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Effect of Carrier Differences on Magnetoresistance in Organic and Inorganic Spin Valves *

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Magnetoresistance in the structure of ferromagnetic/nonmagnetic/ferromagnetic spin valves are studied theoretically from the spin diffusion theory and Ohm's law. The nonmagnetic layer could be an organic or inorganic semiconductor. Carrier mobility and the spin-flip time in organic semiconductors are different from those in inorganic semiconductors, and effects of these differences on the magnetoresistance in organic and inorganic spin valves are discussed. From the calculation, it is found that the magnetoresistance in inorganic spin valves is higher than that in organic spin valves. Effects of the conductivity matching and spin-dependent interfacial resistances between ferromagnetic and nonmagnetic layers, thickness of the nonmagnetic layer, and the bulk spin polarization of the ferromagnetic layer on the magnetoresistance are also discussed.

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Spintronics and spintronic devices are currently hot topics in condensed matter physics, semiconductor materials and devices.[1−4] The main purpose of spintronics is to achieve some special functions in devices by using not the conventional electron's electricity but its spin. Spin injection is a natural way to combine spin dynamics and spin transport. In spin injection, there is no need to excite spin-polarized electrons using a magnetic field or radiation, the only thing one needs is a ferromagnetic electrode which is used as the source of the spin-polarized carriers. The sandwich structure of ferromagnetic/nonmagnetic/ferromagnetic (FM/NM/FM) is the basic structure to achieve spin injection and spin transport in NM materials. The magnetic area moments of the two FM layers will be parallel or antiparallel to each other if their coercive fields are different, so this sandwich structure can be used as a spin valve. The resistance of the spin valve will be changed if the magnetization alignments of the two FM layers are changed, which is called magnetoresistance (MR). The NM interlayer in the sandwich structure could be a superconductor, metal, inorganic semiconductor (ISC) or organic semiconductor (OSC). Spin polarized injection and transport in semiconductors is the emphasis of spintronics because semiconductor devices have the function of signal amplification. OSCs are one kind of the candidate materials of spin transport based on their weak spin-orbit coupling and weak hyperfine interaction, and organic spintronics has been an important branch of spintronics.[5−7]

MR in inorganic spin valves (ISVs) is an interesting research field of spintronics and has been studied and used widely in industry.[1−4] However, unlike ISVs there is a long way to go before OSVs' practical application. There has been much research on how to gain a giant organic MR which is large enough to be used. In 2002, Dediu *et al.*[8] first reported a negative MR across organic material T6. Xiong *et al.* have also observed spin injection and transport in the structure of $La_{0.7}Sr_{0.3}MnO_3/Alq_3/Co$ OSV,[9] remarkable MR could be obtained at low temperature. Santos *et al.*[10] observed that significant tunnel MR can be measured in a $Co/Al_2O_3/Alq_3/NiFe$ magnetic tunnel junction at room temperature, which increased when cooled to low temperatures. Drew *et al.*[11] presented a new depth-resolved technique in an OSV and found that the temperature dependence of the measured spin diffusion length was correlated with the device MR. Sun *et al.*[12] deposited nanodots onto an organic layer using buffer layer assisting growth and this OSV exhibited a giant MR of up to 300%. Barraud *et al.*[13] fabricated nanometrescale $(La,Sr)MnO_3/Alq_3/Co$ magnetic tunnel junctions and the OSV also exhibited an MR response of up to 300%. Theoretically, some differences of OSCs and ISCs were considered when people studied MR in OSVs. Sheng *et al.*[14] studied the effects of spin-orbit interaction on MR in OSVs. Bergeson *et al.*[15] proposed that MR and its inversion are due to the role of hyperfine interaction in a magnetic field. Dong *et al.*[16] investigated organic MR based on Zeeman interaction. Several models for MR by using spin diffusion theory in ISVs and OSVs have also been proposed.[17−19] From the above-mentioned studies, one can find that significant MR in OSVs cannot be easily obtained and is therefore difficult for actual applications, which is different from ISVs. Therefore, it is interesting to study the factors which influence the MR in ISVs and OSVs.

To determine which affects MR in ISVs and OSVs,

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one needs to consider different characteristics of ISCs and OSCs, and one of them is the different carriers. Carriers are extended electrons or holes in ISCs. However, in OSCs, carriers are some charged self-trapped states such as polarons or bipolarons.[20–22] Polarons are effective carriers in spin injection and transport in OSCs.[18,23] Therefore, in this Letter we suppose that carriers are polarons in OSCs and electrons in ISCs. Electrons and polarons have different mobility and spin-flip times, these carrier differences affect the MR in ISVs and OSVs. By considering these carrier differences, we discuss the MR in ISVs and OSVs.

