Spin transport in ferromagnetic/normal-metal tunnel junction arrays

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An array of alternating ferromagnetic and normal-metal islands separated by small tunnel junctions is theoretically investigated in the sequential tunneling regime. A numerical Monte Carlo method is used to calculate the transport properties. The spin-dependent tunneling currents give rise to nonequilibrium spin accumulation on the normal island. The tunneling magneto resistance (TMR) is calculated for a large range of array parameters. The TMR oscillates with bias voltage and can become negative for certain array parameters. We show that the long-range electrostatic interaction in the arrays can significantly increase the TMR; for experimentally accessible parameters the magnitude of the TMR can be as large as 100%.

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I. INTRODUCTION

Electronic devices, which exploit the spin as well as the charge of the electron, have attracted much interest over the last couple of decades. This interest in spintronics is stimulated by industrial applications, such as read-heads in hard disk drives, magnetic random access memory, magnetic-tunnel-junction-based sensors, as well as basic physics, offering new ways to control these two fundamental properties of the electron.[1–3]

One important spintronic device is the ferromagnetic tunnel junction which will have a resistance depending on the relative orientation of the magnetization of the two electrodes. The figure of merit for such a device is the fractional change in resistance when the magnetization configuration changes, known as the tunneling magnetoresistance TMR $= (R_{ap} - R_p)/R_p$, where ap (p) stands for antiparallel (parallel) magnetization of the electrodes. In the original model of a ferromagnetic tunnel junction, the so-called Julliere model,[4] the TMR is only a function of the polarization P of the tunneling electrons (here assumed to be equal for the two ferromagnetic electrodes): TMR $= 2P^2/(1 - P^2)$. If the size of the junctions and electrodes of the device becomes small, two phenomena of mesoscopic physics will begin to play a role—the charging effects of small capacitance tunnel junctions and the nonequilibrium spin accumulation on small-volume electrodes. The transport properties will then be determined by the interplay between spin-dependent tunneling, charging effects, and spin accumulation[5–7] and the tunneling magnetoresistance will no longer be a constant determined by a single material parameter as is the case in the Julliere model.

One of the most studied structures that combines charging effects and spin-dependent transport is the single electron transistor (SET) which consists of two junctions in a series separating a small island. Several material combinations of the SET has been both experimentally and theoretically examined: SETs with ferromagnetic leads and ferromagnetic island (F/F/F) SET,[5] ferromagnetic leads and normal-metal island (F/N/F) SETs,[8–11] as well as superconducting island (F/S/F) SETs.[12–15] In these types of devices the TMR typically oscillates with voltage and can in some cases even change sign. In the case of a nonmagnetic island the spins can accumulate on the island creating a nonequilibrium splitting of the chemical potential thereby affecting the tunneling through the junctions resulting in a difference in current for different magnetization of the outer electrodes.

One can also create devices containing more than two junctions. For example, there has been an experimental interest in devices containing self-assembled granular ferromagnetic nanoparticles displaying both spin-dependent and charging effects. The granular structures are typically large two-dimensional arrays of nanoparticles with varying sizes, but can be fabricated into smaller structures like one-dimensional arrays or SETs.[16] Theoretically, a three-junction array was examined in Ref. 17 where oscillating as well as sign-changing TMR was found. Recently, large TMR was predicted in arrays with normal-metal islands and ferromagnetic outer electrodes.[18]

In this article we numerically examine an array of alternating ferromagnetic and normal-metal islands connected by tunnel junctions. We show that for certain parameters this device can have a very large negative TMR with a magnitude as large as 100%, which is an order of magnitude larger than the TMR of an F/N/F SET with similar parameters. The TMR depends nonmonotonically on the array length with smaller TMR in the limit of short or long arrays. We find that to achieve maximal TMR signal for reasonable array parameters the ideal number of junctions is the order of 10. The large negative TMR will occur when the spin-relaxation time is long, but finite, and when the ratio of junction capacitance to island capacitance to ground is large. The negative TMR is a consequence of the increased sensitivity of the tunneling rates to small shifts in the potential of an individual island; here the shift in potential arises from spin accumulation in the antiparallel state of the ferromagnets. Finally, we estimate the parameters and feasibility of an experimental realization.

