Tunnel magnetoresistance in organic spin valves in the regime of multi-step tunneling

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A model of a spin valve in which electron transport between the magnetized electrodes is due to multistep tunneling is analyzed. Motivated by recent experiments on organic spin valves, we assume that spin memory loss in the course of transport is due to random hyperfine fields acting on electron while it waits for the next tunneling step. Amazingly, we identify the three-step configurations of sites, for which the tunnel magnetoresistance (TMR) is negative, suggesting that the resistance for antiparallel magnetizations of the electrodes is smaller than for parallel magnetizations. We analyze the phase volume of these configurations with respect to magnitudes and relative orientations of the on-site hyperfine fields. The effect of sign reversal of TMR is exclusively due to interference of the spin-flip amplitudes on each site, it does not emerge within commonly accepted probabilistic description of spin transport. Another feature specific to multistep inelastic tunneling is bouncing of electron between nearest neighbors while awaiting a “hard” hop. We demonstrate that this bouncing, being absolutely insignificant for conduction of current, can strongly affect the spin memory loss. This effect is also of interference origin.

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I. INTRODUCTION.

A spin valve is a device the resistance of which, $R_{\uparrow\uparrow}$ or $R_{\uparrow\downarrow}$, depends on the mutual orientation ($\uparrow\uparrow$ or $\uparrow\downarrow$) of magnetization directions in ferromagnetic electrodes. Quantitative measure of the effectiveness of a spin valve is the tunnel magnetoresistance (TMR), which is expressed via the electrode polarizations, $P_1$ and $P_2$, as follows

$$\text{TMR} = \frac{\Delta R}{R_{\uparrow\uparrow}} = \frac{R_{\uparrow\uparrow} - R_{\uparrow\downarrow}}{R_{\uparrow\uparrow}} = \frac{2P_1P_2}{1 - P_1P_2}, \quad (1)$$

If the thickness, $L$, of the active layer is large enough, the spin orientation of injected electrons is “forgotten” in course of transport between the electrodes. Usually this effect is taken into account by multiplying the product $P_1P_2$ by a factor $\exp(-L/l_s)$, where $l_s$ is the spin-diffusion length. The use of the concept of spin diffusion implies that, while traveling between the electrodes, electron experiences many scattering events, and for each event the spin rotation is weak. Under these conditions the spin polarization is a continuous function of the coordinate. More generally, the product $P_1P_2$ should be multiplied by $(1 - 2P_d)$, so that

$$\text{TMR} = \frac{2P_1P_2(1 - 2P_d)}{1 - P_1P_2(1 - 2P_d)}, \quad (2)$$

where $P_d$ is the probability that electron flips its spin over the distance $L$. Then Eq. (2) applies even when the spin rotation in course of a scattering event is not small, i.e. the initial spin orientation is “forgotten” after only a few events. The factor $(1 - 2P_d)$ emerges in Eq. (2) if one takes into account that, as a result of spin-flips in the active layer, the states with spin, say, $\uparrow$, in the left electrode are coupled to the states $\uparrow$ in the right electrode with probability $1 - P_d$ and to the states $\downarrow$ with probability $P_d$. Although Eq. (2), for the particular case $(1 - 2P_d) = \exp(-L/l_s)$ appears in many sources, for completeness, we present its derivation in the Appendix.

FIG. 1: (Color online) (a) Illustration of the regime of transport between ferromagnetic electrodes, $L$ and $R$, dominated by hops via intermediate sites 1 and 2. Spin precession in the hyperfine fields takes place while electron waits for the hops 1 $\rightarrow$ 2 and 2 $\rightarrow$ R. Bias is assumed large, so that all hops are unidirectional; (b) When the sites 1 and 2 are close in energy, electron bounces 2 $\rightarrow$ 1 $\rightarrow$ 2 many times while waiting for the “long” hop 2 $\rightarrow$ R.

