Magnetoresistance of the double-tunnel-junction Coulomb Blockade with magnetic metals

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We have studied the Junction Magnetoresistance (JMR) and the Differential junction magnetoresistance (DJMR) for double tunnel junctions with magnetic metals in the Coulomb Blockade regime. Spikes are seen in both the JMR and the DJMR vs. voltage curves. They occur at those places where the current increases by a step. In all cases the large bias limit can be obtained by adding the resistances of each of the junctions in series. The JMR is positive in all the cases we studied, whereas the DJMR can be positive or negative as a function of the voltage. Moreover, the relative variation of the DJMR as a function of the voltage is larger than the variation of the JMR with the voltage.

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I. INTRODUCTION

Electron tunneling through tunnel junctions which consist of magnetic metals separated by insulators has a long history. Some of the earliest experiments were on tunneling between a superconductor and a ferromagnet. In these experiments, the difference between the spin-up and spin-down density of states of a superconductor in a magnetic field was observed. These experiments also provided a quantitative measure of the spin polarization of the current from the ferromagnetic metal. Later experiments were done on tunneling between two ferromagnetic metals. Because of the spin dependence of the density of states, the conductance changed when the relative orientation of the two magnetizations changed. Tunneling between two ferromagnetic metals has experienced renewed interest because of its potential application in information storage. One of the measurable quantities of practical importance is the junction magnetoresistance, which is defined as the ratio of the change in the conductance from parallel to antiparallel alignment divided by the parallel conductance. Large magnetoresistance response in low magnetic fields has been observed in a variety of tunnel junctions by several groups.

Besides spin, electrons carry charge and at low temperatures in tunnel junctions with small capacitance, the discreteness of electron charge manifests itself through the physics of the Coulomb Blockade. The transfer of an electron by tunneling between two initially neutral regions of capacitance $C$ increases the electrostatic energy of the system by an amount $e^2/2C$. At low temperatures and small voltages, the tunneling current is suppressed because of the charging energy. This energy barrier is called the Coulomb Blockade. The effect of this in a two-junction system is the incremental increase in the current at voltages where it is energetically favorable for an electron to tunnel to the center island. The occurrence of these current steps in the current-voltage characteristics is known as the “Coulomb Staircase.” Advances in technology have led to a number of new experiments which exhibit the Coulomb Blockade phenomenon: granular thin films, lithographically patterned tunnel junctions, narrow disordered quantum wires, scanning tunneling microscopy of small metal droplets, metal-insulator-metal tunnel junctions with small metal particles embedded in the insulator.

Recently there have been experiments where spin polarized tunneling and the Coulomb Blockade have been combined. For example, in discontinuous metal/insulator multilayers consisting of closely spaced ferromagnetic nanoparticles in an insulating matrix, a Coulomb Blockade is seen in the current-voltage characteristic at low temperatures. The purpose of this paper is to further study the nonlinear conductances for parallel and antiparallel magnetization alignment for a double junction with magnetic metals in the Coulomb Blockade regime.

The organization of the rest of the paper is as follows. In the next section we describe the model and the master equation used. The expression for the current, the conductance, the differential conductance and the junction magnetoresistance are then introduced. The results of the calculation are given in the discussion of Sec. III, and summarized in Sec. IV.

II. FORMALISM

The experimental realization of a double tunnel junction consists of three metals separated by oxide layers which act as insulators. The two end leads, denoted by left ($L$) and right ($R$), are connected to a constant voltage source $V$. This double junction can be modeled as a closed loop electrical circuit with two leaky capacitors of capacitances $C_L$ and $C_R$ connected in series (Fig. 1). The resistances of the junctions, $R_L$ and $R_R$, are connected in parallel to $C_L$ and $C_R$ respectively. Depending
on the number of excess electrons, \( n \), in the center metal, the voltage in the central region \( V_M \) fluctuates. The voltage drops, \( (V_L - V_M(n)) \) and \( (V_M(n) - V_R) \), across the left and the right junctions are

\[
V_L - V_M(n) = \frac{C_R}{C} V + \frac{ne}{C} + V_G; \quad (1)
\]

\[
V_M(n) - V_R = \frac{C_L}{C} V - \frac{ne}{C} - V_G; \quad (2)
\]

where \( -e \) is the charge of the electron, \( C \) is the capacitance equal to \( (C_L + C_R) \), and \( V_G \) plays the role of the gate or the offset voltage. To describe the transport of electrons we use a semiclassical model. The different tunneling rates that enter into the dynamics are: the rate for electrons to tunnel onto the center metal from the left (\( \Gamma_{n\rightarrow n+1}^L \)) and right (\( \Gamma_{n\rightarrow n+1}^R \)), and the rate for the electrons to leave the center metal to the left (\( \Gamma_{n\rightarrow n-1}^L \)) and right (\( \Gamma_{n\rightarrow n-1}^R \)). These tunnel rates can be calculated using Fermi’s Golden rule. To express the different tunnel rates in a convenient fashion we introduce a function \( \gamma(e) \):

\[
\gamma(e) = \frac{e}{1 - e^{-\beta e}}. \quad (3)
\]

