Highly Spin-Polarized Room-Temperature Tunnel Injector for Semiconductor Spintronics using MgO(100)

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The spin polarization of current injected into GaAs from a CoFe/MgO(100) tunnel injector is inferred from the electroluminescence polarization from GaAs/AlGaAs quantum well detectors. The polarization reaches 57% at 100 K and 47% at 290 K in a 5 T perpendicular magnetic field. Taking into account the field dependence of the luminescence polarization, the spin injection efficiency is at least 52% at 100 K, and 32% at 290 K. We find a nonmonotonic temperature dependence of the polarization which can be attributed to spin relaxation in the quantum well detectors.

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Devices based on the manipulation of the spin state of electrons and holes within semiconductors are of interest today for possible sensor, memory, and logic applications [1,2]. A prerequisite for the realization of many of these devices is the development of solid state spin injectors at room temperature. The first such injectors used dilute magnetic semiconductors [3–5] but operated only at low temperatures. These materials, however, overcame the conductivity mismatch between the injecting and receiving materials, which had been recognized as an impediment to spin injection [6]. It is now appreciated that highly conducting ferromagnetic (FM) metals can also be used as spin injectors by forming a resistive tunnel contact between the FM metal and the semiconductor [7–14]. Traditional FM metals, such as Fe, Co, and Ni, and their alloys, are thus particularly attractive since they exhibit high Curie temperatures, and their magnetic moments can readily be directed by magnetic engineering concepts developed during the past decade [15]. Using amorphous Al_2O_3 tunnel barriers and FM metals, spin injectors have shown electroluminescent polarization (ELP) of ~40% at 4.5 K [16] and ~20% at 80 K [17] from semiconductor optical detectors. Spin injectors have also been formed from Schottky tunnel barriers with a reported ELP of ~30%, but only at low temperatures [12,14]. In these experiments, the ELP is limited by the tunneling spin polarization (TSP) of the injected electrons. It was predicted that much higher TSP could be realized for certain crystalline ferromagnet/tunnel barrier combinations due to strongly spin-polarized evanescent decay of particular wave functions through the tunnel barrier [18–21]. Recently, Parkin *et al.* have reported TSP values of up to 85% and room-temperature tunneling magnetoresistance values of ~220% in CoFe/MgO(100) tunnel junctions [22], consistent with these predictions. In this Letter we report a spin injector formed from a crystalline MgO(100) tunnel barrier in conjunction with a ferromagnetic CoFe layer. Optical measurements using quantum well (QW) detectors reveal efficient spin injection up to 290 K. The ELP is found to vary nonmonotonically with temperature, which is due to spin relaxation inside the QW detectors.

The spin injection efficiency was measured optically using GaAs/AlGaAs QW light emitting diodes (LEDs). The LEDs were grown using molecular beam epitaxy (MBE). Two LED devices with the following structures are discussed: *p*-GaAs(100) substrate/570 nm *p*-AlGaAs

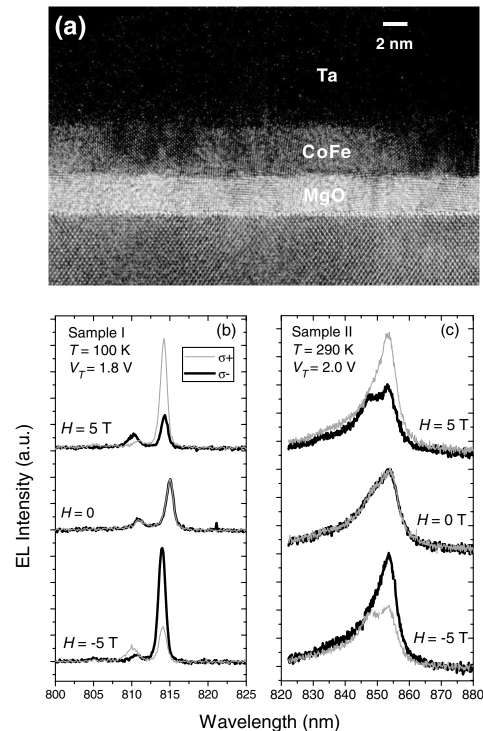

FIG. 1. TEM image of the CoFe/MgO spin injector (a) and electroluminescence (EL) spectrum of samples I (b) and II (c). The thin and thick lines in (b) and (c) represent the left ($\sigma +$) and right ($\sigma -$) circular polarization components of EL, respectively.

