Magnetic Field Modulated Resonant Tunneling in Ferromagnetic-Insulator-Nonmagnetic junctions

Yang Song\textsuperscript{1} and Hanan Dery\textsuperscript{1,2}

\textsuperscript{1}Department of Electrical and Computer Engineering, University of Rochester, Rochester, New York, 14627
\textsuperscript{2}Department of Physics and Astronomy, University of Rochester, Rochester, New York, 14627

We present a theory for resonance-tunneling magnetoresistance in Ferromagnetic-Insulator-Nonmagnetic junctions. The theory sheds light on many of the recent electrical spin injection experiments suggesting that this magnetoresistance effect rather than spin accumulation in the nonmagnetic channel corresponds to the electrically detected spin signal. We quantify the dependence of the tunnel current on the magnetic field by quantum rate equations derived from the Anderson impurity model. Considering the on-site Coulomb correlation, the magnetoresistance effect is caused by competition between the field, impurity spin interactions and coupling to the magnetic lead.

PACS numbers:

Demonstrations of electrically-injected spin accumulation in nonmagnetic materials are considered reliable when measured in a non-local geometry [1, 2]. In this setup, shown in Fig. 1(a), one ferromagnetic electrode injects or extracts spin-polarized electrons and a second one detects the spin accumulation of electrons ($V_{NL}$) that diffuse outside the path of a constant charge current ($I_{T}$). Because the spin diffusion length of many nonmagnetic materials is in the $\lesssim 1$ $\mu$m range, the separation between the injector and detector electrodes is defined by a submicron fabrication process [3, 4]. To mitigate this requirement, many researchers have recently resorted to a local measurement wherein a single ferromagnetic electrode is used for both injection and detection of the spin signal [5–21]. A distinct difference between these geometries is that the detector junction remains unbiased only for the nonlocal setup, and as a result the local detection is highly prone to impurity-assisted tunnel current. Figure 1(b) shows the typically observed change in the detected resistance when applying an external magnetic field. Similar to the Hanle-type experiment of optical spin injection [22–25], the width and amplitude of the Lorentzian-shaped signal ($\delta B$ and $\Delta R$) are frequently used to extract the spin lifetime and accumulation density. One evident result, however, is that $\delta B$ and $\Delta R$ in many of the local experiments noticeably deviate from expected values. This behavior questions many of the recent claims for spin injection.

In this Letter, we present a theory for resonance-tunneling magnetoresistance (MR) in Ferromagnetic-Insulator-Nonmagnetic (F-I-N) junctions. The theory solves the puzzling observations in local spin injection/detection setups, showing that the width and amplitude ($\delta B$ and $\Delta R$) are independent of spin relaxation and accumulation in N. They are set by spin-dependent interplay between the Zeeman energy, on-site Coulomb and spin interactions, and the impurity coupling to F. The MR dependence on orientation of the field agrees with measurements in the local setup.

To quantify the MR, we tailor the Anderson impurity model to our tunneling problem [26–27]. The system Hamiltonian for $s=\frac{1}{2}$ impurity reads

\[
H = \sum_{\ell,k,\sigma} \varepsilon_{\ell k \sigma} a_{\ell k \sigma}^{\dagger} a_{\ell k \sigma} + \left( T_{G \sigma} a_{G k \sigma} d_{\sigma} + \text{h.c.} \right) + U n_{\uparrow} n_{\downarrow} + \varepsilon_1(\theta) n_{\uparrow} + \varepsilon_1(\theta) n_{\downarrow} + \varepsilon_B \sin \theta (d_{\uparrow}^{\dagger} d_{\downarrow} + d_{\downarrow}^{\dagger} d_{\uparrow}),
\]

where $\varepsilon_{\ell k \sigma}$ is the electron energy with wavevector $\mathbf{k}$ and spin $\sigma$ in the $\ell$-th lead (F or N), $a_{\ell k \sigma}^{\dagger}$ ($a_{\ell k \sigma}$) and $d_{\sigma}^{\dagger}$ ($d_{\sigma}$) are the creation (annihilation) Fermi operators in the lead and impurity, respectively. The interaction between the lead and impurity is denoted by $T_{G \sigma}$, assumed here to be $\mathbf{k}$ independent for simplicity. The on-site Coulomb interaction between electrons of opposite spins is denoted by $U$. 

