Spin polarization control through resonant states in an Fe/GaAs Schottky barrier

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Spin polarization of the tunnel conductivity has been studied for Fe/GaAs junctions with Schottky barriers. It is shown that band matching of resonant interface states within the Schottky barrier defines the sign of spin polarization of electrons transported through the barrier. The results account very well for experimental results including the tunneling of photo-excited electrons, and suggest that the spin polarization (from -100\% to 100\%) is dependent on the Schottky barrier height. They also suggest that the sign of the spin polarization can be controlled with a bias voltage.

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One of the main aims in semiconductor (SC) spintronics is to use the spin degree of freedom of electrons for novel electronic devices. The use of ferromagnetic (FM) contacts to inject a spin-polarized current into a SC has been intensively studied as a means to achieve spintronic control in SC devices, and has led to many successful experiments that demonstrate a spin-polarized current through the contact \cite{1, 2, 3, 4, 5, 6, 7}. The spin-injection efficiencies measured in these experiments are impressive with the highest being 57\% at 100K \cite{6}, but they are not as high as first-principles band calculations predict \cite{8}. Quite recently, negative spin polarization (negative $P$) of the tunnel current through the Schottky barrier of FM/GaAs has been reported in several experiments; observation of spin accumulation in lateral Fe/GaAs/Fe \cite{11, 12}, imaging of injected spins in FeCo/GaAs junctions \cite{13}, measurements of tunnel magnetoresistance in Fe/GaAs/Fe junctions \cite{14}, and spin-filtering experiments with photo-excited electrons produced in the GaAs layer \cite{15}. The bias dependence of negative $P$ in these experiments, however, is still controversial as argued by Lou et al. \cite{12}, suggesting that the band structure at the FM/SC interface and the Schottky barrier may play a key role in determining spin transport across the interface.

Several mechanisms of negative $P$ have been proposed for tunneling conductance of Fe junctions; resonant tunneling via extrinsic impurity levels in the barrier region \cite{9}, and interface resonant states (IRSs) appeared intrinsically in the minority spin state of Fe \cite{10}. The extrinsic mechanism may be ruled out for Fe/GaAs junctions since the negative $P$ appears in ideal interfaces \cite{14}. As for IRSs mechanism, Chanits et al. \cite{16} have proposed that IRSs at Fe/GaAs interfaces are responsible for the negative $P$ by performing a first-principles calculation for an Fe/GaAs/Cu junction without the Schottky barrier \cite{17}. On the other hand, Dery and Sham proposed that the sign of $P$ is governed by a competition between conduction electron tunneling with positive $P$ and tunneling of localized electrons with negative $P$ in an overdoped layer near the Fe/GaAs interface \cite{18}. In addition, Dery-Sham have proposed a novel device of spin-switch by a gate voltage control. In spite of these important works, one should examine the spin transport mechanism further, since the role of the Schottky barrier is still unclear and the negative $P$ has been observed also in Fe/GaAs junctions without the overdoped layer \cite{14, 17}.

In this Letter, we will show that the IRSs within the Schottky barrier play an important role for the negative $P$ and its bias dependence. Because of the band symmetry of both Fe and GaAs layers, and symmetry dependent hybridization with the spin-polarized Fe states, down ($\uparrow$) spin IRSs appear near the bottom of GaAs conduction band and vary the thickness of the Schottky barrier effectively. Due to the features of IRSs in the Schottky barrier, a sharp variation of $P$ from $\sim 100\%$ to $\sim -100\%$ occurs when the energy of incident electrons or the applied bias is changed. The present results not only explain the spin-filtering of photo-excited electrons in the GaAs layer semi-quantitatively, but may resolve the controversy about the bias dependence of negative $P$ observed. A strong variation of $P$ in a small bias region may also suggest possible control of spin polarization with a bias voltage, or with Schottky barrier height using FM alloys with different work functions. The former phenomenon could be used to make a new type of spin-switch devices.

