3.45 Paper, Tunneling Magnetoresistance

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1 Introduction

In the past few decades, there have been great strides in the area of magnetoresistance– the effect of magnetic state on the resistance of a material. The magnetization dependence of resistance is of great interest due to the applications to magnetic recording media where the state of a magnetic material needs to be read at high speeds.

Before Magnetoresistance was utilized, the best way to read a magnetic state was to move over the magnetic surface with a coil of wire. This method had two major flaws- the inductance of the circuit inevitably causes a time constant delay in your reading and the currents induced are relatively small and difficult to detect. Magnetoresistance, then, solves both of these problems- there is no inductor, so there is no inherent electronic restriction on reading speed, and because the Magnetoresistive effect frequently causes a change in resistance on the order of at least a few percent, it is easily detectable.

Following Robert O'Handley's textbook[2], by magnetizing a ferromagnet along a particular direction, commonly done by using an anti-ferromagnet to pin the magnetization direction at 45° to the direction of the magnetic field being detected (a magnetic field would either move the ferromagnet's magnetization more parallel or more perpendicular to a current flowing through the device). If the magnetization moves perpendicular to the current it reduces the resistance, and moving parallel to the current increases the resistance.

2 Anisotropic Magnetoresistance

The first notable magneto-resistive effect discovered in ferromagnets was called Anisotropic Magnetoresistance (AMR), and results in a resistance change on the order of several percent. AMR was first discovered by William Thomson in 1857, but was regarded mainly as a curiosity until the advent of magnetic recording technology.[1]

AMR occurs because spin-orbit interactions cause a mixing of the majority and minority spin carriers. By considering a Hamiltonian containing the crystal field splitting, exchange splitting, and spin-orbit coupling, the AMR effect can

be calculated as[1]

$$\frac{\rho_{\parallel} - \rho_{\perp}}{\frac{1}{3}\rho_{\parallel} + \frac{2}{3}\rho_{\perp}} = -\frac{3}{2}\left[\frac{A^2}{\left(E_{ex} - \Delta_{CF}\right)^2} - \frac{A^2}{\Delta_{CF}^2}\right]. \tag{1}$$

3 Giant Magnetoresistance

In 1998, Baibich discovered Giant Magnetoresistance (GMR). Giant Magnetoresistance resulted in resistance changes on the order of 50%, a huge increase over standard AMR. The GMR effect is seen in layered materials where the spin flips from layer to layer, and is a function only of the relative magnetizations of adjacent layers. It has been observed to obey

$$\frac{\Delta\rho\left(\theta\right)}{\rho} = \left(\frac{\Delta\rho}{\rho}\right)_{GMR} \frac{\left[1 - \cos\psi\right]}{2} \tag{2}$$

with ψ as the angle between magnetizations of adjacent layers and $\left(\frac{\Delta\rho}{\rho}\right)_{GMR}$ as the GMR ratio. By changing the $\vec{H}$ applied perpendicular to $\vec{M}$, you can shift ψ, causing large changes in $\Delta\rho$.

The primary mechanism of GMR lies in the two currents (one for spin up and one for spin down electrons) having different scattering probabilities depending upon the magnetization of the layer they are travelling through. In the case where two adjacent layers have opposite magnetizations, spin up electrons will, for example, have a high scattering through the first and a low scattering through the second, giving a total resistance $R_{\uparrow}$. The spin down electrons will have an equivalently low scattering through the first layer and an equivalently high scattering through the second layer, resulting in a resistance $R_{\downarrow}$ equal to $R_{\uparrow}$.

However, in the case where the two layers are magnetized in the same direction, you have for the spin up electrons two sequential low scattering layers to go through, with a total resistance much lower than $R_{\uparrow}$. Alternatively, the resistance of the spin down electrons will be much higher than $R_{\downarrow}$ as they are going through two sequential high scattering layers.

Adding the two resistances of the ferromagnetically coupled layers together in parallel will always result in a lower resistance than the anti-ferromagnetically coupled layers; thus, if you were to keep one layer fixed and rotate the other, you can get very large changes in resistance– the GMR effect.