In terms of the above function the various tunnel rates are:

\[
\Gamma_{n\rightarrow n\pm 1}^{L(R)} = \left( \frac{1}{e^2 R_{L(R)}} \right) \gamma \{ \pm e [V_M(n) - V_{L(R)}] - E_C \}. \quad (4)
\]

Here \( E_C = e^2 / 2C \) is the charging energy, and \( \beta \) is related to the temperature \( T \) via \( \beta = 1/k_B T \).

For magnetic materials, there are two types of carriers: the majority electrons with spins parallel to the magnetization and the minority electrons with spins antiparallel to the magnetization. We make the assumption that the rate of tunneling through the junctions is much smaller than the rate of spin relaxation. Under this assumption the number of majority and minority electrons in the center metal is not conserved separately, but their sum is conserved. This results in a master equation which describes the time evolution of the probability, \( \rho_n(t) \), for \( n \) excess electrons in the center metal:

\[
\frac{d\rho_n(t)}{dt} = [\rho_{n+1} \Gamma_{n+1\rightarrow n} - n \Gamma_{n\rightarrow n+1} - \rho_{n-1} \Gamma_{n-1\rightarrow n}] - [\rho_n \Gamma_{n\rightarrow n-1} - (n-1) \Gamma_{n-1\rightarrow n}], \quad (5)
\]

where the net tunnel rate \( \Gamma_{n\rightarrow j} \) is equal to \( \left( \Gamma_{1\rightarrow j}^L + \Gamma_{1\rightarrow j}^R \right) \). At steady state, \( \frac{d\rho_n(t)}{dt} = 0 \), the current in the left junction equals the current in the right junction due to current conservation, and is given by

\[
I = e \sum_{n=-\infty}^{n=\infty} \rho_n \left( \Gamma_{n\rightarrow n-1}^L - \Gamma_{n\rightarrow n+1}^R \right). \quad (6)
\]

To evaluate the current in general, Eqs. (5) - (8) have to be solved numerically. We follow the same procedure described in Ref. [35]. At low temperature and voltage, the higher charge states \( |n| >> 1 \) are energetically forbidden and therefore we can neglect them. We found that for our choice of parameters it is sufficient to keep the 19 states around \( n = 0 \). The highest and the lowest states are \( n = 9 \) and \( n = -9 \). The total conductance \( G \) is then simply given by dividing the current with the voltage, \( G = I/V \), and the differential conductance \( G_{\text{diff}} \) is obtained by differentiating the current with respect to the voltage, \( G_{\text{diff}} = dI/dV \).

There are two possible choice of materials which lead to non-zero magnetoresistance in a two junction system: (1) all the leads are ferromagnets, i.e. FM/I/FM/I/FM, and (2) the left or the right lead is a paramagnet, i.e. FM/I/FM/I/PM or PM/I/FM/I/FM. Note that with the center lead non-magnetic (FM/I/PM/I/FM) there is no JMR or DJMR because the antiparallel conductance is identical to the parallel conductance within our assumption. For each of these cases, we calculate the conductances for two different relative orientations of the ferromagnet magnetizations: parallel and antiparallel. Here parallel (P) means that the magnetizations of all ferromagnets is in the same direction, and antiparallel (AP) means the magnetization of the center ferromagnetic metal has been reversed. In case (1) of three ferromagnetic metals, other configurations are also possible: the magnetization of the right or the left lead can be reversed keeping the magnetizations of the other two parallel. For case (2) where one of the left or the right lead is non-magnetic, the magnetization of right or the left ferromagnetic lead can also be reversed. However, the qualitative features of the current, conductance, and the magnetoresistance curves remain the same. The junction magnetoresistance (JMR) is then defined as the ratio of the change in the conductance from parallel, \( G^P \), to antiparallel alignments, \( G^{AP} \), divided by the parallel conductance

\[
\text{JMR} = 1 - \frac{G^{AP}}{G^P}. \quad (7)
\]

