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Theory of oblique-field magnetoresistance from spin centers in three-terminal spintronic devices

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We present a general stochastic Liouville theory of electrical transport across a barrier between two conductors that occurs via sequential hopping through a single defect’s spin-0 to spin-1/2 transition. We find magnetococonductances similar to Hanle features (pseudo Hanle features) that originate from Pauli blocking without spin accumulation, and also predict that evolution of the defect’s spin modifies the conventional Hanle response, producing an inverted Hanle signal from spin center evolution. We propose studies in oblique magnetic fields that would unambiguously determine if a magnetoconductance results from spin-center assisted transport.

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I. INTRODUCTION

A current flowing through a magnetic conductor, with carriers flowing into (spin injection) or out of (spin extraction) a nonmagnetic conductor, can produce a nonequilibrium spin population in the nonmagnetic material [1] that can be sensed through a spin-sensitive voltage. An applied magnetic field, intended to precess these nonequilibrium spins and thereby reduce the voltage (the Hanle effect or HE [2]), often produces surprising results challenging to interpret using reasonable spin coherence times [3–7]. Anomalous Hanle features (the inverted Hanle effect or IHE) were found with parallel applied field and magnetization; conventional interpretations attribute IHE to magnetic fringe fields from a nonuniform interface, however detailed structural measurements of the interface failed to correlate these features with measured nonuniformity. For currents and voltages measured using the same contact (three-terminal, or 3T measurements), features mimicking the HE and IHE were found [6,8], originating from magnetic-field-dependent transport through spin centers. However, agreement between some 3T and four-terminal (4T) nonlocal experiments indicates that the impurity effect does not always dominate over direct tunneling [9–11].

Here we analyze sequential spin-center-mediated tunneling between different magnetic or nonmagnetic leads in both the spin injection and spin extraction regimes using the stochastic Liouville equation (SLE) formalism. The SLE framework is highly adaptable to many physical systems, including spin-oriented tunneling through spin centers [12]. We apply the formalism to sequential tunneling involving a spin center that alternates between spin zero and spin one-half as its charge state changes. In the spin extraction regime, our formalism confirms previous studies that demonstrated supposed HE and IHE to be the result of a Pauli blockade [6,13], which we now refer to as “pseudo HE” and “pseudo IHE” phenomena.

To indicate the experimental geometry without assigning a mechanism we refer to “Hanle measurements” or “inverted Hanle measurements.” The combined effects of spin accumulation at the spin center and the coherent evolution of that spin accumulation lead to an altered spin injection process; we predict a previously undescribed, broad IHE (with same physical origin as the conventional HE) accompanying the known conventional HE (that can be broad or narrow). We will refer to these as “conventional HE” and “conventional IHE,” distinct from the pseudo HE and pseudo IHE. We analyze the oblique field data of Ref. [14] to show that a ratio of the angular dependent responses could, in principle, provide incontrovertible evidence of the impurity model by definitively ruling out the fringe-field mechanism for the inverted signal. Our calculations predict distinct magnetic field widths in the magnetoresponse that motivate further measurements to resolve the origin of the observed features. Lastly we show that the effects described herein require charge current and are thus not present for nonlocal (4T) measurements (or spin pumping).

II. INTERFACIAL SPIN CENTERS

This work treats spin injection and spin extraction between a nonmagnetic material (NM) and a ferromagnet (FM); the charge and/or spin current passes through an interfacial trap state through sequential hopping from one lead onto the trap, and then from the trap to the other lead. Figure 1(a) shows spin injection under a forward bias. The transition level of the spin center within the insulating barrier is labeled as (0 1/2 0) to denote the two possible spin states: 0 here refers to the spin when no charge has hopped onto the level whereas 1/2 is the spin when a charged has hopped onto the level. Figure 1(b) shows the same center in the spin extraction regime (reverse bias, or with the nonmagnetic and ferromagnetic contacts reversed). We differentiate between the two configurations of Fig. 1 by defining (a) a spin transport center (since spin will accumulate in the NM) and (b) a spin bottleneck center since a bottleneck forms at the right junction. For both situations,
The current operators are formulated by identifying, e.g., in Fig. 1(b), the probability for a charge to pass from the barrier trap to the FM to be \((1 + P_R \cdot \sigma(t))\). The generalization of this probability for our current operator is \(\hat{M}_R \rho(t)\) which yields, for the “right” current (trap to right lead \(R\)) [12]