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buffer layer/75 nm undoped AlGaAs/10 nm undoped GaAs/15 nm undoped AlGaAs/100 nm AlGaAs upper layer/5 nm undoped GaAs, where AlGaAs is $Al_{0.08}Ga_{0.92}As$ for sample I and $Al_{0.16}Ga_{0.84}As$ for sample II. The AlGaAs upper layer is doped n type (5×10^{16} cm^{-3}) and p type (1×10^{17} cm^{-3}) for samples I and II, respectively. The LEDs were passivated with arsenic in the MBE chamber, and then transferred in air into a magnetron sputtering chamber to grow the spin injector, where they were heated to 550 °C to remove the arsenic cap. After the samples cooled down to ambient temperature, shadow masks were used to deposit the tunnel barrier (~3 nm MgO) and the FM electrode (~5 nm $Co_{70}Fe_{30}$ capped with ~10 nm Ta to prevent oxidation) which form the spin injector. The MgO barrier was deposited by reactive magnetron sputtering in an argon-oxygen gas mixture [22]. The CoFe and Ta layers were sputtered in pure argon gas. The active area of the spin injector was $\sim 100 \times 300\ \mu m^2$. Finally, the LEDs were annealed in vacuum at 300 °C for 1 h.

Figure 1(a) shows a high resolution transmission electron microscopy image of the CoFe/MgO spin injector. Both the MgO and CoFe layers are very smooth and are polycrystalline with a strong (100) texture along the growth direction. Such a crystallographic orientation is consistent with the theoretically predicted orientation which gives rise to a high tunneling spin polarization [19–21].

The electroluminescence (EL) was measured in a superconducting magnet cryostat. With a bias voltage (V_T) applied across the LED structure, spin-polarized EL was collected from the front side of the sample, i.e., through the CoFe and MgO films. The ELP was measured at various temperatures and bias voltages in a perpendicular magnetic field (H). In this measurement geometry the electron spin polarization is simply related to the ELP by the optical selection rules [23].

Figures 1(b) and 1(c) show the EL spectrum of sample I at 100 K ($V_T = 1.8$ V) and sample II at 290 K ($V_T = 2.0$ V), respectively. The EL peaks at longer and shorter wavelengths are due to the heavy hole (HH) and light hole (LH) emissions [24], respectively. For both samples, the EL intensities of the left (I^+) and right (I^-) circular components are magnetic field dependent, giving rise to a significant ELP, as the CoFe moment is rotated out of the film plane. The sign of the ELP indicates majority spin injection from CoFe. Henceforth, we focus only on the HH emission and refer to its ELP as P_{EL}, which is equal to the electron spin polarization just prior to recombination in the QW [23]. In this sense, P_{EL} sets a lower bound for the spin injection efficiency since the electrons will very likely undergo some spin relaxation before recombination. For sample I, the HH emission is well resolved from the LH emission due to its narrow linewidth (~1 nm). Therefore, it is rather straightforward to determine P_{EL}. In contrast, the HH and LH peaks for sample II are broad at 290 K and are thus less well resolved. In order to extract P_{EL} for this sample, we fit the EL spectrum with two Lorentzians and calculate P_{EL} from the fit, using the integrated area under the HH peak.

