spin-relaxation rate at lower temperatures and is responsible for a broad maximum in the spin polarization between 100 and 200 K. The spin injection efficiency of the $Fe/Al_xGa_{1-x}As$ Schottky barrier decreases at higher temperatures, although a steady-state spin polarization of at least 6% is observed at 295 K.

Ferromagnetic metals such as iron are natural sources of spin-polarized electrons, and semiconductors have been shown to be an ideal host for the transport and manipulation of spin. The demonstration of electrical spin injection from conventional ferromagnetic metals[1–5] has addressed the possibility of purely electronic control of spin transport in semiconductors. For example, the steady-state spin polarization electrically injected into a quantum well from an $Fe/Al_xGa_{1-x}As$ Schottky contact has been shown to be as high as 32% at 2 K.[6] Improved efficiencies have been achieved for injection through an artificial tunnel barrier,[7,8] and evidence for electrical spin injection at room temperature has been reported.[1,9] In spite of these successes, no experiment on ferromagnet-semiconductor heterostructures has addressed the properties of these devices over a wide range of temperatures and electrical bias conditions.

In this article we report on a comprehensive study of spin injection in $Fe/Al_xGa_{1-x}As/GaAs$ heterostructures from 2 to 295 K. When a shallow GaAs quantum well (QW) is used as a spin detector, three distinct temperature regimes for spin transport and relaxation are identified. Below 70 K, the bias dependence of the spin polarization in the QW is clearly influenced by excitonic effects. A pronounced peak appears in the steady-state polarization over a narrow bias range. This peak disappears rapidly with increasing temperature. Between 70 and 150 K, the spin polarization *increases* with temperature over a wide range of bias voltages. We show that the temperature dependence of the polarization signal from 2 to 150 K can be understood in terms of a crossover from excitonic to free-electron spin relaxation in the quantum well. Above 180 K, the steady-state spin polarization decreases in all heterostructures that we have studied but is at least 6% at 295 K. Measurements using a bulk GaAs spin detector indicate that the decrease at higher temperatures is due in part to a reduction in the spin injection efficiency of the Schottky barrier.

Each of the epitaxial ferromagnet-semiconductor heterostructures used for these measurements consists of a Schottky diode in series with a n-i-p junction.[1,2] The design of the Schottky tunnel barrier follows the approach of

Hanbicki *et al.*[6] Three samples will be discussed in detail in this paper. The first two, denoted I and II, use quantum wells as optical detectors. Sample I is grown on a p-type ($p=1\times10^{18}$ cm^{-3}) GaAs (100) substrate and consists of 300-nm p-GaAs ($p=1\times10^{17}$ cm^{-3}), 150-nm p-$Al_{0.1}Ga_{0.9}As$ ($p=1\times10^{16}$ cm^{-3}), 25-nm i-$Al_{0.1}Ga_{0.9}As$, 10-nm i-GaAs QW, 25-nm i-$Al_{0.1}Ga_{0.9}As$, followed by a 100-nm n-$Al_{0.1}Ga_{0.9}As$ ($n=1\times10^{16}$ cm^{-3}) drift layer. The Schottky junction is then formed by growing a $n\rightarrow n^+$ transition layer going from $n=1\times10^{16}$ cm^{-3} up to 5×10^{18} cm^{-3} over a thickness of 15 nm. This is followed by 15-nm n^+-$Al_{0.1}Ga_{0.9}As$ ($n^+=5\times10^{18}$ cm^{-3}), 5-nm Fe, and a 2.5-nm Al capping layer. The Fe and Al layers are grown at a temperature of 0 °C. Sample II is identical to sample I except for a lower doping ($p=3\times10^{15}$ cm^{-3}) in the p-$Al_{0.1}Ga_{0.9}As$ layer immediately beneath the QW structure. Sample III differs from sample I only in that the 10-nm i-GaAs QW is eliminated. The optical emission from this sample is dominated by GaAs band-edge luminescence emitted from the substrate. The samples are processed into light-emitting diodes by photolithography and wet etching and are contacted by a gold wire bond on top of each device and a diffused In contact to the p layer. After processing, each device is annealed at 250 °C in a N_2 atmosphere for 1 h. A schematic of a sample is shown in the inset of Fig. 1. Light is collected through the top of the device.