\[
\hat{i}_R(t) = \frac{e}{2} \gamma_R [\hat{M}_R, \rho(t)],
\]

where curly braces denote the anticommutator. The “left” current (left lead \(L\) to trap) is derived similarly after constraining the center to at most singly occupancy:

\[
\hat{i}_L(t) = e\gamma_L [1 - \text{Tr} \rho(t)] \hat{M}_L.
\]

Charge conservation demands that the left charge current equal the right charge current.

Charge currents, spin currents, and spin accumulations are calculated from the spin center’s steady-state density matrix. The center undergoes several interwoven processes (e.g., charge hopping on and off, applied and local fields, charge and spin blocking) so unraveling the physics is not intuitive, however the SLE is well suited for this type of problem. It avoids complications from the choice of a spin quantization axis (see concluding remarks). The SLE [16–19]:

\[
\frac{\partial \rho(t)}{\partial t} = -\frac{i}{\hbar} [\mathcal{H}, \rho(t)] - \gamma_R [\hat{M}_R, \rho(t)] + 2\gamma_L [1 - \text{Tr} \rho(t)] \hat{M}_L,
\]

where \(\gamma_{RL}\) are the spin dependent hopping rates to the right (left) electrode. The spin Hamiltonian at the spin center site is \(\mathcal{H} = \frac{\hbar}{2} (b_0 + b_{hf}) \cdot \sigma = \frac{\hbar}{2} b \cdot \sigma\), where \(b_0 = g\mu_B B_0 / \hbar\), \(b_{hf} = g\mu_B B_{hf}/\hbar\), \(b = b_0 + b_{hf}\), \(B_0\) is a uniform magnetic field, and \(B_{hf}\) is the hyperfine field at the spin center. The hyperfine fields are assumed to be distributed as a Gaussian function with width \(\tilde{\gamma}_{hf}\). The first term of the SLE represents the coherent evolution of the center’s spin, the second term denotes the spin-selective nature of tunneling into the FM. The third term describes hopping onto the impurity site from the left contact. These latter two terms are responsible for the stochastic nature of the SLE; they represent random hopping (with average rates \(\gamma_R\) and \(\gamma_L\), respectively) from the center to the FM and from the NM to the center.

We assume the spin lifetimes and coherence times of the trap spin are longer than the transport processes producing the current.

### III. Theory

Operators for the static magnetizations of each electrode are \(\hat{M}_{L,R} = \frac{1}{2} (I + P_{L,R} \cdot \sigma)\). The time-evolution of the density matrix of the spin center, \(\rho(t)\), is determined by the SLE. A \(2 \times 2\) matrix for the current \(\hat{i}\) fully describes the flow of charge and coherent spin. Diagonal elements represent the movement of charge with up or down spins, and off-diagonal elements describe the flow of charge with up and down spin superpositions. Charge currents are \(i = \text{Tr} \hat{i}\), and spin currents are \(i_{L,R} = \text{Tr} [\hat{M}_{L,R} \hat{i}]\).

