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CONTRIBUTION FROM MOLDCULAR FIELD TO THE TEMPERATURE DEPENDENCE OF TUNNELING MAGNETORESISTANCE

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Based on the nearly free-electron approximation, we have investigaed the temperature (T) dependence of spin-polarized tunneling in the magnetic tunnel junction with an asymmetrical barrier, with emphasis on the variation of molecular field with T in the same way as that of surface magnetization. It is found that the Slonczewski model can describe well the T dependence of spin-polarized tunneling, while the Julliere model only describes the T dependence of *JMR* qualitatively, but does accurately that of the difference of tunneling conductance between the parallel and antiparallel alignments for the magnetizations of *FMs*; Differing from the previous finding, we find the electron spin polarization is not strictly proportional to the surface magnetization, for the former decreases with the increasing T more rapidly than the latter does.

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Tunnel magnetoresistance in ferromagnetic junctions (*JMR*), first observed more than two decades ago[1−2] has attracted much recent attention due to the application potential in digital storage and magnetic sensor technologies[3−6] Recently, Moodera *et al.*[7] obtained a *JMR* of 27.1% at 77 K, and 20.2% at 295 K in the high quality $Co/Al_2O_3/Ni_{80}Fe_{20}$ junction; Earlier, Lu *et al.*[8] even found a *JMR* of 83% in an epitaxial tunnel junction of $La_{0.67}Sr_{0.33}MnO_3/SrTiO_3/La_{0.33}MnO_3$. However, such large *JMR* exhibits both a temperature (T) dependence and a dc bias dependence[9], the magnitude of which depends on the quality of the junctions: the lower the *JMR* is the larger the T and dc bias dependence are. In addition to *JMR*, the tunneling conductance (G) also exhibits a strong T dependence. Likewise, for the junctions with amorphous barriers, such as α-Si and α-Ge, there is also a substantially higher T dependence of conductance G[10]. So far, this effect has been interpreted successfully by the hopping transport model[11−12]. However, as for the case of normal junction, such as that with Al_2O_3 barrier, theoretical understanding to the T dependences of G and *JMR* remains to be incomplete, although Shang *et al.*[13] discussed them phenomenologically by extending the Julliere's model[1] to the case of spin-independent tunneling, as this model gives a opposite T dependence of paralell conductance G_p with the experimental results.

So far, there are two theoretical models that are mostly used to describe the spin-polarized tunneling phenomena: one is the Julliere model, which is based on arguments of

Tedrow and Meservey[14]. It assumes that the spin for each channel is conserved in the tunneling process, and the tunneling current is proportional to the product of the "effective tunneling density of states". Although this simple model lacks firm theoretical foundation, it is consistent with many large *JMRs* observed recently[7]. Parkin pointed out that this model provides a reasonable upper bound to the *JMR*; The other theory is the Slonczewski model[15], which analyses the transmission of charge and spin current through the rectangular barrier separating two free-like *FM* metals. It predicts that the *JMR* depends not only on the spin-polarizations of two *FMs*, as that of Julliere's model, but also on the barrier height. MacLaren *et al.*[16] verified that, in the case of free electron, this model provides a good approximation to the exact expressions for large barrier thickness and for small barrier height.

As far as the two models are concerned, both of them do not involve the T dependence of the spin-polarized tunneling. In the present paper, based on the nearly free-electron model, we ascribe mainly the T dependence of spin-polarized tunneling to the variation of molecular field h in the *FMs* versus T, owing to the proportionality of h to the surface magnetization of *FMs*. Moreover, we extend the Slonczewski's model to case of asymmetrical barrier[17], which is attributed to the different work functions of two *FMs*. During calculation, we do not make any approximation such as small bias and large barrier thickness as well as the replacement with Fermi energy to the electron energy.

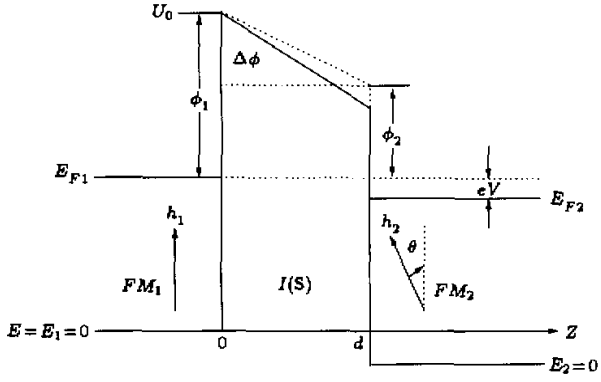

Fig.1. Schematic potential for $FM_1/I/FM_2$ junction with the asymmetrical barrier in the presence of positive bias, eV. The dotted lines in the barrier corresponds to the zero-bias case. The zero of energy is taken at the middle of bottoms for majority-spin band and minority-spin one in the FM_1.