The spin detection measurements are carried out using the electroluminescence polarization (ELP) technique in the Faraday geometry.[10] Light is emitted by electrons that tunnel into the semiconductor from the Fe film and recombine with unpolarized holes from the substrate. The electroluminescence polarization $P=(I_+-I_-)/(I_++I_-)$, where I_+ and I_- are the intensities of right and left circularly polarized light, is measured as a function of magnetic field, temperature, and the bias voltage across the device. For samples I and II, the electroluminescence (EL) at low temperatures is dominated by the QW heavy-hole exciton, for which P is equal to the steady-state electron spin polarization in the QW. The polarization for these samples below 200 K is determined from
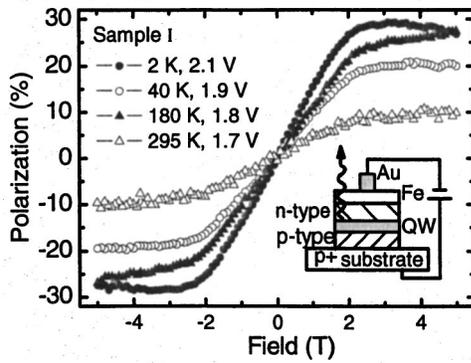
FIG. 1. Electroluminescence polarization (ELP) as a function of magnetic field for sample I at the temperatures and bias voltages indicated in the legend. A schematic of the structure for samples I and II is shown in the inset. The quantum well is omitted in sample III.
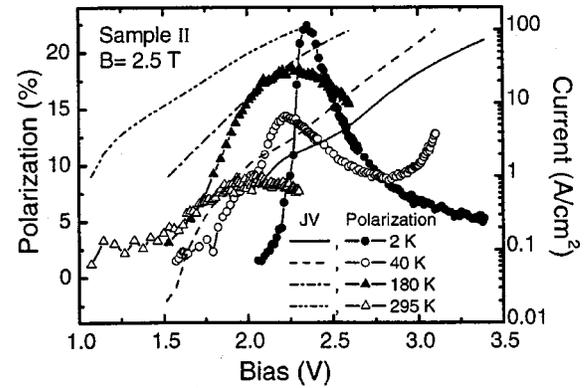

FIG. 2. The polarization (symbols) is shown as a function of the bias voltage for sample II in a field of 2.5 T at the temperatures indicated in the legend. The curves are the corresponding current-voltage characteristics.

the intensities integrated over a window 3-meV wide surrounding the heavy-hole exciton peak. At higher temperatures, the electroluminescence from samples I and II becomes dominated by recombination in the substrate and the data are windowed over a 5-meV window around the EL maximum. The EL from sample III is due to band-edge recombination in GaAs at all temperatures, and in this case P, which is determined from the spectrum integrated over a 40-meV window, is expected to be equal to half of the steady-state spin polarization in the detection region.[11] Only the raw optical polarization P will be shown in this paper. This includes small contributions from magnetoabsorption in the Fe film (less than 2% in all cases discussed here) and, at very low temperatures, the Zeeman splitting of electron and hole states in the semiconductor.

The electroluminescence polarization P for sample I is shown as a function of magnetic field in Fig. 1. The data are obtained at temperatures ranging from 2 to 295 K at the bias voltages indicated in the legend. As demonstrated in previous work,[1,2] P is approximately proportional to the magnetization of iron, which saturates at an applied field of $H=4\pi M=2.1$ T. This magnetic-field dependence is observed for all three samples. For samples I and II, a polarization of 8% at 2.5 T (6% after background subtraction) is observed even at 295 K.