The differential junction magnetoresistance (DJMR) is defined in the same fashion as the JMR but with the total conductances replaced by the differential conductances, \( G_{\text{diff}} \).
\[ DJMR = 1 - \frac{G_{AP}}{G_{PAP}}. \] (8)

**III. DISCUSSION**

The different parameters of our problem are: the capacitances \( C_L \) and \( C_R \), the parallel resistances \( R_L^P \) and \( R_R^P \), the antiparallel resistances \( R_L^{AP} \) and \( R_R^{AP} \), the temperature \( T \), and the gate or the offset voltage \( V_G \). To estimate the antiparallel resistances, we use Julliere’s model for FM/I/FM tunnel junctions, where the junction magnetoresistance is expressed as a product of the magnitudes of the polarizations of the ferromagnets.

\[ \frac{\Delta R}{R_{AP}} = \frac{2P_1P_2}{(1 + P_1P_2)}. \] (9)

In the above equation, \( \Delta R \) is the change in resistance from antiparallel to parallel orientations, \( R_{AP} \) is the junction resistance when the magnetizations of the ferromagnets are antiparallel, and \( P_1 \) and \( P_2 \) are the spin polarizations of the two FMs.

In Fig. (2) the currents and the total conductances are plotted for parallel and antiparallel orientations of the magnetizations. Asymmetric junctions and low temperature are used to obtain the steps in the current. Also, we take the offset or the gate voltage to be zero, \( V_G = 0 \). The effect of finite gate voltage is only to shift the \( I - V \) curves. As expected in the Coulomb blockade regime, the current increases by steps (case a), and the conductance shows oscillations with decreasing amplitude (case b).

Finally, for large enough voltage, the current becomes linear with voltage and the conductance approaches the classical limiting value which is the inverse of the resistance of two resistors connected in series

\[ G = \frac{1}{(R_L + R_R)}. \] (10)

The effect of temperature dependence is shown in Fig. (3). With the rise in temperature, the number of accessible states increases. The steps in the current round off (case a), and the amplitude of the oscillations in the conductance vs. voltage curve (case b) decreases. The peaks in the JMR (case c) and the DJMR vs. voltage curves (case d) become broader and reduced in size.

Restricting ourselves to \( k_B T \ll E_C \) and zero gate or offset voltage \( (V_G = 0) \), in Fig. 4 we plot the junction magnetoresistance for four different cases: (a) FM\(_1\)/I/FM\(_1\)/I/FM\(_2\), (b) FM\(_2\)/I/FM\(_1\)/I/FM\(_1\), (c) FM\(_1\)/I/FM\(_1\)/I/PM, and (d) PM/I/FM\(_1\)/I/FM\(_1\), where FM\(_1\) and FM\(_2\) are different ferromagnetic metals. In the high bias regime for all four cases, the current is linearly proportional to the voltage

\[ I = \frac{e}{(R_L + R_R)C} \left[ \frac{CV}{e} - 1 \right]. \] (11)

It then follows from Eq. (11), that the plateaus in Figs. 4(a) - 4(d) are given by the difference in the total resistances from antiparallel to parallel alignments divided by the total resistance for the antiparallel alignment,

\[ \text{JMR(plateau)} = 1 - \frac{R_L^P + R_R^P}{R_L^{AP} + R_R^{AP}}. \] (12)

For asymmetric junctions in the low voltage, low temperature regime we see spikes in the JMR vs. voltage curves in Fig. (4). The occurrence of the first spike can be understood analytically. We confine ourselves to only three states: the \( n = 0 \) state and the two states \( n = \pm 1 \) immediately accessible from it. At very low temperature, the state which is most likely occupied is the \( n = 0 \) state for \( V_G = 0 \). The tunnel rates \( \Gamma_{0\rightarrow 1} = \Gamma_{1\rightarrow 0} \) do not enter in the expression for the tunneling current at zero temperature, but at finite temperature they do contribute.