In the following, we calculate the spin-dependent tunnel conductance for photo-excited electrons through an Fe/GaAs contact with a Schottky barrier using a full-orbital tight-binding model and the linear response theory. The present model is sufficiently realistic to reproduce the previous results obtained in the first principles \cite{8}, and feasible to deal with the thick tunnel barrier formed by a Schottky barrier. The model is also appropriate to study the effects of the IRSs on the tunnel conductance \cite{15}.

When we restrict our discussion to absolute zero tem-
temperature and neglect a thermally excited Schottky current, the tunnel current $I_{L(R)}$ of electrons excited by left(right) circularly polarized light in GaAs can easily be obtained by using the selection rule, the symmetry of the valence and conduction bands \cite{19}, and spin ($\sigma = \uparrow, \downarrow$) dependent tunnel conductance $\Gamma_\sigma(E)$ at an energy $E$. When the Schottky barrier is sufficiently high and thick, the tunnel currents should be governed by the tunneling probability at the excitation energy that $E_1 = E_{\text{ph}} - E_g$ and $E_0 = E_{\text{ph}} - E_g - \Delta$, from $P_{1/2}$ and split-off $P_{1/2}$ valence bands, respectively, where $E_{\text{ph}}$, $E_g$ and $\Delta$ are the photon energy, the band gap energy and the spin splitting of the valence band, respectively. Then, the difference between $I_L$ and $I_R$ under a forward bias $V_F$ is given by

$$\Delta I \equiv I_L - I_R \sim 2 \left[ P(E_1)\Gamma_\uparrow(E_1) - P(E_0)\Gamma_\downarrow(E_0) \right] V_F, \quad (1)$$

where $P(E) = [\Gamma_\uparrow(E) - \Gamma_\downarrow(E)] / [\Gamma_\uparrow(E) + \Gamma_\downarrow(E)]$ is the spin polarization of the tunneling conductance.

Three different photon energies were used to excite the valence electrons, $E_{\text{ph}} = 1.58$, 1.85 and 1.96eV, which give $E_1 = 0.15$ ($\equiv \epsilon_1$), 0.42 ($\equiv \epsilon_2$), and 0.53eV ($\equiv \epsilon_3$), respectively. Since the values of $E_0$ are smaller than $E_1$ by $\Delta = 0.35$eV, we expect $\Gamma_\uparrow(E_1) \gg \Gamma_\downarrow(E_0)$ unless the Schottky barrier is too low. The experimental results of the differential tunnel conductance $\Delta I/V_F$ show that the sign of $\Delta I/V_F$ for $E_{\text{ph}} = 1.85$eV is different from that for $E_{\text{ph}} = 1.85$ and 1.96eV, and that $\Delta I/V_F$ begins to decrease when $V_F \lesssim 0.2$V \cite{15}.

The tunnel conductance has been calculated by using a full-orbital tight-binding model; $s$, $p$ and $d$ orbitals for Fe, and $s$ and $p$ orbitals for GaAs. The hopping parameters are determined by fitting the calculated energy dispersion curves to those obtained by the other calculations \cite{21, 22}. The local density of states (DOS) at each layer and the tunnel conductance at an energy $E$ are calculated by using recursive Green’s function method. We calculate the tunnel conductance for an Fe(9)/Ga(9)(9)/Fe contacts, neglecting the mismatch of the lattice constants between Fe and GaAs.

We adopt a model in which the shape of the Schottky barrier (the position dependence of the bottom of the conduction band) is given by $E_C(\ell) = \Delta_S e^{-\ell/\lambda}$, where $\Delta_S$ and $\ell$ are the Schottky barrier height and the distance measured from the interface. The value of $\lambda$ is determined in such a way that $E_C(\ell) \to 10^{-4}$eV at $\ell = L_S$, the Fermi level $E_F$ of bulk GaAs is taken to be the bottom of the conduction band $E_C(\ell = L_S)$ assumed for highly doped n-type GaAs. The forward bias dependence is taken into account by shifting the GaAs bands by $eV_F$, i.e., $E_C(\ell) \rightarrow (\Delta_S - eV_F) e^{-\ell/\lambda} + eV_F$. Bias dependence of the barrier thickness is neglected since its effect is much smaller than that of the reduction of the effective barrier height. In the practical calculations, the Schottky barrier is included as a position-dependent shift of the atomic potential of Ga and As atoms. Calculated results of the tunnel conductance and spin polarization for incident electrons normal to the layer plane agree semi-quantitatively with those obtained in the first principles \cite{8}.