![FIG. 1: (Color online) (a) Nonlocal and local electrical setups for detecting spin accumulation. (b) The measured signal is a change in junction resistance when applying in-plane or out-of-plane magnetic fields. The Lorentzian due to in-plane field is typically observed only in the local setup. (c)/(d) Resonant tunneling via type A/B impurities for electrons flow from F to N.](image-url)
and \( n_\sigma = d_\sigma^\dagger d_\sigma \) is the impurity occupation operator. The \( \sigma = \uparrow (\downarrow) \) component is parallel to the majority (minority) spin population of F. The second line in [1] denotes the impurity Zeeman terms where \( \theta \) is the angle between B and the spin quantization direction. The energy terms are \( \varepsilon_\uparrow (\theta) = \varepsilon_0 + \varepsilon_B \cos \theta \) and \( \varepsilon_\downarrow (\theta) = \varepsilon_0 - \varepsilon_B \cos \theta \) where \( \varepsilon_0 \) is the resonance energy of the singly occupied state and \( \varepsilon_B = g_\mu_B B/2 \). The off-diagonal operators, \( d_\sigma^\dagger d_\sigma' \), appear when \( \sin \theta \neq 0 \).

We briefly describe the procedure for deriving the resonance current. Using the Heisenberg picture, the equation of motion for density-matrix operators is

\[
-i\hbar \frac{d}{dt} \rho_{\sigma\sigma'} = \sum_{\ell,k} T_{\ell\sigma\sigma'}\rho_{k\ell} - T_{k\ell\sigma}\rho_{\ell\sigma'} + \varepsilon_B \sin \theta (d_\sigma^\dagger d_\sigma' - d_\sigma' d_\sigma) \pm 2\varepsilon_B \cos \theta d_\sigma^\dagger d_\sigma \delta_{\sigma\sigma'}.
\]

Henceforth, the +/- sign refers to the case that \( \sigma = \uparrow / \downarrow \). To form a closed set of equations, we use the Langreth theorem and recast the averages of the sum terms into lesser and retarded Green functions on the impurity [28–30],

\[
\sum_{\ell,k} T_{\ell\sigma\sigma'} a_{\ell k\sigma}^\dagger a_{\ell k\sigma'} = \sum_{\ell,\kappa} \int \frac{d\varepsilon}{2\pi} \Gamma_{\ell\kappa} \left( G_{R,\kappa\sigma}^\ell f_{\ell\sigma} + \frac{1}{2} G_{L,\kappa\sigma}^\ell \right),
\]

\( f_{\ell\sigma} (\varepsilon) \) is the Fermi-Dirac electron distribution in the \( \ell \)th lead and \( \Gamma_{\ell\kappa} \) is the Fermi-Dirac electron distribution in the \( \ell \)th lead and \( \Gamma_{\ell\kappa} = 2\pi \sum_k |T_{\ell\kappa}|^2 \delta (\varepsilon - \varepsilon_{k\ell \kappa}) \) is its coupling to the impurity. The analysis is greatly simplified outside the Kondo regime and by assuming weak coupling (\( \Gamma \ll \{k_B T, eV\} \)). We focus on two impurity types wherein the population of the resonance state fluctuates between zero and one electron [type A; see Fig. 1(c)] or between one and two electrons [type B; see Fig. 1(d)]. This classification is motivated by the dependence of the Green functions on the impurity population. It is justified when considering together the broad energy distribution of mid-gap impurity defects at oxide tunnel barriers [34–38], the large on-site Coulomb repulsion (\( U \gg k_B T \)), and the common conditions of local-setup experiments where \( eV \gg k_B T \gg \varepsilon_B \). With these considerations, we can replace \( f_{\ell\sigma} (\varepsilon) \) by 1 (0) for the injector (extractor) lead and the analysis becomes independent of spin accumulation in the leads. Furthermore, the Green functions assume simple form under these considerations, thereby enabling simple integration in Eq. [3] where \( \Gamma (\varepsilon) \) varies slowly on the scale of \( \varepsilon_B \) (e.g. \( G_{R,\kappa\sigma}^\ell = 2i\pi (n_\uparrow + n_\downarrow - 1) \delta_{\kappa\sigma} \) for type B; see [30] for details). Putting these pieces together, we reach a concise equation set [30, 32]. For spin extraction (electrons flow from N to F) via type A impurities we get