4 General Theory of TMR

Following now M. Ziese's review of Magnetoresistance[1], if a tunneling barrier is placed between the two ferromagnetic layers, you have tunneling Magnetoresistance (TMR). If you assume that spin is conserved upon tunneling through

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the barrier, there are some interesting effects caused by the two-current model. The crux of the problem is that the tunneling probability is governed by the density of states at the Fermi level not only for an electron, but for an electron of the appropriate spin. Thus, you end up with a conductance that is proportional to the density of states for each spin on either side. (Figure 1)

This gives you a conductance proportional to the products of the densities of states[1]

$$G_{\uparrow\uparrow} \propto (1+P)(1+P') + (1-P)(1-P') \tag{3}$$

$$G_{\uparrow\downarrow} \propto (1+P)(1-P') + (1-P)(1+P') \tag{4}$$

with P and P' as the spin-polarizations of the ferromagnetic electrodes (where the spin-polarization is defined as $P \equiv \frac{(N_\uparrow - N_\downarrow)}{(N_\uparrow + N_\downarrow)}$).

Using equations 3 and 4, we find we can write

$$\frac{\Delta G}{G} \equiv \frac{G_{\uparrow\uparrow} - G_{\uparrow\downarrow}}{G_{\uparrow\uparrow}} = \frac{2PP'}{1+PP'} \tag{5}$$

$$\frac{\Delta R}{R} = \frac{2PP'}{1-PP'}, \tag{6}$$

where $\frac{\Delta R}{R}$ is the Tunneling Magnetoresistance (TMR) ratio.

In 1989, Slonczewski used the free electron model with the Landauer-Büttiker formula to find that the conductance was of the form

$$G \propto \left[1 + P_{fb}^2 \cos(\Theta)\right] \tag{7}$$

where Θ is the total angle that the exchange fields in the ferromagnets span and P_{fb} is an effective polarization dependant on the field splitting, such that for a

single band, $P_{fb} = 1$, which gives you

$$G \propto [1 + \cos(\Theta)], \tag{8}$$

the conductance relation for a simple spin valve; however, with two bands he calculated the effective polarization as

$$P_{fb} = \left[\frac{(k_\uparrow - k_\downarrow)}{(k_\uparrow + k_\downarrow)}\right] \cdot \left[\frac{(\kappa^2 - k_\uparrow k_\downarrow)}{(\kappa^2 + k_\uparrow k_\downarrow)}\right] \equiv PA_{fb}, \tag{9}$$

where $k_\uparrow$ and $k_\downarrow$ are the wavevectors of the electrons and κ is the decay length of the wavefunction ($\kappa = \sqrt{2\,(U_0 - E_F)}$ with U_0 the barrier height and E_F the Fermi energy).

Later on, A.M. Bratkovsky derived a slightly altered equation more rigorously.[3] He directly solved the Schrödinger equation for a single-particle Hamiltonian with an exchange energy

$$\mathscr{H} = -\left(\frac{\hbar^2}{2\mu_\alpha}\right) + U_\alpha - \mathbf{h} \cdot \hat{\sigma} \tag{10}$$

with α representing either the left terminal, barrier, or right terminal and μ_α representing the reduced mass in the α region.

Bratkovsky directly calculated the tunneling coefficients for the barrier and used them to calculate

$$G = \frac{e^2}{\pi\hbar}\frac{\kappa_0}{\pi w}\left[\frac{\kappa_0\,(\kappa_\uparrow + \kappa_\downarrow)\left(\kappa_0^2 + \mu_{barrier}^2\kappa_\uparrow\kappa_\downarrow\right)}{\left(\kappa_0^2 + \mu_{barrier}^2\kappa_\uparrow^2\right)\left(\kappa_0^2 + \mu_{barrier}^2\kappa_\downarrow^2\right)}\right]^2 e^{-2\kappa_0 w} \tag{11}$$

with $\kappa_0 = [2\mu_{barrier}(U_0 - E)/\hbar^2]^{1/2}$, assuming the effective mass of an electron in each electrode is the same (same material), and w is the barrier width. Clearly, by choosing carefully the material in our barrier, we can significantly affect the tunneling conductance.