In the limit of zero temperature, where we replace the function \( \gamma(e) \) in Eq. (3) by \( \gamma(e) = e\theta(e) \), the steady state current can be expressed in terms of the tunnel rates in the regime \( (e/2) \lesssim C_R V \lesssim (C_R/C_L)(e/2) \) as

\[ I = e \frac{\Gamma_{0\rightarrow 1}^L \Gamma_{1\rightarrow 0}^R}{(\Gamma_{0\rightarrow 1}^L + \Gamma_{1\rightarrow 0}^R)}. \] (13)

In the derivation of Eq. (13) we have assumed that the
FIG. 3. Temperature dependence of the (a) Current, (b) Conductances, (c) JMR, and (d) DJMR for a FM/I/FM/I/FM tunnel junction. As in Fig. 2(b), the solid and the dashed lines represent the current and the conductance for parallel and antiparallel alignments. With increase in temperature, the steps in the current vs. voltage curve round off (case a), resulting in reduced amplitude of oscillations in the total conductance (case b). The peaks in both the JMR (case c) and the DJMR (case d) curves broaden. Also the peaks in the JMR and the DJMR curves decreases with increase in temperature. The parameters are the same as used in Fig. 2, except for the temperature which is $k_B T = 0.1 E_C$.

capacitance of the right junction is larger than the capacitance of the left junction, i.e, $C_R > C_L$. Near the special point $C_R V \approx e/2$, where the current increases by a step at zero temperature, the height of the spike is simply given by the difference,

$$\text{JMR}(C_R V = \frac{e}{2}, T \rightarrow 0) = 1 - \frac{R_L^{P}}{R_L^{AP}}. \quad (14)$$

However, at finite temperature, the height of the spikes depend on the temperature, the capacitances, and the resistances of the junctions. We also notice that spikes go up for cases 4(a) and 4(c) whereas spikes go down for cases 4(b) and 4(d). Cases 4(a) - 4(b) or cases 4(c) - 4(d) differ only by the exchange of the metals in the left and the right leads. In all the above cases we keep the resistance of the left junction fixed, $R_L^{P} = 0.01(R_L^{P} + R_R^{P})$. By only exchanging the metals of the left and the right leads, we go from spikes which go up to spikes which go down and vice versa. The interchange of the spikes from up to down or down to up can be explained in the following way. At zero temperature, whenever the JMR obtained from Eq. (14) is greater than the height of the plateau (Eq. (12)), we see an up-spike (Fig. 4(a) and 4(c)), otherwise we see a down-spike (Figs. 4(b) and 4(d)). Within Julliere's model(4), we should mention that to see the spikes, the left or the right metal should be of a different ferromagnetic material. When all three metals are of the same ferromagnetic materials, the steady state solution $\rho_n^P(t)$ is the same for parallel and antiparallel orientation of magnetizations since the solution depends only on the ratio of the left and the right resistances.

Comparing Figs. 4(a) - 4(b) with Figs. 4(c) - 4(d), we see that the variation of the JMR as a function of voltage is larger when one of the leads is non-magnetic. Following Eq. (12) and Eq. (14), which give the height of the plateau and the height of the first spike at zero temperature, we see that the height of the plateau for case (c) is lower than for case (a). On the other hand, the heights of the first spikes are same for both cases because the metals in the left and the center are same. Thus the variation of the JMR for case (c) is more than case (a). Cases (b) and (d) can be understood similarly.

We plot the differential junction magnetoresistance as a function of the voltage in Fig. 5 for the same cases as in Fig. 4 and in the same parameter regime. Many of the
FIG. 5. Differential Junction magnetoresistance (DJMR) as a function of voltage for the same cases as in Fig. 4. We see spikes in the DJMR at those places where the current increases by a step. As in Fig. 4, the variation in the DJMR is larger as a function of voltage for cases (c) and (d) compared to cases (a) and (b). For case (c) the DJMR changes sign. Also compared to Fig. 4, the change in the DJMR is larger than the JMR. The metals are the same as used in Fig. 4 and the parameters are identical to Fig. 2.

features of the DJMR are the same as for the JMR. The spikes are at the same places. The height of the spikes is also the same as in Eq. (14). The DJMR at large voltages is obtained from the classical value with two resistors in series. Finally, the variation of the DJMR with the voltage is larger in cases 5(c) and 5(d) than in cases 5(a) and 5(b). There are, however, some differences. The DJMR can be negative, as shown in Fig. 5(c), while the JMR is positive as long as $R_P < R_P$ and $C_R > C_L$. Finally, the variation of the DJMR is larger as a function of voltage than that of the JMR.

In conclusion, double tunnel junction systems which have magnetic metals exhibit rich Coulomb blockade conductance vs. voltage curves. These curves provide a signature of both the Coulomb charging effects and the spin polarization of the tunneling. By measuring the JMR or DJMR, one can extract information about both the capacitance charging energies and the spin polarization of the tunneling electrons, testing both theories of spin polarized tunneling and the Coulomb blockade. Furthermore, there are cases where the JMR can be dramatically enhanced near one of the Coulomb blockade steps, meaning that there may be some applications of these effects. In any case, for small enough systems, both charging and spin-polarization effects will be important.

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