Figure 1(a) shows the calculated results of the spin resolved conductance $\Gamma_\sigma(\sigma = \uparrow, \downarrow)$ for an Fe-As contact with a Fe-As barrier with $L_S = 400ML$ and $\Delta_S = 0.8eV$. We see that $\Gamma_\uparrow$ increases nearly monotonically, while $\Gamma_\downarrow$ shows a sharp peak around $E - E_C = 0.4eV$. Therefore, the spin polarization of the tunnel conductance becomes negative in a specific energy window. $\Gamma_\uparrow$ is nearly constant for $E - E_C \gtrsim 0.8eV$ (not shown) until the energy $E$ touches the $\Delta_1$ band in the Fe minority spin states. When $E - E_F \sim 1eV$, $\Gamma_\uparrow$ increases rapidly as the $\Delta_1$ band of the Fe minority spin state begins to contribute the tunneling, resulting in an abrupt decrease of the spin polarization at the energy. Figure 1(b) shows the spin polarization of the tunnel conductance for various values of $\Delta_S$. We find that $P$ can be $\sim -100\%$ for a certain energy window, and that the energy window shifts in proportion to $\Delta_S$. It should be noted that the calculated conductance is less accurate when $E - E_C \sim 0$, since the thickness of the Schottky barrier at this energy region is too thick for numerical calculations.

Calculated results of $P$ for the Fe-As and Fe-Ga contacts with $L_S = 200ML$ are shown in Figs. 2(a) and 2(b), respectively. In Fig. 2(a) the negative spin polarization becomes less perfect when $L_S = 200ML$. This is because $L_S$ is small, and more states in the Fermi surface begin to contribute to the tunneling. Similar to the results for $L_S = 400ML$, the peaks of the negative $P$ shift to the lower energy region with increasing $\Delta_S$. Thin curves in Fig. 2(a) show the bias dependence of the spin polarization for the Fe-As contact with $L_S = 200ML$ and $\Delta_S = 0.5eV$. We see the energy window with negative $P$ shifts towards the lower energy region in proportion to the bias voltage $V_F$.

The energy dependence of $P$ for the Fe-Ga contact is essentially the same with that for the Fe-As contacts, however, there are a few differences to be noted: (i) The
energy windows for the negative $P$ are wider for the the Fe-Ga contacts than those for the Fe-As contacts. (ii) The negative spin polarization for the Fe-Ga contacts can always be perfect irrespective to $L_S$. (iii) Most importantly, a large value of $\Delta_S$ is necessary to realize the negative $P$ for the Fe-Ga contacts.

The above mentioned results can well be accounted for in terms of the IRSs in the Schottky barrier of the GaAs layer. Figure 3(a) presents the local DOS on the As and Ga layers at the Fe-As and Fe-Ga contacts, respectively, with the Schottky barrier of $L_S = 200ML$ and $\Delta_S = 0.5eV$. We find many sharp peaks appear in both the As and Ga local DOS, which may be identified to be the IRSs. These IRSs are spin dependent due to the hybridization with the spin-polarized Fe bands. The existence of an IRS at $E - E_C \sim 0.2eV$ in the $\uparrow$ spin state may explain the negative value of $P$ calculated for the Fe-As contact with these parameter values. As $\Delta_S$ increases, the IRS is shifted by nearly the same amount of the increase of $\Delta_S$ as shown in Fig. 3(b). These results are in good accordance with the shift of the energy window where $P < 0$. A schematic figure of the IRSs in the present model is shown in Fig. 3(c). When a forward bias $V_F$ is applied, the chemical potential of the GaAs layer (in other words, $E_C$) shifts by $eV_F$, and therefore the energy window of negative $P$ is shifted to the lower energy region by $\sim eV_F$ as shown in Fig. 2(a).

