Magnetoresistance in ferromagnetic-metal/graphene/ferromagnetic-metal lateral junctions

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(Received 29 May 2010; published 8 July 2010)

Based on a numerical study of a full-orbital tight-binding model, we show that magnetoresistance (MR) in ferromagnetic metal (FM)/graphene/FM lateral junctions originates from a spin-dependent shift of the Dirac points (DPs) caused by the presence of FM electrodes. Graphene with finite length L is in contact with FM at a zigzag edge and the spin-dependent conductance is calculated in the ballistic transport regime. We find that the tunneling conductance near DPs influences the features of the MR considerably. We further show that a slight change in the electronic structure near the DPs strongly affects the spin-dependent conductance resulting in a large MR ratio for an FM made of certain ferromagnetic transition metal alloys.

DOI: 10.1103/PhysRevB.82.033402 PACS number(s): 72.80.Vp, 73.22.Pr, 85.75.−d

Graphene, a two-dimensional honeycomb lattice composed of carbon atoms, has attracted much interest after the discovery of a novel fabrication method and the material's characteristic transport properties,[1–4] which are caused by a linear dispersion relation near the Fermi level. The states at which the conduction and valence bands meet are called Dirac points (DPs); at these points, electrons behave as massless Dirac fermions in the graphene (G). The massless electrons give rise distinctive features of electrical transport unlike those in conventional metals and semiconductors. The two-dimensionality of graphene offers advantage over other materials in gate controllability of the current. The long spin-diffusion length caused by a negligibly small spin-orbit interaction in carbon is promising for magnetoresistance (MR) devices. Thus, graphene has a huge potential for application to next-generation devices.

Many theoretical studies have been performed to clarify the characteristics of electronic transport in graphene,[5] coherent transport in finite-size graphene,[6–8] disordered effects,[9,10] and tunable conductance with a gate,[11] in addition to a study of electronic states with metallic contacts.[12] Novel mechanisms for the MR effect have also been proposed theoretically, use of half-metallicity at the zigzag edges of graphene nanoribbons[13,14] and a valley valve that uses a difference in the symmetry of two DPs in the Brillouin zone.[15] Experimentally, the spin diffusion length in graphene has been successfully measured to be $1-1.5$ μm by using nonlocal four-terminal devices.[16] More recently, a longer spin-diffusion length of 8 μm has been reported.[17] MR has also been observed in conventional two-terminal graphene junctions,[18,19] as well as in nonlocal measurement.[20] Nevertheless, MR in two-terminal junctions remains to be studied because the reported MR ratios are not large enough for device applications and no clear mechanism for the MR has been presented. Therefore, theoretical study of MR in two-terminal G junctions with ferromagnetic (FM) electrodes (FM/G/FM junctions) is desirable to clarify the mechanism and to provide a guiding principle for junction design.

In this Brief Report, we first demonstrate a simple mechanism of MR in matching/mismatching of conduction channels, and then propose a mechanism of MR using a realistic tight-binding (TB) model. In the latter mechanism, spin-dependent shift of effective DPs caused by band mixing between the graphene and the electrodes plays an important role. That is, the MR originates from a conflict between the DP shift in the up- and down-spin states in the antiparallel alignment of the FM magnetization. The MR ratio is independent of the graphene length because of a characteristic dependence of the tunneling conductance via states near the DP. The mechanism of the MR effect is different from those previously described,[14,15] in that it is caused by contact with the electrodes. We further show that a huge MR effect may appear in junctions with electrodes made of FeCo, FeCr, and FeV alloys. The result is attributed to a suppression of conducting states in the majority spin state near the junction contacts, which indicates matching between DPs and conduction channel of electrodes becomes worse in the spin state.

