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Magnetic tunnel junctions consisting of a periodic grating barrier and two half-metallic electrodes

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Keywords: magnetic tunnel junction, tunneling magnetoresistance, half-metallicity, effect of temperature, Heusler alloys, spintronics

Abstract

We have developed a spintronic theory for magnetic tunnel junctions consisting of a single-crystal barrier and two half-metallic ferromagnetic electrodes. Radically different from the conventional theories, the barrier is now regarded as an optical diffraction grating, and treated by the traditional optical scattering method, i.e. Bethe theory and two-beam approximation. After tunneling, the electrons can thus possess high coherence. In the case that the electrodes are both half-metallic, the conventional theories give an infinite tunneling magnetoresistance (TMR). By contrast, in the Bethe theory and two-beam approximation, there can exist the scattering channels of nonconservation of energy. Therefore, the TMR can still be far away from infinity, which is in accordance with experiments. Also, we find that, due to the half-metallicity of the electrodes, the parallel conductance oscillates with temperature whereas the antiparallel conductance will increase rather than oscillate with temperature. That is in agreement with experiments, too. Finally, two applications of the present theory are discussed with regard to the material design and engineering: one is how to choose appropriate materials for the barrier to realize infinite TMR; the other is a criterion for judging whether a material is half-metallic or not.

1. Introduction

Highly spin-polarized current source is essential for spintronic devices [1–6]. For magnetic tunnel junctions (MTJs), highly spin-polarized ferromagnetic electrodes can achieve large tunneling magnetoresistance (TMR) [7]. In particular, a complete spin polarization will lead to almost infinity TMR near zero bias and low temperature in conventional theories [8], e.g. Jullièr’s model and Slonczewski’s model [9, 10]. For applications, the material in which all conduction electrons have the same spin (100% spin-polarized) is called as a half-metallic ferromagnet (HMF) [11, 12]. There are numbers of HMFs, e.g. Heusler alloys [13–15], the manganite (La2/3Sr1/3MnO3, La0.9Pr0.1Ca0.3MnO3, La0.97Ca0.03MnO3 etc) [16–18], the magnetite (Fe3O4) [19], ZnFeS−δO4 [20], and Cr2O3 [21] etc. Among numerous prospective HMFs, Heusler alloys have been widely studied and theoretically predicted to be the most favorable candidates [22–26]. To our knowledge, Co2MnSi, Co2FeSi, and Co2FeAl0.5Si0.5 have been experimentally proved to be HMFs up to now [13–15]. And thus, these Heusler alloys can be used as completely spin-polarized ferromagnetic electrodes for the MTJs. Apart from the electrodes, the barrier is also crucial for MTJs to have large TMR. In last century, the barriers are usually fabricated with the amorphous aluminum oxide. Because there exist much disorders in amorphous barrier, the TMR is depressed highly by the scattering of those disorders. Since 2000, single-crystal MgO has come into use as barriers for the MTJs. Due to the good single crystallinity, the TMR is significantly improved [27]. As such, much effort has been dedicated to fabricating the MTJs with Heusler alloys as electrodes and MgO as barriers [1, 2, 5, 28–30]. Unfortunately, the TMR in these MTJs has still been far away from infinity up to now. This is in contradiction with the conventional theories. Physically, there should be three possible reasons: (1) Some Heusler alloys may
not be half-metallic. (2) The quality of the barrier needs to be improved. (3) The conventional theories are inapplicable to the experiments. However, the former two reasons can be excluded in [2, 28] where the half-metallic Co2MnSi electrodes are used and the barriers are high-quality single crystalline. That is to say, only the third reason is valid for [2, 28]. Therefore, it is necessary to develop a more rational theory to describe the TMR results of experiments. On the other hand [2], finds another intriguing phenomenon with regard to the conductances: the parallel conductance \( G_\| \) oscillates with the temperature whereas the antiparallel conductance \( G_\perp \) increases other than oscillates with the temperature. This phenomenon has not been understood theoretically as yet. In sum, there are two important problems in MTJs consisting of single-crystal barriers and half-metallic ferromagnetic electrodes: (1) the contradiction between conventional theories and experiments. (2) The unusual temperature dependence of the conductances. The physical mechanism for them needs to be clarified, which is just the aim of this letter.