The magnetic field dependences of P_{EL} for sample I at 100 K and sample II at 290 K are depicted in Figs. 2(a) and 2(b) (open circles), where P_{EL} is calculated as $P_{EL} = (I^+ - I^-)/(I^+ + I^-)$. In each case the polarization increases rapidly with field up to ~2 T, when the CoFe moment is rotated completely out of plane. Above 2 T, P_{EL} continues to increase with field approximately linearly, but at a much lower rate, reaching 57% and 47% at 5 T for samples I and II, respectively. A linear variation of P_{EL} with field above 2 T (referred to as background polarization hereafter) is observed for both samples over a wide temperature range. The slope of the background usually varies gradually from a negative value at low temperatures to a positive value at high temperatures, crossing zero at ~40–50 K. Several factors may contribute to the background polarization. At low temperatures, thermalization of electron spins in the QW due to Zeeman splitting could give rise to a negative background since GaAs has a negative g factor. At high temperatures, however, the Zeeman energy is negligible compared to kT, and therefore cannot explain the observed background polarization. It is likely due to a field dependent spin relaxation rate and/or electron-hole recombination time. It is well known that a perpendicular magnetic field can suppress D'yakonov-Perel' (DP) spin relaxation in GaAs [23], which would therefore give rise to a positive background. Moreover, we

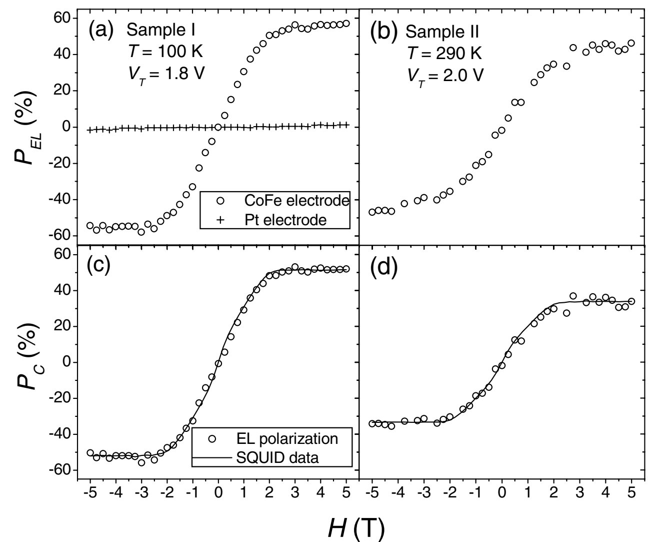

FIG. 2. Magnetic field dependence of P_{EL} [(a) and (b)] and P_C [(c) and (d)] for sample I at 100 K and sample II at 290 K (open circles). The crosses in (a) are P_{EL} of a control sample with a Pt electrode. The solid lines in (c) and (d) show the field dependence of the CoFe moment measured with a SQUID magnetometer at 20 K, which has been scaled to allow comparison with P_C.

found that the light intensity from the QW increased with increasing field, implying a shorter recombination time at higher fields which would also give rise to a positive background.

The EL polarization after subtraction of the linear background (P_C) is shown in Figs. 2(c) and 2(d) (open circles), which is a measure of spin polarization when the magnetic field influence on the ELP is excluded. P_C values as high as 52% and 32% were obtained at 100 and 290 K, respectively. The solid lines in Fig. 2(c) and 2(d) show the field dependence of the CoFe moment measured in a perpendicular magnetic field with a superconducting quantum interference device (SQUID) magnetometer at 20 K. The excellent agreement between the EL and SQUID data further confirms that the large ELP originates from spin injection.

To rule out possible artifacts of our measurement setup, we measured P_{EL} of a control sample, which had the same QW detector as sample I, but had a nonmagnetic Pt electrode in place of CoFe [crosses in Fig. 2(a)]. The polarization at 100 K was small, ~1%, and had a very weak field dependence. Photoluminescence experiments with linearly polarized pump light also gave a small polarization (<2%) and a weak field dependence. These results proved that the effects of polarization-dependent light absorption or reflection by the metal and semiconductor layers were very small.