This new form for conductance leads to the altered form of the effective polarization,

$$P_{fb} = \left[\frac{(k_\uparrow - k_\downarrow)}{(k_\uparrow + k_\downarrow)}\right] \cdot \left[\frac{(\kappa_0^2 - \mu_{barrier}k_\uparrow k_\downarrow)}{(\kappa_0^2 + \mu_{barrier}k_\uparrow k_\downarrow)}\right] (\equiv PA_{fb}) \tag{12}$$

Either of these equations allow us to use the free electron model to replace

$$P = \frac{(n_\uparrow - n_\downarrow)}{(n_\uparrow + n_\downarrow)}, \tag{13}$$

the standard definition of the spin-polarization. A_{fb} is a function of interface properties (which affect κ primarily) and the effective mass. It ranges from -1 to 1. In addition, it is interesting that as κ^2 becomes larger or smaller than the

product $\mu k_\uparrow k_\downarrow$, A_{fb} switches signs. In experiments, Slonczewski's expression for the TMR ratio was a good approximation for wide, low barriers. In 1997, MacLaren solved the TMR problem taking into account the band structure of the electrodes. This also found a dependence on the barrier height and thickness.

As with any tunneling problem, there is a dependence of conductance on the applied voltage. Guinea found a functional dependence for the conductivity that goes as

$$G(V) \propto \begin{cases} (V/J_{AF})^2 & V \ll J\,(a/d) \\ (a/d)^2 & V \gg J\,(a/d) \end{cases} \tag{14}$$

with a the lattice constant of the crystal, V the applied bias, d the thickness of the barrier, and J_{AF} the interface exchange constant.

5 Half-metallic tunneling junctions

A very interesting aspect of the tunneling conductance can be seen by looking at the case where the magnetic electrodes are anti-parallel and are both totally polarized. This case is known as half-metallic because the Fermi energy is in the conduction band for one spin, but above it for the other spin– thus, the material is conductive for one spin, but insulating for the other. Two materials with this property are CrO_2 and Fe_3O_4. (Figure 2)

In this case, the two polarization terms, P and P' are 1 and -1. Clearly, from equation 5, in this case, the TMR goes to infinity.

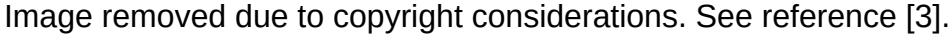
Bratkovsky solves numerically the Hamiltonian for these half-metallic systems with the inclusion of image forces and finds a TMR of 3000%(!!!), compared to 20% for an Iron-based junction, even with the electrodes at a 20° angle from one another (taking into account the idealization of perfectly aligned spins). (Figure 3)

Taking into account impurities in the barrier which allow sequential tunneling in a resonance, we find that the conductance increases sharply when the impurity density of states

$$\nu \gtrsim \left(\frac{\kappa_0}{\pi}\right)^3 \epsilon_i^{-1} \exp(-\kappa_0 w)$$

with $\epsilon_i = \hbar^2\kappa_0^2/(2\mu_{barrier})$.

For standard ferromagnetic electrodes, the conductance increases, but the TMR is reduced. In the case of half-metallics, however, the TMR is still only limited by spin-flip processes in the interfaces or spin misalignments in the two electrodes. Unfortunately, creating a resonant tunnel diode structure will also increase the spin-flip rate, so having too large a concentration of defects in the barrier will decrease the TMR. Regardless, it is possible to reach both high conductivities and very large magnetoresistance in these half-metallic materials.