Previously, a spintronic theory for the MTJs with single-crystal barrier has been developed by us [31, 32]. The theory is founded on traditional optical scattering theory [33]. Within it, the barrier is treated as a diffraction grating with intralayer periodicity. It is found that the periodic grating will bring strong coherence to the tunneling electrons. So far, the theory has successfully explained two important properties of the MgO-based MTJs with normal (not half-metallic) electrodes. One is that the parallel resistance \( R_\| \), the antiparallel resistance \( R_\perp \) and TMR all oscillate with the barrier thickness [1, 29, 34]; the other is that the decrease of TMR with rising temperature is most carried by the change of \( R_\perp \), while the \( R_\| \) changes so little that it seems roughly constant [35]. In this letter, we shall try to use the previous theory to handle the MTJs consisting of single-crystal barriers and half metallic ferromagnetic electrodes. As will be seen in the following, the theory can solve well the two fundamental problems stated above.

2. Theory

To begin with, we employ a periodic potential \( U(r) \) to describe the perfect single-crystal barrier of the MTJs. It can be written as

\[
U(r) = \sum_{l=0}^{n-1} \sum_{a_0} v(r - R_h - l_3 a_3),
\]

where \( v(r) \) represents the atomic potential of the barrier; \( n \) denotes the total number of the layers of the barrier; \( R_h = l_1 a_1 + l_2 a_2 \), with \( a_1 \) and \( a_2 \) being the intralayer primitive vectors of barrier, and \( l_1 \) and \( l_2 \) the corresponding integers; \( a_3 \) is the interlayer primitive vector of the barrier, with \( l_3 \) the corresponding integer.

As for the tunneling process in the present model, we do not consider the case of spin flip, just as in the conventional theories [9, 10]. Let the \( z \)-axis be antiparallel to the tunneling current. The transmission coefficient for the channel of the spin-up to spin-up tunneling can be written as follows [31, 32]

\[
T_\|^\parallel(k) = \frac{1}{8k_z} \{ p_\|^\parallel e^{i(k - (p'_\|^\parallel - p''\|^\parallel)l_3)} + p'_\|^\parallel e^{i(p'_\|^\parallel - (p''\|^\parallel - q^\|^\parallel)l_3)} + q^\|^\parallel e^{i(q^\|^\parallel - (q'^\|^\parallel - q''\|^\parallel)l_3)} + q''\|^\parallel e^{i(q''\|^\parallel - (q'^\|^\parallel - q''\|^\parallel)l_3]} + c.c.,
\]

where \( k \) represents the incident wave vector of tunneling electrons, and \( k_z \) its \( z \)-component, \( d \) denotes the thickness of MgO barrier, and

\[
p_\|^\parallel = |k^2 - k_h^2| + 2m \hbar^2 v(K_h)|^2 / 2,
\]

\[
q^\|^\parallel = |k^2 - (k_h + K_h)^2| + 2m \hbar^2 v(K_h)|^2 / 2.
\]

Here, \( k_h \) is the intralayer component of \( k \), \( K_h \) is the intralayer reciprocal lattice vector, and \( v(K_h) \) is the Fourier transformation of \( v(r) \). From \( T_\|^\parallel(k) \), the conductance \( G_\|^\parallel \) can be expressed as follows

\[
G_\|^\parallel = \frac{e^2}{16\pi^2} \int_0^{\pi/2} d\theta \int_0^{2\pi} d\varphi \ k_z^2 \sin(2\theta) T_\|^\parallel(k_h, \theta, \varphi),
\]