The system considered here comprises of two semi-infinite FM layers ($x < 0$, FM1 and $x \geq x_0$, FM2) and one NM interlayer will be ISCs or OSCs with the length x_0. The two FM layers are in parallel or antiparallel magnetization alignment. Spin-polarized electrons in the left FM layer are injected into the NM layer. Carriers will be electrons if the NM layer is ISCs and polarons if the NM is OSCs. Here we do not consider the polaron formation process. Based on the above picture, the spin diffusion equation for the electrochemical potential μ_s ($s =\uparrow, \downarrow$) in three layers can be written as[17–19,24]

$$\frac{\partial^2(\mu_\uparrow - \mu_\downarrow)}{\partial x^2} = \frac{\mu_\uparrow - \mu_\downarrow}{\lambda^2}, \tag{1}$$

The difference between up and down electrochemical potentials ($\mu\uparrow-\mu\downarrow$) decays exponentially inside the materials, approaching zero at $\pm\infty$, e.g. $\mu\uparrow(\pm\infty) =\mu\downarrow(\pm\infty)$. Here λ is the spin diffusion length and it can be expressed as[25]

$$\lambda = \sqrt{\frac{k_B T \tau_{\rm sf} w}{2e}}, \tag{2}$$

where $k_{\rm B}$ is Boltzmann's constant, T is temperature, $\tau_{\rm sf}$ is spin-flip time and w is mobility of carriers.

The solutions to Eq. (1) can be written as

$$\begin{aligned}
&\mu_{\rm FM1\uparrow} - \mu_{\rm FM1\downarrow} = Ae^{x/\lambda_{\rm FM}}, \quad x < 0,\\
&\mu_{\rm NM\uparrow} - \mu_{\rm NM\downarrow} = B_1 e^{-x/\lambda_{\rm NM}} + B_2 e^{(x-x_0)/\lambda_{\rm NM}},\\
&\qquad 0 \leq x < x_0,\\
&\mu_{\rm FM2\uparrow} - \mu_{\rm FM2\downarrow} = De^{-(x-x_0)/\lambda_{\rm FM}}, \quad x \geq x_0,
\end{aligned} \tag{3}$$

where $\lambda_{\rm FM}$ and $\lambda_{\rm NM}$ are the corresponding spin diffusion lengths in the two FM layers and the NM interlayer, respectively. Coefficients A, B_1, B_2, and D are determined by the continuity of the electrochemical potential at the interfaces.

We assume that there is no spin-flip scattering and no space charge distribution at the interfaces, so that from Ohm's law, the current densities can be written as

$$j_s = -\frac{\sigma_s \cdot \partial(\mu_s/e)}{\partial x}, \tag{4}$$

where σ_s is the conductivity of spin-polarized carriers (electrons in ISCs or polarons in OSCs). In the NM interlayer, we have approximately $\sigma_{\rm NM\uparrow} = \sigma_{\rm NM\downarrow} = \sigma_{\rm NM}/2$, where $\sigma_{\rm NM\uparrow}$ ($\sigma_{\rm NM\downarrow}$) is the conductivity for carriers with spin $\uparrow$ ($\downarrow$), and $\sigma_{\rm NM}$ is the total conductivity of the NM layer. The bulk spin polarization of the FM is $\beta_0 = (\sigma_{\rm FM\uparrow} - \sigma_{\rm FM\downarrow})/(\sigma_{\rm FM\uparrow} + \sigma_{\rm FM\downarrow})$, so conductivities for spin-up and spin-down carriers in the FM layer can be written as $\sigma_{\rm FM\uparrow} = (1+\beta_0)\sigma_{\rm FM}/2$, $\sigma_{\rm FM\downarrow} = (1-\beta_0)\sigma_{\rm FM}/2$, where $\sigma_{\rm FM} = \sigma_{\rm FM\uparrow} + \sigma_{\rm FM\downarrow}$ is the total conductivity of the FM layer.

At the interfaces of the FM/NM or NM/FM, the total current is written as

$$j = -\sum_s \frac{\Delta\mu_s/e}{R_s}, \tag{5}$$

where R_s is the spin-related interfacial resistance.

