

Spin polarization control through resonant states in an Fe/GaAs Schottky barrierS. Honda,¹ H. Itoh,² J. Inoue,¹ H. Kurebayashi,³ T. Trypiniotis,³ C. H. W. Barnes,³ A. Hirohata,⁴ and J. A. C. Bland³¹*Department of Applied Physics, Nagoya University, Nagoya 464-8603, Japan*²*Department of Pure and Applied Physics, Kansai University, Suita 564-8680, Japan*³*Cavendish Laboratory, University of Cambridge, J. J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom*⁴*Department of Electronics, University of York, Heslington, York YO10 5DD, United Kingdom*

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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 photoexcited 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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I. INTRODUCTION

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.¹⁻⁷ The spin-injection efficiencies measured in these experiments are impressive with the highest being 57% at 100 K ,⁶ but they are not as high as what first-principles band calculations predict.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,^{9,10} imaging of injected spins in FeCo/GaAs junctions,¹¹ measurements of tunnel magnetoresistance in Fe/GaAs/Fe junctions,¹² and spin-filtering experiments with photoexcited electrons produced in the GaAs layer.¹³ The bias dependence of negative P in these experiments, however, is still controversial as argued by Lou *et al.*,¹⁰ 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 region¹⁴ and interface resonant states (IRSs) appeared intrinsically in the minority-spin state of Fe.¹⁵ The extrinsic mechanism may be ruled out for Fe/GaAs junctions since the negative P appears in ideal interfaces.¹² As for IRS mechanism, Chantis *et al.*¹⁶ 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. On the other hand, Dery and Sham¹⁷ 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. In addition, Dery and Sham¹⁷ 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.^{12,13}

In this paper, 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 (\downarrow) 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 in 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 photoexcited electrons in the GaAs layer semiquantitatively but may resolve the controversy about the bias dependence of negative P observed. A strong variation in 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 photoexcited 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 principles⁸ 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.¹⁸ Although we perform our analysis to interpret the experimental results given by Kurebayashi *et al.*,¹³ we will present results for wide range of parameter values, estimate reasonable parameter values which reproduce the experimental results, and give predictions for transport properties in high-quality samples. It is also noted that the present work is an attempt to make comparison specifically between experiments and theoretical results obtained for sufficiently realistic models.

II. EXPERIMENTS AND THEORETICAL FORMALISMS

When we restrict our discussion to absolute zero 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,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 $P_{3/2}$ and split-off $P_{1/2}$ valence bands, respectively, where E_{ph} , E_g , and Δ 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

$$\Delta I \equiv I_L - I_R \sim 2[P(E_1)\Gamma(E_1) - P(E_0)\Gamma(E_0)]V_F, \quad (1)$$

where

$$P(E) = [\Gamma_\uparrow(E) - \Gamma_\downarrow(E)] / [\Gamma_\uparrow(E) + \Gamma_\downarrow(E)] \quad (2)$$

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 1.96 eV, which give $E_1 = 0.15 (= \varepsilon_1), 0.42 (= \varepsilon_2),$ and 0.53 eV ($= \varepsilon_3$), respectively. These photon energies are those used in our experiment.¹³ Since the values of E_0 are smaller than E_1 by $\Delta = 0.35$ eV, we expect $\Gamma(E_1) \gg \Gamma(E_0)$ 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 $E_{\text{ph}} = 1.58$ eV is different from that for $E_{\text{ph}} = 1.85$ and 1.96 eV and that $|\Delta I / V_F|$ begins to decrease when V_F exceeds 0.2 V.¹³

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.^{20,21} 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/GaAs(001) interface with both Fe-As and Fe-Ga contacts, neglecting the mismatch of the lattice constants between Fe and GaAs since both contacts might be realized at an Fe/GaAs interface due to imperfections of the interface. In the end, we will show that the results for Fe-Ga contact are unreasonable to explain the experimental results, and therefore, we concentrate mainly on the results for Fe-As contact. It might be noted, however, that Fe/GaAs samples are often fabricated under an excess As pressure in chambers, and in that case Fe-As contacts may probably realized.