In the present paper we assume that the underlying mechanism responsible for $P_d$ is the spin rotation in hyperfine magnetic fields. This situation is generic for organic spin valves. In Ref. [11] experimental data on...
spin valves with an organic active layer was analyzed. The results were interpreted within a model in which the tunnel transport through the active layer proceeds in two steps: first tunneling from the left electrode \( L \) (see Fig. 1) to a localized state in the middle, and, subsequently, to the right electrode \( R \). This “stop” near the middle of the active layer increases the overall tunnel probability from \( \exp(-L/a) \) to \( \exp(-L/2a) \), where \( a \) is the under-barrier tunneling length. At the same time, while electron waits to tunnel into \( R \), its spin is subject to a hyperfine magnetic field created by surrounding nuclei. If the average waiting time is \( \tau \), the expression for \( P_{st} \) takes the form
\[
P_{st}^{(0)} = \frac{1}{2} \left( \frac{\Omega^2 - \Omega_\perp^2}{1 + \Omega^2 \tau^2} \right),
\]
where \( \Omega \) is the total magnetic field at the site (in frequency units), and \( \Omega_\perp \) is the projection of this field on the direction of magnetization; \( z \)-direction is determined by the magnetization in the electrode \( L \).

Upon gradual increase of the thickness, the transport will be dominated by three-step tunneling, then four-step tunneling, and so on. Rigorous treatment demonstrates that the number of steps, \( N \), grows with the thickness, \( L \), as \( N = \sqrt{L/a} \). In the present paper we study in detail the domain of lengths where the transport is via three-step tunneling, as illustrated in Fig. 1. This regime is still analytically tractable, and yet reveals fundamental features which are germane to multistep transport and are lacking in the two-step regime. These features are:

(i) TMR is strongly affected by the fact that the amplitude for the net spin rotation is the sum of amplitudes for the rotations taking place when electron waits for the hop on site 1 and on site 2. We show that this addition of amplitudes rather than probabilities can lead to negative TMR, and explore the domain in which the sign reversal of TMR occurs.

(ii) If the waiting time for the hop 2 \( \rightarrow R \) is long, the electron bounces between the sites 1 and 2 while awaiting the hop 2 \( \rightarrow R \). This bouncing, which has absolutely no effect on the current, can strongly affect the spin rotation.

Both above findings have quantum interference at their core. In this regard note, that, while electron hops are incoherent, the spin evolution in course of these hops remains fully coherent. The fact that the times spent by electron on each site are random tends to average out the interference effects. It is thus nontrivial that interference effects survive this averaging, and manifest themselves in the limit \( \Omega \tau \gg 1 \), when the typical spin rotation is strong.

The paper is organized as follows. In Sect. II we consider the transport via two sites at high bias when electron moves only forward. In Sect. III, we relax this condition and allow fast backward hops while awaiting the slow forward hop. For both situations we calculate \( P_{st} \) averaged over the random durations of the waiting periods, which should be substituted into Eq. (2). We pay special attention to \( P_{st} \) in the presence of external magnetic field in view of mysterious absence of the Hanle effect in spin valves reported recently. In Sect. IV we discuss the implications of our findings for true multistep or bulk transport.

II. INTERFERENCE CORRECTION TO THE TWO-STEP SPIN-FLIP PROBABILITY

A. Analytical expression for \( P_{st} \)

Under a strong applied bias the motion of the electron is unidirectional. The hops proceed in a sequence \( L \rightarrow 1 \rightarrow 2 \rightarrow R \). Denote with \( t_1 \) and \( t_2 \) the random times spent by electron on sites 1 and 2, respectively. The evolution of spin is described by the product of the unitary matrices \( U(t_2)U(t_1) \), where the matrix \( U(t) \) is defined as
\[
U(t) = \left[ \begin{array}{cc} \cos \alpha - i \frac{\Omega_\perp}{\Omega} \sin \alpha & -i \frac{\Omega_\perp}{\Omega} \sin \alpha \\ -i \frac{\Omega_\perp}{\Omega} \sin \alpha & \cos \alpha + i \frac{\Omega_\perp}{\Omega} \sin \alpha \end{array} \right], \quad \alpha = \frac{\Omega t}{2},
\]
where \( \Omega_\perp = \Omega_z \pm i \Omega_y \). The spin-flip amplitude is given by a non-diagonal element, \( A_{13} = -i \frac{\Omega_\perp}{\Omega} \sin \left( \frac{\Omega t_1}{2} \right) \). Averaging of \( p_{t_1} = |A_{13}|^2 \) over the Poisson distribution, \( \frac{1}{t} \exp(-t/\tau) \), of the waiting time, \( t \), reproduces Eq. (3).