From the above equations, the individual electrochemical potential for spin-up carriers (electrons or polarons) across the device can be obtained,

$$\begin{aligned}
\mu_{\rm FM1\uparrow}(x) =& -\frac{ejx}{\sigma_{\rm FM}} - \frac{ej\lambda_{\rm FM}}{\sigma_{\rm FM}}\frac{\alpha_0-\beta_0}{1+\beta_0}(e^{x/\lambda_{\rm FM}} - 1),\\
& x < 0,\\
\mu_{\rm NM\uparrow}(x) =& -\frac{ejx}{\sigma_{\rm NM}} - \frac{ej\lambda_{\rm NM}\alpha_0}{\sigma_{\rm NM}}[-(e^{-x/\lambda_{\rm NM}} - 1)\\
& \pm e^{-x_0/\lambda_{\rm NM}}(e^{x/\lambda_{\rm NM}} - 1)][1 + e^{-x_0/\lambda_{\rm NM}}]^{-1}\\
& + \Delta\mu_\uparrow(0), \quad 0 \leq x < x_0,\\
\mu_{\rm FM2\uparrow}(x) =& -\frac{ej(x-x_0)}{\sigma_{\rm FM}} \pm \frac{ej\lambda_{\rm FM}}{\sigma_{\rm FM}}\frac{\alpha_0-\beta_0}{1\pm\beta_0}\\
& \cdot (e^{-(x-x_0)/\lambda_{\rm FM}} - 1) + \mu_{\rm NM\uparrow}(x_0)\\
& + \Delta\mu_\uparrow(x_0), \quad x \geq x_0,
\end{aligned} \tag{6}$$

where $\pm$ corresponds to the parallel and antiparallel magnetization alignment of the two FM layers; $\alpha_0 = (j_\uparrow - j_\downarrow)/(j_\uparrow + j_\downarrow)|_{x=0}$ is the interfacial current spin polarization; $\Delta\mu_\uparrow(0)$ and $\Delta\mu_\uparrow(x_0)$ are the electrochemical potential discontinuities at the two interfaces. The corresponding electrochemical potentials for the spin-down carriers can also be obtained.

From the electrochemical potentials we can obtain the resistances of the FM/NM/FM device,

$$R_{\rm anti\,(par)} = \frac{\mu_{0\rm anti\,(par)}(-\lambda_{\rm FM}) - \mu_{0\rm anti\,(par)}(x_0 + \lambda_{\rm FM})}{e \cdot j}, \tag{7}$$

where $R_{\rm anti}$ and $R_{\rm par}$ are resistances corresponding to antiparallel and parallel FM magnetization alignment. Here μ_0 is the spin-independent component of the electrochemical potential, which is related to the measured voltage drop across the device. To obtain μ_0, we consider the expression for either $\mu\uparrow(x)$ (Eq. (6)) or $\mu\downarrow(x)$ and extract the linear (ohmic) part.[17] Finally from Eq. (7), the MR of spin valves is given by

$$\mathrm{MR} = \frac{\Delta R}{R_{\rm anti}} = \frac{R_{\rm anti} - R_{\rm par}}{R_{\rm anti}}. \tag{8}$$

The parameters are set as follows: $\lambda_{\rm FM} = 100\,\mathrm{nm}$, $k_{\rm B} = 1.38 \times 10^{-23}\,\mathrm{J{\cdot}K^{-1}}$, $T = 300\,K$, $e = 1.6 \times$

10^{-19} C. For the spin-flip time and the mobility of the carriers, $\tau_{sf} = 10^{-10}$ s, $w = 10^4 \text{cm}^2/\text{V}\cdot\text{s}$ in ISCs and $\tau_{sf} = 10^{-6}$ s, $w = 10^{-2}\text{cm}^2/\text{V}\cdot\text{s}$ in OSCs, respectively.[7] It has been indicated that a suitable conductivity matching favors an increase of the spin polarized injection. We discuss the effect of the conductivity matching on the MR in OSVs and ISVs by considering the carrier differences between ISCs and OSCs, as shown in Fig. 1 with $x_0 = 500$ nm, where the solid line is that for ISVs and the dashed line is for OSVs. The spin related resistances are $R_\downarrow = 10^{-3}\,\Omega\text{cm}^2$ and $R_\uparrow = R_\downarrow/100$. It is found that MR increases with σ_{NM} approaching σ_{FM} for both OSVs and ISVs. In ISVs, the MR increases remarkably and tends to a constant when σ_{NM} approaches σ_{FM}. However, the MR in OSVs is small and nearly constant when σ_{NM} is comparable with σ_{FM}. Therefore, a matching conductivity of the NM layer is favorable for gaining large MR in OSVs and ISVs, and the effect of the conductivity matching on the MR in ISVs is larger than that in OSVs.