A detailed picture of the spin transport properties of these devices can be obtained by measuring the optical polarization as a function of the bias voltage between the ferromagnetic electrode and the substrate. For this measurement, the magnetic field is held fixed at 2.5 T, just above the saturation field of Fe. Results at several different temperatures are shown for sample II in Fig. 2. These data show three distinguishing features. The first is the pronounced peak in the polarization as a function of bias that is observed at 2 K. Second, the maximum polarization at 180 K is *higher* than that measured at 40 K. Finally, there is a significant decrease in the polarization signal between 180 and 295 K.

It is evident from Fig. 2 that the temperature and bias dependence of the polarization signal are unusual. Complete maps of the polarization as a function of temperature and bias voltage are provided in Figs. 3(a)–3(c) for the three

samples discussed in this paper. The closed symbols in Fig. 3(d) show the polarization at the voltages along the solid curves in each of the first three panels. The data in Fig. 3(d) approximate the maximum polarization at each temperature.

It is clear from Fig. 3 that there are two regions of maximum polarization signal for QW detectors. The first is at low temperature over a narrow bias range. The second maximum occupies a much wider bias range at intermediate temperatures, between 70 and 200 K. For the bulk GaAs detector of sample III, there is a single maximum at low temperature, and the polarization signal decreases with increasing temperature above 20 K for all biases. The temperature dependence of the maximum polarization that we observe with QW detectors below 150 K agrees with recent results obtained with an artificial tunnel barrier as an injector.[8]

The polarization signal P measured in these experiments can be related to the injected spin by $P=\alpha S_i/(1+\tau_r/\tau_s)$, where S_i (maximum value=1/2) is the spin that is injected into the quantum well, τ_r is the recombination time, τ_s is the spin-relaxation time, and α is a factor determined by the optical selection rules. For the two QW samples below 200 K, the EL is dominated by the heavy-hole exciton, and $\alpha=2$. For sample III, there is no confinement and $\alpha=1$ at all temperatures.[11] We focus first on the QW samples. The fact that the polarization signal always increases with bias near threshold can be related to a decrease in τ_r with increasing bias, as would be expected due to the flattening of the bands in the n-i-p junction. The sharp peak in the response at low temperature occurs at the bias where the ratio τ_r/τ_s is smallest. This peak disappears with increasing temperature because τ_r increases, as is expected for heavy-hole excitons in shallow quantum wells[12] and verified for our QW's using Hanle effect measurements.[11]

There are, however, important features of the QW data in Fig. 3 that cannot be due simply to variations in the recombination time. As can be seen in Figs. 3(a) and 3(b), the rapid decrease in P from 10 to 70 K occurs only over a narrow bias range. For higher bias voltages, the polarization signal actually increases with temperature from 10 K up to 150 K. These unusual effects are due to the dependence of the spin-relaxation time τ_s on bias voltage and temperature.
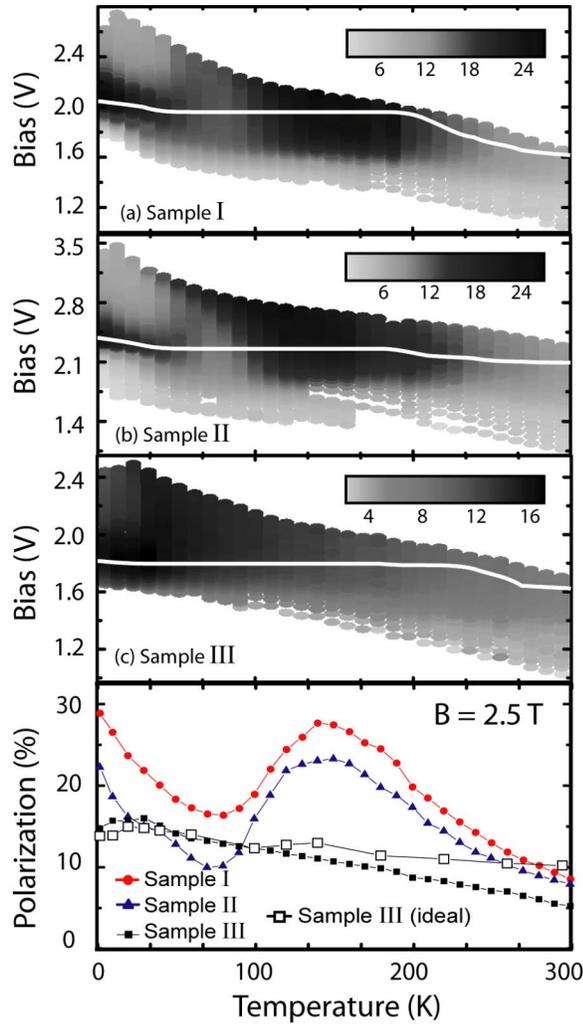
FIG. 3. (Color online) (a)–(c) The polarization measured at the electroluminescence peak is shown as a function of the temperature and bias voltage for samples I, II, and III in a field of 2.5 T. The grayscales are indicated in each panel. (d) Optical polarization (closed symbols) is shown as a function of temperature for each of the samples in this study. The data are shown at points along the solid white curves in panels (a)–(c). The maximum polarization expected for sample III for the ideal case of 100% injection efficiency is shown using open symbols. This is based on the calibration procedure described in the text.