Let us consider a $FM_1/I/FM_2$ sandwiched structure in the presence of dc bias, as illustrated in Fig.1, where the *FMs* are of semi-infinite thickness, ϕ_1 and ϕ_2 are the work functions of the FM_1 and FM_2, respectively, and $\Delta\phi$ is their difference. It should be pointed out that, in the present paper, the zero energy is taken at the middle of the bottoms for the majority-spin band and that of minority spin in the FM_1 (the left-hand *FM*). According to the Simmons's work[17], this difference would result in an barrier asymmetry $\Delta U = \Delta\phi$, even at the zero bias.

Within the *FMs*, the conduction electrons, being described well by the nearly free-electron model, have the *s-d* exchange interaction with the localized electrons[18]:

$$E_{\rm ex} = -\sum_i J(|\boldsymbol{r} - \boldsymbol{R}_i|)\hat{S}_i \cdot \hat{s}\,, \tag{1}$$

where $\hat{s}$ is the spin operator of a conduction electron, $\hat{S}_i$ that of localized electrons at site i in the lattice, and $J(|\boldsymbol{r} - \boldsymbol{R}_i|)$ the exchange integral. Taking the approximation of average field, Eq.(1) becomes

$$E_{\rm ex} \approx \boldsymbol{h} \cdot \sigma\,, \tag{2}$$

where $\boldsymbol{h}$, refered as molecular field, takes value of

$$h = -\frac{\hbar}{2} < \sum_i J(|\boldsymbol{r} - \boldsymbol{R}_i|)\hat{S}_i > \propto M_s\,. \tag{3}$$

It should be pointed out that the molecular field h introduced here is not the conventional Weiss moleuclar field, which represents the exchange interaction between the localized magnetic moments.

It is known from spin wave theory[19] and experimental observation[20] on clean surface in systems such as $FeNiB_{0.5}$, $Ni_{80}Fe_{20}$, and Fe that the T dependence of surface magnetization $M_S(T)$ follows $M_S(0)(1 - B_S T^{3/2})$, where the value of B_S is seen up to two or more than the bulk value, depending on the surface cleanliness. Therefore, the T dependence of h may be expressed as:

$$h(T) = h(0)(1 - B_S T^{3/2})\,. \tag{4}$$

Correspondingly, for the nearly free electrons in the *FMs*, the wave vectors may be written as:

$$k_\sigma(T) = \sqrt{E \pm h(T)} = \sqrt{E \pm h(0)(1 - B_S T^{3/2})}\,, \tag{5}$$

where $\pm$ correspond to $\uparrow$, $\downarrow$ spins respectively.

According to the Slonczewski's model, in the case of positive bias, the longitudinal (along the z direction) part of the effective single-electron Hamiltonian may be written as:

$$H = -\frac{\mathrm{d}^2}{\mathrm{d}z^2} + U(z) + F(z) + E_{\rm ex}\,, \tag{6}$$

with

$$U(z) + F(z) + E_{\rm ex} = \begin{cases} \pm h_1(T), & z < 0 \\ U_0 - \dfrac{\Delta\phi + eV}{d} z, & 0 \le z < d \\ \pm h_2(T) - eV, & z \ge d \end{cases} \tag{7}$$

where $U(z)$ represents the potential prifile, $E_{\rm ex}$ is the *s-d* exchange energy discussed before. Although the transverse momentum $\hbar k_{/\!/}$ is omitted from the above notations, the effects of summation over $k_{/\!/}$ will be considered in our calculation.