where \( e \) represents the electron charge, \( \theta \) the angle between \( k \) and \( e_x \), \( \varphi \) the angle between \( k_0 \) and \( a_0 \), and \( k_\|^\parallel \) the Fermi wave vector of the spin-up electrons. Similarly, \( G_\|^\perp \), \( G_{\|^\parallel \|^\perp} \), and \( G_{\|^\perp \|^\perp} \) can be obtained. As usual, \( G_P = G_\|^\parallel + G_\|^\perp \), \( G_{AP} = G_{\|^\parallel \|^\perp} + G_{\|^\perp \|^\perp} \), and TMR = \( G_P / G_{AP} - 1 \). Now, let us consider the effect of the half-metallicity of ferromagnetic electrodes. As well known, the half-metallicity means that the chemical potential \( \mu \) is smaller than the half of the exchange splitting \( \Delta \). Therefore, for the incident waves, there dose not exist spin-down electrons, only spin-up electrons can remain, i.e. \( G_{\|^\perp} = G_{\|^\parallel} = 0 \). Accordingly, \( G_P = G_\|^\parallel \) and \( G_{AP} = G_{\|^\parallel \|^\perp} \). In conventional theories [9, 10], \( G_{AP} = G_{\|^\parallel \|^\perp} = 0 \), and TMR = \( G_P / G_{AP} - 1 \). That is because, in those theories, the energy of each tunneling electron is conserved, and the incident spin-up electrons can not tunnel to the spin-down states in the lower electrode.
However, the present theory is based on the Bethe theory and the two-beam approximation [31–33], according to them, the energy of the tunneling electron can be non-conserved in the tunneling process. To be specific, in present theory the transmitted electron waves include four components, the two of them will acquire an energy of $v(K_h)$, the other two will lose an energy of $v(K_h)$ [31]. The four components are equi-probalistic, therefore, due to this nonconservation of energy, there will be half of the incident spin-up electrons that can get enough energy to overcome the barrier and transit into the spin-down band of the lower electrode when $v(K_h) > \Delta - \mu$. This tunneling channel leads to $G_{\uparrow\downarrow} = 0$, so $\text{TMR} = G_{\uparrow\uparrow}/G_{\uparrow\downarrow} - 1 < +\infty$, i.e. the TMR will be finite other than infinite. This is quite different from the result of conventional theories. On the contrary, when $v(K_h) < \Delta - \mu$, the energy will be insufficient for all the incident spin-up electrons to transit into the spin-down band of the lower electrode, so $G_{\uparrow\downarrow} = 0$ and $\text{TMR} = +\infty$, which is the same as the conventional result. In a word, within the framework of the present theory, the TMR can be finite or infinite for the MTJ with single-crystal barriers and half-metallic ferromagnetic electrodes, which depends on whether $v(K_h) > \Delta - \mu$ or not. This physical picture has been sketched diagrammatically in figure 1 where the z-axes of spin for the two ferromagnetic electrodes are respectively chosen as their own.

3. Results and discussion

First, we would like to investigate the effect of $v(K_h)$ on the oscillation of conductances and TMR with barrier thickness. Here, we only discuss the case that $v(K_h) > \Delta - \mu$, because otherwise the TMR will be infinite as stated above. The theoretical results are depicted in figure 2 where the chemical potential $\mu$ is 7 eV, the half of the exchange splitting $\Delta$ is 12 eV, and $v(K_h)$ is set sequentially as 12, 16 and 20 eV. As can be seen from figure 2(b),
the $G_{AP}$ dose not oscillate with barrier thickness. Physically, that is because, for $G_{AP}$, $p^z$ and $q^z$ will always be imaginary in half-metallic electrodes ($\mu < \Delta$), which can be easily seen from equation (3). This results in that $T$ does not contain oscillating term, as stated in [31]. Unlike $G_{AP}$, the $G_P$ oscillates with the barrier thickness if $v(K_h) < \Delta + \mu$, and does not if $v(K_h) > \Delta + \mu$, as can be seen in figure 2(a). This is also because, for $G_{AP}$, $p^z$ and $q^z$ will be imaginary if $v(K_h) > \Delta + \mu$. Combining the above results of $G_P$ and $G_{AP}$, the TMR will oscillate with the barrier thickness if $v(K_h) < \Delta + \mu$, and does not if $v(K_h) > \Delta + \mu$, which are shown clearly in figure 2(c). This theoretical result can explain the oscillation of TMR on barrier thickness found in [28]. By the way, as shown in figure 2(c), the amplitude and period of TMR will both increase with decreasing $v(K_h)$, which has been discussed and explained in [31].