\[
\dot{n}_\sigma = \Gamma_N P_0 - (1 \pm p) \frac{\Gamma_F n_{\sigma\sigma}}{2 - \varepsilon_B \sin \theta \Im (n_{\sigma\sigma})},
\]

\[
\dot{n}_{\sigma'} = \varepsilon_B \sin \theta \Im (n_{\sigma\sigma'}) - \Gamma_F n_{\sigma\sigma'},
\]

where \( n_{\sigma\sigma'} = \langle d_\sigma^\dagger d_\sigma \rangle \). \( P_0 = 1 - n_{\uparrow\uparrow} - n_{\downarrow\downarrow} \) is the probability for zero occupation and the coupling parameters \( \Gamma (\varepsilon) = (\Gamma_{\ell\uparrow} + \Gamma_{\ell\downarrow})/2 \) are evaluated around the impurity’s energy level \( \varepsilon = \varepsilon_0 \). The junction polarization is given by \( p = (\Gamma_{F\uparrow} - \Gamma_{F\downarrow})/\Gamma_F \). The master equations for injection conditions (electrons flow from F to N) are obtained by exchanging \( \Gamma_{N,\sigma} \equiv \Gamma_N \) with \( \Gamma_{F,\sigma} \). Similarly, the equations for type B impurities are obtained by evaluating \( \Gamma_F \) and \( \Gamma_N \) around \( \varepsilon = \varepsilon_0 + U \), by considering double rather than zero occupancy (\( P_2 = n_{\uparrow\uparrow} + n_{\downarrow\downarrow} \)), and by noting that type A and B impurities flip roles in extraction and injection conditions [30]. This feature reflects their symmetry and can be understood when regarded from the viewpoint of electron tunneling in type A and hole tunneling in type B [39].

The resonance currents are found from the steady state solution of the master equations using \( i_A = 2e\Gamma_N P_0/h \) and \( i_B = 2e\Gamma_N (1 - P_2)/h \) for extraction, or \( i_A = -2e\Gamma_N (1 - P_0)/h \) and \( i_B = -2e\Gamma_N P_2/h \) for injection. We get

\[
i_{N\rightarrow F}^A = -i_{F\rightarrow N}^A = \frac{2e}{h} \frac{\Gamma_F \Gamma_N}{2\Gamma_F^2 + \Gamma_N^2} \frac{1 - p^2 \chi (B)}{1 - p^2 \chi (B)},
\]

\[
i_{N\rightarrow F}^B = -i_{F\rightarrow N}^B = \frac{2e}{h} \frac{\Gamma_F \Gamma_N}{2\Gamma_F^2 + \Gamma_N^2},
\]

where

\[
\chi (B) = \frac{B_F^2 + B_{\cos}^2 \theta}{2}, \quad \alpha = \frac{\Gamma_F}{2\Gamma_F + \Gamma_N}, \quad B_F = \frac{\Gamma_F}{g_\mu_B}.
\]

Most relevant to our analysis, the resonance current across type A/B impurities depends on the magnetic field in extraction/injection conditions (via \( \chi (B) \)). This dependence is best perceived when considering half-metallic F and out-of-plane magnetic field (\( p = 1 \) and \( \theta = \pi/2 \)). Without magnetic field, extraction via type A or injection via type B are completely blocked, \( i_{N\rightarrow F}^A = i_{F\rightarrow N}^B = 0 \). In extraction via type A, electrons tunnel from N into the impurity and have equal probability to be parallel or antiparallel to the spin orientation in the half-metal. The tunnel conductance is blocked once an antiparallel spin settles on the impurity since it cannot be extracted by the half-metal. For injection via type B we get that once the lower impurity level is filled with an electron from the half-metal, the upper resonant level can only accept the electron of opposite spin which the half metal cannot provide. In a large out-of-plane field, the blockade is completely lifted in both cases due to depolarization of the impurity spin (Larmor precession). Finally, from [5] we get that \( i_A + i_B \) merely flips sign when reversing the bias direction. Therefore, the MR effect in injection (\( F \rightarrow N \)) and extraction (\( N \rightarrow F \)) is similar if the densities of type A and B impurities are similar.