#### A. Spin extraction

We now apply this theory to spin extraction [15] [Fig. 1(b)]. The current,

\[
i \equiv i (P_{R,0}) = e \frac{(1 - P_R \chi(b)) \gamma_L \gamma_R}{(1 - P_R \chi(b)) \gamma_R + 2\gamma_L},
\]

with \(\chi(b) = (\gamma_R^2 + (b \cdot P_R)^2)/(\gamma_R^2 + b^2)\) agrees with Refs. [6,13], but differs from Ref. [20] (see concluding remarks).
The spin polarization of the impurities, $P_d = \text{Tr}(\sigma \rho)$, is
\[
P_d = \frac{2\gamma_L}{\gamma_R + 2\gamma_L} \left[ \frac{\gamma_R^2 P_p + \gamma_R b \cdot P_R + (b \cdot P_R)b}{\gamma_R^2 + b^2} \right].
\] (6)

Spins parallel to the FM preferentially leave the barrier trap, thus a polarization opposite to the FM develops on the impurity site. This can be seen for $b_{hf} = 0$ and $b_{hf}|P_R$, which leads to $P_d \propto -P_p$. This accumulation of defect spins, due to requiring spins leaving the defect to be parallel to $P_R$, is called spin filtering [12]. There can be no spin accumulation in the NM since the impurity is only filled from the NM when it is empty; therefore no preferred spin is taken out of the NM [15].

B. Spin injection

There is no spin filtering effect for the spin injection geometry [Fig. 1(a)], so the current, $i(P_L, P_R = 0) = e\gamma_L/(\gamma_R + 2\gamma_L)$, is independent of $b_0$. The steady state impurity spin polarization,
\[
P_d = \frac{2\gamma_L}{\gamma_R + 2\gamma_L} \left[ \frac{\gamma_R^2 P_p + \gamma_R b \cdot P_R + (b \cdot P_R)b}{\gamma_R^2 + b^2} \right].
\] (7)

The spin current, $i = e\gamma_L P_d/2$, leads to an excess (above an assumed unpolarized background) spin polarization of NM carriers per unit volume, which we call the spin accumulation density, $p_0$, at the interface with area $A$. The spin accumulation density decays further within the NM as $p = p_0 \exp(-x/\lambda_s)$, where $\lambda_s$ is the NM spin diffusion length. By setting the spin gained due to the spin current equal to the spin loss/evolution in NM [15], $i_0/e = \lambda_s (\frac{1}{2} P_0 - b_0 \times P_0)$, the spin accumulation density can be solved analytically for a general spin current:
\[
p_0 = \frac{\tau_s \gamma_R P_d + \tau_r b_0 \times P_d + \tau_s^2 (b_0 \cdot P_d)b_0}{1 + \tau_s^2 b_0^2},
\] (8)

where the field dependence is hidden within $P_d$ [see Eq. (7)] and $\tau_s$ is the spin relaxation time of carriers in the NM. This expression is identical to expressions for the oblique Hanle effect for direct tunneling, replacing the spin polarization injected into the NM with the defect polarization $P_d$ [21].

Canting the direction of the spin current away from the magnetization direction produces a previously unknown effective IHE. The origin of the canting is seen by investigating Eq. (7) for scenarios where $b_{hf}|P_L|b_0$ and $b_{hf} \perp P_L|b_0$. When $b_{hf}|P_L|b_0$, $P_d \perp P_L$ (i.e., no canting) and upon substituting Eq. (7) into Eq. (8) we find no field dependence. For $b_{hf} \perp P_L|b_0$, $P_d$ develops components transverse to $P_L$ which indicate canting and the component of $p||P_L$ will be smaller than for a parallel hyperfine field. As the applied field is increased the transverse hyperfine component’s effect is minimized and the maximum $p_s$ is restored. This is an IHE but in reality its origin is the same as the conventional HE except that a “hidden” transverse field (a component of the hyperfine field $\perp P_L|b_0$) exists at the trap site. This transverse hyperfine component rotates the spin that is injected into the NM which results in a smaller accumulation of $p_s$. Prior theories of inverted Hanle measurements have either been (1) solely attributed to the spatially inhomogeneous fringe fields of the ferromagnetic contact [22], or (2) assigned to the pseudo IHE [6,8,13].