The behavior above 100 K can be understood in terms of the D'yakonov-Perel (DP) mechanism,[13,14] in which the electron spins precess incoherently about the spin-orbit field. In a manner similar to motional narrowing, this process can be suppressed if the momentum scattering time τ_p is short enough. For the case of electron spin relaxation in quantum wells, the simplest theory predicts $\tau_s^{-1} \propto \tau_p T$,[14] and so one would expect τ_s to *increase* with temperature if the momentum scattering time decreases with temperature faster than $1/T$. This argument would appear to apply above the onset of optical-phonon scattering (≈ 70 K), and has been advanced to explain the increase in the spin-polarization signal above 70 K observed by Jiang *et al.*[8] In practice, however, the spin-relaxation time measured for free

electrons in GaAs/Al$_x$Ga$_{1-x}$As quantum wells is either constant or decreases with increasing temperature, as is found in more exact calculations.[15] Therefore, while the DP mechanism would explain the relatively weak temperature dependence between 100 and 200 K in Fig. 3, it cannot explain the pronounced temperature and bias dependence that we observe below 100 K. We have considered various models that treat consistently the dependence of the DP relaxation rate on temperature and the kinetic energy of the injected carriers. Most importantly, none of the common models for free-electron spin relaxation predicts the increase in P with temperature that is observed at high biases. As noted above, this trend starts at progressively lower temperatures (far below the onset of optical-phonon scattering) at the highest bias voltages. Clearly some other process besides the DP mechanism is contributing to the electron spin relaxation at low temperatures.

The key to understanding the low-temperature behavior observed in Figs. 2 and 3 is the formation of excitons. The electron-hole exchange interaction has been shown to enhance the spin-relaxation rate significantly compared to that observed for free electrons.[16–20] The exchange interaction can be tuned by controlling the spatial overlap of the electron and hole wave functions. For example, a factor of 5 decrease in the spin-relaxation rate in a GaAs QW at 20 K was observed by Vinatierri *et al.* as the electric field was increased from 0 to 30 kV/cm.[17] Any other parameter that decreases the electron-hole overlap, such as an increase in temperature or a decrease in the confining potential, should also suppress the excitonic contribution to the electron spin-relaxation rate.

The experimental situation is complicated by the fact that the polarization signal depends on both the recombination and spin-relaxation rates. For this reason, it is extremely difficult to model the full bias dependence at low temperatures. As noted above, the sharp decrease in the *maximum* signal with increasing T between 2 and 70 K is consistent with the observed increase in the excitonic recombination time. However, the fact that the polarization signal increases with T at higher biases is due to a crossover from excitonic spin relaxation at low temperatures to slower free-electron spin relaxation at higher temperatures. The electron-hole overlap can be suppressed either by increasing temperature or by increasing the electric field at the quantum well. An example of the latter effect can be seen in the data for sample II at 40 K in Fig. 2. P is actually increasing at the highest biases, for which the measured Stark shift indicates an electric field in the QW of order 10^4 V/cm. Although the details of the low-temperature behavior will depend on both τ_s and τ_r, the clear boundary separating the low-temperature regime from the broad maximum observed at intermediate temperatures in Figs. 3(a) and 3(b) is associated with the suppression of the electron-hole exchange.

