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Reversal of Spin Polarization in Fe/GaAs (001) Driven by Resonant Surface States: First-Principles Calculations

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A minority-spin resonant state at the Fe/GaAs(001) interface is predicted to reverse the spin polarization with the voltage bias of electrons transmitted across this interface. Using a Green's function approach within the local spin-density approximation, we calculate the spin-dependent current in a Fe/GaAs/Cu tunnel junction as a function of the applied bias voltage. We find a change in sign of the spin polarization of tunneling electrons with bias voltage due to the interface minority-spin resonance. This result explains recent experimental data on spin injection in Fe/GaAs contacts and on tunneling magnetoresistance in Fe/GaAs/Fe magnetic tunnel junctions.

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Ferromagnetic metal or nonmagnetic semiconductor contacts have recently attracted significant interest due to the possibility to generate nonequilibrium electron spin distributions in normal semiconductors and hence are practical for spintronics applications [1]. The contact structures in which electron tunneling dominates transport properties are used to achieve a sizable spin polarization of the electric current and produce spin accumulation in the semiconductor. The spin polarization originates from the spin dependence of the wave functions and densities of states of the ferromagnetic contact. As a result, the tunneling transmission coefficients are different for majority- and minority-spin electrons.

Among various ferromagnet or semiconductor structures, Fe/GaAs contacts have been extensively studied, showing that spin-dependent tunneling through Schottky barriers formed by delta doping is an efficient method for injecting nonequilibrium spin distributions in a semiconductor [2–4]. These experiments showed that in biased Fe/GaAs contacts the net spin of electrons injected from Fe into GaAs is parallel to the magnetization of the ferromagnetic Fe electrode. This implies that majority-spin electrons tunnel through the Schottky barrier more efficiently that minority-spin electrons.

Recently, however, Crooker *et al.* [5] and Lou *et al.* [6] observed an anomalous bias dependence of the transport spin polarization in Fe/GaAs Schottky barrier structures. They found that both the magnitude and the sign of the spin polarization depend on applied bias voltage producing either majority- or minority-spin accumulation in GaAs. Moser *et al.* [7] observed a related phenomenon in Fe/GaAs/Fe magnetic tunnel junctions. They found that tunneling magnetoresistance (TMR) changes sign with bias voltage, reflecting the reversal of the transport spin polarization at the Fe/GaAs interface. To explain the experiments by Crooker *et al.* [5], Dery and Sham [8] developed a model suggesting that the reversal of spin polarization is due to localized states in the semiconductor formed by electrostatic confinement of doping profiles.

In this Letter, we demonstrate that the observed reversal of the spin polarization in Fe/GaAs(001) tunnel contacts is intrinsic to their interface electronic structure. The Fe/GaAs(001) interface supports a minority-spin interface band lying in the vicinity of the Fermi energy [9–11]. This interface band is reminiscent of the Fe(001) surface band observed experimentally using scanning tunneling spectroscopy [12]. Because of the coupling to continuum bulk states in Fe the Fe/GaAs(001) interface band evolves into an interface resonant band and strongly contributes to the tunneling conductance. The minority-spin character of this resonant band leads to the reversal of the spin polarization from positive to negative in the relevant range of electron energies. This explains the experimental findings of anomalous bias dependence of the spin polarization in experiments on spin injection [5,6] and TMR [7].

The results of experiments [5–7] reflect features of spin transmission across the Fe/GaAs(001) interface. This is due to the transport spin polarization in tunneling geometry being largely controlled by the interface atomic and electronic structure [9]. In the case of spin injection [5,6], electrons injected from Fe into GaAs tunnel through the GaAs barrier, then experience scattering by a defect or impurity, and further propagate diffusively producing spin accumulation in GaAs. Since diffusive transport in a nonmagnetic material is independent of electron spin, the spin polarization established within GaAs is entirely due to asymmetry in the spin transmission across the Fe/GaAs(001) interface. A similar argument applies to spin extraction from GaAs into Fe. In case of magnetic tunnel junctions, variations in TMR are expected to reveal spin polarizations of the two interfaces [13]. However, Moser *et al.* [7] observed a reversed TMR only for those Fe/GaAs/Fe tunnel junctions in which one interface was "ideal" epitaxial, whereas the other was either oxidized or cleaned by a H^+ plasma. Therefore, their findings reveal features of spin transmission across the epitaxial Fe/GaAs(001) interface only.