II. MODEL

We consider an array of N identical tunnel junctions separating alternating ferromagnetic (F) and normal (N) metal islands. In Fig. 1(a) the schematic of an F/N/F/... array is shown. Each island is capacitively coupled to a ground plane through a ground capacitor C_0. A transport voltage is applied across the array by the external voltages $+V/2$ and $-V/2$. We assume that the total capacitance of each island is sufficiently small in order to have the charging energy of the islands larger than the thermal energy k_BT, and that the tunnel resistance is

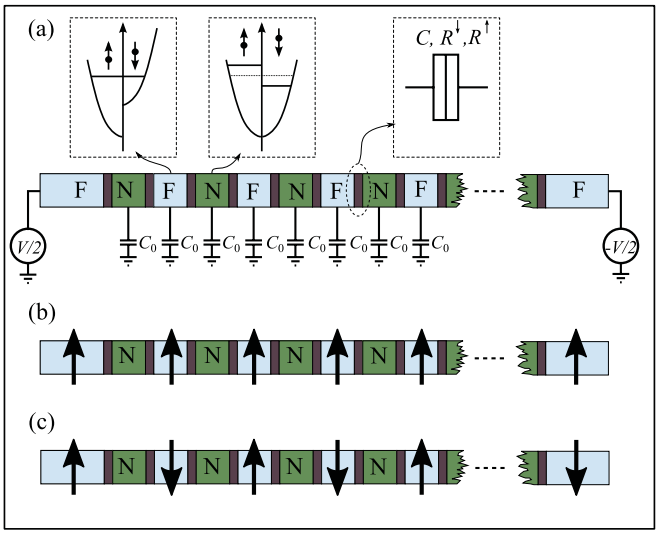

FIG. 1. (Color online) Schematics of the F/N/F/N. . . array. (a) A schematic of the array circuit showing the external voltages $\pm V/2$ and the ground capacitance C_0. The two left insets shows cartoons of the density of states in the ferromagnet and the normal metal, respectively. The density of states in the normal metal show the split chemical potential due to a nonequilibrium spin accumulate on the island. The right inset shows the tunnel junction characterized by the capacitance C, the spin-down resistance $R^{\downarrow}$, and the spin-up resistance $R^{\uparrow}$. The array is shown in the parallel state (b) and in the antiparallel state (c).

much larger than the quantum resistance:

$$R \gg R_{\mathrm{K}} = h/e^2 \approx 25.8\ \mathrm{k}\Omega. \tag{1}$$

Furthermore, we assume that the energy relaxation is significantly faster than all other dynamics, so the electrons are described by a Fermi distribution determined by the temperature (i.e., $\tau_{\mathrm{in}} \ll \Delta t$ where τ_{in} is the inelastic scattering time and Δt is the average time between successive tunnel events). These conditions ensure that the system is always in a well-defined charge state and hence the "orthodox" theory of single electron tunneling[19] applies. In this work we neglect co-tunneling, which is justified by the fact that the co-tunneling rate over N junctions is suppressed by a factor $(R_{\mathrm{K}}/R)^N$. However, for very small currents co-tunneling can still be the dominant transport mechanism but our results (e.g., the TMR) are in the regime of finite current from sequential tunneling and here the co-tunneling will be negligible due to the condition in Eq. (1).

The consequence of having ferromagnetic metals in the array is that the tunneling resistance for the two spin orientations will depend on the magnetization direction of the ferromagnet. This difference in resistance for the two spin orientations has the effect that a current does not only carry charge but also a net magnetization. The magnetization accumulates on the islands, creating a split chemical potential for different spin orientations [see middle inset of Fig. 1(a)]. We assume that the ferromagnets are single domained with a uniform magnetization which ensures that there will exist only two types of carriers: the majority electrons with spin parallel to the magnetization and the minority electrons with spin antiparallel to the magnetization. See left inset of Fig. 1(a) for a schematic of the density of states of the ferromagnet. We consider only collinear magnetization of the electrodes and restrict ourselves to two magnetic configurations of the array; the magnetization is either parallel (p) or the magnetization of the ferromagnets is pairwise antiparallel (ap) [see Figs. 1(b) and 1(c), respectively]. We follow the Julliere model[4] to calculate the spin-dependent resistances: $R^{\sigma} = 2R_0/(1+\sigma P)$ where $\sigma = +(-)$ for majority (minority) spins, P is the polarization of the tunneling electrons, and $R_0 = (1/R^{+} + 1/R^{-})^{-1}$ is the spin-*independent* total resistance of the junction.