We therefore find that the observed polarization signal in the quantum well systems below 150 K can be understood in terms of a crossover from an excitonic regime at low temperatures to the regime above 100 K in which free-electron spin relaxation by the DP mechanism applies. Above 150 K, however, the polarization signal begins to decrease at all biases. This can be attributed in part to a crossover from QW to bulk-dominated emission, but a more fundamental question

is whether the spin injection efficiency, which we have assumed to be constant for the purposes of the preceding discussion, decreases with increasing temperature. Sample III, which does not have a QW, provides an opportunity to test this assumption. In this case, recombination occurs in the p-GaAs layer at all temperatures, and excitonic effects are relatively weak. The maximum ELP at low temperatures is approximately 15%, which corresponds to a steady-state spin polarization of 30%.

The advantage of using a bulk recombination region is that the ratio τ_r/τ_s can be measured over the entire temperature range by means of the Hanle effect,[11] thus allowing us to calibrate the spin detector.[9] From the Hanle curve at each temperature, we calculate the ideal value $P=S_i/(1+\tau_r/\tau_s)$ of the optical polarization signal for the case $S_i=0.21$, which corresponds to a spin injection efficiency of 100% from Fe. The results are shown as the open symbols in Fig. 3(d). The relative agreement with the results found for sample III at low temperatures suggests that the maximum spin injection efficiency achieved with the Schottky barrier is nearly unity. At temperatures above 100 K the measured values start to drop faster than the ideal case, falling 50% below the limiting value at room temperature. This suggests that some other mechanism, such as field-assisted thermionic emission, contributes significantly to the injection current above 100 K.

Our results demonstrate that the spin injection efficiency of the $Fe/Al_xGa_{1-x}As$ Schottky barrier remains extremely high up to 150 K and is of order 50% at room temperature. The bias and temperature dependence of the steady-state polarization are attributed primarily to changes in the sensitivity of the spin detector, and steady-state spin polarizations greater than 20% can be reached over a large range of temperature and bias voltage. Our discussion has ignored the possibility that the injection efficiency itself may depend on the bias conditions, as discussed in several theoretical proposals.[21–23] These approaches might explain some of the extremely strong bias dependence observed at low temperatures, but they cannot be addressed satisfactorily until a spin detector is developed that can be calibrated over a wide range of bias conditions. The experiment discussed here has identified several of the factors that must be considered in order to achieve this goal.

*Electronic address: crowell@physics.umn.edu

[1] H. J. Zhu, M. Ramsteiner, H. Kostial, M. Wassermeier, H. P. Schonherr, and K. H. Ploog, Phys. Rev. Lett. **87**, 016601 (2001).

[2] A. T. Hanbicki, B. T. Jonker, G. Itskos, G Kioseoglou, and A. Petrou, Appl. Phys. Lett. **80**, 1240 (2002).

[3] V. F. Motsnyi, J. De Boeck, J. Das, W. Van Roy, G. Borghs, E. Goovaerts, and V. Safarov, Appl. Phys. Lett. **81**, 265 (2002).

[4] X. Jiang, R. Wang, S. van Dijken, R. Shelby, R. Macfarlane, G. S. Solomon, J. Harris, and S. S. P. Parkin, Phys. Rev. Lett. **90**, 256603 (2003).

[5] J. Strand, B. D. Schultz, A. F. Isakovic, C. J. Palmstrøm, and P. A. Crowell, Phys. Rev. Lett. **91**, 036602 (2003).

[6] A. T. Hanbicki, O. M. J. van't Erve, R. Magno, G. Kioseoglou, C. H. Li, B. T. Jonker, G. Itskos, R. Mallory, M. Yasar, and A. Petrou, Appl. Phys. Lett. **82**, 4092 (2003).

