Giant Coulomb blockade magnetoresistance in magnetic tunnel junctions with a granular layer

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We show that the Coulomb blockade voltage can be made to depend strongly on the electron spin in a discontinuous magnetic granular layer inserted in the middle of an insulating layer of a tunnel junction. This strong spin dependence is predicted from the local intergranular magnetoresistance effects, including giant magnetoresistance (GMR), tunneling magnetoresistance, colossal magnetoresistance, or GMR through a polymer spacer. The resulting Coulomb blockade magnetoresistance (CBMR) ratio can exceed the magnetoresistance ratio of the granular layer itself by orders of magnitude. Unlike other magnetoresistance effects, the CBMR effect does not require magnetic electrodes.

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Since the discovery of the giant magnetoresistance (GMR) (Refs. 1 and 2) effect in magnetic multilayers, each discovery of a magnetoresistance effect, from the tunneling magnetoresistance (TMR) (Refs. 3 and 4) effect, to the colossal magnetoresistance (CMR) (Ref. 5) effect, or the GMR effect with a polymer spacer,[6] brings forth a significant progress in our understanding of the physics of spintronics and properties of materials. The GMR and TMR effects in particular have also led to breakthroughs in the electronics and computer technologies. As the magnetoresistance ratio (MR) of today's best TMR junctions[7–9] rapidly approaches the theoretically predicted value,[10] one is compelled to look for magnetoresistance mechanisms that will shatter such theoretical limit and at the same time are as practical as GMR and TMR effects for device applications.

In this paper, we propose strongly spin-dependent Coulomb blockade (CB) voltage as a possible mechanism for producing very large MR. To be consistent with the literature of CB, we refer to the metal islands of the granular film as quantum dots (QDs). The spin dependence of the CB voltage is realized through the coupling between the QDs due to spin-dependent electron transport, as illustrated in Fig. 1. The basic mechanism is similar to the so-called "collective Coulomb blockade."[11] The collective blockade voltage is determined by the interdot tunneling conductance and does not rely upon other factors such as intradot energy level spacing.[12,13] As the interdot conductance is enhanced, the collection of the QDs that are coupled through tunneling act together as a single, larger QD. This causes a reduction in the CB voltage. When interdot coupling is controlled by the magnetic moments of the QDs, the Coulomb blockade magnetoresistance (CBMR) effect is produced. We will show that the CBMR can reach tens of thousands of percent, easily surpassing the best single barrier TMR junctions available today. We further show that some of the published experimental data that exhibit very large TMR[18–20] may have already contained the CBMR effect. Finally, we will suggest candidate materials and suitable parameter range for realizing this effect.

The CBMR effect can be explained using a double dot tunneling system as illustrated in Fig. 1. The CB voltage of this system depends strongly on interdot tunneling conductance, with larger interdot conductance corresponding to smaller CB voltage.[12,13] When the magnetic moments of two neighboring QDs are antiferromagnetically oriented, the interdot conductance is small and the CB voltage is large, as in Fig. 1(a). When the moments are aligned parallel under an applied magnetic field, the interdot conductance is large and the CB voltage is small, as in Fig. 1(b). In the extreme case, if the interdot conductance is less than the quantum conductance e^2/h for the antiparallel alignment and greater than e^2/h for the parallel alignment, the CB voltage may change by as much as a factor of two.[12] Even if the change in the CB voltage is much less than that, when the applied bias voltage is between the two blockade voltages, the tunneling current switches from nearly zero to finite under a magnetic field leading to a nearly infinite magnetoresistance.

The mechanism proposed here is different than the previous considerations of spin-dependent Coulomb blockade effect.[14–16] In previous models the Coulomb blockade voltage is assumed to be a constant and the magnetoresistance arises from the spin-dependent tunneling resistance between the electrodes and the quantum dots. In our model the Coulomb blockade voltage itself is spin dependent which leads to a much larger magnetoresistance. Below we write out in explicit form the tunneling conductance in order to illustrate this difference. We consider the zero temperature $T=0$ case of the model in Ref. 14. The current for the spin channel σ through a single QD (labeled by i) is given by,[17]