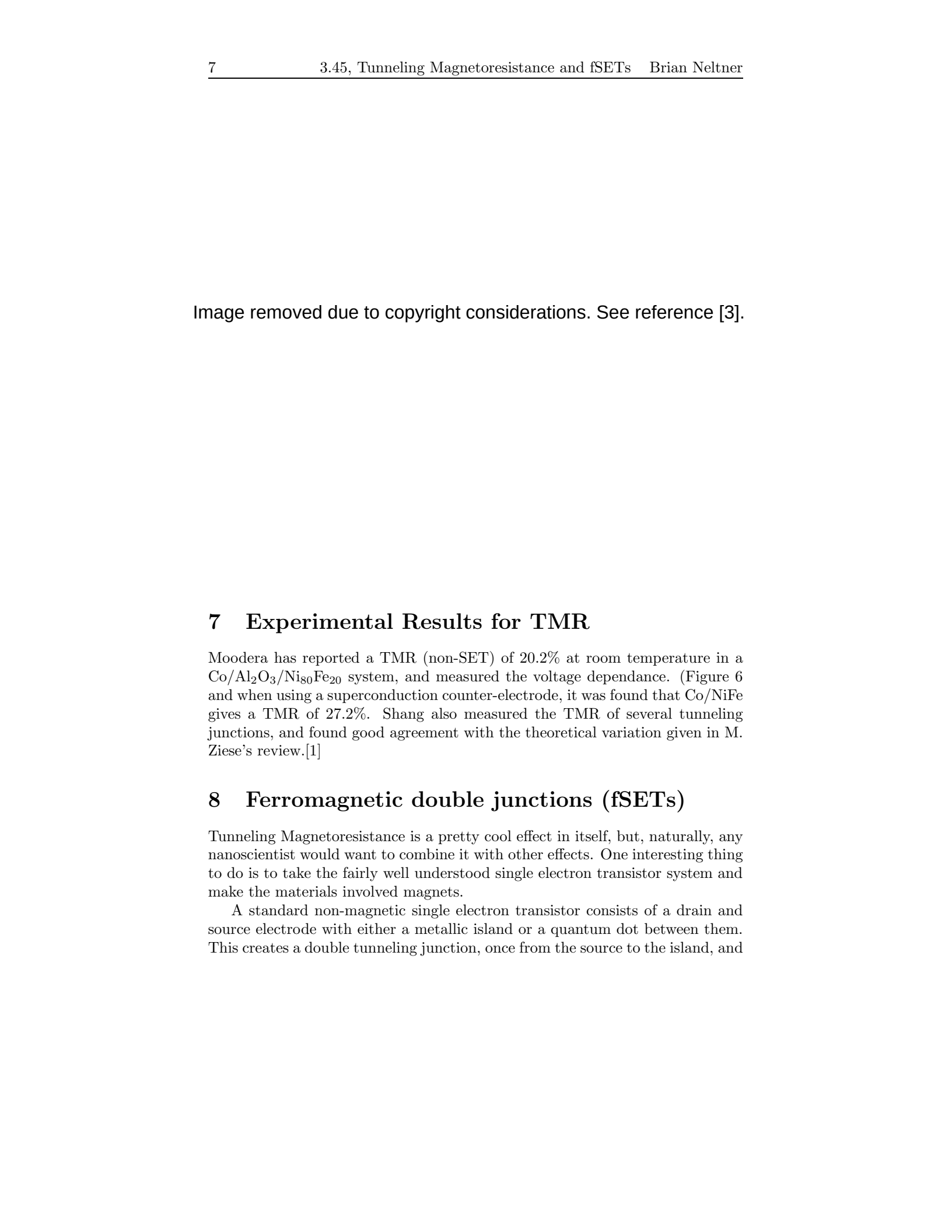


6 Coherent electron transport in the junction

It is also possible to create coherent electron transport by making the characteristic length scale of the system less than the phase coherence length. In the Kondo and Fano regimes of resonance, it has been shown that the difference between the resistance for the parallel and antiparallel cases can be very large due to the large change in number of conducting channels when magnetization changes direction.[8]

B.R. Bulka attempts to calculate the coherent electronic transport between ferromagnetic electrodes separated by a nonmagnetic nanoparticle.[9] By using various methods, he finds the currents and magnetoresistances of a model system with identical electrodes. Figure 4 uses a slave-boson method within the mean-field approximation, while Figure 5 uses an equation of motion method.

As can clearly be seen, the method of calculation drastically effects the results found, but in the Kondo and empty state regimes, they both give the same results. Bulka predicts a large magnetoresistance in the mixed valence regime with a strong temperature dependance.



7 Experimental Results for TMR

Moodera has reported a TMR (non-SET) of 20.2% at room temperature in a $Co/Al_2O_3/Ni_{80}Fe_{20}$ system, and measured the voltage dependance. (Figure 6 and when using a superconduction counter-electrode, it was found that Co/NiFe gives a TMR of 27.2%. Shang also measured the TMR of several tunneling junctions, and found good agreement with the theoretical variation given in M. Ziese's review.[1]

8 Ferromagnetic double junctions (fSETs)

Tunneling Magnetoresistance is a pretty cool effect in itself, but, naturally, any nanoscientist would want to combine it with other effects. One interesting thing to do is to take the fairly well understood single electron transistor system and make the materials involved magnets.