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 Δ_S and ℓ are the Schottky barrier height and the distance measured from the interface. A schematic figure of the contact and the shape of the Schottky barrier are shown in Figs. 1(a) and 1(b), respectively. The value of λ is determined in such a way that $E_C(\ell)$ becomes 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

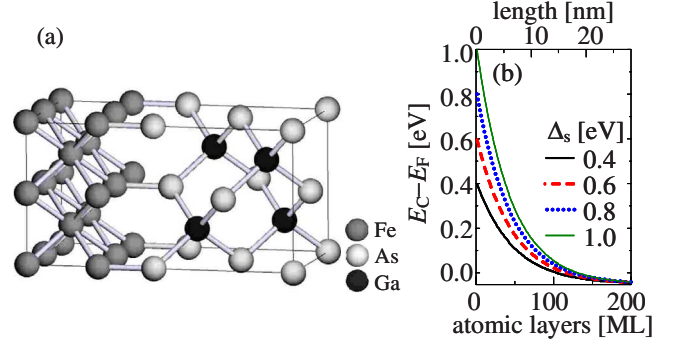


FIG. 1. (Color online) (a) A schematic figure of the contact of Fe and GaAs layers, in which Fe atoms contact with Ga atoms at the interface. (b) Model of the Schottky barrier used in the calculation. Here, E_C denotes the bottom of the conduction band.

$= E_C(\ell > L_S)$ assumed for highly doped n -type GaAs. An effective width of the Schottky barrier may be estimated from the shape of the Schottky barrier shown in Fig. 1(b).

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 in 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 semiquantitatively with those obtained in the first principles.⁸

III. THEORETICAL RESULTS

A. Spin polarization of conductance

Figure 2(a) shows the calculated results of the spin-resolved conductance Γ_σ ($\sigma = \uparrow, \downarrow$) for an Fe-As contact with a Schottky barrier with $L_S = 400$ ML and $\Delta_S = 0.8$ eV. We adopt the value of Δ_S since it is a value estimated from the

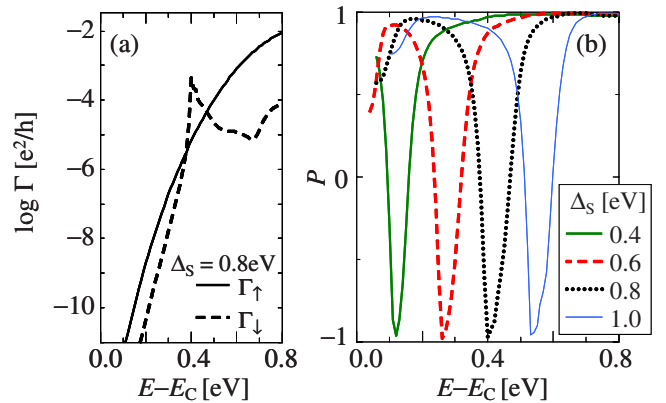


FIG. 2. (Color online) (a) Calculated results of conductance as a function of an energy for the Fe-As contact with $\Delta_S = 0.8$, $L_S = 400$ ML, and zero bias and (b) those of the spin polarization for various values of Δ_S . Here $E - E_C$ denotes the energy of photoexcited electrons measured from the band bottom E_C of the conduction band.

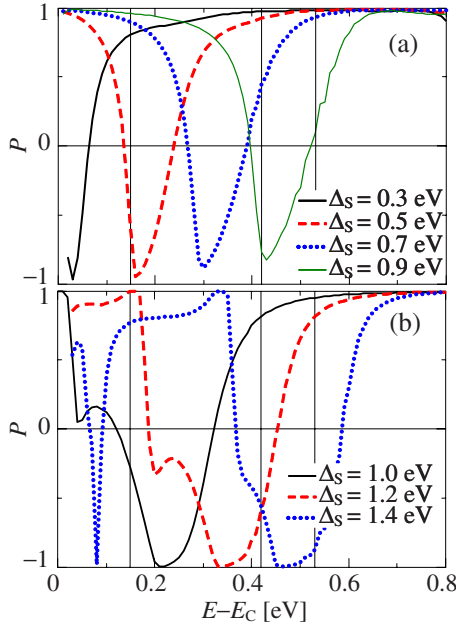


FIG. 3. (Color online) Calculated results of the spin polarization of the tunnel conductance as a function of an energy for (a) the Fe-As contact and (b) the Fe-Ga contact with $L_S=200$ ML and various values of Δ_S . The vertical lines correspond to the energy $E-E_C=E_{ph}-E_{gap}$ for three photon energies E_{ph} used in experiments.