We assume that the tunneling process preserves the spin orientation but the spin can relax with a characteristic spin-relaxation time on the islands. On the ferromagnetic island, coupling between the spins and the magnetization is very strong and the spins will instantly relax to the equilibrium spin distribution. However, on the normal-metal islands, the spin-relaxation time (τ) is finite and spins will accumulate here provided that the spin-relaxation time is not shorter than the time between successive tunneling events. The spin-dependent shift in the chemical potential from spin accumulation is given by

$$\Delta\mu_i^{\sigma} = n^{\sigma}[N_0\Omega]^{-1}, \tag{2}$$

where $[N_0\Omega]^{-1}$ gives the single-particle level spacing, N_0 is the density of states per spin, Ω is the island volume, and $\sigma = \uparrow, \downarrow$ is the spin index. Here, we have assumed that the spin distribution is uniform across the normal-metal islands which is the case as long as the spin diffusion length ($\lambda_s = \sqrt{D\tau}$ where D is the diffusion constant and τ is the spin-relaxation time) is not much smaller than the island dimensions.[20,21]

The state of the array is characterized by the configuration of spin-up ($n^{\uparrow}$) and spin-down ($n^{\downarrow}$) electrons on each island. The charge on an island is given by $ne = e(n^{\uparrow} + n^{\downarrow})$. A charge residing on an island in the array will influence the potential of the neighboring islands because of the capacitively coupling of the islands. The ratio of the ground capacitance to junction capacitance determines this length scale and is approximately $2\lambda^{-1}$ where $\lambda = \mathrm{acosh}(1 + C_0/2C)$ is known as the inverse soliton length. In the case $C_0 \ll C$ the soliton length is given by $\lambda^{-1} \approx (C/C_0)^{1/2}$.

To calculate the charge and spin transport properties of the array we need the spin-dependent tunneling rates. The tunneling rate for an electron to tunnel from island i to island $i \pm 1$ is given by[19]

$$\Gamma_{i,i\pm1}^{\sigma}(n^{\uparrow},n^{\downarrow}) = \frac{1}{e^2R_i^{\sigma}} \frac{\Delta E_{i,i\pm1}^{\sigma}(n^{\uparrow},n^{\downarrow})}{\exp\left[\Delta E_{i,i\pm1}^{\sigma}(n^{\uparrow},n^{\downarrow})/k_BT\right] - 1}, \tag{3}$$

where $\Delta E_{i,i\pm1}^{\sigma}(n^{\uparrow},n^{\downarrow})$ is the change in free energy for the tunneling process consisting of a spin-independent electrostatic part, $\Delta E_{i,i\pm1}^{c}(n)$, and the spin-dependent difference in chemical potentials,

$$\Delta E_{i,i\pm1}^{\sigma}(n^{\uparrow},n^{\downarrow}) = \Delta E_{i,i\pm1}^{c}(n) + \Delta\mu_{i\pm1}^{\sigma} - \Delta\mu_i^{\sigma}. \tag{4}$$

The electrostatic contribution to the free energy is is only a function of the charge configuration in the array, the capacitances, and the bias voltage, and can be expressed as[22,23]

$$\Delta E_{i,i\pm1}^{c} = \frac{e}{2}[(\varphi_{i\pm1} + \varphi'_{i\pm1}) - (\varphi_i + \varphi'_i)], \tag{5}$$

where the island electrostatic potential before (after) tunneling φ_i (φ_i') is related to the charges through the continuity equation:

$$-C\varphi_{i-1} + (2C + C_0)\varphi_i - C\varphi_{i+1} = en_i. \tag{6}$$

The spin dependence of the tunneling rates is taken into account through the spin-dependent resistance and the different chemical potential for the two spin bands. With Eqs. (2)–(6) all tunneling rates can be calculated. A Monte Carlo scheme is used to simulate the flow of charges:[22] A tunneling event is randomly chosen with probability proportional to the tunneling rate for that event. The time between tunneling events, Δt, depends on the tunneling rates through the probability (P_0) to preserve the current charge configuration: $P_0 = \exp[-\Gamma_\Sigma \Delta t]$, where Γ_Σ is the sum of all tunnel rates. The spin flip process is then accounted for by letting $n^\sigma \to [n^\sigma \exp(-\Delta t/\tau)]$, where τ is the spin flip time and $[\cdots]$ denotes rounding to the closest integer in a way that preserves $n^+ + n^-$ (i.e., the charge is preserved in the spin flip process). This approach is the standard way to treat spin relaxation on small islands and follows from the self-consistent equation for the shift in the chemical potential:

$$\frac{I_i^\sigma - I_{i+1}^\sigma}{e} = \frac{N_0 \Omega \Delta \mu_i^\sigma}{\tau}, \tag{7}$$

where I_i^σ is the current carried by up/down ($\sigma = \uparrow / \downarrow$) spins through junction i. The procedure is repeated, keeping track of all quantities, until good statistics are obtained.