$$I_\sigma^i = e\sum_{n=-\infty}^{\infty} p_n^i \Gamma_\sigma^i(n), \tag{1}$$

where p_n^i is the probability of QD i occupied by n electrons, and $\Gamma_\sigma^i(n)$ is the forward tunneling rate. The backward tunneling rate is zero at $T=0$ under a finite bias voltage V. The total current is obtained by summing over all QDs and both spin channels,

$$I = \sum_{i\sigma} I_\sigma^i. \tag{2}$$

To further simplify the model, we consider only the $n=0$ term. The forward tunneling rate is calculated from,[14]

$$\Gamma_\sigma^i(0) = \frac{\hbar}{2\pi e^4 R_{i\sigma}^2}\int_0^{eV} dE E(eV-E)\left|\frac{1}{E+E_C^i-\frac{1}{2}eV+i\gamma_\sigma}+\frac{1}{-E+E_C^i+\frac{1}{2}eV+i\gamma_\sigma}\right|^2, \tag{3}$$

which yields,

$$\Gamma_\sigma^i(0) = \frac{\hbar}{2\pi e^4 R_{i\sigma}^2}\left\{\left[E_C^i+\frac{\frac{1}{4}(eV)^2+\gamma_\sigma^2}{E_C^i}\right]\ln\frac{\left(E_C^i+\frac{1}{2}eV\right)^2+\gamma_\sigma^2}{\left(E_C^i-\frac{1}{2}eV\right)^2+\gamma_\sigma^2}+2\left[\frac{\frac{1}{4}(eV)^2-(E_C^i)^2}{\gamma_\sigma}-\gamma_\sigma\right]\left[\tan^{-1}\frac{E_C^i+\frac{1}{2}eV}{\gamma_\sigma}-\tan^{-1}\frac{E_C^i-\frac{1}{2}eV}{\gamma_\sigma}\right]\right\}, \tag{4}$$

where E_C^i is the Coulomb energy of an electron on the QD and $R_{i\sigma}$ is the tunneling resistance between one of the electrodes and the QD i. The decay rate of the QD charge state γ_σ is given by,

$$\gamma_\sigma = \frac{\hbar E_C^i}{e^2 R_{i\sigma}}. \tag{5}$$

By taking the limit of $\gamma_\sigma \ll E_C^i$ (or equivalently $R_{i\sigma} \gg \hbar/e^2$) which is valid for most magnetic tunnel junctions, we arrive at,

$$\Gamma_\sigma^i = \begin{cases} 0, & V < V_C^i; \\ \dfrac{\frac{1}{4}(eV)^2-(E_C^i)^2}{e^2 R_{i\sigma} E_C^i}, & V > V_C^i; \end{cases} \tag{6}$$

where $V_C^i = 2E_C^i/e$ is the Coulomb blockade voltage. Here the small cotunneling current at small voltages is set to zero for convenience but will be restored later in Eq. (13). The differential conductance is $G_i(V) = \Sigma_\sigma \partial I_\sigma^i/\partial V$ or,

$$G_i(V) = \begin{cases} 0, & V < V_C^i; \\ \dfrac{V}{V_C^i R_i}, & V > V_C^i; \end{cases}, \tag{7}$$

where $1/R_i = \Sigma_\sigma 1/R_{i\sigma}$. Unlike previous models, we neglect the dependence of R_i on the moment alignment of the QD with respect to that of the electrodes, focusing instead on the much larger effect due to the spin dependence of V_C^i.

For a double dot system the magnetoresistance is simply determined by the change in V_C^i when the moments of both QDs are aligned by an external magnetic field. In real samples instead of a double dot we have a discontinuous middle layer with a wide distribution of QDs. In this case we need to integrate the tunneling current contribution over the entire distribution. We assume that each QD (labeled by i) has the shape of a flat disk with a diameter d_i, and is oriented parallel to the film layers. The blockade voltage for QD i is given by,[21]

$$V_C^i = \frac{8eD}{\pi\varepsilon_0\varepsilon d_i^2}, \tag{8}$$

where D is the effective thickness of the barriers on both sides of the QD, ε is the dielectric constant of the insulating layer and ε_0 is the vacuum permittivity. We further assume that the conductance $1/R_i$ is proportional to the cross-section area A_i of the QD, $1/R_i = (e^2/h)(A_i/S)$, so that S is a constant independent of the QDs. The total conductance is the sum over all particles,

$$G(V) = \sum_i G_i(V) = \frac{e^2}{h}\sum_{V \geq V_C^i}\frac{VA_i}{V_C S}. \tag{9}$$