Schematics of lateral FM/G/FM junctions are shown in Fig. 1(a). We adopt a zigzag edge contact in the junction, as shown in Fig. 1(b), because the contact structure is easily modeled, as shown in the figure. It is also expected that the zigzag contact may strongly affect the electronic states of the graphene. Two models are adopted for the electrodes: one is a thin film with bcc structure [Fig. 1(c)], typically an Fe film (Fe/G/Fe junction) with s, p, and d orbitals, and the other is a square lattice with a single s orbital [s/G/s junction, Fig. 1(d)]. A periodic boundary condition is used for the junction

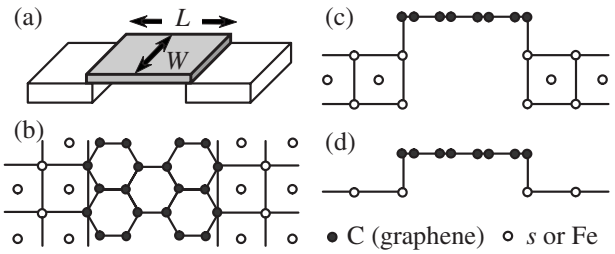

FIG. 1. (a) Schematic of a junction, (b) top view of a junction with electrodes made of bcc Fe, (c) side view of a bccFe/graphene/bccFe junction, and (d) side view of a junction with electrodes made of a square lattice.

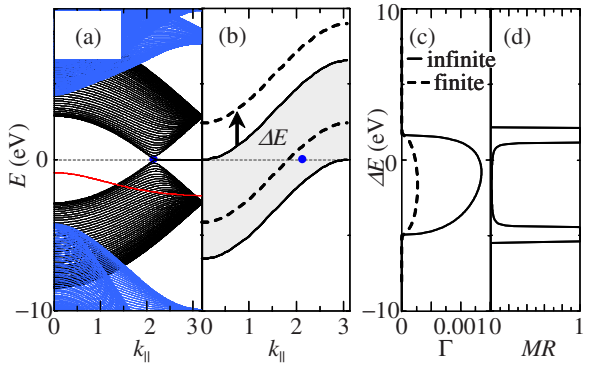

FIG. 2. (Color online) (a) Electronic structure of an isolated graphene sheet with zigzag edges, (b) band structure of a simple square lattice with an s orbital, and calculated results of (c) conductance, and (d) MR ratio for $s/\mathrm{G}/s$ junctions with a shift ΔE of the s band of the right electrode. $k_\parallel$ at Dirac point is shown by a dot. Γ is in units of e^2/h per atom.

width. The graphene length L is taken to be less than $\sim$20 000 monolayers (ML) typically 1000 ML, where the number is taken in such a way that one zigzag chain corresponds to two MLs. No roughness is included in the junction, and therefore the momentum $k_\parallel$ parallel to the contact line is conserved.

The band parameters of the TB model we use are taken from Harrison's textbook.[21] The parameters for graphene are $ss\sigma=-4.926$ eV, $pp\pi=-2.850$ eV, and $sp\sigma=6.474$ eV. Nearest-neighbor hopping in the square lattice is taken to be $t_s=ss\sigma/3$, and the hopping parameter between graphene and the electrodes is assumed to be $t_I\equiv\alpha\times sp\sigma$, where α is a parameter.

Figure 2(a) shows the energy dispersions $E(k_\parallel)$ calculated for an isolated graphene sheet with $L=1000$ ML in the sp^3 TB model. The DP (the K' point in this case) appears at $k_\parallel \simeq 2.094/\pi a$ indicated by a dot in the figure, where a is the lattice constant of the graphene. An edge state also appears at $E=0$ for $k_\parallel > 2.094/\pi a$. The figure shows not only the π band but also the σ bands. We confirmed that an interface (in-gap) state of the σ band appearing around $E=-1.5$ to -2.5 eV would not affect the results given below. Figure 2(b) shows the energy dispersion of an infinite square lattice used for the electrode.