The bias and temperature dependence of P_C are shown in Fig. 3 for the two samples. The relatively small confinement potential of the $GaAs/Al_{0.08}Ga_{0.92}As$ QW resulted in weak EL signals at high temperatures and consequently limited the measurements on sample I to below 100 K. In contrast, measurements on sample II were possible up to room temperature due to the use of a deeper $GaAs/Al_{0.16}Ga_{0.84}As$ QW. For both samples, P_C decreased with increasing bias at a given temperature. A similar bias dependence was observed in optical experiments and was attributed to spin relaxation through the DP mechanism before photoexcited electrons reached the QW [25,26]. In semiconductors lacking inversion symmetry, DP spin relaxation occurs due to spin precession about an effective magnetic field whose orientation and magnitude depends on the electron momentum. Larger electron momentum at higher bias results in a bigger effective field and consequently more rapid spin relaxation [23].

A nonmonotonic temperature dependence of the ELP was found for both samples, which is illustrated most clearly in Fig. 4. The bias voltage is $V_T = 1.8$ and 2.0 V for samples I and II, respectively. The ELP depends on the

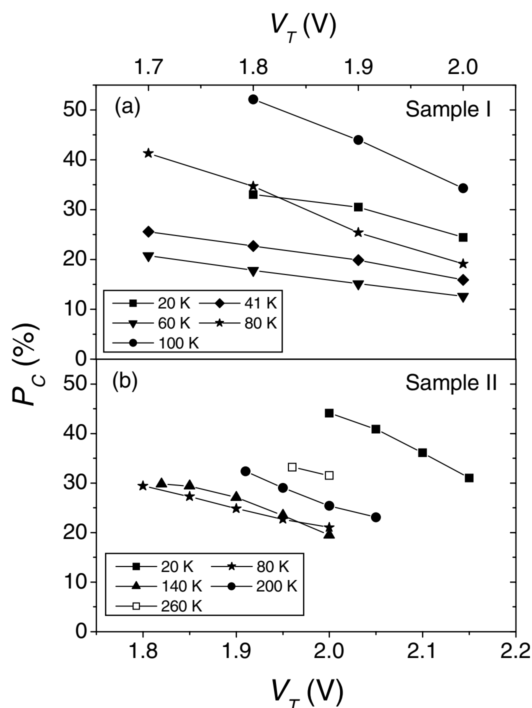

FIG. 3. Bias and temperature dependence of P_C of samples I (a) and II (b). Note the different bias ranges for (a) and (b).