To compare the analysis with experimental findings we study the magnetic field components. When comprised of the external field alone, \( \mathbf{B} = \mathbf{B}_e \), the modulation amplitude \( \Delta i (\theta_e) \) is

\[
\Delta i (\theta_e) = \frac{\sin^2 \theta_e}{1 - \alpha p^2 \cos^2 \theta_e} \frac{(1 - \alpha p^2)}{1 - \alpha p^2},
\]

where \( \Delta i (\theta_e) \) is the resonance current. Using the Heisenberg picture, the equation of motion for density-matrix operators is

\[
-i\hbar \frac{d}{dt} \rho_{\sigma\sigma'} = \sum_{\ell,k} T_{\ell\sigma\sigma'}\rho_{k\ell} - T_{k\ell\sigma}\rho_{\ell\sigma'} + \varepsilon_B \sin \theta \delta (\varepsilon - \varepsilon_{k\ell \kappa}) \pm 2\varepsilon_B \cos \theta d_\sigma^\dagger d_\sigma \delta_{\sigma\sigma'}.
\]
where \( i_0 = 2\alpha e \Gamma_N / h \). Throughout this work, \( B_i \) is assumed smaller than the large out-of-plane coercive field of F. We see that the MR effect vanishes for in-plane external field \( (\phi = 0) \) in contrast to most measurements where the in-plane field modulation is larger than that of the out-of-plane. Furthermore, for \( B = B_i \), the signal width stems from the coupling to F \( (\delta B \sim B_F) \), thereby decreasing exponentially with increasing oxide thickness. In virtually all local-setu measurements of F-I-N structures, on the other hand, \( \delta B \sim 0.1 - 1 \) kG regardless of the oxide details [6,19]. To explain these aforementioned observations, we examine the ubiquitous spin interactions which tend to randomize the spin orientation at the impurities. They include, for example, hyperfine fields due to interaction with the nuclear spins and exchange interactions between nearby impurities. Invoking mean-field approximation, an effective internal magnetic field can be written by \( B_i = B_{hf} + B_{ex} = (\langle A(1) + (J_{nn}(S)) \rangle / 4\mu_B \) where \( A \) is the hyperfine coupling constant with nuclear spin I and \( J_{nn} \) is the exchange coupling with an electron of spin \( S \) on the nearest neighbor impurity. Considering ferromagnet-oxide-silicon as a case study, unpaired electrons on \( ^{29}\text{Si} \) dangling bonds would experience hyperfine fields of a few hundred Gauss [36,37,40–42]. Similar defects can exist in \( \text{Al}_2\text{O}_3 \) barriers [38,43,45] or perovskite interfaces [46]. The defect densities can be controlled by oxide preparation techniques [19]. The sources for \( B_i \) also include stray fields whose amplitude and direction depend on the interface roughness [11].

With \( B = B_i + B_F \), we can complete the analysis using [4] and write the extraction/injection tunnel current via a type A/B impurity, \( i = i_A = -i_B \),

\[
\frac{i}{i_0} = 1 - (1 - \alpha) p^2 \int B_i^2 \int d \phi \int \cos \theta_i \frac{B^2_F(B_i, \theta_i, \phi_i)}{B^2_F + (1 - \alpha p^2)B^2_i} \tag{7}
\]

\( i \) is averaged over \( F(B_i, \phi, \phi) \), the probability density function for the internal field. The components of the effective field along and normal to F are \( B^2_i = B^2_{fi} + (B_{fi} + B_{ei})^2 \) and \( B^2_F = B^2_{fi} + B^2_{ei} + 2B_{fi}B_{ei} \cos(\phi_i - \phi_e) \), respectively. Figures 2(a) and (b) show the solution of \( \phi_e = 0 \) with hyperfine fields of common defect centers in Si/Oxide interfaces. The tunneling involves unpaired electrons on \( ^{29}\text{Si} \) dangling bonds next to oxygen vacancy \( V_0 \) in the barrier \( (E' \text{ center}) \) or in \( \text{Si}_3 \) configuration on the atomic interface \( (P_d \text{ center}) \). The hyperfine field of \( E' \) is assumed isotropic with amplitude of 420 G [40], and that of \( P_d \) has axial symmetry with an out-of-plane (in-plane) amplitude of 160 G (90 G) [12]. Figure 2(c) shows the solution for internal fields due to exchange between nearest-neighbor impurities. The localization length and impurity density in the tunnel barrier are chosen \( \ell_i = 4.4 \text{ Å} \) and \( n_i = 8 \times 10^{18} \text{ cm}^{-3} \), respectively. Further details are provided in the supplemental material [20]. In all three cases we assume \( (B_i) > B_F \) so that the width of the signal is set by internal fields rather than by coupling with F (i.e., \( \delta B \) is essentially independent of barrier thickness). The modulation amplitude in the regime that \( B_i \sim B_F \) is realized from \( \Delta i(\theta_e) = (B_i \sim B_F) - i(B_i \sim B_F) \). For isotropic internal field distribution \[ F(B_i, \theta_i, \phi_i) = F(B_i)/4\pi \], we get