We begin by presenting a simple picture of MR in $s/\mathrm{G}/s$ junctions. The conductance calculated for the junction, $\Gamma=\Sigma_{k_\parallel}\Gamma_{k_\parallel}/N_k$ is finite because states near the DP contribute to tunneling currents, as explained below. However, this is correct only when the K' point is included in the available $k_\parallel$ states of the electrodes. Values of Γ calculated for $s/\mathrm{G}/s$ junctions with $L=1000$ ML, $\alpha=0.1$ are shown in Fig. 2(c) as a function of the band shift ΔE of the right electrode. Here, ΔE is a parameter representing the position of the conduction band in the right electrode relative to that in the left electrode. This is a demonstration that Γ is finite only in a certain energy range of the band. The dotted curve in Fig. 2(c) shows the conductance for graphene with a finite size, $W=102$ ML. We see that, like junctions with a periodic boundary condition, the finite-width graphene junctions also show finite conductance.

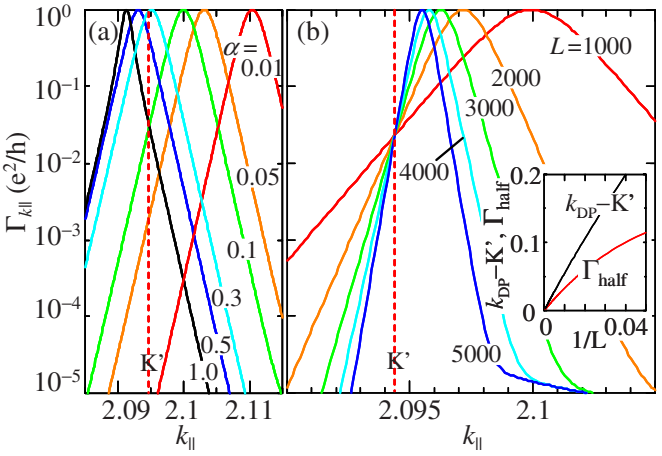

FIG. 3. (Color online) Calculated results of conductance resolved by momentum parallel to the interface (a) with various values of interlayer hopping integrals and with $L=1000$ ML and (b) various values of L.

MR appears when the electrodes are ferromagnetic and ΔE is spin dependent. The MR ratio is given in Fig. 2(d), in which $\Delta E\rightarrow\Delta E-(+)0.5\times sp\sigma$ for up ($\uparrow$) spin (down ($\downarrow$) spin) state. Here, the MR ratio is defined as $MR=(\Gamma_\mathrm{P}-\Gamma_\mathrm{AP})/(\Gamma_\mathrm{P}+\Gamma_\mathrm{AP})$, where Γ_P and Γ_AP are the conductance with parallel (P) and antiparallel (AP) alignment of the electrode magnetization, respectively. Note that in AP alignment, the band shifts of $\uparrow$ and $\downarrow$ spin states are reversed. We see that the MR ratio is either 1 or 0, as expected. The MR effect appears only when the K' point is included in either up- or down-spin conduction state of the electrode. In other words, a matching/mismatching of the conduction pass between the graphene and the leads is essential for finite MR in this case.

The simple mechanism explained above may not work for electrodes with realistic FMs because the spin splitting of bands in realistic FMs is not as large as demonstrated above. So we study the relationship between the conductance and electronic states near the DP putting an emphasis on the role of the band mixing between the graphene and the electrodes.

Figure 3(a) shows the $k_\parallel$-resolved conductance $\Gamma_{k_\parallel}$ calculated for $s/\mathrm{G}/s$ junctions with various values of α and with $L=1000$ ML. We find that the calculated conductance per spin takes the quantum conductance e^2/h at a certain $k_\parallel(\equiv k_\mathrm{DP})$ point away from K' shown by a broken line in the figure, and that $\Gamma_{k_\parallel}$ shifts with α. We identify the state with $\Gamma_{k_\parallel}=e^2/h$ at k_DP as an effective DP. We attribute the shift of the effective DP to band mixing between the s band of the electrodes and the π band of the graphene, after calculating the details of the electronic states in finite-size junctions. With $\alpha\rightarrow 0$, the graphene is isolated and k_DP goes to a value slightly larger than the bulk value.