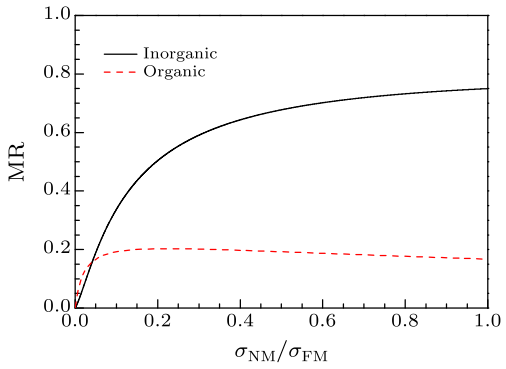

Fig. 1. Dependence of MR on the conductivity ratio σ_{NM}/σ_{FM} with interfacial resistances $R_\downarrow = 100R_\uparrow = 10^{-3}\,\Omega\cdot\text{cm}^2$. The other parameters are $\beta_0 = 0.9$, λ_{FM}=100 nm.

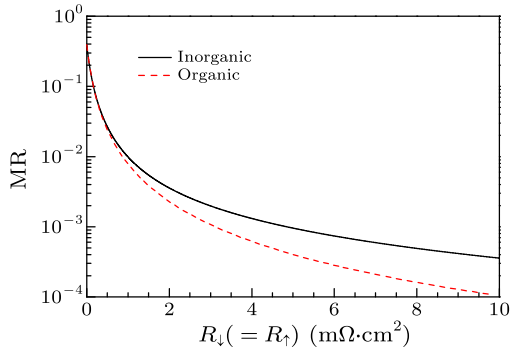

Fig. 2. Dependence of MR on the equivalent interfacial resistances. Conductivities for FM and NM layer are $\sigma_{FM} = 1\,\Omega^{-1}\cdot\text{cm}^{-1}$ and $\sigma_{NM} = 0.2\,\Omega^{-1}\cdot\text{cm}^{-1}$, thickness of the nonmagnetic layer is $x_0 = 100$ nm. The other parameters are the same as those in Fig. 1.

When the interfacial resistances are included, the MR depends upon the absolute conductivity values of the OSCs and ISCs. In the following calculations, we choose $\sigma_{FM} = 1\,\Omega^{-1}\cdot\text{cm}^{-1}$, $\sigma_{NM} = 0.2\,\Omega^{-1}\cdot\text{cm}^{-1}$ in order to gain apparent MR and make comparisons between OSVs and ISVs. Figure 2 shows the dependence of the MR on equivalent interfacial resistances ($R_\downarrow = R_\uparrow$). It is found that MR decreases rapidly with increasing interfacial resistances for both OSVs and ISVs. There will be a large MR when the equivalent interfacial resistances are small and it tends to zero for large equivalent interfacial resistances. From the figure, it is also found that the MR in ISVs is larger than that in OSVs when other parameters are the same. Therefore, it is beneficial for gaining large MR when equivalent interfacial resistances are small.

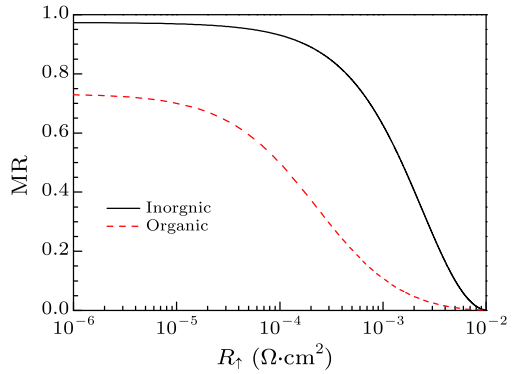

Fig. 3. Dependence of MR on the up-spin interfacial resistance with the condition $R_\downarrow = 10^{-2}\,\Omega\cdot\text{cm}^2$. The other parameters are the same as those in Fig. 2.