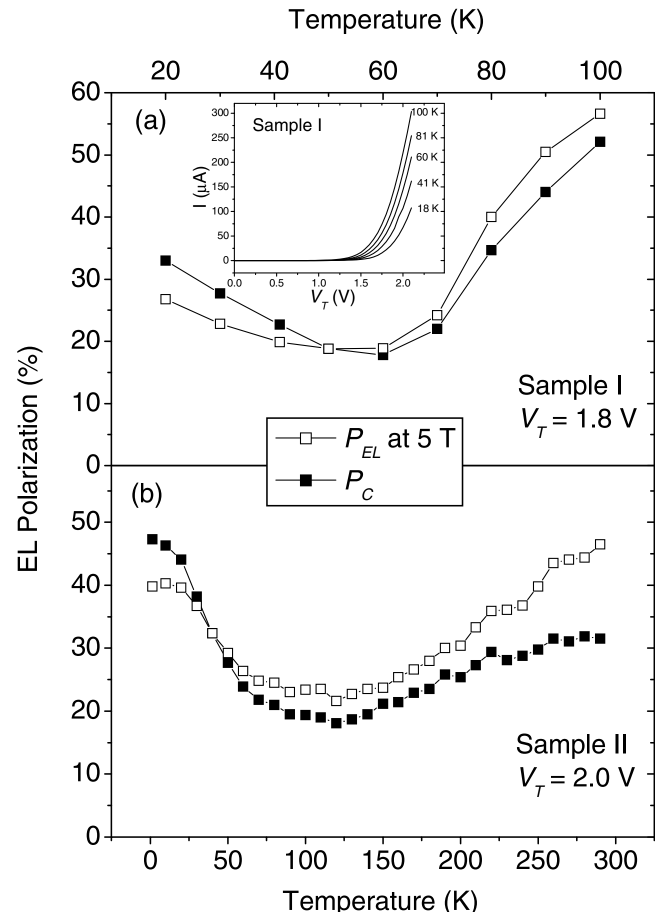

FIG. 4. Temperature dependence of the EL polarization of samples I (a) and II (b). The open and closed squares correspond to values of P_{EL} at 5 T and of P_C, respectively. Note the different temperature ranges for (a) and (b). The inset of (a) shows the *I-V* curves of sample I at various temperatures.

spin relaxation rate and the electron recombination time in the QW detectors. The DP spin relaxation rate in a QW is given by $\tau_s^{-1} \propto \tau_p T$, where τ_p is the momentum relaxation time and T is the temperature [27]. At very low temperatures, τ_p is dominated by ionized impurity scattering which has a weak temperature dependence, so that $\tau_p T$ and, consequently, τ_s^{-1} increase with temperature. At higher temperatures, when polar optical phonon scattering dominates the momentum scattering, $\tau_p T$ and, therefore, τ_s^{-1} decrease with increasing temperature [28]. As a result, the ELP tends to increase with temperature. The electron recombination time also varies with temperature [29,30] and could contribute to the temperature dependence of the ELP. Both the spin relaxation rate and the electron recombination time are dependent on the details of the QW detectors, which likely accounts for the quantitative differences between samples I and II.

A few subtle points require further discussion. First, the threshold voltage of the LED device decreases with increasing temperature [see the inset of Fig. 4(a)]. However, the light emission efficiency drops rapidly at high temperatures. As a result, larger currents are required to obtain enough EL signal at high temperatures. Second, the applied bias V_T is across the entire LED structure. As the temperature changes, the total voltage drop across the MgO barrier and the n- or p-AlGaAs depletion region (V_1) can vary slightly even if V_T remains constant. However, changes in V_1 would give rise to a monotonic temperature dependence of the ELP and thus cannot account for the experimental results. In addition, current-voltage measurements suggest that the change of V_1 with temperature at a given V_T is small and, therefore, could not significantly influence the temperature dependence of the ELP. Spin relaxation mechanisms other than the DP mechanism, such as the Elliot-Yafet (EY) and Bir-Aronov-Pikus (BAP) mechanisms [2,23], cannot account for the increase of the ELP with temperature at higher temperatures. The EY spin relaxation rate is proportional to the momentum scattering rate and would, therefore, give rise to a decreased ELP with increasing temperature, while BAP relaxation is weak in undoped QWs and cannot give rise to the observed temperature dependence. Finally, we note that DP spin relaxation in bulk semiconductors has a rate proportional to T^3 [23] and so such relaxation in the GaAs and AlGaAs layers between the injector and the QW is unlikely to give rise to the pronounced nonmonotonic temperature dependence which we found.

The observation of efficient spin injection up to 290 K using a CoFe/MgO tunnel injector is consistent with the high Curie temperature of CoFe and the weak temperature dependence of spin-dependent tunneling. The actual spin injection efficiency will be higher than that inferred from the polarization of the QW electroluminescence because of spin relaxation in the QW detector. Moreover, the spin relaxation is strongly temperature dependent, thus giving rise to a nonmonotonic temperature dependence of the ELP. The MgO based spin injector can readily be fabricated by sputter deposition. Moreover, the MgO barrier prevents intermixing of the FM metal and semiconductor, leading to improved device thermal stability [31]. These desirable features make MgO based tunnel spin injectors attractive for future semiconductor spintronic applications.

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