The spin-flip amplitude after two steps is given by non-diagonal elements of \( U(t_2)U(t_1) \). It can be written in the form
\[
\hat{A}_{13} = A_{13}^{(1)} A_{31}^{(2)} + A_{13}^{(1)} A_{13}^{(2)},
\]
where \( A_{1,2}^{(1,2)} \) are the corresponding elements of the matrices \( U(t_1) \) and \( U(t_2) \). Averaging of \( P_{st} = |A_{13}|^2 \) over random times \( t_1, t_2 \) can be easily carried out. First, it is convenient to present \( P_{st} \) in the form
\[
P_{st} = P_{\text{incoh}} + \delta P_{\text{int}}
\]
\[
\text{(6)}
\]
of the sum of incoherent and interference contributions defined as
\[
P_{\text{incoh}} = p_{st}^{(1)} \left( 1 - p_{st}^{(2)} \right) + \left( 1 - p_{st}^{(1)} \right) p_{st}^{(2)},
\]
\[
\text{(7)}
\]
where \( p_{st}^{(1)} \) and \( p_{st}^{(2)} \) are the partial probabilities given by Eq. (3), and
\[
\delta P_{\text{int}} = 2 \left< \text{Re} \left( A_{13}^{(1)} A_{31}^{(2)} A_{1\uparrow}^{(1)*} A_{1\downarrow}^{(2)*} \right) \right>_{t_1, t_2}.
\]
\[
\text{(8)}
\]
Averaging of \( \delta P_{\text{int}} \) over \( t_1 \) and \( t_2 \) can be performed independently. The product of the terms depending on \( t_1 \) is
\[
A_{13}^{(1)*} A_{1\uparrow}^{(1)*} = \left[ -\frac{\Omega_\perp}{\Omega} \sin \left( \frac{\Omega t_1}{2} \right) \right] \times \left[ \cos \left( \frac{\Omega t_1}{2} \right) + i \frac{\Omega_\perp}{\Omega} \sin \left( \frac{\Omega t_1}{2} \right) \right].
\]
\[
\text{(9)}
\]
Denote with $\tau_i$ the average waiting time for the hop $1 \to 2$. Averaging of Eq. (9) over $t_i$ yields a compact expression

$$\langle A_1^{(1)} A_2^{(1)*} \rangle_{t_1} = \frac{1}{2} \Omega_{1z} \tau_1 \left( -i + \Omega_{1z} \tau_1 \right).$$

The same expression with $\tau_2$ instead of $\tau_1$ and $\Omega_2$ instead of $\Omega_1$ together with an additional complex conjugation describes the result of averaging over $t_2$. Altogether, the expression for $\delta P_{\text{int}}$ can be cast in the form

$$\delta P_{\text{int}} = \frac{1}{2} \text{Re} \left( \frac{\Omega_{1z} \Omega_{2z} \tau_1 \tau_2 (1 + i \Omega_{1z} \tau_1)(1 - i \Omega_{2z} \tau_2)}{(1 + \Omega_{1z}^2 \tau_1^2)(1 + \Omega_{2z}^2 \tau_2^2)} \right).$$

At this point note that, within the probabilistic approach, the result for $P_d$ would be simply $P_{\text{incoh}}$. Indeed, within this approach, the net spin flip corresponds to flipping on the first site and preserving spin on the second site or vice versa. Since these are mutually exclusive events their probabilities simply add. Because of this, $\delta P_{\text{int}} = P_d - P_{\text{incoh}}$ is a measure of quantum interference of the amplitudes of two rotations that took place at site 1 and at site 2.