The hyperfine-field averaged NM spin accumulation is shown in Fig. 2. The combined dephasing evident in Eqs. (7) and (8) changes the conventional quadratic fall off in field to $p_s \sim 1/(\gamma_R^2 + b^2)(1 + \tau_s^2 b_0^2)$. The width of the effects are primarily determined by $\gamma_R$, $b_{hf}$, or $\tau_s^{-1}$; the width of the IHE is governed by the hyperfine field, and the HE by $\tau_s^{-1}$ for $\tau_s^{-1} < b_{hf}$, leading to very different widths. The discrepancy should offer a means of distinguishing if this trap-mediated spin accumulation leads to observed spin voltages. However, if $\tau_s^{-1} < b_{hf}$, the widths are the same.

Experiments [11] on Fe/MgO/Si do observe a sharper peak of the HE superimposed upon a broader peak, and broader inverted Hanle peak. The spin lifetime of the narrow peak is comparable to the one obtained in an accompanying nonlocal four-terminal experiment, which suggests an origin in direct tunneling. The presence of similarly broad peaks in the Hanle and inverted Hanle curves point to additional charge hopping through spin bottleneck sites, though contributions from hopping through spin transport sites cannot be ruled out. Other experiments [23] on Fe/SiO$_2$/Si displayed behavior in oblique fields which suggests the importance of stray fields.

C. Oblique fields

Recently, the effect of oblique magnetic fields was measured to help distinguish spin accumulation and magnetic-field-dependent transport though barrier traps [14]. A strong field applied parallel to the magnetization suppresses any stray fields and returns the spin accumulation to its stray-field-free
value, which for oblique fields is proportional to \( \cos^2 \theta \) for direct tunneling [21]. Reference [14] compared oblique measurements (solid symbols in Fig. 3) to both the direct tunneling model and the trap model (spin bottleneck sites); the result was inconclusive as both models adequately described the data.

We propose alternate quantities to distinguish the two models: the ratio of the current or spin accumulation at zero and high fields at angle \( \theta \). The barrier trap model predicts a universal response whereas the stray-field model depends on the details of the magnetic layer’s roughness. Specifically,

\[
i_{\text{ratio}} = \frac{\langle i(b_0 \to 0) \rangle - \langle i(b_0 \to \infty, \theta) \rangle}{\langle i(b_0 \to 0) \rangle}
\]

(9)

for spin extraction and

\[
p_{\text{ratio}} = \frac{\langle p_s(b_0 \to \infty, \theta) \rangle - \langle p_s(b_0 \to 0) \rangle}{\langle p_s(b_0 \to \infty, \theta = 0\) \rangle - \langle p_s(b_0 \to 0) \rangle}
\]

(10)

for spin injection. Angular brackets denote averaging over the gaussian distribution of hyperfine fields.

In general, \( i_{\text{ratio}} \) can only be computed numerically but \( p_{\text{ratio}} \) is analytically found to be [15]

\[
p_{\text{ratio}} = \frac{3C(\cos^2 \theta - 1)}{2} + 1
\]

(11)

with

\[
C = \left[ 1 - \frac{e^2}{\hbar c} + \frac{\sqrt{2\pi} \gamma_L \gamma_R}{\hbar c} e^{1/2\gamma_R} \text{erfc}(\frac{1}{\sqrt{2\gamma_R}}) \right]^{-1},
\]

(12)

where remarkably, there is no dependence on \( \gamma_L \) nor \( \tau_s \), and no approximations have been made on the relative size of the various rates. Meanwhile \( i_{\text{ratio}} \) is independent of \( \tau_s \). As shown in the Supplement Material [15], for either metric, if \( \gamma_L \) is nearly identical to \( \gamma_R \) for spin injection. Angular brackets denote averaging over the various rates. Meanwhile \( i_{\text{ratio}} \) is independent of \( \tau_s \). As shown in the Supplement Material [15], for either metric, if

\[\gamma_L \approx \gamma_R \approx \gamma_L \text{ then } i_{\text{ratio}} = p_{\text{ratio}} \equiv r(\theta) = (3\cos^2 \theta - 1)/2.\]