\[
\Delta i(\theta_e) = \frac{1}{\alpha} \left[ \frac{\arctanh(\sqrt{\alpha p})}{\sqrt{\alpha p}} - \frac{1}{1 - \alpha p^2 \cos^2 \theta_e} \right] i_0. \tag{8}
\]

The in-plane field modulation \( (\theta_e = 0) \) can exceed that of the out-of-plane \( (\theta_e = \pi/2) \), and their ratio as \( |\Delta i(0)/\Delta i(\pi/2)| \) is

\[
r = \frac{2 + 2q(\alpha p^2)}{1 - 2q(\alpha p^2)}, \quad q(z) = \sum_{m=1}^{\infty} \frac{z^m}{2m + 3}. \tag{9}
\]

\( r \geq 2 \) and the lower bound increases for internal fields that point mostly in the out-of-plane direction. For example, \( r \geq n + 2 \) when \( F(B_i, \theta_i, \phi_i) \propto \sin^n \theta_i F(B_i) \).

We can now quantify the total voltage change that one measures in the local geometry \( |\Delta R| \) in Fig. 1(b)]. Denoting the total tunneling current used in experiments [Fig. 1(a)] as \( I_T \), for small MR effect, we simply have

\[
\frac{\Delta R}{R} = \sum_n \frac{\Delta i_n(0) - \Delta i_n(\pi/2)}{I_T} = \frac{1}{I_T} \sum_n \frac{(1 - \alpha_n p^2) i_0}{1 - \alpha_n p^2}, \tag{10}
\]

where \( n \) runs over type A (B) impurities in the tunnel barrier for spin extraction (injection). The much larger total current is by tunneling via larger impurity clusters of which \( U \lesssim eV \) and a background direct tunneling. The MR effect is enabled by the nonzero polarization of a F-I-N junction \( (p \neq 0) \), rendering it distinct from resonant tunneling MR in N-I-N junctions where \( p \mu_B B > (k_B T, eV) \) [14,55]. The quadratic-in-\( p \) dependence signifies the fact that the modulation is achieved when both F and the impurity are polarized. Since \( \Delta R \) measures the effect in the regime that \( B_F \gg B_i, B_F \), its amplitude is independent of the details of the internal field distribution. Accordingly, one can use either [4] or [3] to get [14]. The amplitude of \( \Delta R \) depends on the junction’s polarization, the impurities density and their coupling to the leads (via \( \alpha \) and \( i_0 \)).
Conclusion and discussion. The magnetoresistance effect in F-I-N junctions comes from spin precession of the electron in impurities whose population fluctuates between zero and one (type A) in spin extraction (electrons flow into F), or between one and two (type B) in spin injection (electrons flow from F). The effect is stronger for impurities that are located closer to N than to F. This feature is manifested by the increase of the effect when \( \alpha \rightarrow 0 \) (i.e., \( \Gamma_N \gg \Gamma_F \)). We can understand this physics by recalling that if a type A/B impurity is closer to F in extraction/injection then it will be mostly empty/doubly occupied. Therefore, spin precession becomes meaningless and the modulation cannot be observed. An additional feature of the MR effect is a more resistive junction when applying in-plane magnetic field for both injection and extraction. The physics is explained by the blockade of the current in this magnetic field configuration. Finally, we have made connection between the measured voltage change in local geometry experiments and the MR effect via \( |\Delta R|/R \). From this expression, we see that temperature-dependent changes in \( \Delta R/R \) can reflect those of the junction resistance (\( R \)) rather than of the MR effect itself (\( \Delta R \)). Specifically, since the resonance tunneling was studied for \( eV \gg k_B T \gg g\mu_B B \), the population of the impurity is independent of temperature. One important consequence is that when direct or resonant tunneling is the dominant transport mechanism across a F-I-N junction, one should expect a small \( \Delta R \) component that does not change appreciably with temperature.