To study spin-polarized transport across the Fe/GaAs(001) interface, we consider a Fe/GaAs/Cu(001) tunnel junction with a bcc Cu counterelectrode, which serves as a detector of spin polarization, in the spirit of Ref. [14]. The bcc Cu electrode has a spin-independent free-electron-like band structure and a featureless surface transmission function [13], making it a perfect spin detector. This implies that variations in the spin polarization of the tunneling current with bias voltage found in the calculation performed for the Fe/GaAs/Cu(001) tunnel junction are entirely due to the changes in the spin transmission across the Fe/GaAs(001) interface. This makes the results of our calculations relevant to experiments [5–7].

The particular junction studied consists of a semi-infinite Fe electrode, 8 monolayers of GaAs barrier, and a semi-infinite bcc Cu electrode. We consider an As-terminated interface, motivated by the experiments on spin injection [5,6] where the epitaxial Fe/GaAs interfaces were grown in As-rich environment [15]. Since intermixing of Fe and As atoms at this interface is not energetically favorable [11,16], we assume that the interface is abrupt. The small change of the As-Ga interplane distance of about 0.14 Å due to relaxation [11] is not taken into account.

Calculations are performed using the Green's function representation of the tight-binding linear muffin-tin orbital method in the atomic sphere approximation [17]. We apply third-order parametrization for the Green's function [18]. The electronic structure problem is solved within the scalar-relativistic density functional theory where the exchange and correlation potential is treated in the local spin-density approximation. The conductance is calculated using the principal-layer Green's function technique [19,20] within the Landauer-Büttiker approach [21]. Charge self-consistency is achieved before performing the transport calculations.

The spin-dependent transmission coefficient $t^{\sigma}(E, \mathbf{k}_{\|})$ is calculated for a given spin $\sigma = \uparrow, \downarrow$ (where $\uparrow$ and $\downarrow$ denote majority and minority spin, respectively) as a function of energy E and the transverse wave vector $\mathbf{k}_{\|}$ which is conserved due to the transverse periodicity of the junction. The total transmission for a given energy and spin is obtained by integrating over $\mathbf{k}_{\|}$ within a two-dimensional Brillouin zone (2DBZ): $T^{\sigma}(E) = \int t^{\sigma}(E, \mathbf{k}_{\|}) d^2\mathbf{k}_{\|}/(2\pi)^2$. A uniform 200 × 200 mesh is used for the integration. The current density associated with this transmission is obtained from $J^{\sigma}(V) = (e/h) \int_{E_F}^{E_F+eV} T^{\sigma}(E) dE$, where E_F is the Fermi energy and V is the applied bias voltage. This is a reasonable approximation for small voltages considered in this work. This definition of $J^{\sigma}(V)$ implies that for a *negative* voltage electrons tunnel *from* Fe across GaAs. The spin polarization is defined as $P = (J^{\uparrow} - J^{\downarrow})/(J^{\uparrow} + J^{\downarrow})$.

Figures 1(a) and 1(b) show the calculated local density of states (DOS) at the interface Fe monolayer and the integrated transmission as a function of energy for the Fe/GaAs/Cu junction. The energies are given with respect to E_F, which is found to be in the middle of the GaAs band gap in agreement with previous calculations [10,11]. As is seen from Fig. 1(a), the minority spin dominates the interface DOS in the vicinity of the Fermi energy throughout the energy interval shown. There is a sharp peak in the minority-spin DOS between −50 and −160 meV. The majority-spin transmission [Fig. 1(b)] exhibits a featureless free-electron-like energy dependence mirroring the featureless majority-spin DOS [Fig. 1(a)]. In contrast, the minority-spin transmission is nonmonotonic and dominates in two energy windows, between −130 and −110 meV and between +50 and +175 meV [Fig. 1(b)]. The former local maximum corresponds to the sharp peak in the minority-spin interface DOS, whereas the latter maximum has no distinct analog in the DOS.