A standard non-magnetic single electron transistor consists of a drain and source electrode with either a metallic island or a quantum dot between them. This creates a double tunneling junction, once from the source to the island, and

another from the island to the drain. When the capacitative charging energy ($E_C = e^2/2C$) of the island is larger than the thermal energy, it is found that charge must build up in quantized chunks. This essentially forces the current through the double junction to increase in 2 electron steps every time the Fermi energy of the electrons in the source electrode gets high enough to allow electrons to tunnel through the barrier to the island.

The situation becomes much more interesting with the introduction of ferromagnetic materials– spin is assumed conserved in the tunneling event, but once on the island, it can flip to tunnel to the anti-ferromagnetic drain (which merely requires a relaxation time), continue on it's merry way if the drain is also ferromagnetic, or, in the Kondo limit, you can even have coherent electron transport going on. In addition, if your island itself is ferromagnetic, you have the possibility of the spin of an electron which has tunneled from the source to the island now affecting the magnetization of the island, resulting in second order effects in the system.

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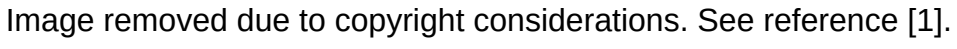
9 Quantum Dot or Single-electron Islands

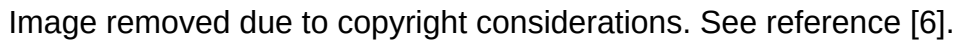
W. Rudziński found theoretically[6] that for a system of ferromagnetic electrodes with a nonmagnetic quantum dot or single atom as an island, the TMR was strongly related to the factor Ω, defined by

$$\Omega = \frac{1}{\gamma_0 \tau_S},$$

where γ_0 is a parameter for a material related to the ratio of the spin-flip and tunneling rates. Ω=0 corresponds to no spin relaxation, $\Omega \gg 1$ corresponds to fast spin relaxation (Figure 7) It can be clearly seen that using a material in which spins have a long spin relaxation time have a much larger TMR ratio. This is particularly interesting because this model not only applies to an island, but also to any other form of impurity between the two tunneling electrodes.

Later, Ping Zhang included spin-orbit coupling while an electron is on the quantum dot in conjunction with prior results for the spin-flip mechanisms in





GaAs quantum dots to calculate resistances. The general gist of the derivation is that the electrons are being forced into a superposition of spin up and spin down states relative to the electrodes as the quantization axes are fixed by the internal magnetization. That coherence needed to be taken into account to solve more exactly the resistance of the system.

This method eventually found that the Kondo resonance for spin up electrons is suppressed while the resonance for spin down electrons is enhanced. In addition, when there is a large spin-flip transition, the Kondo resonance is actually split into three energies for parallel electrodes, while splitting into only two energies when the electrodes are antiparallel.(Figure 8)

Zhang ends up calculating that when the dot level is higher than the chemical potential at the electrodes, the TMR is enhanced up to 160%.

10 Spin Accumulation on a Ferromagnetic Island

Another interesting case is where you allow the island to be ferromagnetic as well. By including spin accumulation on the island (the spin relaxation time is long on the island), J. Barnas found that you can enhance TMR, generate TMR when the central electrode is naturally non-magnetic, cause a negative differential resistance, or even reverse the sign of the tunnel Magnetoresistance. [5]

The key difference between having a non-magnetic and ferromagnetic island is that with a ferromagnetic island, the electron energy levels are no longer degenerate (spin up and spin down electrons are split in energies).

Following the Barnas derivation, there are a source and drain, both ferromagnetic, and either parallel or anti-parallel in magnetization, with the island ferromagnetic and parallel to the source in magnetization. There is also a gate electrode (sufficiently far away as to avoid tunneling) which controls the energy of the electrons on the island through a gate voltage applied capacitively.

You then assume that spin is conserved upon tunneling, and that the resistances for each spin through the tunneling junctions are greater than $2R_Q$, the quantum resistance.