Let us consider a spin-up normal incident plane wave having unit particle flux in the region of FM_1. In the barrier region, i.e., $0 \le z < d$, the Schrödinger equation can be

simplified by a coordinate transformation[21], whose solution is the linear combination of Airy function $Ai[\rho(z)]$ and its complement $Bi[\rho(z)]$:

$$\psi_{2\sigma} = A_{2\sigma} Ai[\rho(z)] + B_{2\sigma} Bi[\rho(z)], \tag{8}$$

where

$$\begin{cases} \rho(z) = \dfrac{z}{\lambda_0} + \beta_0 \\ \lambda_0 = -\left(\dfrac{d}{\Delta\phi + eV}\right)^{1/3} \\ \beta_0 = \dfrac{d(E - U_0)}{(\Delta\phi + ev)\lambda_0}, \end{cases} \tag{9}$$

After deriving the eigenfunctions for the three layers, the transmissivity $T_{p\sigma}$, at the position $z = 0$, can be derived analytically by

$$T_{p\uparrow} = \mathrm{Im} \sum_{\sigma} \psi_{\sigma}^{*} \frac{\mathrm{d}\psi}{\mathrm{d}z} \tag{10}$$

As for the case of spin-down incidence, $T_{p\downarrow}$ is given by the same expression but with $\uparrow$ and $\downarrow$ interchanged. According to the Duke's theory[22], for the nonzero temperature, the expression for the net tunneling current density

$$j_e = \sum \frac{4\pi m e k_B T}{h^3} \int_0^{\infty} \frac{T_{p\uparrow} + T_{p\downarrow}}{2} \ln\left\{\frac{1 + \exp[(E_F - E)/k_B T]}{1 + \exp[(E_F - E - eV)/k_B T]}\right\} \mathrm{d}E, \tag{11}$$

where k_B is the Boltsman constant, T the temperature, and E_F the Fermi Energy.

The differential tunneling conductance G is defined as: $G = \mathrm{d}j_e/\mathrm{d}V$, and the *JMR* can be calculated according to

$$JMR = \frac{\Delta G}{G_p} = \frac{G_p - G_{ap}}{G_p}, \tag{12}$$

where G_p is the conductance for parallel alignment of the two *FM*'s magnetizations, while G_{ap} is the that for antiparallel alignment of the two *FM*'s magnetizations.

As for the Julliere's theory[1], the tunneling conductance for the θ angle orientation of the two *FMs* magnetizations may be written as:

$$G(\theta) = G_T[1 + P_1 P_2 \cos(\theta)], \tag{13}$$

where G_T is the prefactor for direct elastic tunneling, P_1 and P_2, denoting the spin polarization ratios of the effective tunneling density of states of the *FMs*, may be expressed as: $P = (N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow)$, where $N_\uparrow$ and $N_\downarrow$ are the effective tunneling densities of states. So the *JMR* can be written as

$$JMR = \frac{\Delta G}{G_p} = \frac{2P_1 P_2}{1 + P_1 P_2}, \tag{14}$$

For the nearly free-electron model, the polarization P may be written as

$$P = \frac{k_{F\uparrow} - k_{F\downarrow}}{k_{F\uparrow} + k_{F\downarrow}}. \tag{15}$$

where $k_{F\uparrow}$ and $k_{F_\downarrow}$ are the Fermi wave vectors of the *FMs* for the $\uparrow$, $\downarrow$ spins.

Owing to the proportionality of h to the surface magnetization, as the discussed before, the molecular field h and the wave vector k_σ vary considerably with T as Eq.(4) and (5), thus the $k_{F\uparrow(\downarrow)}$ in Eq.15 should be regarded as functions of temperature T as that in Eq.(5). With regard to G_T, according to the Stratton's theory[23], when temperature changes from 0 K to 300 K, G_T only increases a few percent, which is much less than experimental measurement, so we may neglect the contribution to T dependence arising from G_T. Therefore, we may focus our attention on the T dependence of molecular field h. Hereafter, we refer to this mechansim as the Julliere model-I.

According to Shang's view[13], it is the spin polarization P that is proportional to the surface magnetization, so P varies with T in the same way as that of $M_S(T)$, i.e., $P(T) = P(0)(1 - B_S T^{3/2})$. So the T dependences of spin-polarized tunneling is mainly ascribed to variation of h versus T. Hereafter we refer to the Juliere model based on Shang's mechanism as Julliere model-II.

Taking the $Co/Al_2O_3/Fe_{20}NI_{80}$ junction as an concrete example, we investigate the T dependence of G_p, ΔG and *JMR*, using the Slonczewski model and two Julliere ones, and compare them with the experimental results. Here we refer to $Fe_{20}Ni_{80}$ electrode as FM_1 and Co electrode as FM_2. For Al_2O_3 barrier, according to the Moodera's estimation[7], the average height $\bar{\phi}$ (above the Fermi energy) is taken as 3.4 eV, and the thickness d is 11 Å. The barrier asymmetry $\Delta\phi$, which can be derived from the work functions[24] of Co and $Fe_{20}NI_{80}$, is taken as 0.85 eV.