III. RESULTS

In Fig. 2 the transport characteristics of six arrays with different numbers of junctions is shown. The current-voltage characteristics in Fig. 2(a) display the typical Coulomb blockade with exponentially suppressed current up to the threshold voltage (V_t). The threshold voltage is independent of the magnetic state, which is consistent with spin-dependent transport relying on spin accumulation on the nonmagnetic islands; no current = no spin accumulation.[24] In Fig. 2(b) the tunneling magnetoresistance, TMR $= (R_{ap} - R_p)/R_p = (I_p - I_{ap})/I_{ap}$, is shown. The magnetoresistance displays a complex behavior with large negative TMR developing for intermediate array lengths.

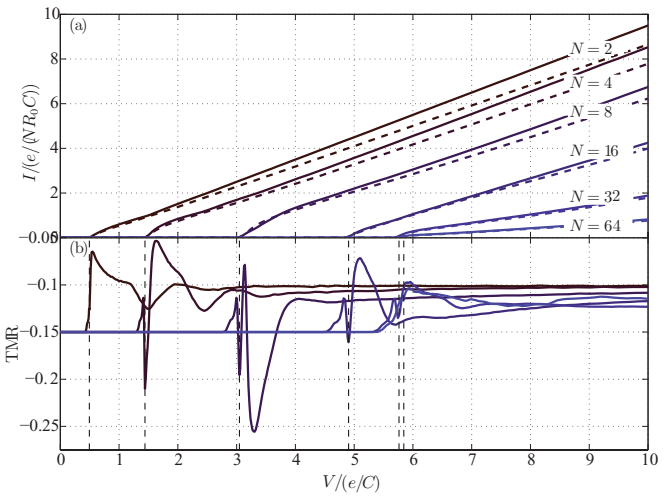

FIG. 2. (Color online) (a) The current-voltage characteristics for different array lengths. Solid (dashed) line is parallel (antiparallel) configuration of the array. Note that the current is scaled by the number of junctions. (b) The TMR for different array lengths. The horizontal dashed lines indicate the zero temperature threshold voltage. The parameters used are $P = 0.3$, $C_0/C = 2.5 \times 10^{-3}$ ($\lambda \approx 0.05$), $\tau = 9 \times 10^3\ R_0C$, $[N_0\Omega]^{-1} = 10^{-3}\ e^2/C$, and $k_BT = 2 \times 10^{-3}\ e^2/C$.

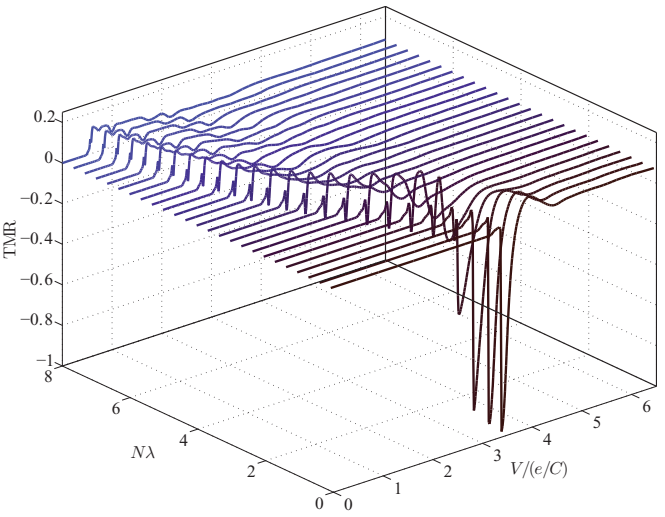

FIG. 3. (Color online) The TMR of an eight-junction array as a function of voltage and the product $N\lambda$. The parameters used are $P = 0.3$, $\tau = 9 \times 10^3\ R_0C$, $[N_0\Omega]^{-1} = 10^{-3}\ e^2/C$, and $k_BT = 2 \times 10^{-3}\ e^2/C$.