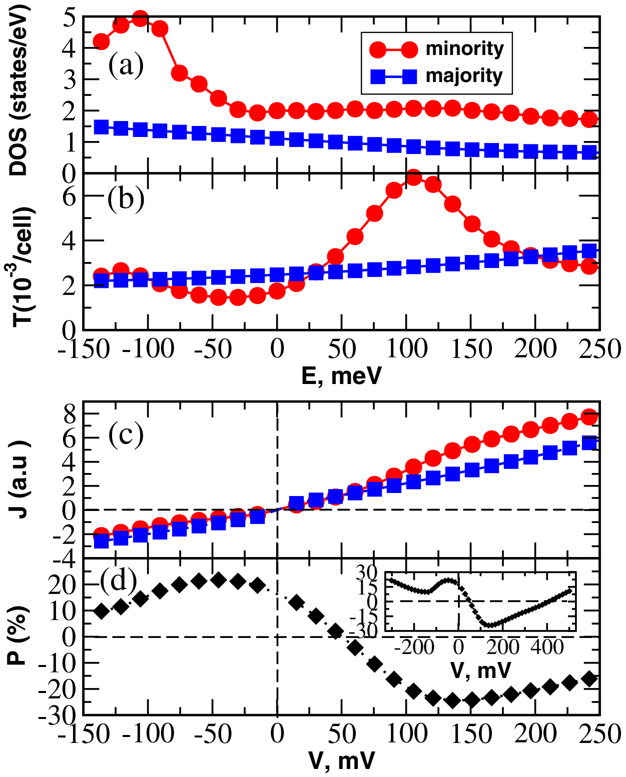

FIG. 1 (color online). Results of calculations for a Fe/GaAs/Cu tunnel junction: (a) spin-resolved local DOS for the interface Fe monolayer; (b) spin-resolved integrated transmission as a function of energy; (c) spin-resolved current density as a function of bias voltage; (d) spin polarization as a function of bias voltage. The inset shows the spin polarization over an extended range of bias [22]. In (a) and (b), the Fermi level is set at zero energy.

The energy dependence of the transmission is reflected in the voltage dependence of the spin-resolved current density shown in Fig. 1(c). It is seen that, while for negative bias voltages majority-spin electrons dominate the current density, there is a crossover at about +50 mV that makes the minority-spin current dominating at higher voltages up to $V = +400$ mV (see inset of Fig. 1(d)]. This leads to the reversal of spin polarization at about $V = +50$ mV seen in Fig. 1(d). At $V = +400$ mV the spin polarization changes sign again, reversing from anomalous (negative) to normal (positive). At $V = 0$ we obtain high positive spin polarization similar to earlier work [23]The transmission peak between −130 and −110 meV [Fig. 1(b)] is too small to change the sign of the spin polarization and only leads to a reduction of the spin polarization by about 10%. The reversal of the spin polarization with bias voltage is the central result of this Letter. In the following we show that an interface resonant band is responsible for this anomalous behavior.