[7] O. M. J. van 't Erve, G. Kioseoglou, A. T. Hanbicki, C. H. Li, B. T. Jonker, R. Mallory, M. Yasar, and A. Petrou, Appl. Phys. Lett. **84**, 4334 (2004).

[8] X. Jiang, R. Wang, R. M. Shelby, R. M. Macfarlane, S. R. Bank, J. S. Harris, and S. S. P. Parkin, Phys. Rev. Lett. **94**, 056601 (2005).

[9] V. F. Motsnyi, P. Van Dorpe, W. Van Roy, E. Goovaerts, V. I. Safarov, G. Borghs, and J. De Boeck, Phys. Rev. B **68**, 245319 (2003).

[10] R. Fiederling, M. Kelm, G. Reuscher, W. Ossau, G. Schmidt, A. Waag, and L. W. Molenkamp, Nature (London) **402**, 787 (1999); Y. Ohno, D. K. Young, B. Beschoten, F. Matsukura, and H. Ohno, *ibid.* **402**, 790 (1999).

[11] *Optical Orientation*, edited by F. Meier and B. P. Zakharchenya (North-Holland, New York, 1984).

[12] J. P. Bergman, P. O. Holtz, B. Monemar, M. Sundaram, J. L. Merz, and A. C. Gossard, Phys. Rev. B **43**, 4765 (1991); V. Srinivas, J. Hryniewicz, Y. J. Chen, and C. E. C. Wood, *ibid.* **46**,

10 193 (1992).

[13] G. E. Pikus and A. N. Titkov, in *Optical Orientation*, edited by F. Meier and B. P. Zakharchenya (North-Holland, New York, 1984), and references therein.

[14] M. I. D'yakonov and V. Yu. Kachorovski, Fiz. Tekh. Poluprovodn. (S.-Peterburg) **20**, 178 (1986) [Sov. Phys. Semicond. **20**, 110 (1986)].

[15] W. H. Lau, J. T. Olesberg, and M. E. Flatté, Phys. Rev. B **64**, 161301(R) (2001); cond-mat/0406201 (unpublished). Relatively weak temperature dependence of τ_s has also been observed in time-resolved optical experiments: J. M. Kikkawa, I. P. Smorchkova, N. Samarth, and D. D. Awschalom, Science **277**, 1284 (1997); A. Malinowski, R. S. Britton, T. Grevatt, R. T. Harley, D. A. Ritchie, and M. Y. Simmons, Phys. Rev. B **62**, 13 034 (2000); R. S. Britton, T. Grevatt, A. Malinowski, R. T. Harley, P. Perozzo, A. R. Cameron, and A. Miller, Appl. Phys. Lett. **73**, 2140 (1998).

[16] M. Z. Maialle, E. A. de Andrada e Silva, and L. J. Sham, Phys. Rev. B **47**, 15 776 (1993).

[17] A. Vinattieri, Jagdeep Shah, T. C. Damen, D. S. Kim, L. N. Pfeiffer, M. Z. Maialle, and L. J. Sham, Phys. Rev. B **50**, 10 868 (1994).

[18] E. Blackwood, M. J. Snelling, R. T. Harley, S. R. Andrews, and C. T. B. Foxon, Phys. Rev. B **50**, 14 246 (1994).

[19] L. Muñoz, E. Pérez, L. Viña, and K. Ploog, Phys. Rev. B **51**, 4247 (1995).

[20] I. Ya. Gerlovin, Yu. K. Dolgikh, S. A. Eliseev, V. V. Ovsyankin, Yu. P. Efimov, I. V. Ignatiev, V. V. Petrov, S. Yu. Verbin, and Y. Masumoto, Phys. Rev. B **69**, 035329 (2004).

[21] J. D. Albrecht and D. L. Smith, Phys. Rev. B **66**, 113303 (2002).

[22] Z. G. Yu and M. E. Flatté, Phys. Rev. B **66**, 235302 (2002).

[23] V. V. Osipov and A. M. Bratkovsky, Phys. Rev. B **70**, 205312 (2004).