Throughout this subsection we implicitly identified $P_d$ with the spin-flip probability which appears in Eq. (9). It is however not entirely obvious that the quantumechanical quantity $P_d(t_1, t_2)$ averaged over the Poisson distribution of the waiting times is the same quantity which appears in Eq. (4). Formal justification is presented in the Appendix.

In the next subsection we analyze several particular cases when the interference term has dramatic consequences for TMR.

### B. Limiting cases

It is instructive to express the result Eq. (9) via the partial probabilities $p_d^{(1)}$ and $p_d^{(2)}$ as follows

$$\delta P_{\text{int}} = \sqrt{p_d^{(1)} \left( 1 - 2p_d^{(1)} \right) p_d^{(2)} \left( 1 - 2p_d^{(2)} \right)} \cos \phi,$$

where the phase $\phi$ is defined as

$$\phi = \varphi_1 - \varphi_2 + \tan^{-1}(\Omega_{1z} \tau_1 \cos \vartheta_1) - \tan^{-1}(\Omega_{2z} \tau_2 \cos \vartheta_2).$$

The angles $\vartheta_1$, $\varphi_1$ ($\vartheta_2$, $\varphi_2$) are the spherical angles describing the polar and azimuthal orientations of the vector $\Omega_1$ ($\Omega_2$). Eqs. (12), (13) indicate that interference can be either constructive or destructive depending on the mutual orientations of the fields $\Omega_1$, $\Omega_2$. When $\Omega_1 \tau_1$ and $\Omega_2 \tau_2$ are of the same order, the interference correction is of the order of $P_{\text{incoh}}$.

1. **Identical fields**, $p_d^{(1)} = p_d^{(2)}$

The role of interference is maximal when the vectors $\Omega_1$ and $\Omega_2$ are collinear and $\Omega_1 \tau_1 = \Omega_2 \tau_2$. Then we have

$$P_d = 3p_d(1 - p_d) + 2p_d(1 - 2p_d) = 3p_d - 4p_d^2.$$ 

To illustrate the non-triviality of Eq. (14), note that the single-scattering value, $p_d$, never exceeds $1/2$. Equally the incoherent part of the two-scattering probability, $P_{\text{incoh}}$, never exceeds $1/2$. The physical meaning of these restrictions is obvious: $p_d = 1/2$ implies a full loss of the spin memory. Therefore, if either of two values of $p_d$ in Eq. (7) is equal to $1/2$, we get $P_{\text{incoh}} = 1/2$ regardless of the value of the other $p_d$. Interestingly, the exact $P_d$ does not satisfy this restriction. Similarly to $P_{\text{incoh}}$, Eq. (14) does yield $1/2$, for $p_d = 1/2$, when the interference term vanishes. However, the value of $P_d$ can actually exceed $1/2$ for smaller $p_d$. Namely, at $p_d = 3/8$, Eq. (14) has a maximum and assumes the value $P_d = 9/16$. This

![Graph showing the cumulative spin-flip probability for different values of $p_d$ and $\phi$](image)
implies that the TMR, defined by Eq. (11), is negative for this $p_{st}$. Moreover, it retains negative value within the domain $1/4 < p_{st} < 1/2$. Physically, this means that the resistance for antiparallel orientations of magnetization in the electrodes is smaller than for the parallel orientation.

In fact, negative values of TMR happen not only when the vectors $\Omega_1$ and $\Omega_2$ coincide. For illustration, assume that the product $\Omega_1 \tau_1$ is still equal to $\Omega_2 \tau_2$, but the vectors $\Omega_1$ and $\Omega_2$ are skewed by an angle $\phi$. The domain $P_{st} = 1/2$ on the $(p_{st}, \phi)$-plane is shown in Fig. 2. The “allowed” values of $\phi$ range from 0 at $p_{st} = 1/4$ to $\pm \pi/2$ at $p_{st} = 1/2$.