Since the IRSs are formed by an interference effect between the incident and reflected waves of the conduction band of GaAs at the interface, they are dominated by the $\Delta_1$ symmetry for the Fe/GaAs(001) interface. Therefore, they hybridize stronger with $\uparrow$ spin Fe bands which have the $\Delta_1$ symmetry band near $E_C$ than with $\downarrow$ spin Fe bands. Strong hybridization in the $\uparrow$ spin states pushes down (up) the bonding (anti-bonding) state of the IRSs, resulting in a weak intensity of the IRSs near $E_C$. The IRSs in the $\downarrow$ spin state with $k_\parallel \neq (0,0)$, where $k_\parallel$ is a momentum parallel to the layer plane, hybridize with the $\Sigma_1$ band of Fe mainly, and have rather strong intensity near $E_C$ as shown in Fig. 3(a). Although the IRSs are evanescent states, they make the effective barrier thickness thinner significantly, therefore giving rise to the negative $P$. It should also be noted the nature of the IRSs is changed with different layer stacking orientation, since the IRSs are symmetry dependent.

![Figure 2](image2.png)

**FIG. 2**: (color online) Calculated results of the spin polarization of the tunnel conductance as a function of an energy for (a) the Fe-As contact with $L_S = 200ML$ and various values of $\Delta_S$ at a bias voltage $V_F$, and for (b) the Fe-Ga contact with $L_S = 200ML$ and various values of $\Delta_S$ at zero bias. Insets of (a) and (b) are the bias dependence of $P$ at $E = E_C + 0.005eV$ for the Fe-As contact with $\Delta_S = 0.5eV$ and the Fe-Ga contact with $\Delta_S = 1.0eV$.

![Figure 3](image3.png)

**FIG. 3**: (color online) (a) Calculated results of the local DOS of As and Ga at the interface for the Fe-As and Fe-Ga contacts, respectively, with $\Delta_S = 0.5eV$, $L_S = 200ML$ and zero bias. (b) Enlargement of the $\downarrow$ spin local DOS for the Fe-As contact, and (c) a schematic figure of the resonant states in the Schottky barrier.

![Figure 4](image4.png)

**FIG. 4**: (color online) (a) Calculated results of the local DOS for the (a) $\uparrow$ and (b) $\downarrow$ spin states at $E - E_C = 0.2eV$, and (c) a momentum-resolved conductance for the $\uparrow$ and $\downarrow$ spin states for the Fe-As contact, where $k'$ indicates the momentum along $k_x - k_y$. Parameter values are $L_S = 200ML$ and $\Delta_S = 0.5eV$. 
Above consideration is justified by the calculated results of the \(k\)\(_F\)-resolved local DOS and conductance, which are shown in Fig. 4. Figures 4(a) and (b) are the local DOS of the ↑ and ↓ spin states of the As layer at the Fe-As contact. The both local DOS spread over the whole Brillouin zone, however, the ↓ spin local DOS is much larger than the ↑ spin one near \(k\| = (0, 0)\). Since the Schottky barrier is thick, the tunnel conductance is governed by the states near \(k\| = (0, 0)\), and as a result \(\Gamma_\uparrow\) becomes much larger than \(\Gamma_\downarrow\) as shown in Fig. 4(c). It should be noted that the \(\Gamma_\downarrow\) is precisely zero at \(k\| = (0, 0)\) by symmetry.