We now emphasize several important aspects regarding the interpretation of many recent experiments that aimed at demonstrating spin injection to semiconductors by using local geometry measurements (the so-called three-terminal Hanle setup). The finite bias on the detecting electrode in these experiments enables magnetic field modulation of the impurity-assisted resonance tunnel current. The so-far overlooked MR effect that we studied in this work can explain the experimental results without invoking spin accumulation or spin relaxation in the non-magnetic side of the junction. In this context, we mention the previous phenomenological rate-equation model of Tran et al. who conjecture that the competition between hopping time and impurity sub-ns spin relaxation may be the origin for the observed signal \( |\Delta R|/R \). We find this speculation to conflict with the commonly found case of ultra-long spin relaxation of localized electrons in impurities at low temperatures. Furthermore, unless the spin-orbit coupling of the impurity is strong, there is no apparent reason for the spin relaxation to be in the sub-ns scale even at elevated temperature \( T \).

An additional important aspect is that the studied MR effect is independent of doping considerations in the semiconductor, and therefore it can equally explain the experimental results for \( n \)-type and \( p \)-type doped F-I-N junctions. So far, however, the detected spin signal in junctions with \( p \)-type semiconductors were attributed to spin accumulation and spin relaxation of holes \([6, 11, 13]\). These claims contradict known physics in typical bulk semiconductors that are not subjected to large strain. Specifically, the spin relaxation rate of holes is governed by scattering between heavy and light holes. Due to the band degeneracy in the top of the valence band, the strong spin mixing of light-hole states renders this scattering ultrafast irrespective of the amplitude of the spin-orbit coupling. The result is that spin and momentum relaxation rates of holes are in the same ballpark (typically sub one-picosecond regime). That the observed signals in F-I-N structures with \( n \)-type and \( p \)-type semiconductors have comparable amplitudes and widths \( (\hbar/g\mu_B B \sim 0.1 \text{ ns}) \) strengthens the argument that the proposed MR effect corresponds to the measured signal. Another noteworthy aspect is that in F-I-N junctions where I is an oxide and N is a semiconductor, the majority of defects are typically formed on the atomic interface between the oxide and the semiconductor. Accordingly, \( \Gamma_F \) and \( \Gamma_N \) scale with the conductance of the oxide and Schottky barriers, respectively. As long as the oxide is more resistive, the impurity-assisted resonant current and direct tunneling show similar exponential decay with increasing the oxide thickness in accord with experimental findings \( [15] \).

Finally, further experiments are needed to fully characterize the MR effect in F-I-N junctions. Scanning tunneling spectroscopy, for example, allows one to resolve the contributions from a single type A (B) impurity in extraction (injection) bias conditions. Combination of this technique and electron paramagnetic resonance has already been used to detect spin precession of single electron defects \( [48] \). Further experiments are also needed to unravel the internal-field sources and their dependence on properties of the tunnel barrier. Lastly, we note that direct contacts between magnetic metals and \( n \)-type semiconductors (i.e., Schottky barriers) can eliminate the MR effect. These contacts are not prone to the multitude of defects in typical oxides and subjected mainly to mid-gap defect states at the interface with F where the MR effect is disabled \( (\Gamma_F \gg \Gamma_N; \alpha \rightarrow 1) \). Furthermore, direct contacts are less resistive and thereby enable better operation of F-I-N based spintronic devices \( [49, 50] \), as well as clearer detection of spin accumulation in \( n \)-type semiconductors in local-setup geometries \( [20, 25, 51] \).

We are indebted to Felix Casanova, Oihana Txoperena, Kohei Hamaya, and Ian Appelbaum for insightful discussions and for sharing invaluable data prior to their publication. This work is supported by NRI-NSF, NSF, and DTRA Contract numbers DMR-1124601, ECCS-1231570, and HDTRA1-13-1-0013, respectively.