Secondly, we shall study the effect of temperature on the conductances and TMR, the method is the same as [32]. Here, the corresponding model parameters are chosen as follows: The defect concentration $\sigma$, the effect of strain of the barrier $K_h \cdot \alpha_0$, the recovery temperature $T_a$, the Fourier transform of the atomic potential of ideal perfect barrier $v_0(K_h)$, the chemical potential $\mu$ and the half of the exchange splitting $\Delta$ are set as $0.08$, $\pi/3$, $800$ K, $15.3$ eV, $7$ eV and $12$ eV, respectively. The results are displayed in figure 3 where the barrier thickness $d$ is set sequentially as 1, 2 and 3 nm. Figure 3(a) shows that the $G_P$ will oscillate with the temperature. The phenomenon has been explained physically in [32]. This theoretical result is in agreement with the experimental results of [2] on the unusual temperature dependence of $G_P$. However, quite different from [32], the $G_{AP}$ now increases other than oscillate with the temperature. First, the non-oscillation of $G_P$ comes physically from the half-metallicity of the electrodes: for HMFs, $\mu < \Delta$. As stated above, all the wave vectors of $p^z$ and $q^z$ are imaginary for $G_{AP}$. Correspondingly, $T$ does not include any oscillating term. This results in that the $G_{AP}$ does not oscillate. Secondly, the thermal activation, i.e. the $G_{AP}$ increases with temperature, can be explained physically as follows: according to equation (5) of [32], $v(K_h)$ increases monotonously with temperature. As stated above, the large the $v(K_h)$, the more energy the tunneling electrons will acquire. Therefore, the increasing

![Figure 2](image-url)
of $v(K_h)$ will make more tunneling electrons to get enough energy to transit into the spin-down band of the lower electrode. Therefore, the tunneling current as well as $G_{AP}$ will increase remarkably as temperature goes high. Those theoretical results are in agreement with the experimental results of [2] on the unusual temperature dependence of $G_{AP}$.

Finally, we would like to conclude the parameter dependence of the present theory, which are shown in table 1. It includes not only the present results but also the previous ones [31, 32]. In table 1, the parameters are partitioned into five regions. Here, the oscillation contains two meanings: (1) The oscillating with barrier thickness. (2) The oscillating with temperature. In each region, the two kinds of oscillating behavior are identical: if $G_P$, or $G_{AP}$, or TMR oscillates with barrier thickness, it will also oscillate with temperature. If it does not oscillate with barrier thickness, it will not oscillate with temperature. The first two columns correspond to $G_P$, Oscillation | No oscillation | Oscillation | No oscillation | $=0$

| Relation | $\mu > \Delta$ | $\Delta + \mu > v(K_h)$ | $\Delta + \mu < v(K_h)$ | $\Delta - \mu < v(K_h)$ | $\Delta + \mu < v(K_h)$ | $\Delta - \mu > v(K_h)$ |
|----------|----------------|-----------------|----------------|----------------|----------------|----------------|
| $G_P$    | Oscillation    | No oscillation  | Oscillation    | No oscillation | $=0$           |                |
| $G_{AP}$ | Oscillation    | No oscillation  | Oscillation    | No oscillation | $=0$           |                |
| TMR      | Oscillation    | No oscillation  | Oscillation    | No oscillation | $=0$           | Infinity       |

Figure 3. (a)$G_P$, (b) $G_{AP}$ and (c) TMR as functions of the temperature. The curves A, B and C correspond to $d = 1$ nm, 2 nm and 3 nm, respectively.
the normal cases where the electrodes are not half-metallic, the other three columns correspond to the half-metallic cases.