difference of work functions between Fe and GaAs. We see that Γ_{\uparrow} increases nearly monotonically, while Γ_{\downarrow} shows a sharp peak around $E-E_C=0.4$ eV. Therefore, the spin polarization of the tunnel conductance becomes negative in a specific energy window. Γ_{\downarrow} is nearly constant for $E-E_C \geq 0.8$ eV (not shown) until the energy E touches the Δ_1 band in the Fe minority-spin states. When $E-E_F \sim 1$ eV, Γ_{\downarrow} increases rapidly as the Δ_1 band of the Fe minority-spin state begins to contribute the tunneling, resulting in an abrupt decrease in the spin polarization at the energy. Figure 2(b) shows the spin polarization of the tunnel conductance for various values of Δ_S calculated using expression (2). We find that P can be $\sim -100\%$ for a certain energy window and that the energy window shifts in proportion to Δ_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 $L_S=200$ ML are shown in Figs. 3(a) and 3(b), respectively. In Fig. 3(a) the negative spin polarization becomes less perfect when $L_S=200$ ML. 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 $L_S=400$ ML, the peaks of the negative P shift to the lower energy region with decreasing Δ_S . We see that the calculated result of P for $\Delta_S=0.3$ eV, which is close to a Schottky barrier height of 0.23 eV estimated from I - V relation, disagree with any experimental results at zero bias. The reason will be discussed later.

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 Fe-Ga contacts

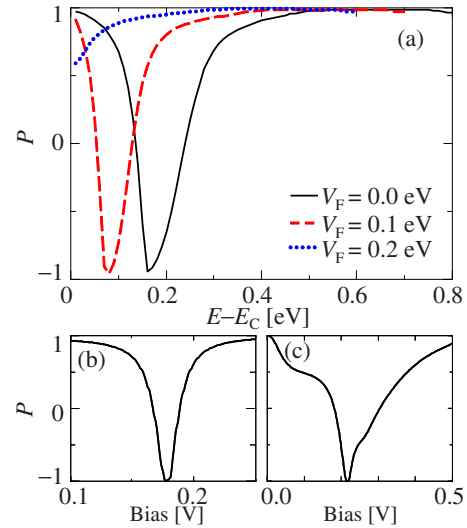


FIG. 4. (Color online) (a) Calculated results of spin polarization P as a function of an energy with various forward bias voltages V_F for an Fe-As contact with $\Delta_S=0.5$ eV and $L_S=200$ ML and bias dependence of P at $E=E_C+0.005$ eV for (b) an Fe-As contact $\Delta_S=0.5$ eV and (c) an Fe-Ga contact with $\Delta_S=1.0$ eV.

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 , and (iii) most importantly, a large value of Δ_S is necessary to realize the negative P for the Fe-Ga contacts. It is also noted that a value of $\Delta_S=1.4$ eV is unreasonably large for Fe/GaAs junctions.

Calculated results of the voltage dependence of P are shown in Fig. 4 for parameter values which give a reasonable agreement between the theoretical and experimental results. Figure 4(a) shows the energy dependence of P for various values of forward bias voltage V_F for the Fe-As contact with $L_S=200$ ML and $\Delta_S=0.5$ eV. We see the energy window with negative P shifts toward the lower energy region in proportion to the bias voltage V_F . Figures 4(b) and 4(c) show the bias dependence of P at $E=E_C+0.005$ eV for an Fe-As contact $\Delta_S=0.5$ eV and an Fe-Ga contact with $\Delta_S=1.0$ eV, respectively. In both cases, the results show a rapid change in P with increasing the bias voltage. It should be noted that no energy integral between 0 and V_F is included in the calculation since the tunneling of electrons at the bottom of the conduction band is assumed in the present model.