Note that shape of $\Gamma_{k_\parallel}$ has a finite width. The width is determined by the *tunneling of electrons* through the graphene and is related to the tunneling probability, which is $T(k_\parallel,L)\propto\exp[-cL(k_\parallel-k_\mathrm{DP})]$ near the effective DP. By integrating $T(k_\parallel,L)$ over $k_\parallel$, we get the well-known result $\Gamma\propto 1/L$.[5,7] Moreover, $\Gamma_{k_\parallel}$ depends on the length L, as shown in Fig. 3(b). We find that both the relative shift of the DP from the K' point and the half-width of $\Gamma_{k_\parallel}$ are proportional to

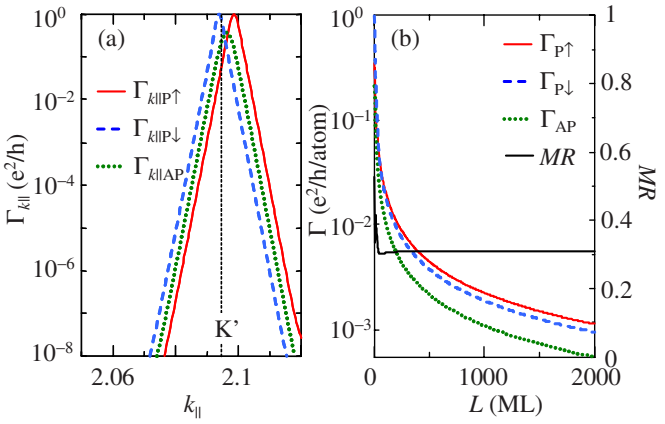

FIG. 4. (Color online) (a) Momentum-resolved conductance $\Gamma_{k_\|}$ for parallel and antiparallel alignment of Fe magnetization and (b) Γ and MR ratio as functions of graphene length L.

$1/L$, as shown in the inset of Fig. 3(b). An L-independent conductance at K' can be explained by noting $T(K',L)$ is independent of L since $K'-k_{DP} \propto 1/L$.

Now we propose a possible mechanism for MR in graphene junctions using spin-polarized s, p and d bands in Fe electrodes. Since band mixing at the contacts is generally spin dependent, we expect the shift of the DP to become spin dependent and MR to appear. We show below that MR does appear in Fe/G/Fe junctions.

The structure of an Fe/G/Fe junction is shown in Figs. 1(b) and 1(c). The bcc Fe layer is a thin film with three atomic layers. Figure 4(a) shows the calculated results of the $\Gamma_{k_\|}$ with P and AP alignments of Fe magnetization in a junction with $L=1000$ ML. We find that the up ($\uparrow$) spin conductance in the P alignment $\Gamma_{k_\|P\uparrow}$ shifts from the K' point more than the down ($\downarrow$) spin conductance $\Gamma_{k_\|P\downarrow}$ does. The shift in $\Gamma_{k_\|}$ is attributed to spin-dependent band mixing at the contact, as explained above. In the AP alignment, on the other hand, the maximum value of Γ_{AP} is smaller than e^2/h. (Note that Γ is plotted in logarithmic scale.) This is because the spin dependence of the hopping integrals of the left and right contacts is reversed in the AP alignment, and the $\uparrow$ and $\downarrow$ spin shifts of the DP point conflict with each other in the graphene junction.

As a result, MR occurs in FM/G/FM junctions, as shown in Fig. 4(b), in which the L dependence of $\Gamma_{P\uparrow(\downarrow)}$, Γ_{AP}, and the MR ratio are presented. The conductance decays in proportion to $1/L$, as expected, and the spin dependence is maintained independently of L; therefore, the MR ratio is constant except for very short L. The latter result is attributed to the L dependence of $\Gamma_{k_\|}$ shown in Fig. 3(b): both the shift of the DP and the half-width of $\Gamma_{k_\|}$ are proportional to $1/L$. This is a characteristic of the linear dispersion near the DP.

