RAPID COMMUNICATIONS

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Spin injection across the Fe/GaAs interface: Role of interfacial ordering

B. D. Schultz,[1] N. Marom,[2] D. Naveh,[2] X. Lou,[3] C. Adelmann,[4,*] J. Strand,[3] P. A. Crowell,[3] L. Kronik,[2] and C. J. Palmstrøm[5,†]

[1]*International Technology Center, Raleigh, North Carolina 27617, USA*
[2]*Department of Materials and Interfaces, Weizmann Institute of Science, Rehovoth 76100, Israel*
[3]*School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455, USA*
[4]*Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, Minnesota 55455, USA*
[5]*Electrical and Computer Engineering and Materials, University of California–Santa Barbara, Santa Barbara, California 93106, USA*

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Spin injection efficiency is shown to strongly depend on the interfacial structure between Fe contacts and $\mathrm{Al_xGa_{1-x}As}$ in spin-based light emitting diodes. Both the magnitude and sign of the injected carriers are dependent on the atomic structure of the contacts and can be controlled through changes in temperature both during and following growth. We propose that the observed dependence is due to phase formation resulting from Fe/GaAs interfacial reactions. This proposed mechanism is consistent with electronic structure calculations, which show that thin layers of $\mathrm{DO_3}$ $\mathrm{Fe_3Ga}$ at the Fe/GaAs interface can produce the observed sign reversals in the spin polarization of injected carriers.

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Injection of spin-polarized currents into nonmagnetic materials is a key aspect of spintronics.[1] Many methods for achieving this injection have been suggested, including, use of ferromagnetic semiconductors[2] or hot-electron injection.[3] Interestingly, a contact of elemental Fe on a substrate of $\mathrm{Al_xGa_{1-x}As}$ has proven to be sufficient for generating spin-polarized currents in an array of spintronic devices.[4–9] Highly polarized optical signals, corresponding to high spin polarizations, have been measured from the electrical injection of spins for such junctions.[6,10] Based on the polarization of Fe and the efficiency of the detection schemes, electrical spin injection signals have been found to approach their theoretical limit at temperatures below 100 K.[10]

The physical origins of the successful spin injection are not obvious. Fe contacts for spin injection are typically grown at or below room temperature specifically to avoid interfacial reactions between the metal and semiconductor.[11] But the highest spin injection efficiencies have only been observed following postgrowth anneals of the device heterostructures at temperatures around 250 °C.[12–14] This annealing temperature is well below the growth temperature of the semiconductor structure; therefore, the annealing-induced spin injection enhancement must result from modification of the Fe/semiconductor contact. The simplest explanation would be that annealing promotes a more ordered and abrupt Fe/GaAs interface.[14] However, Fe and GaAs are not thermodynamically stable in contact with one another.[15] Therefore, annealing should promote interfacial reactions and an intermixed interfacial region rather than an ordered-phase-separated interface. The sluggishness of the Fe-GaAs reaction kinetics combined with similar diffusivities for each of the reaction species confines the formation of any reactions to a few monolayers at the interface for these anneals.[16] But the mechanisms by which spin injection and ejection signals are altered at the interface between ferromagnetic metal contacts and nonmagnetic semiconductors remain highly controversial and a convergence of theory and experiment has yet to occur.

In this Rapid Communication, we correlate spin injection signals with identifiable changes in the atomic structure of the contact interface, induced through thermal manipulations during the formation of $\mathrm{Fe/Al_xGa_{1-x}As}$ junctions. Importantly, we provide experimental evidence for polarization *reversal* during spin injection. We propose a physical mechanism, in which ultrathin interfacial layers can seriously affect, and even dominate, spin injection, through changes in *local* spin polarization. We further provide first-principles calculations based on density-functional theory (DFT) that strongly support this mechanism. The understanding afforded here may also explain the behavior of other ferromagnetic metal contacts and can provide means for controlling and manipulating spin injection processes through careful interface design.