Figure 2 shows the $\mathbf{k}_{\|}$-resolved minority-spin local DOS for two monolayers of Fe at the Fe/GaAs(001) interface in comparison to the bulk DOS of Fe. The upper three panels correspond to the energy $E = -121$ meV at the maximum in the interface DOS [Fig. 1(a)] and the matching local peak in the transmission [Fig. 1(b)]. The lower three panels correspond to the energy $E = +106$ meV at the maximum in the transmission [Fig. 1(b)]. It is seen that for both energies the interface DOS is strikingly different from the respective bulk DOS [compare Figs. 2(a), 2(c), 2(d), and 2(f)]. As is evident from Figs. 2(c) and 2(f), for both energies the interface DOS are characterized by features that have the C_{2v} symmetry intrinsic to the atomic structure of the Fe/GaAs(001) interface. The topology of these features is preserved at the subinterface monolayer, but their intensity drops down by a factor of 5 [compare Figs. 2(b), 2(c), 2(e), and 2(f)]. This behavior clearly points to the presence of minority-spin interface states at energies $E = -121$ meV and $E = +106$ meV. The integral DOS for the state at $E = -121$ meV is much higher than that for the state at $E = +106$ meV and consequently the former produces the sharp peak in Fig. 1(a), whereas the latter is relatively broad.

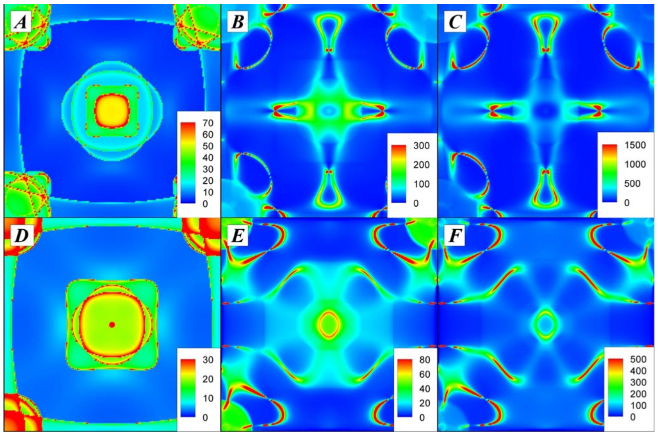

FIG. 2 (color online). Minority-spin Fe local density of states resolved in the two-dimensional Brillouin zone by $\mathbf{k}_{\|}$ with abscissa along [10] and ordinate along [01] direction. The upper three panels are for $E = -121$ meV corresponding to the local maximum of the minority-spin transmission: (a) bulk, (b) subinterface monolayer, (c) interface monolayer. The lower three panels are for $E = 106$ meV corresponding to the maximum of the minority-spin transmission: (d) bulk, (e) subinterface monolayer, (f) interface monolayer.

The analysis of the character of the interface states (bands) shows that they arise from a mixture of $d_{3z^2-r^2}$ and d_{xy} orbitals on the interface Fe sites. These states moderately hybridize with bulk Fe minority-spin bands and develop into interface resonances. The latter fact is evident from their finite width that allows these states to be resolved in $\mathbf{k}_{\|}$ space [see Figs. 2(c) and 2(f)].

The interface resonances contribute to the tunneling conductance. The magnitude of their contribution, however, strongly depends on their distribution across the 2DBZ, because the decay of evanescent states in GaAs depends on $\mathbf{k}_{\|}$. By analyzing the complex band structure of GaAs, Mavropoulos *et al.* [24] demonstrated that the decay constant κ for the evanescent states has a rather deep parabolic global minimum at the $\bar{\Gamma}$ point ($\mathbf{k}_{\|} = 0$). This feature strongly suppresses the transmission through the resonant states at $E = -121$ meV, because they are located far from the $\bar{\Gamma}$ point [Fig. 2(c)]. In contrast, the resonance at $E = +106$ meV corresponds to the opening of a parabolic pocket at the $\bar{\Gamma}$ point, which is seen as an ellipse in the surface DOS [Fig. 2(f)]; the proximity to the $\bar{\Gamma}$ point allows these electrons to tunnel efficiently across the GaAs barrier.