The MR obtained for Fe/G/Fe junctions is rather moderate because it originates from the spin dependence of the hopping integrals at the contacts. It would be interesting, however, to examine whether a large MR ratio can be realized in any ferromagnetic alloys due to spin-dependent matching/mismatching of the conductive states between graphene and electrodes, as explained for the simple MR mechanism. In

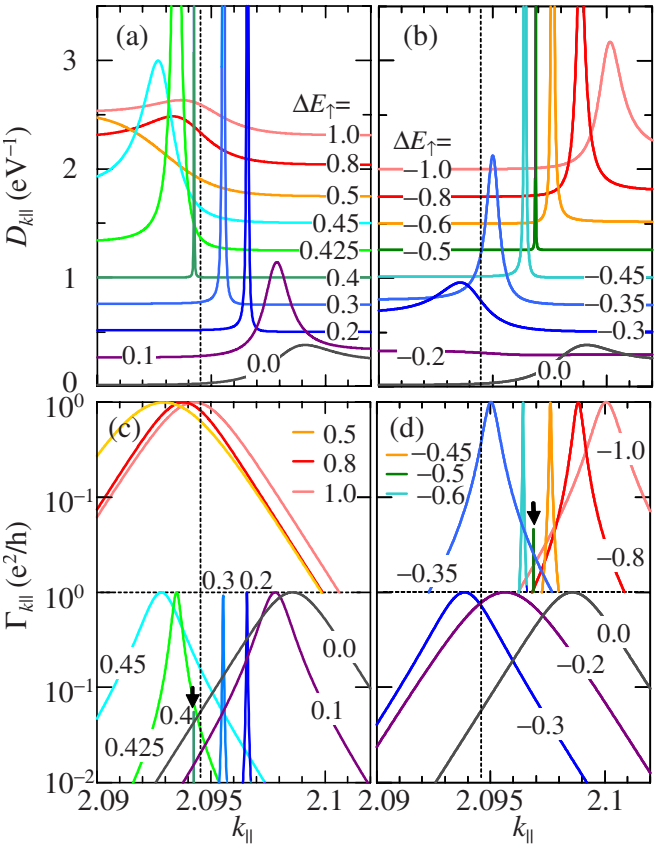

FIG. 5. (Color online) Calculated momentum-resolved local DOS of Fe at an interface obtained by shifting the up-spin band of Fe for (a) $\Delta E_\uparrow > 0$ and (b) $\Delta E_\uparrow < 0$: (c) and (d) show the corresponding conductance values.

Fe-based ferromagnetic alloys, a rigid band model may be applicable: the electronic structure of an Fe alloy is given by shifting the up-spin Fe bands by $\Delta E_\uparrow$ (in electron volt) while keeping the down-spin bands unchanged. For example, FeCo alloy corresponds to $\Delta E_\uparrow < 0$, and FeCr or FeV corresponds to $\Delta E_\uparrow > 0$.

Calculated results of $k_\|$-resolved local density of states (DOS), $D_{k_\|}(E=E_F)$, at the contact with the electrode are shown in Figs. 5(a) and 5(b) for $\Delta E_\uparrow > 0$ and <0, respectively. We find that the shape of $D_{k_\|}$ changes systematically with $\Delta E_\uparrow$ and becomes extremely sharp around $\Delta E_\uparrow = -0.5$ and 0.2–0.4. The corresponding results for $\Gamma_{k_\|\uparrow}$ are shown in Figs. 5(c) and 5(d) in a logarithmic scale. As expected, $\Gamma_{k_\|\uparrow}$ shifts with $\Delta E_\uparrow$ since the hopping parameter at the contact depends on the band structure at E_F, and $\Gamma_{k_\|\uparrow} \propto e^{-cL(k_\|-k_{DP})}$. However, $\Gamma_{k_\|\uparrow}$ becomes extremely small around $\Delta E_\uparrow = -0.5$ and 0.2–0.4, as pointed by arrows in the figure. Because of a strong correlation between $D_{k_\|}$ and $\Gamma_{k_\|\uparrow}$, the change in $\Gamma_\uparrow(k_\|)$ shown in Figs. 5(c) and 5(d) may be attributed to a change in the local DOS at the contact. $D_{k_\|}$ of the leads nearly vanishes for $\Delta E_\uparrow \sim -0.5$ and 0.2–0.4 in the region of $k_\|$ near K'. The sharp peaks in $D_{k_\|}$ may be caused by mixing with the graphene states.