Using Eq. (8), the restriction on the sum over i is equivalent to,

$$G(V) = \frac{\pi}{4}\frac{e^2}{hSx_V^2}\sum_{d_i \geq \sqrt{\frac{8e}{\pi\varepsilon_0\varepsilon}\frac{D}{V}}} d_i^4 = \frac{\pi}{4}\frac{e^2}{hSx_V^2}\int_{x_V}^{\infty} x^4 F(x)dx, \tag{10}$$

where the size distribution of the QDs is given by $F(x)$ and $x_V = \sqrt{(8e/\pi\varepsilon_0\varepsilon)(D/V)}$. Under zero magnetic field, we assume that there is no coupling between the QDs therefore the effective size distribution is the actual size distribution of the granular film, $F_0(x)$. With an applied magnetic field H, the effective size distribution is changed to $F_H(x)$, which is obtained by combining neighboring pairs of QDs into single QDs. Assuming that there is no correlation between the nanoparticles,

$$F_H(x) = \int_0^x dx_1 \int_0^x dx_2 F_0(x_1)F_0(x_2)\delta(x_1+x_2-x). \tag{11}$$

The net MR of the entire junction is expressed in terms of the conductance ratio,

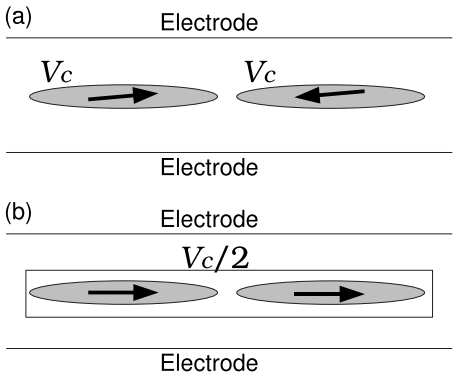

FIG. 1. (a) When the moments of two magnetic QDs are antiferromangetically oriented, the CB voltage is V_c for each dot. (b) An external magnetic field aligns the moments of the two dots, which are now coupled electronically due to enhanced interdot tunneling, and the CB voltage is reduced to $V_c/2$.

$$\frac{G_H(V)}{G_0(V)} = \frac{(\lambda_0/\lambda_H)^2 \int_{x_V}^{\infty} x^4 F_H(x)dx}{\int_{x_V}^{\infty} x^4 F_0(x)dx}, \tag{12}$$

with $(\lambda_0/\lambda_H)^2 \int_0^\infty x^2 F_H(x)dx = \int_0^\infty x^2 F_0(x)dx$ to keep total area unchanged, and λ_H and λ_0 are the effective mean island size with and without the magnetic field, respectively. One of the main features of Eq. (12) is its independence of the magnetic configuration of the electrodes. It suggests that the CBMR effect can be realized *even with nonmagnetic electrodes*. This is a significant advantage over other magnetoresistance effects in device applications.

We tested several forms of the functions $F_0(x)$ and $F_H(x)$ and found that the CBMR is robust as long as the two functions are peaked at different voltages. As an illustration, we show the results for the following distributions $F_0(x) = 2x \exp(-x^2/\lambda_0^2)/\lambda_0^2$ and $F_H(x) \approx 2x \exp(-x^2/\lambda_H^2)/\lambda_H^2$,

$$\frac{G_H(V)}{G_0(V)} = \frac{(\lambda_0/\lambda_H)^2 (x_V^2 + 2\lambda_H^2 + 2\lambda_H^4/x_V^2)\exp(-x_V^2/\lambda_H^2) + aV^2}{(x_V^2 + 2\lambda_0^2 + 2\lambda_0^4/x_V^2)\exp(-x_V^2/\lambda_0^2) + aV^2}, \tag{13}$$

where aV^2 accounts for the cotunneling current at small voltages[17] with a as a spin-independent fitting parameter. Neglecting the spin dependence of a leads to zero TMR at V $=0$ but does not affect the main features of CBMR.