The Fermi energy and molecular field can be derived from the d-band width and the spin polarization measurement near the Fermi surface: the d-band widths[25] are 4.2 eV and 4.55 eV for Co and $Fe_{20}Ni_{80}$ respectively; the polarizations[7] are 35% and 45% for Co and $Fe_{20}Ni_{80}$, respectively. Thus, we derive $E_{F1} = 2.6$ eV, $h_1 = 1.95$ eV; $E_{F2} = 2.58$ eV, $h_2 = 1.61$ eV.

According to the surface magnetization measurement[20], for $Fe_{20}Ni_{80}$, about 15% decrease in M_S has been observed as T increases from 77 K to 300 K. From this fact, we can derive $B_{S1} = 3.25 \times 10^{-5}\,K^{-3/2}$ for the surface magnetization, which is nearly three times of the bulk value[26], $1.23 \times 10^{-5}\,K^{-3/2}$. As for the case of Co, the bulk value[27] of B_S is $1.5 \times 10^{-6}\,K^{-3/2}$, in the same ratio as $Fe_{20}Ni_{80}$, we take the surface value $B_{S2} = 4 \times 10^{-6}\,K^{-3/2}$. Morever, the magnitude of bias is taken as 0.001 V, which is so small that it can be regarded as zero bias.

In Fig.2, we plot the normalized T dependence of G_p, using the Slonczewski model and two Julliere ones. One can find that, for the Slonczewski's model, G_p increases with the increasing T, this is qualitatively consistent with the experimental result, but there still exists a discrepancy to some extent. We infer that, in addition to the spin-polzried tunneling, there is also a spin-independent tunneling, which plays an important role in the tunneling process, but has no influence on the of ΔG (see Fig.3). As for the two Julliere models, both of them derive a opposite T dependence with the experimental results. In other words, for Julliere models, theoretical G_p decreases with the increasing T, while the experimental measurement keeps increasing. Even though, based on this model, Shang *et al.* still derived

fitted T dependences of the conductance G, JMR and ΔG, by taking into account the contribution of the spin-independent tunneling to the total tunneling conductance.

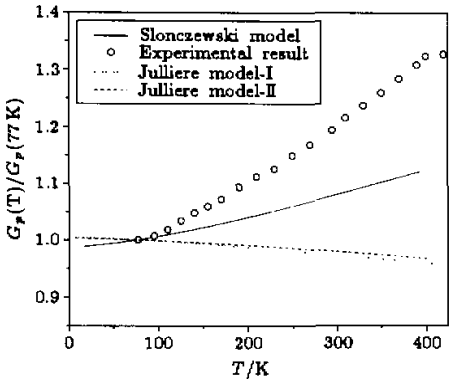

Fig.2. The normalized temperature dependence of $G_p(T)$ for $Co/Al_2O_3/Fe_{20}Ni_{80}$ junction. The circles represent experimental results of Ref.13, the solid, dotted, and dashed lines denote the theoretical results with Slonczewski model, Julliere model-I, and Julliere model-II, respectively.

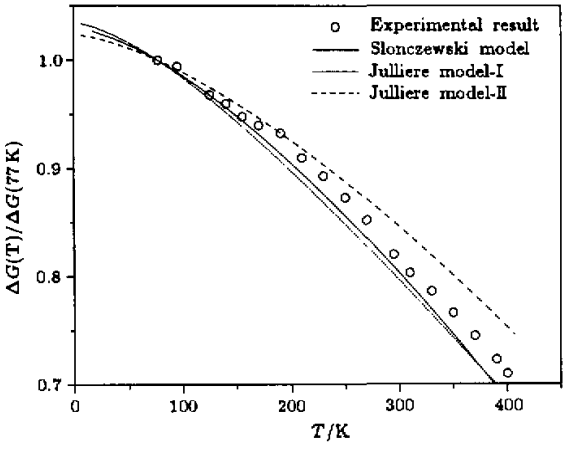

Fig.3. The normalized temperature dependence of $\Delta G(T)$ for $Co/Al_2O_3/Fe_{20}Ni_{80}$ junction. The representation of each symbol is the same as that in Fig.2.