We can gain some understanding of this behavior by examining the effect of the array parameters in a few limits. We first turn to the effect of the soliton length and array length. Figure 3 shows a plot of the TMR versus the ratio of array length to soliton length. $N\lambda$ is a measure of what fraction of the array that gets electrically polarized by an electron residing in the array, or alternatively, how many solitons one can have simultaneously in the array. For short solitons compared to the array length, $N\lambda > 1$, there is a week-long-range interaction in the array (most of the voltage drop is over the ground capacitance) and the array behaves more like noninteracting islands connected in a series. In the opposite limit, $N\lambda \ll 1$, any charge in the array will influence the potential on all islands and the transport properties will generally become more complicated due to the long-range interactions.

There is a crossover of the behavior of the magnetoresistance around $N\lambda = 1$. When $N\lambda \gtrsim 1$ the TMR is positive and the TMR oscillations are a consequence of the alignment of the split chemical potential on the islands. In this limit the spin transport reassembles that of the F/N/F SET. As $N\lambda$ decreases a dip in the magnetoresistance develops close to the threshold voltage and when $N\lambda < 1$ the TMR becomes negative for voltages just above the threshold voltage. If we examine the tunneling rates close to the threshold voltage we find that when $N\lambda \ll 1$ the current is predominantly determined by the tunneling rate through the two end junctions[25] and once an electron enters into the array there is no barrier to tunnel all the way through the array. Because of this the transport is

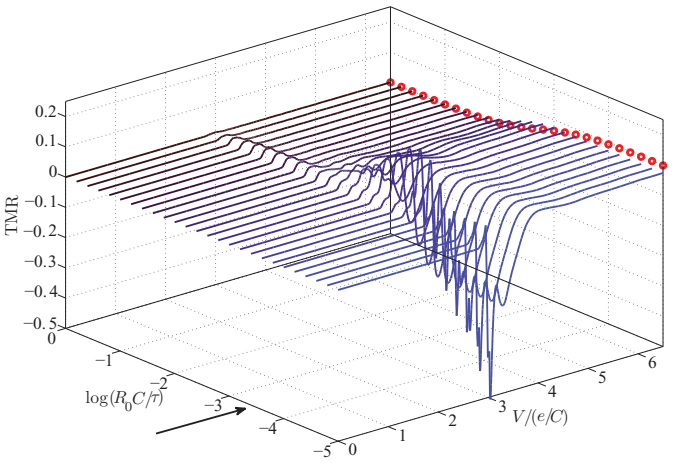

FIG. 4. (Color online) The TMR of an eight-junction array as a function of voltage and spin-relaxation time. The arrow indicates the time between tunneling events close to the threshold voltage (e/I @ $1.05 \times V_t$). The circular symbols shows the large voltage TMR from Eq. (8). The parameters used are $P = 0.3$, $C_0/C = 2.5 \times 10^{-3}$ ($\lambda \approx 0.05$), $[N_0\Omega]^{-1} = 10^{-3}\ e^2/C$, and $k_BT = 2 \times 10^{-3}\ e^2/C$.

very sensitive to small shifts in the chemical potential of the outermost islands and the splitting of the chemical potential due to spin accumulation will strongly affect these tunneling rates giving a larger current in the antiparallel state. In the opposite limit, $N\lambda \gg 1$, the tunneling rate through a junction is primarily dependent on the charges and potentials on the two islands involved in the tunneling. Therefore, a change in chemical potential only affects the local tunneling rates and one has the standard situation with the smaller current in the antiparallel state (i.e., positive TMR). This behavior of increased sensitivity to small changes of the island potential is related to the gate voltage dependence in arrays.[26] When $N\lambda \ll 1$ one needs to induce a very small charge via the gate to almost completely suppress the Coulomb blockade, but in the opposite limit one needs to induce a charge close to $e/2$ to achieve a large change in current. The $N\lambda \gtrsim 1$ case is similar to the single island (SET) case where an induced charge of half an electron is needed to suppress the Coulomb blockade.