The negligibly small local DOS at certain momentum states may result in huge MR because the matching of the

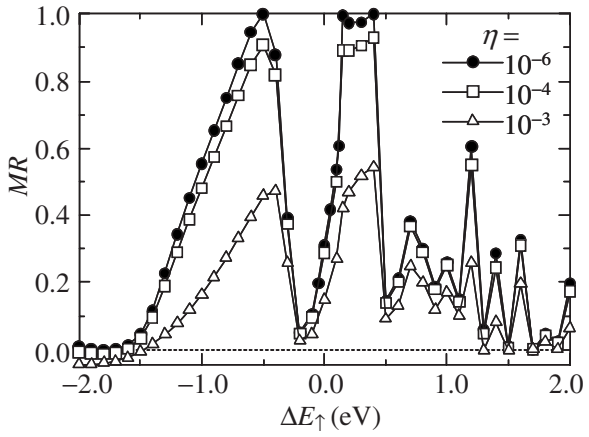

FIG. 6. Calculated results of MR as a function of the shift in the up-spin Fe bands. η is an imaginary part of the energy (in electron volt) and shows lifetime broadening.

conductive states in the graphene and the electrodes becomes worse. Figure 6 shows the calculated MR ratio as a function of $\Delta E_{\uparrow}$. We see that a large MR occurs for $0.15 < \Delta E_{\uparrow} < 0.4$ and at around $\Delta E_{\uparrow} \sim -0.5$. The former corresponds to $Fe_{0.9}Cr_{0.1}$ alloy and the latter to $Fe_{0.7}Co_{0.3}$. Since these alloys are ferromagnetic with well-defined magnetic moments, and the electronic states of down-spin electrons are not very different from those of Fe, large MR is expected to occur in graphene junctions.

Several issues remain to be studied; effects of doping and roughness[8–10] on the conductance and MR, transport properties of graphene junctions with an energy gap,[22–24] such as a double-layer graphene,[25] realistic graphene/metal contacts,[12] and effects of hydrogen absorption. Roughness may give rise to nonconservation of $k_{\parallel}$, breaking the matching/mismatching condition of the conductive states. In fact, a lifetime broadening effect decreases the MR, as shown in Fig. 6. Our preliminary results for more realistic contacts with finite overlap of graphene with electrodes with a triangular or fcc lattice show basically the same tendency as those shown in Fig. 3. Details will be reported elsewhere.

In conclusion, we have shown that MR appears in FM/G/FM junctions with zigzag-edged contacts due to a spin-dependent shift of the Dirac points caused by spin-dependent band mixing between the graphene and the electrodes. The MR is independent of graphene length because of a characteristic dependence of the tunneling conductance near a Dirac point. We further suggest that the MR can be quite large for FM electrodes made of certain ferromagnetic transition metal alloys because their electronic structure is strongly modified at the interface. The electronic states of both electrodes and graphene near the contacts are crucial for the MR effect.

This work was supported by a Grant-in-Aid for Scientific Research in Priority Area "Creation and control of spin current" from MEXT (Japan) and the Next-Generation Supercomputing Project, Nanoscience Program, MEXT (Japan).

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