In Fig. 2(a) we plot the ratio $G_H(V)/G_0(V)$ given by Eq. (13) for different values of λ_H/λ_0 and with $a/\lambda_0^2 = 5 \times 10^{-3}$ V^{-2} and $V_C = 8eD/(\pi\varepsilon_0\varepsilon\lambda_0^2) = 1$ V. If we assume MgO thickness $D = 2.5$ nm and $\varepsilon = 9.6$, then the mean island diameter $\lambda_0 = 3.4$ nm. The figure shows that a modest $\lambda_H/\lambda_0 = 1.1$ can lead to a giant CBMR. If the ratio is improved to $\lambda_H/\lambda_0 = 2$, then the resulting CBMR ratio exceeds even the best GMR and TMR devices today by orders of magnitude. Note the double peak feature with a suppressed MR at low voltages. These features are in qualitative agreement with the recent double barrier measurement that yielded over 1 000% TMR at room temperature.[19]

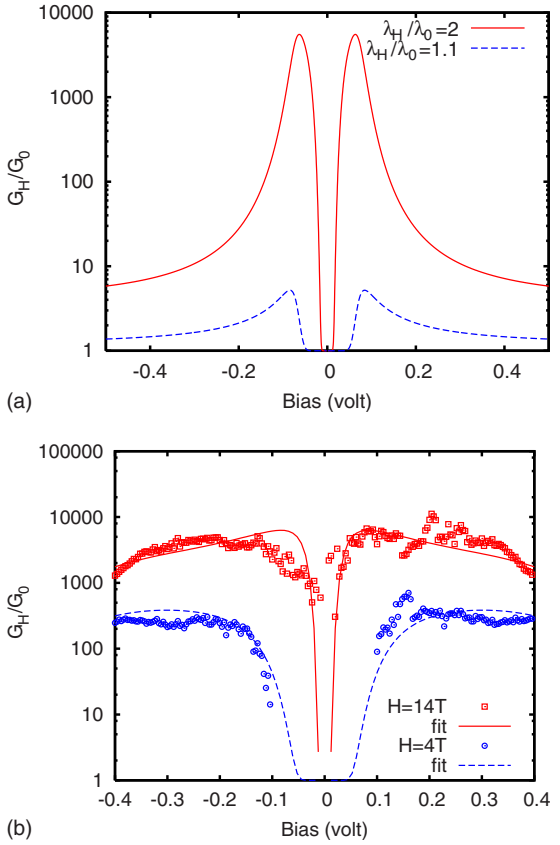

FIG. 2. (Color online) (a) Ratio $G_H(V)/G_0(V)$ given by Eq. (13) for two spin-dependent QD size distributions, $\lambda_H/\lambda_0 = 1.1$, and $\lambda_H/\lambda_0 = 2$. (b) Comparison of Eq. (13) with experiment (Ref. 18) for a LaSrMnO/SrTiO/LaSrMnO junction with the fitted parameters $\lambda_H/\lambda_0 = 2.33$ for $H = 4$ T and $\lambda_H/\lambda_0 = 4.67$ for $H = 14$ T.

A single nanometer QD is likely superparamagnetic and does not exhibit magnetic hysteresis. In a conventional magnetic tunnel junction this leads to a diminished MR. But the CBMR relies on the change in the collective CB voltage which does not require magnetic hysteresis. This explains the sizeable MR in Ref. 19 for the samples with 0.8 nm and 1.0 nm middle layers (note the absence of hysteresis in Fig. 1 of Ref. 19). In addition, the field for in-plane moment rotation is small, also evident from the right-hand curves, especially one for the 1.2 nm sample.

In Fig. 2(b) we compare Eq. (13) with recently measured TMR (Ref. 18) of a LaSrMnO/SrTiO/LaSrMnO tunnel junction at 4 K. In that work, although the blockade effect due to impurities inside the barrier layer is identified, the huge MR that does not saturate with the applied magnetic field and the unusual voltage dependence of the TMR remain unexplained. Here we offer CBMR as a possible explanation of the extraordinary magnetoresistance effect. Using Eq. (13), the fitted parameters are for $H = 4$ T, $\lambda_H/\lambda_0 = 2.33$, and for $H = 14$ T, $\lambda_H/\lambda_0 = 4.67$, and both with $a/\lambda_0^2 = 1.22 \times 10^{-2}$ V^{-2} and $V_C = 8eD/(\pi\varepsilon_0\varepsilon\lambda_0^2) = 4.11$ V. Figure 2(b) establishes a quantitative agreement between the model and the experiment for the bias voltage dependence. There is a strong field dependence of the effective QD size. The MR continues to rise long after the moments in both electrodes

are already aligned with the field, indirectly supporting our proposal that the CBMR does not rely on the magnetic configuration of the electrodes thus can be realized with nonmagnetic electrodes.