Here, we would first like to apply table 1 to MgO-based MTJs. As has been stated above, for the MTJ consisting of both single-crystal barrier and half-metallic electrodes, its TMR is finite or infinite will depend on whether $v(K_0) > \Delta - \mu$ or not. In present theory, $v(K_0)$ is proportional to the energy gap of the barrier [31, 32]. For MgO barrier, the band gap is as large as 8 eV or so [36], the corresponding $v(K_0)$ is about 16 eV according to our theoretical calculations [31, 32]. It satisfies the condition of $v(K_0) > \Delta - \mu$ for usual Heusler alloys, and thus the TMR is of course finite according to table 1. This completely explain why the experimental TMR has still been far away from infinity up to now. On the other hand, according to table 1, the TMR can be infinite if such a material is used as barrier that $v(K_0) < \Delta - \mu$. This is certainly an exciting goal for the application of MTJs. Some candidate materials have been proposed in [31], e.g. graphite and MoS2. Finally, we would like to discuss a potential application of table 1: it can be conversely used to judge the relation among $\mu$, $\Delta$ and $v(K_0)$. For example, the behaviors of $G_P$, $G_{AP}$ and TMR in [29] belong to the case of the first column of table 1. And thus, it can be deduced that $\mu > \Delta$ for the Co2Cr0.6Fe0.4Al electrodes which is used in [29]. That is to say, the Co2Cr0.6Fe0.4Al should not belong to half-metallic Heusler alloy, this theoretical assertion is expected to be verified in the future. In short, the behaviors of conductances and TMR can be used to judge whether the electrode is half-metallic or not. This could be a potential criterion for half-metallicity of Heusler alloys.

In the present theory, the ferromagnetic electrodes are handled by two exchange-splitting bands, i.e. Stoner model. In principle, the theory is valid for all the half-metallic ferromagnetic electrodes which can be described approximately by Stoner model, such as the manganite (La2/3Sr1/3MnO3, La0.3Pr0.7Ca0.3MnO3, La0.7Ca0.3MnO3 etc) [16–18], the magnetite (Fe3O4) [19], ZnFe3-xO4 [20], and CrO2 [21].

4. Conclusions

In this letter, we have developed a tunneling theory to take into account the half-metallic characteristics of the electrodes. With present theory, the energy of the tunneling electrons can be non-conserved, they can acquire or lose an energy of $v(K_0)$. Due to this nonconservation, the incident spin-up electrons can transit into the spin-down band of the lower electrode if $v(K_0) > \Delta - \mu$, and thus $G_{1f} \approx 0$. Consequently, the TMR can be finite other than infinite, which is radically different from the conventional theories. That can explain the problem why the experimental TMR has still been far away from infinity up to now even both electrodes are half-metallic. Meanwhile, we also study the effect of temperature on the conductances. We find that the $G_{AP}$ increases other than oscillates with temperature due to the half-metallicity of the electrodes, which can explain the problem why in [2] the $G_P$ oscillates but $G_{AP}$ increases with temperature. These two results answer completely the two fundamental problems posed in the field of MTJs consisting of both single-crystal barrier and half-metallic electrodes. Besides, we have listed the partitions of model parameters in table 1. According to it, we give a suggestion of how to realize infinite TMR for MTJs consisting of a single-crystal barrier and two half-metallic electrodes, and propose a criterion for judging whether the material of electrode is half-metallic or not.

Acknowledgments

This work is supported by the National Natural Science Foundation of China (11704197, 61574079, 51971109, 51771053), National Key Research and Development Program of China (2016YFA0300803), the NUPTSF (NY217046).

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