The MR is studied further when the interfacial resistances are spin-dependent. The result is shown in Fig. 3. It is found that MR could be improved if the interfacial resistances are spin-dependent for both OSVs and ISVs. For example, for equivalent value of interfacial resistances, $R_\downarrow = R_\uparrow = 10^{-2}\,\Omega\cdot\text{cm}^2$, the MR is nearly zero for both OSVs and ISVs. However, when $R_\downarrow = 10R_\uparrow$, the MR will be about 0.1 in OSVs and 0.62 in ISVs. The stronger the split of the interfacial resistances, the larger the MR. When $R_\downarrow = 10^3 R_\uparrow$, the MR will be about 0.7 in OSVs and 0.97 in ISVs. From the figure, it is also found that MR in ISVs is larger than that of OSVs because of the different carriers in them. Owing to the self-adjusting capability of the OSCs, an FM/OSC interface should have a low contact resistance, and the interfacial resistances can be spin-dependent if a thin insulating layer is added between the FM and the NM or by using a doping profile.[26] Self-assembly techniques can be used to grow ordered monolayers at an FM/OSC structure so OSCs may be particularly attractive for the fabrication of the ordered tunnel barriers.[27] Therefore, an enhanced MR in OSVs could be expected.

Figure 4 shows the dependence of MR on the bulk polarization of the FM layer. It can be seen that the MR increases slowly when the bulk polarization of the FM layer is small and quickly increases when $\beta_0 \to 1.0$. Again, it is found that MR will be larger in

ISVs than that in OSVs if all the other parameters are the same because of the different mobility and spin-flip time of the carriers.

The thickness of the NM interlayer in spin valve is an important factor that affects the MR. Figure 5 gives the dependence of the MR on the NM interlayer thickness. From the figure, one finds that the MR ratio decreases with the increasing of the NM layer thickness and also MR in ISVs is larger than that in OSVs. For a thick interlayer, the MR ratio tends to zero, which means that there is no apparent spin correlation between the FM layers. Therefore, to get an apparent MR in spin valves, the thickness of the NM interlayer should be not longer than hundred nanometers.

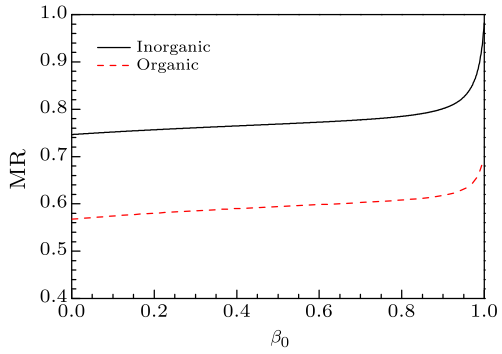

Fig. 4. Dependence of MR on the bulk spin polarization β_0 of the FM layer. Interfacial resistances are $R_\downarrow = 100R_\uparrow = 10^{-3}\,\Omega{\cdot}\mathrm{cm}^2$. Other parameters are the same as those in Fig 1.

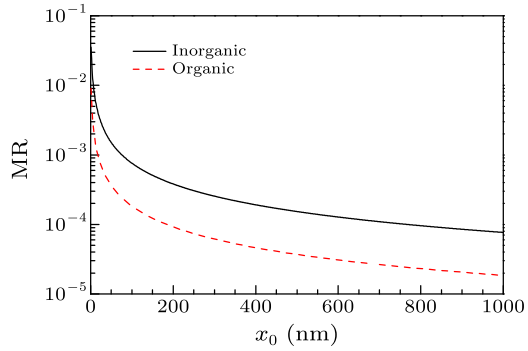

Fig. 5. Dependence of MR on the thickness of the nonmagnetic layer. Values of the spin-dependent interfacial resistances are $R_\downarrow = 100R_\uparrow = 10^{-3}\,\Omega{\cdot}\mathrm{cm}^2$. The other parameters are the same as those in Fig 2.

In summary, we have theoretically studied the MR in ISVs and OSVs based on Ohm's law and the spin diffusion theory and discussed the factors which affect the MR. Carriers in OSCs are supposed to be self-trapped polarons, and their mobility and spin-flip time are different from electrons in ISCs. It is found that conductivity matching is favorable for gaining remarkable MR and the MR increases with the decreasing equivalent interfacial resistance. Spin-dependent interfacial resistances and large bulk spin polarization of the FM materials are beneficial for achieving efficient MR. The MR ratio will tend to zero when the thickness of the NM interlayer is larger than hundred nanometers. Especially, for the effect of the different carriers, it is predicted that the different mobility and spin-flip time for polarons and electrons induce a larger MR in ISVs than that in OSVs.

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