The single-electron energy levels are then calculated for the island taking into account all the magnetic energy contributions except for the Zeeman energy which is assumed to be small. Barnas then assumes that the energy spectrum of the electrons is shifted rigidly when the gate voltage is changed. This allows them to calculate the tunneling rate of electrons from the drain to the level i of the electrode as

$$\Gamma_{i_\sigma}^{l(r)} = \frac{2\pi}{\hbar}|M_{i_\sigma}^{l(r)}|^2 D_\sigma^{l(r)}. \tag{15}$$

with $M_{i_\sigma}^{l(r)}$ as the average matrix element for transitions from the drain electrode

to the i_σ level in the island and $D_\sigma^{l(r)}$ is the spin-dependent density of states in the drain.

The magnetic moment on the island is $M = N_\uparrow - N_\downarrow$ and the excess magnetic moment is $M^* = M - M_0$, with M_0 the equilibrium magnetization of the island. Then, the probability P is determined by minimizing $\frac{\partial P(\{n\})}{\partial t} = 0$, which includes all possible tunneling contributions to the distribution growth or decays along with internal relaxation processes on the island and the influence of the gate voltage.

If you then restrict to a short electronic relaxation time while maintaining a much longer spin relaxation time, you find that the electrons are thermalized. This gives you a formula for electric current as a function of $P(N_\uparrow, N_\downarrow)$, the probability that the island has $N_\uparrow$ spin up electrons and $N_\downarrow$ spin down electrons–

$$\begin{aligned} I = e \sum_{N_\uparrow, N_\downarrow} \sum_{\sigma} \sum_{i_\sigma} P(N_\uparrow, N_\downarrow) \\ \times \left\{ \left[1 - F(E_{i_\sigma}|N_\uparrow, N_\downarrow)\right] \Gamma^r_{i_\sigma} f(E_{i_\sigma} + E^{r-}_{N*} - E_F) - \right. \\ \left. - F(E_{i_\sigma}|N_\uparrow, N_\downarrow) \Gamma^r_{i_\sigma} \left[1 - f(E_{i_\sigma} + E^{r-}_{N*} - E_F)\right] \right\}. \end{aligned} \tag{16}$$

where

$$F(E_{i_\sigma}|N_\uparrow, N_\downarrow) = \left\{ 1 + \exp\left[\frac{E_{i_\sigma} - (1/2)(E_{1_\sigma} + E_{2_\sigma})}{(1/2)k_B T} \right] \right\}^{-1}. \tag{17}$$

This gives a probability in the state space as shown in Figure 9, as well as a current and TMR as shown in Figure 10.

In Figure 10, there are two interesting features, best seen on the graph of the derivative of the current with respect to voltage. The first is the large peaks, which are from the coulomb blockade behaviour. The second is the small peaks between the large peaks– these represent the opening of a tunneling channel with a new value of excess spin on the island. This predicted behaviour is similar to what is seem in experiments with Al particles or C_{60} molecules. In addition, it can be seen from the plot of TMR versus V that the TMR oscillates with the bias voltage, and decreases overall as V increases.

If it is assumed that the island to drain junction is more resistive than the source to drain junction, it is found that electrons (and thus spin) accumulate on the island. For overall charge accumulation, the parallel and antiparallel configurations are nearly identical, but the antiparallel configuration has a far greater spin accumulation than the parallel configuration. (Figure 11)

As temperature increased, the small peaks disappeared quickly, followed by the coulomb charging peaks at around 60 K for this theoretical system.

11 Very small barriers

In 1999, X.H. Wang did an analysis of fSETs in the regime where the tunnel resistance is smaller than the quantum resistance h/e^2. Following his work[4],

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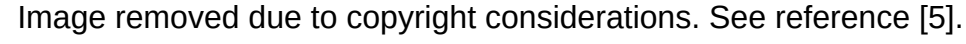
we attempt to solve the tunneling Hamiltonian in a nonperturbative manner by using path integrals. By using the standard single-electron tunneling Hamiltonian with a spin-dependent density of states, we find that the high-temperature TMR ratio is simply

$$\gamma_{classical} = \frac{2PP'}{1 - PP'}, \tag{18}$$

the same equation we had calculated before for a simple ferromagnetic tunneling junction.