Figure 3 shows the $\mathbf{k}_{\|}$-resolved transmission for three energies around the transmission maximum at $E = +106$ meV. It is seen that around the $\bar{\Gamma}$ point the resonance band is parabolic and anisotropic, reflecting the C_{2v} symmetry of the interface. Owing to its location near the zone center and a relatively large DOS, this minority-spin band dominates the transmission near $E = +106$ meV. For lower energies [Fig. 3(a)] the resonant band only partially

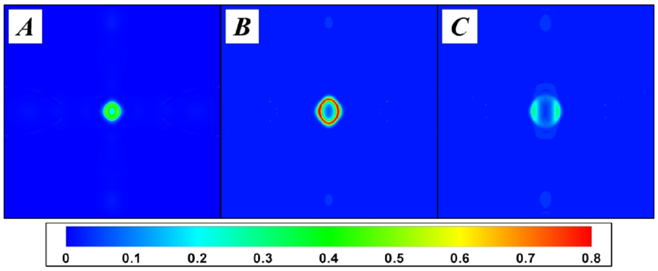

FIG. 3 (color online). $\mathbf{K}_{\|}$-resolved minority-spin transmission through a Fe/GaAs/Cu (001) tunnel junction for three energies near the maximum at $E = 106$ meV: (a) 45 meV, (b) 106 meV, (c) 166 meV.

crosses the Fermi level providing fewer states to the tunneling current, while for higher energies [Fig. 3(c)] the crossings occur for larger $\mathbf{k}_{\parallel}$ which reduces the resonant band contribution due to $\mathbf{k}_{\parallel}$ filtering in GaAs. This leads to the dominant contribution of the minority-spin resonant states in the tunneling conductance in a finite energy window [Fig. 1(b)] and results in the reversal of the spin polarization at bias voltages from +50 to +400 mV [Fig. 1(d)].

Our results explain the experimental data on spin injection by Crooker *et al.* [5] and Lou *et al.* [6]. In our calculations the reversal of the spin polarization occurs at positive applied bias voltage. This corresponds to electrons incoming from GaAs into unoccupied states of Fe, that is *forward* applied bias (spin extraction) in the experiments [5,6]. We find positive spin polarization for negative applied bias voltage, corresponding to electrons incoming from Fe into GaAs, that is *reverse* applied bias (spin injection) in experiments [5,6]. These results are in agreement with the experimental data by Lou *et al.* [6] (samples A and C). We note, however, that the energy of the interface states is sensitive to details of the sample preparation and may be affected by interdiffusion and disorder resulting in energy shifts of the order of several tenths of an eV. Such a shift may explain why the reversal of the spin polarization occurs when electrons are injected from Fe into GaAs in sample B of Lou *et al.* [6]. Magnetic anisotropy due to Rashba effect at the interface can also result in energy shifts of a few tenths of an eV [14]. This suggests that the bias dependence of spin polarization may depend on the orientation of magnetization as well.

Our results also agree with the TMR data of Moser *et al.* [7]. They observed a TMR reversal for Fe/GaAs/Fe tunnel junctions with one epitaxial interface and the other one either oxidized or cleaned with H^+ plasma. Since no anomalies are observed when both interfaces are disordered, the reversed TMR is entirely due to the reversal of the spin polarization at the epitaxial interface, which occurs at bias voltages from −90 to about +400 mV. The minimum in TMR is at $V = +50$ mV, which corresponds to electrons transmitted across GaAs to the epitaxial interface. This is consistent with our results shown in Fig. 1(d). When a sample was annealed at 150 °C for 1 h the first reversal of TMR occurred at +20 mV instead of −90 mV, while the shape of the trace remained essentially unchanged [25]. This supports our view concerning the sensitivity of interface states to sample preparation.

In conclusion, we have demonstrated that the minority-spin resonant states at the Fe/GaAs(001) interface are responsible for the reversal of spin polarization of electrons transmitted across this interface. This explains experimental data on spin injection in Fe/GaAs contacts and on TMR in Fe/GaAs/Fe magnetic tunnel junctions.

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Note added in proof.—In a new experiment Kotissek *et al.* [[26]] found no bias-induced reversal of the spin polarization in a $Fe_{32}Co_{68}$/GaAs spin injection device with a doping profile similar to Refs. [5,6]. The FeCo alloy has a larger band filling that removes the resonant interface band from the Fermi level; this result is therefore consistent with the mechanism described in this Letter.

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