B. Interfacial resonant states

The above mentioned results can well be accounted for in terms of the IRSs in the Schottky barrier of the GaAs layer. Figure 5(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 $L_S=200$ ML and $\Delta_S=0.5$ eV. We find many sharp peaks appear in both the As and Ga local DOSs, 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 $E-E_C \sim 0.2$ eV in the \downarrow spin state may explain the negative value of P calculated for the Fe-As contact with these parameter values. As Δ_S

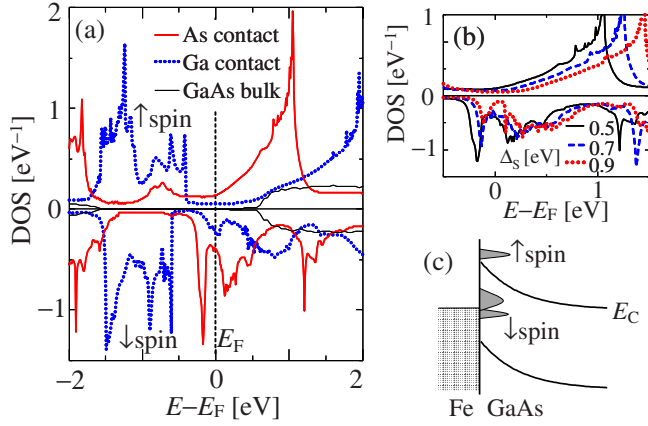


FIG. 5. (Color online) (a) Calculated results of the local DOS of As and Ga at the interface for the Fe-As and Fe-Ga contacts, respectively, with $\Delta_S=0.5$ eV, $L_S=200$ ML, and zero bias. (b) Enlargement of the \downarrow spin local DOS for the Fe-As contact and (c) a schematic figure of the resonant states in the Schottky barrier.

increases, the IRS is shifted by nearly the same amount of the increase in Δ_S as shown in Fig. 5(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. 5(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. 3(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 Δ_1 symmetry for the Fe/GaAs(001) interface. Therefore, they hybridize more strongly with \uparrow spin Fe bands which have the Δ_1 symmetry band near E_C than with \downarrow spin Fe bands. Strong hybridization in the \uparrow spin states pushes down (up) the bonding (antibonding) 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 Σ_1 band of Fe mainly and have rather strong intensity near E_C as shown in Fig. 5(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 that the nature of the IRSs is changed with different layer stacking orientations since the IRSs are symmetry dependent.

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 Δ_5 symmetry, among which only p_x and p_y orbitals hybridize with the Σ_1 band when $k_{\parallel} \neq (0,0)$. We may confirm that the resonant states in the Fe minority-spin state stay at almost the same energy position when Δ_S is increased as shown in Fig. 6. For comparison, the bulk DOS of Fe and the local DOS at the surface of Fe are shown in the inset. We see the peaks of DOS near $E=0$ are caused by the interfacial states at the Fe surface. Since it is difficult to explain the shift of the energy window proportional to Δ_S by the resonant states in the Fe layer, it would less contribute to

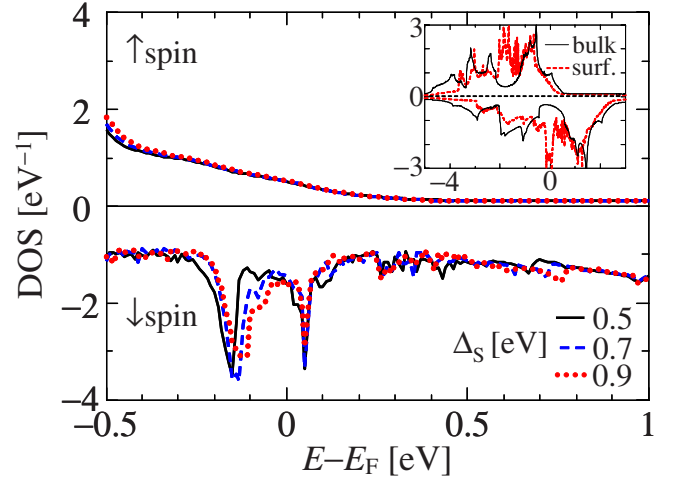


FIG. 6. (Color online) (a) Calculated results of the local DOS of Fe at the interface for the Fe-As contacts with $\Delta_S=0.5$ eV, $L_S=200$ ML, and zero bias. The inset is the bulk DOS of Fe and a local DOS at a surface.

the origin of the negative spin polarization calculated here.