Taking the coulomb charging effects into account as we lower the temperature, we find that the action of our system is given by

$$S_\eta[\phi] = \int_0^\beta \mathrm{d}\tau \frac{\dot{\phi}^2(\tau)}{4\mathrm{E_C}} - \alpha_\eta^{cl} \int_0^\beta \mathrm{d}\tau \int_0^\beta \mathrm{d}\tau' \chi(\tau - \tau') \times \cos[\phi(\tau) - \phi(\tau')]. \tag{19}$$



where η represents either the parallel or antiparallel state of the two leads, α_η^{cl} represents the normalized classical conductance, and ϕ represents the angle of the magnetism at a particular τ. The boundary condition is that $\phi(\beta) = \phi(0) + 2\pi k$.

Wang uses a Monte Carlo simulation to solve this path integral, finding that when the tunnel resistance is close to the quantum resistance, the result is the same as for the cotunneling solution for a SET. He then chose to use Ni leads with a spin polarization of P=0.23 and a Co island where P=0.35, along with a typical value of α_η^{cl}=10, as this was one of the few setups that had been experimentally observed at the time.

They found that the TMR is a strong function of temperature and reaches the cotunneling limit when $E_C/k_BT \approx$15. Including higher order terms give an enhancement of 90%(!!!) to the TMR when $E_C/k_BT \approx$40. Fitting their

numerical results to a second order function in E_F/k_BT gives

$$\gamma^{SE} = \gamma^{cl} + \mu^{SE}\left(1+\gamma^{cl}\right)\left(\frac{E_C}{k_BT}\right)^2, \tag{20}$$

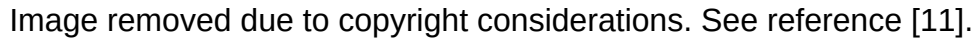
with

$$\mu^{SE} = 0.04\alpha_M\left[\frac{1+PP'}{(1+P)(1+P')}\right].$$

Clearly, this allows for a great deal of control over the TMR through varying the materials the leads are made of– by simply choosing an alloy with a different spin-polarization or α, you can drastically change the TMR in the low temperature limit.

12 Experimental Results

It was found by L.F. Schelp [11] that in a fSET formed from Co electrodes with an Al_2O_3 barrier containing a layer of small, disconnected Co clusters (Figure 12), coulomb blockade effects were induced by tunneling through the clusters. They found that the TMR did not change as temperature was varied from 50K to room temperature. They did, however, find that if they applied a strong external field (which saturated the electrodes and islands), the resistance



went down drastically. (Figure 13). In addition, they found a dependance on the applied voltage. (Figure 14)

13 Conclusions

Magnetoresistance is of great interest to modern nanoscience as it offers unique abilities in reading magnetic memory, storing magnetic bits in MRAM, and spintronics. Originally, Magnetoresistance was seen as a curiosity of magnetic materials which caused a fluctuation of a few percent in resistance as a material became magnetized. Recently, the discovery of Anisotropic Magnetoresistance, Giant Magnetoresistance, and Tunneling Magnetoresistance have all made this property much more practical for use in our daily lives. Anisotropic Magnetoresistance gave us resistance fluctuations which were dependant upon the direction of magnetization, allowing us to read the direction of magnetic bits on magnetic recording material. Then, Giant Magnetoresistance gave us a mechanism to again read bits stored in magnetic orientation, but with a much greater change

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in resistance. Much more recently, Tunneling Magnetoresistance has produced systems where rotating the magnetic field of one electrode can result in a change in resistance of 20% easily, and with the potential of a several thousand percent change in resistance in the case of half-metallic electrodes.

As our ability to manipulate materials on a nanoscale improves, we will undoubtably be able to produce tunneling magnetic junctions and even ferromagnetic single-electron transistors with extremely high changes in resistance that operate at room temperature. As this technology becomes available, the density of magnetic material will become even greater, and as costs come down, Giant Magnetoresistance and even Tunneling Magnetoresistance will be used

in Magnetic RAM, a non-volitile replacement for traditional RAM which uses magnetic instead of capacitative storage– conveniently alse removing the time constant limitation inherant in all LRC circuits.

All in all, the various forms of Magnetoresistance are one of the most useful areas of research in magnetic materials today.

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