Figure 3 shows the oblique field data from Ref. [14], along with \( r(\theta) \), and the agreement with the barrier trap prediction is remarkable. The fixed ratio between currents at 90° and 0° \( r(90°) = -1/2 \) was already noted in STO/LAO/Co structures [6]. Ratios near this value hold in other experiments as well, but not all [5,24,25]. The common occurrence of the ratio is a strong indicator that stray fields rarely are the IHE mechanism. Different structures and magnets possess varying degrees of roughness and thus varying stray field distribution which have no cause to yield \( r(90°) = -1/2 \).

Distinguishing spin transport centers from spin bottleneck centers is difficult solely from the angle-dependent amplitudes. The width for spin bottleneck centers, determined by the hyperfine coupling, is the same for parallel and perpendicular applied fields [15]. This is not the case for spin transport centers when \( \tau_s \hbar c \gg 1 \); in this instance, the width is governed by \( \tau_s^{-1} \). We expect that both impurity types occur (in addition to direct tunneling) so disentangling their contributions in measurements is not feasible within the scope of this paper. For \( \gamma_R \ll \gamma_L \ll \gamma_L \), both \( i_{\text{ratio}} \) and \( p_{\text{ratio}} \) approach \( r \) but outside that strict constraint the two ratios are not identical [Fig. 3(a) versus Fig. 3(b)]. \( p_{\text{ratio}} \) is more sensitive to \( \gamma_R \) than \( i_{\text{ratio}} \).

Figure 4 summarizes the magnitudes of the conventional and pseudo HE and IHE for either spin injection (red) or spin extraction (orange). The blue curves (red dashed line) indicate slow (fast) \( \gamma_R \) approximations for extraction (injection). Three regions are represented by red (forbidden), cyan (IHE < HE), and green (IHE > HE). The quantity \( -x_{\text{ratio}}^{-1}(90°) \) should be interpreted as the ratio between IHE and HE: \( e.g. -i_{\text{ratio}}^{-1}(90°) = \Delta i(\theta = 0°)/\Delta i(\theta = 90°) \). The large-\( \gamma_R \) asymptotic behavior
of \(-p_{\text{ratio}}^{-1}(\theta)\) is \(2\beta^2 \csc \theta / \gamma^2\) which is the dashed red line in Fig. 4.

IV. RAMIFICATIONS FOR NONLOCAL SPIN DETECTION

For smaller voltage bias, hops occur back and forth between each lead and the barrier trap. At zero bias (zero charge current), no spin current is produced [15]. Thus the described magnet effects for spin transport centers or spin bottleneck centers cannot occur without a bias, so such effects are not expected in nonlocal measurements. Reference [11] observes impurity-assisted signatures in three-terminal but not four-terminal devices. Thus the trap effects here will not confuse spin pumping or thermal spin transport experiments involving FM/NM interfaces without charge currents. The traps will not alter the spin currents without charge currents, nor can unbiased spin centers mediate ferromagnetic proximity polarization [26–28].

V. CONCLUDING REMARKS

The ambiguity in the spin-dependent magnetoresistance in a resonant tunneling formulation originates from the dependence of the Hubbard Hamiltonian \((U_{\uparrow\downarrow}, n_{\uparrow\downarrow})\) on the spin quantization axis [13,20]. Our results are independent of the quantization axis and agree with Ref. [13], although the reason why this is the “proper” choice is unclear; we thus suggest the SLE approach is more robust as no assumption of an axis is required.

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[15] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.103.035310 for further description of spin centers in spin injection and extraction regimes, details of the Hanle effect calculation, additional results for spin extraction regime, details on the calculations of \(p_{\text{ratio}}\) and \(p_{\text{ratio}}\), and analysis of the junction with zero bias which is relevant for nonlocal spin detection techniques.