A spin-based light emitting diode (spin-LED) device was used to quantify the level of spin injection from Fe contacts into the $\mathrm{Al_xGa_{1-x}As}$ semiconductor.[10] The electroluminescent polarization (ELP) emanating from the spin LED was measured in the Faraday geometry at 20 K with the magnetic field applied perpendicular to the sample surface. The selection rules in a quantum well allow the ELP to be directly mapped onto the spin polarization of the electrons at the time of recombination. The ELP signals collected from Fe contacts grown at −5 °C, 95 °C, 175 °C, and 250 °C are plotted in Fig. 1(a). The ELP signal saturates around 7% for the Fe contacts grown at −5 °C but decreases to approximately 2% as the growth temperature is increased to 50 °C (not shown) and 95 °C. An increase in the growth temperature to 175 °C results in ELP signals rising to ~7% again, but with the sign of the polarization *opposite* to that observed for lower growth temperatures, implying a change from majority-spin injection from the Fe contacts grown at lower temperatures to minority-spin injection at 175 °C. On further increase to 250 °C, the sign is no longer reversed, but the ELP saturation field along with the perpendicular magnetization of the contact have both decreased below 2 T, which is not consistent with Fe, indicating the bulk of the contact is being dominated by a reacted Fe-GaAs phase(s).[16] Therefore,

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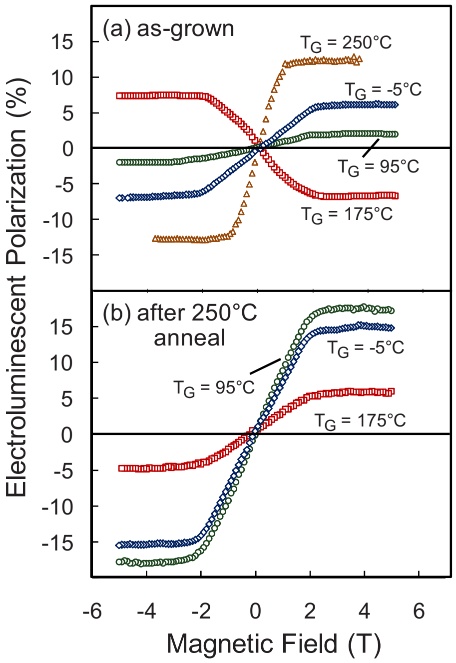

FIG. 1. (Color online) ELP as a function of magnetic field for Fe contacts grown at −5, 95, 175, and 250 °C (a) following growth and (b) following a postgrowth anneal at 250 °C for 1 h.

at 250 °C growth, we are no longer affecting solely, or even primarily, the interface.

ELP signals for the same Fe contacts following 1 h postgrowth anneals at 250 °C in a N_2 atmosphere are shown in Fig. 1(b). The magnitude of the saturated ELP signals increases dramatically after annealing for the −5 °C and 95 °C grown contacts. For the 175 °C grown contacts, the sign of the injected spin polarization flips and becomes consistent with that of the lower growth temperature contacts, albeit with a lower magnitude.

The extent of the interfacial reactions has been previously determined to be 5, 9, and 16 Å for Fe grown on GaAs(100) surfaces at −15 °C, 95 °C, and 175 °C.[11] Despite the difference in interfacial reactions, the perpendicular magnetization of the Fe contacts grown at both −5 °C and 175 °C is consistent with that of a pure Fe film and, as shown in Fig. 1(a), the ELP curves saturate at the saturation field (~2 T) of the perpendicular magnetization, ruling out a bulk effect (unlike growth at 250 °C, where the saturation field is ~1 T). Furthermore, photoemission spectra acquired from Fe/GaAs interfaces show no detectable changes in the bonding states of Ga as a result of postgrowth anneals at 250 °C, ruling out significant Fe/GaAs interdiffusion as a possible mechanism. But the Schottky barrier height is found to rise from 0.77 to 0.82 eV for Fe/$Al_{0.1}Ga_{0.9}As$ ($n=1\times10^{16}/\text{cm}^3$) contacts,[12] again indicating interfacial reactions. This makes the inverted sign of the spin injection at 175 °C surprising: reacted interfacial layers are typically expected to result in additional spin scattering at the interface and to be detrimental to spin injection without changing its sign. Here, the magnitude of the ELP signals for 175 °C is similar to that obtained for −5 °C, but of opposite sign, ruling out simple disorder arguments.