It should also be noted that all of the resonant states in the Fe minority spin state do not contribute to the tunneling via the IRSs formed at an Fe/GaAs(001) interface, since the former states may have \(\Delta_\downarrow\) symmetry, among which only \(p_x\) and \(p_y\) orbitals hybridize with the \(\Sigma_1\) band when \(k\| \neq (0, 0)\). We have confirmed that the resonant states in the Fe minority spin state (not shown) stay at almost the same energy position when \(\Delta_\downarrow\) is increased. Since it is difficult to explain the shift of the energy window proportional to \(\Delta_\uparrow\) by the resonant states in the Fe layer, it would less contribute to the origin of the negative spin polarization calculated here.

Now let us compare the calculated results with experimental ones. As mentioned, the experiments have used three excitation energies \(\varepsilon_1, \varepsilon_2\) and \(\varepsilon_3\), which are shown by vertical lines in Figs. 2(a) and (b). The experimental results suggest that the sign of the differential conductance \(\Delta I/V_F = (I_L - I_R)/V_F\) at \(E = \varepsilon_1\) is different from that at \(E = \varepsilon_2\) and \(\varepsilon_3\). One of the conditions which agree with the experimental observation is \(\Delta_\uparrow \sim 0.5eV\) for the Fe-As contact irrespective to the barrier thickness, where \(P(\varepsilon_1) < 0, P(\varepsilon_2) > 0\) and \(P(\varepsilon_3) > 0\). When the bias voltage is increased to 0.2V, \(P(\varepsilon_1)\) calculated changes the sign, and \(P(\varepsilon_1), P(\varepsilon_2)\) and \(P(\varepsilon_3)\) are all close to 1. The latter result may not agree with the experimental one in which \(|\Delta I/V_F|\) begins to decrease above \(V_F \sim 0.2V\). The discrepancy may be attributed to the quality of the Schottky barrier of the measured sample. The height of the Schottky barrier estimated experimentally for our sample is 0.23eV, and hence the electron conduction becomes metallic-like when \(V_F > 0.2V\), leading to a decrease of the spin polarization across the interface. In addition, the estimated barrier height 0.23eV can be the lower limit, assuming an in-plane barrier height distribution where lower barrier (less resistive) parts would dominate the electron transport property. Consequently, higher barrier regions in our junction would still give rise to spin-polarized tunneling, though its weight may decrease. Actually we observed no sign change in \(\Delta I\) for a sample with the lower barrier height of 0.1eV. We expect that the negative spin polarization of the spin-filtering effect should be clearly seen for high-quality samples with the higher barrier as estimated to be 0.46eV by Hanbicki et al.[2].

Since the value of \(P\) is strongly dependent on the energy, bias voltage as well as Schottky barrier height as shown in Figs. 2(a), (b) and the insets, the results could shed light on the enigmatic topic on \(P\) at Fe/GaAs interfaces mentioned in the introduction, and propose a feasible control of \(P\) at an FM/GaAs junction. The inevitable variation of Schottky barrier heights in experimental samples may explain the observed differences in bias dependence of \(P\) in these cases. Since the value of \(P\) varies from \(-100\%\) to \(+100\%\) with the bias voltage, the complete spin polarization tuning by the bias voltage can be realized in ideal FM/GaAs interfaces. Such devices should be promising since they require no overdoped layers nor complex structures with gate terminals for switching \(P\). The proposed spin-switch devices can operate in low bias voltage regions due to the switchings seen in the insets of Fig. 2. Control of the interface spin polarization with different Schottky barrier heights may also be possible by using FM alloys with different work functions as performed in FM/Si interfaces.

In conclusion, we have calculated the spin polarization of the tunnel conductivity using a full-orbital tight-binding model, and have shown that the interface resonant states within the Schottky barrier in the GaAs layer influence significantly the spin-dependent tunneling across the interface. It has been clearly shown that the band matching of the IRSs plays a crucial role on the spin polarization. The theoretical results account well for earlier experimental results including the tunneling of photo-excited electrons. The present results suggest that the spin polarization can be controlled by the Schottky barrier heights, and that a spin-switch device with bias control may also be promising. Quantitative performance of the device, however, needs more quantitative calculations including effects of atomic disorder for example.

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