Next we look at the dependence on the spin-relaxation time; in Fig. 4 the TMR is plotted versus spin-relaxation time and voltage. As expected for a structure containing normal-metal islands, the TMR vanishes when the spin-relaxation time approaches zero. We need $\tau \gg e/I$ to have an appreciable spin accumulation, where e/I gives the average time between successive tunneling events. In Fig. 4 the time between successive tunneling events close to the threshold voltage (e/I @ $1.05 \times V_t$) is indicated with an arrow, and it can be seen that it is in this region the negative TMR starts to develop. For long arrays the current is small due to the large series resistance of the array and spin accumulation and TMR is suppressed in the vicinity of the threshold voltage (see the $N = 32$ and $N = 64$ curve in Fig. 2). Changing the polarization P leads to similar effects as changing the spin-relaxation time with decreasing TMR with decreasing P. We also note that the parity of N only has a small effect on the TMR. For example, if we increase the number of junctions by one but keep $N\lambda$ constant we find only a slight reduction of the TMR that can be traced to the reduction of the current, and thereby the spin accumulation, due to the increase in the total resistance by one R_0.

At voltages much larger than the threshold voltage the TMR is only dependent on the spin-relaxation time and polarization. An analytic expression for the TMR can be derived in the limit of large bias voltages and long solitons compared to the array length ($N\lambda \lesssim 1$):

$$\mathrm{TMR} = \frac{\alpha P^2}{\alpha(1 - P^2) - 1/2}, \tag{8}$$

where $\alpha = \tau/(2e^2 N_0 \Omega R_0)$ is the dimensionless spin-relaxation time introduced in Ref. 8. Interestingly, the large voltage TMR is independent of the length of the array and equivalent to the large voltage TMR found for the F/N/F SET.[27] The large voltage TMR is plotted with symbols in Fig. 4.

To estimate the feasibility of an experimental realization we first note that the standard shadow evaporation technique to fabricate ultrasmall tunnel junction arrays[28,29] is well suited for fabricating arrays with alternating materials since neighboring islands will be deposited in different steps. To estimate the parameters we assume cobalt ($P \sim 30\%$–35%) as the ferromagnet and copper as the normal metal and an island volume $\Omega = 500$ nm $\times$ 60 nm $\times$ 20 nm, junction resistance $R_0 = 10^5\ \Omega$, and $N_0 = 9 \times 10^{27}$ [eV]$^{-1}$m^{-1}. With these parameters we arrive at a system close to what was used in the simulations presented here: $C \approx 0.1$ fF (60 nm $\times$ 40 nm junction with a standard 40 fF/μm^2 specific capacitance for tunnel junctions), charging energy $E_c = e^2/C/k_B \approx 18$ K, and characteristic time scale $R_0C \approx 10$ ps. With these parameters the spin-relaxation time in Fig. 4 goes from 10 ps to 1 μs which is still much faster than the recently measured spin-relaxation time of 10 μs in MnAs nanoparticles.[30] One issue that must be addressed is random background charges which are known to render tunnel junction arrays more or less useless as working devices.[26,31] Here we find that random background charges reduce the magnetoresistance around the threshold voltage and completely destroy the large negative TMR (data not shown). As expected, the large voltage TMR is not affected by the background charges. However, for small arrays ($N \sim 10$) it is possible to tune the background charges to zero[32] and it should be possible to recover the behavior shown in Fig. 2.

IV. CONCLUSION

We considered an array of small tunnel junctions consisting of alternating ferromagnetic and normal-metal islands. A Monte Carlo method is used to calculate the transport properties and the tunneling magnetoresistance in the sequential tunneling regime. The spin-dependent tunneling resistance leads to a nonequilibrium spin accumulation on the normal islands. The amount of spin accumulation is determined by balancing the spin injection from the ferromagnetic islands with spin relaxation. We investigated the case with nonzero spin-relaxation time on the normal-metal islands and zero spin-relaxation time on the ferromagnetic islands. However, it is straightforward to extend the model to include finite

spin accumulation on the ferromagnetic islands as well, but taking into account that spin relaxation is much stronger in ferromagnets than in normal metals (typically by orders of magnitude) the shift in chemical potential will be reduced by this factor according to Eq. (7) and the transport properties will be dominated by spin accumulation in the normal islands as long as the normal and the ferromagnetic islands have similar density of states and volume.

The resulting TMR displays oscillations with bias voltage which is typical for devices exploiting both spin-dependent tunneling and Coulomb blockade. We show that when the soliton length is comparable to or longer than the array the TMR is negative close to the threshold voltage provided that the spin-relaxation time is long enough. Not only the sign of the TMR changes but the magnitude of the TMR can be substantially increase in this regime. The negative TMR is an effect of the long-range interaction in the array and in this limit the transport is qualitatively different than the TMR for a single island F/N/F device.

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