Interfacial Fe-GaAs reactions progress toward an intermixed interfacial region composed primarily of the two stable binary phases Fe_3Ga and Fe_2As, although ultrathin intermediate phases of $Fe_3Ga_{2-x}As_x$ are possible during the reaction process.[16] Fe_3Ga is a ferromagnetic phase with a Curie temperature of 1040 K,[17] Fe_2As is an antiferromagnetic phase with a Néel temperature of 367 K,[18] and $Fe_3Ga_{2-x}As_x$ is a ferromagnetic phase with Curie temperatures as high as 644 K.[19] The Fe_3Ga phase is a likely candidate for producing observable spin injection signals and therefore DFT calculations of the Fe/Fe_3Ga/GaAs structure were performed to examine the plausibility of a thin Fe_3Ga interface layer producing a flip in the spin injection. Specifically, the spin-polarized density of states (DOS) was calculated by solving the Kohn-Sham equations of DFT using the plane-wave approach, as implemented in the Vienna *ab initio* simulation package (VASP) (Refs. 20 and 21). The generalized gradient approximation exchange-correlation functional of Perdew, Burke, and Ernzerhof[22] was used throughout. Fe/GaAs supercells comprising 7 layers of Fe and 11 atomic layers of GaAs, with a varying number of Fe_3Ga monolayers in the DO_3 structure[23] at the Fe-GaAs interface, were studied. In order to simulate epitaxial growth, the lattice constant of GaAs 5.65 Å was assumed throughout and the structure was not relaxed. An example for the structure of the central region of a junction with four atomic layers of Fe_3Ga between Fe and As-terminated GaAs is shown schematically in Fig. 2(a).

Figure 2(b) shows a grayscale map of the local spin polarization, i.e., the difference between the majority DOS and the minority DOS as a function of energy and position. Importantly, the local DOS near the Fermi level E_F is dominated by *majority* carriers within the Fe layer (note the white "stripe" across layers 1–3 near E_F). However, across the entire interface, i.e., from the last Fe layer to the first As layer, the local DOS near E_F is dominated by *minority* carriers (note the "black spots" across layers 4–9). This is further elucidated with the aid of cross sections of the local DOS at various locations shown in Figs. 2(c)–2(h). A cross section through the Fe layer farthest from the interface [Fig. 2(c)] shows a local DOS very similar to that of bulk Fe, whereas a cross section through the Fe layer closest to the interface [Fig. 2(d)] reveals a significant deviation from the DOS of bulk Fe: the majority DOS in the vicinity of E_F is considerably lower than that of bulk Fe, whereas the minority DOS is considerably higher. The cross section through Fe_3Ga in Fig. 2(e) is similar to the DOS obtained for bulk DO_3 Fe_3Ga. It reveals a minority peak at E_F that is a characteristic feature of the DO_3-Fe_3Ga DOS. A cross section through the Fe_3Ga layer adjacent to the GaAs [Fig. 2(f)] shows further evolution of the minority peak at E_F and an additional increase in the number of minority states. Cross sections through the first and second GaAs layers from the interface [Figs. 2(g) and 2(h)] deviate significantly from that of bulk GaAs due to metal-induced gap states[24,25]—electronic states of the metal that extend into the forbidden gap of the semiconductor. A qualitatively similar behavior of DOS variations across the different interface layers was also found for other interface configurations, i.e., for As- or Ga-terminated GaAs, for Fe_3Ga terminated by a Fe-only or a mixed Fe-Ga atomic