The normalized T dependence of ΔG is shown in Fig.3, where one can find that all the three models can give the T dependence consistent with experimental observation. But comparatively, the Slonczewski model and Jullere model-I are more accurate than the JUlliere model-II. To our surprise, although the Julliere models are in contradiction with the experiments with respect to the T dependence of G_p, it still describes well the T dependence of ΔG. Thereofre, we can infer that, for the Julliere model, both G_p and G_{ap} perhaps lose a same part of spin-polarized tunneling, thus ΔG still remains being consistent with experiments. Of course, this inference needs to be verified further by the other theory.

In Fig.4, we plot the normalized T dependence of JMR. It is found that the Slonczewski model describes well the T dependence of JMR, for it provides a good approximate result,

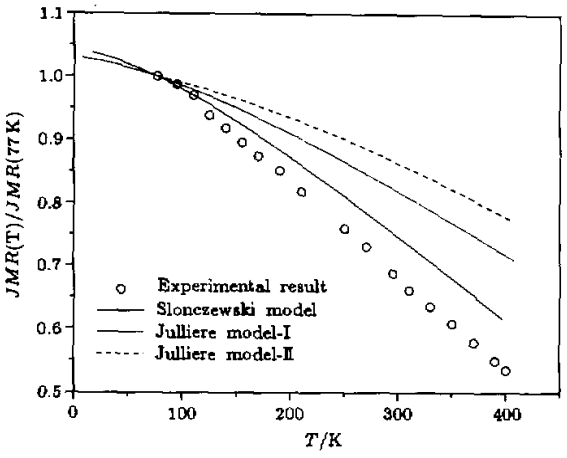

Fig.4. The normalized temperature dependence of $JMR(T)$ for $Co/Al_2O_3/Fe_{20}Ni_{80}$ junction. The representation for each symbol is the same as that in Fig.2.

which is slightly less than the experimental measurement. Therefore, we can conclude that although the spin-independent tunneling contributes to the total conductance a comparable part, the spin-polarized tunneling still dominates the T-dependence of JMR. As for the two Julliere models, one can find that both of them work bad, owing to the large discrepancies between these models and the experiments. According to the definition of JMR in Eq.(14) and the calculating G_p with the Julliere models.

In order to achieve some insight into the Julliere model-II, we plot the T dependences of P_1 and P_2 in Fig.5, as described in Eq.15. For comparison, the T dependences of M_{si} is is also shown there. It can be seen that, with the increasing T, $P_i(T)$ decreases more rapidly than $M_{si}(T)$. In other words, the polarization P_i follows a $1 - B_{pi}T^{3/2}$ law, with B_{pi} being slightly larger than B_S. For Co, this difference is not too great, but it is comparatively large for $Fe_{20}Ni_{80}$ FM. Combining this feature of $P_i(T)$ and the behaviors of Julliere model-II in B-subsection, we can draw a conclusion that the spin polarization is not strictly proportional to the surface magnetization, and the viewpoint of Shang[13] based on the Julleire model is only a rough approximation.

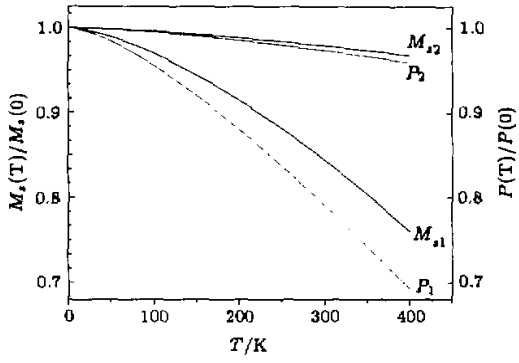

Fig.5. Comparison of the temperature dependence of $P(T)$ with that of $M_S(T)$ for Co and $Fe_{20}Ni_{80}$.

In summary, as the proportionality of molecular field to the surface magnetization, h varies with T as $h(0)(1 - B_m T^{3/2})$. It is just this T dependence of molecular field that is mainly responsible for the variations of JMR, ΔG and G_p versus T. The Slonczewski model can describe well the spin-polarized tunneling, while the Julliere ones only describe qualitatively the T dependence of JMR, but do accurately that of ΔG. The spin-polarized tunneling dominates the T dependence of JMR, although the spin-independent tunneling contributes to the T dependence of tunneling conductance a considerable part. Moreover, differing from other's finding, we find that the spin polarization is not strictly proportional to the surface magnetization.

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