We now discuss other intergranular magnetoresistance mechanisms for CBMR. Polymer GMR is similar to TMR but with the insulating layer replaced by a spacer layer made of organic polymer materials. Organic materials have the advantage of very long spin diffusion length due to their weak spin-orbit coupling.[22] The polymer layer can be up to several hundred nm thick. If we use $D=100$ nm and $\varepsilon=3$, then for $V_C=4$ V one needs $\lambda_0=20$ nm. The nanoparticles can be made into spherical shape to reduce the magnetic anisotropy and allow a smaller switching field. A recent work[20] on CoFe nanoparticle superlattices in an organic medium shows a possible collective CB effect with a large MR. Although the phenomenon in that material is much more complex than the CBMR predicted here, it provides a supporting evidence that CBMR is possible in an organic medium.

In the case of intergranular GMR coupling, let us consider a granular layer consisting of Cu and Co nanoparticles, specifically, a composite nanoparticle made of two Co particles separated by a Cu particle. This is a nanoscale version of a GMR spin valve. If we neglect spin-flip scattering, then for the majority spin electrons all three regions of the spin valve have nearly the same potential when the moments of the two Co particles are aligned, leading to a nearly uniform charge distribution. But when the moments of the two Co particles are antiparallel to each other, the majority spin electrons see different potentials in the two Co particles. This difference creates a spin-dependence in the charge distribution across the nanoparticles. Although the value shall depend on the shape and the relative sizes of the Co and Cu nanoparticles, if we assume a modest spin dependence of the effective size distribution $\lambda_H/\lambda_0 \approx 1.1$, from Fig. 2 we can see that the MR can reach close to one hundred percent. Note that this is achieved without any magnetic electrodes. The parameter requirement is similar to TMR.

The last example is the case of CMR. Instead of embedding QDs into the barrier layer as in the previous cases, in this case we exploit the inherent inhomogeneity of the manganite materials. La(Sr/Ca)MnO can be an antiferromagnetic insulator within a certain concentration range of Sr or Ca doping.[23,24] However, these compounds are dominated by fluctuations of mixed phases.[25] Furthermore, the local tunneling conductivity between these phase domains has a strong dependence on the external magnetic field.[26] In addition, such conductance fluctuation is nonpercolative, allowing electrons to remain localized when the magnetic field is increased. Thus, using manganite materials as the insulating layer of a tunnel junction may provide a CBMR mechanism with a continuously increasing MR as a function of the applied magnetic field, similar to the LaSrMnO/SrTiO/LaSrMnO junction discussed above. The magnitude of the MR is also expected to be similarly large.

In conclusion, we propose a CBMR effect based on a strongly spin-dependent CB voltage arising from intergranular magnetoresistance (GMR, TMR, CMR, or GMR with a polymer spacer) effects. This CBMR effect is expected to be robust in the sense that with sufficiently small QD size, the CB voltage is large enough to overcome thermal smearing so that the CBMR persists up to the room temperature. The magnitude of the CBMR is expected to exceed MR of the underlying spin-dependent coupling mechanism within the granular layer. Thus the CBMR based on TMR could yield MR up to tens of thousands of percent. The CBMR is distinct from previous attempts to combine the CB effect and the TMR effect to yield an enhanced MR.[14,16] In previous works, the CB voltage was not made to depend on the electron spin, therefore the MR enhancement relies on higher order effects. Experimentally, whereas the measured TMR for MgO based single barrier tunnel junctions can easily reach several hundred percent, even with the TMR enhancement similar measurements for double barrier junctions yield significantly lower MR.[16] The lack of spin-dependent CB voltage is the fundamental difference between previous works and the CBMR. An additional practical difference is that the CBMR does not require the use of magnetic electrodes. This feature shall open up broader applications for the CBMR.

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