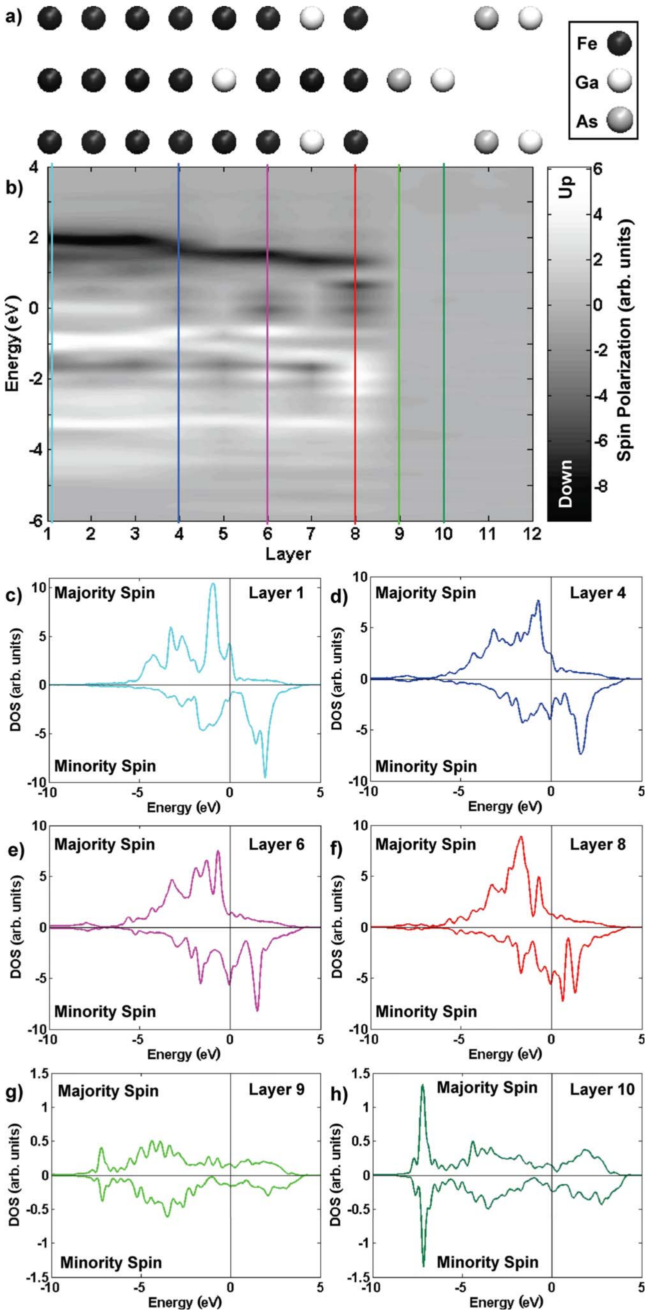

FIG. 2. (Color online) Calculated electronic structure of the epitaxial Fe/Fe_3Ga/GaAs interface for an As-terminated GaAs surface with four atomic layers of Fe_3Ga in the DO_3 structure. (a) A schematic representation of the interface region. (b) Grayscale map of the local spin polarization, i.e., the difference between the majority DOS and minority DOS as a function of energy and position, for the structure shown in a. [(c)–(h)] Cross sections of the local DOS through various atomic layers.

layer, and for varying thickness of the Fe_3Ga layer. Thus, the combination of the experimental and theoretical results of Figs. 1 and 2 suggests that the interface minority population is strongly enhanced at and near E_F due to the presence of DO_3-Fe_3Ga at the Fe/GaAs interface and that it could induce polarization reversal for spin injection.[26] This may provide a natural explanation for the spin-polarization reversal observed in the ELP experiments of Fig. 1. It also rationalizes how a modest change in growth temperature can result in spin reversal. Because the injected carriers originate from the vicinity of the interface, an increase in the thickness of the interface Fe_3Ga layer will enhance minority-spin injection, eventually to the point of spin reversal.