Above consideration is justified by the calculated results of the k_{\parallel} -resolved local DOS and conductance, which are shown in Fig. 7. Figures 7(a) and 7(b) are the local DOS of the \uparrow and \downarrow spin states of the As layer at the Fe-As contact. Both local DOSs spread over the whole Brillouin zone; however, the \downarrow spin local DOS is much larger than the \uparrow spin one near $k_{\parallel}=(0,0)$. Since the Schottky barrier is thick, the tunnel conductance is governed by the states near $k_{\parallel}=(0,0)$, and as a result Γ_{\downarrow} becomes much larger than Γ_{\uparrow} as shown in Fig. 7(c). It should be noted that Γ_{\downarrow} is precisely zero at $k_{\parallel}=(0,0)$ by symmetry.

C. Comparison between experimental and theoretical results and perspective

Now let us compare the calculated results with experimental ones. As mentioned, the experiments have used three excitation energies ε_1 , ε_2 , and ε_3 , which are shown by vertical lines in Figs. 3(a) and 3(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 ε_3 .

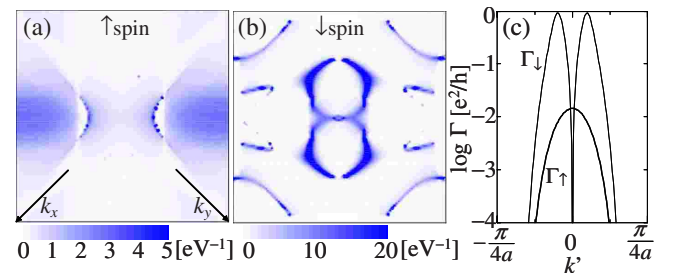


FIG. 7. (Color online) Momentum-resolved density of states for the (a) \uparrow and (b) \downarrow spin states at $E-E_C=0.2$ eV and (c) momentum-resolved conductance for the \uparrow and \downarrow spin states for the Fe-As contact, where k' indicates the momentum along k_x-k_y . Parameter values are $L_S=200$ ML and $\Delta_S=0.5$ eV.

One of the conditions which agree with the experimental observation is $\Delta_S \sim 0.5$ eV 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 0.2 V, $P(\varepsilon_1)$ 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 $V_F \sim 0.2$ V. 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 0.23 eV, and hence, the electron conduction becomes metalliclike when $V_F > 0.2$ V, leading to a decrease in the spin polarization across the interface. In addition, the estimated barrier height, 0.23 eV, can be the lower limit, assuming an in-plane barrier height distribution where lower barrier (less resistive) parts would predominate the electron-transport properties. Consequently, higher barrier regions in our junction would still give rise to spin-polarized tunneling, although its weight may decrease. Actually we observed no sign change in ΔI for a sample with the lower barrier height of 0.1 eV. 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 0.46 eV by Hanbicki *et al.*² The value of $\Delta_S \sim 0.5$ eV is also close to a value estimated by work functions of Fe and GaAs.

Since the value of P is strongly dependent on the energy, bias voltage, as well as Schottky barrier height as shown in Figs. 4(b) and 4(c), the results could shed light on the enigmatic topic on P at Fe/GaAs interfaces mentioned in Sec. I and propose a feasible control of P at an FM/GaAs junction. The inevitable variation in 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 neither overdoped layers nor complex structures with gate terminals for switching P .¹⁷ The proposed spin-switch devices can operate in low bias voltage regions due to the switching seen in the insets of Fig. 4(b) for Fe-As contacts. 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.²²

IV. CONCLUSION

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 photoexcited 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.^{23,24}

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