Turning to annealing effects, recent high-angle annular dark-field scanning transmission electron microscopy studies showed that postgrowth anneals of a 50 °C grown Fe/GaAs interface result in an epitaxial Fe layer on an As-terminated GaAs surface with a partially occupied plane inserted between the Fe film and the As-terminated GaAs layer, which is likely Fe bonded to As.[27] The rearrangement of the interface into this structure does not require additional reactions to occur between the Fe and GaAs, which is consistent with photoemission results. For the abrupt Fe/GaAs interface, additional calculations (not shown for brevity) show that a peak in the minority DOS at E_F emerges in the Fe layer adjacent to the interface. This peak has also been observed in previous studies.[28–31] For the abrupt interface, two recent theories propose either a resonant interface band[29] or localized bound states in an interfacial quantum well within the semiconductor[32] to explain the reversal of spin polarization in the spin extraction regime but not the spin injection regime. Additionally, bias-dependent calculations of the Schottky diode[33] and experimental interface doping profile investigations[34] have shown spin transport to depend strongly on the electronic structure of the Fe/GaAs interface. The minority peak at E_F can be strongly affected by the precise atomic arrangement at the interface. For example, our calculations suggest that it is absent from the (unrelaxed) interface structure suggested by Zega *et al.*,[14] which features one atomic layer of intermixed Fe and As, as well as from an interface in which the last Fe atomic plane contains half the number of Fe atoms. If annealing does promote majority-spin injection by suppressing the interface minority peak, then even with significant concentrations of Fe_3Ga present, postgrowth anneals may eliminate the spin flip. This is consistent with Fig. 1(b), where the saturated ELP signal for 175 °C-grown contact changes sign after annealing, but its magnitude is still significantly smaller than that from samples grown at low temperature, possibly due to the competition between growth and annealing related minority- and majority-spin currents, respectively.

Perhaps surprisingly, neither the direct deposition of Fe_3Ga as an injection contact[35] nor the inclusion of a thin 10 Å Fe_3Ga interlayer grown between the Fe contact and (Ga,Al)As surface shows spin reversal in ELP measurements. But it is entirely likely that the growth conditions were outside the "window," leading to spin flip in Fig. 1, and we have already concluded that even in the presence of Fe_3Ga a specific interface arrangement is needed to support minority-spin injection. Preliminary high-angle annular dark-field scanning transmission electron microscopy studies of Fe_3Ga films grown on Ga-rich and As-rich GaAs(001) surfaces reveal various interfacial atomic structures, which are distinctly different from the postgrowth annealed Fe/GaAs interface. Furthermore, when a 10 Å Fe_xAs interlayer is grown between the Fe contact and (Ga,Al)As surface, the resulting ELP signals are essentially zero before a 250 °C anneal (consistent with the antiferromagnetic nature of Fe_2As) but increase to nearly the same levels found for the equivalent 10 Å Fe_3Ga interlayers after annealing. These examples further illustrate the intrinsic link between interfacial structure and spin transport and their dependence on deposition conditions and postgrowth annealing.

In conclusion, we showed that it is the interface composition and bonding between Fe-based contacts and the underlying GaAs-based LED structures that determine the magnitude and sign of spin-injected carriers, more than the bulk properties of the contact itself. In particular, a thin interface layer, e.g., of Fe_3Ga, can result in complete reversal of the injected spin, but this reversal can be mitigated or even overcome by additional changes in the precise interface arrangement that may occur during annealing or growth at elevated temperatures.

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*Present address: IMEC vzw., B-3001 Heverlee, Belgium.

[†] Author to whom correspondence should be addressed; cpalmstrom@ece.ucsb.edu

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