To what degree is the assumption that the field magnitudes are precisely equal to each other crucial for negative TMR? To answer this question we have plotted in Fig. 2, the contour plot of $P_{st}$ for configurations with $\phi = 0$ when $p_{st}^{(1)}$ and $p_{st}^{(2)}$ vary over their allowed values. We see that negative TMR corresponds to the domain above the diagonal of the square. This domain shrinks upon increasing $\phi$.

2. Identical fields, many hops

In the example considered above the TMR was “most negative” when both hyperfine fields were equal, i.e. the hopping of electron does not interrupt the spin precession at all. It might seem that this case should be reducible to the precession in one given field for which the result Eq. (3) never goes above 1/2. The resolution lies in the fact that Eq. (3) was obtained upon averaging over exponential distribution of the waiting times. When two hops are performed in the same magnetic field, the distribution function of the two-hop waiting times is different: $F_2(T) = T/\tau^2 \exp(-T/\tau)$. It is because of this difference that $P_{st} > 1/2$ emerges. In this regard, it is interesting to consider what happens if an electron performed $N > 2$ steps in the same magnetic field. Then the distribution function of the waiting time is

$$F_N(T) = \frac{T^{N-1}}{\tau^N(N-1)!} \exp(-T/\tau).$$

With this distribution, the expression for spin-flip probability can be easily shown to take the form

$$P_{st} = \frac{\left|\Omega_1^2\right|^2}{2\Omega^2} \left[1 - \frac{\cos\left(N \sin^{-1} \frac{\Omega}{\sqrt{1+\Omega^2 \tau^2}}\right)}{(1 + \Omega^2 \tau^2)^{N/2}}\right].$$

The situation most favorable for negative TMR is an in-plane orientation of magnetic field, when the prefactor in Eq. (16) is equal to 1/2. Then we have $P_{st} > 1/2$ in the domains of $\Omega$ when the cosine is negative. These domains are shown in Fig. 3 for several values of $N$. It is apparent from Eq. (12) that, since $\cos \phi$ is zero on average, the interference correction to $P_{st}$ vanishes upon averaging over hyperfine field distribution. This explains why the D’yakonov-Perel result for the spin relaxation time derived from probabilistic treatment remains valid in spite of the fact that spin rotations for subsequent electron steps are strongly correlated; large number of electron collisions each of which is accompanied by a small spin rotation guarantees that the averaging takes place. Equally, the averaging happens for a spin valve with large area of the active layer. For a given path through the layer $\delta P_{\text{int}}$ can be of the order of $P_{st}$.
but it will not contribute to the average spin-flip probability coming from many channels. If the area is finite, so that the number of channels, $N \gg 1$, is also finite, the averaging will be incomplete. The TMR will acquire a random correction of the order $\Delta/\sqrt{N}$, where $\Delta^2$ is the variance of $P_{st}$, which we calculate below.

It follows from Eqs. (6), (12) that the variance has two contributions

$$
\Delta^2 = \overline{P_{st}^2} - \left(\overline{P_{st}}\right)^2 = \Delta_{\text{incoh}}^2 + \Delta_{\text{int}}^2.
$$

(18)

where $\Delta_{\text{incoh}}^2$ and $\Delta_{\text{int}}^2$ are the variances of the incoherent and coherent contributions, respectively. The overline stands for hyperfine averaging over the gaussian distribution, $\frac{1}{\sqrt{2\pi}b_0}\exp[-b_i^2/b_0^2]$, of the hyperfine-field components, $b_i$. Then the variance $\Delta_{\text{incoh}}^2$ can be expressed through averages $\overline{p_{1,2}}$ and partial variances $\Delta_{1,2}^2 = \overline{p_{1,2}^2} - (\overline{p_{1,2}})^2$ as follows

$$
\Delta_{\text{incoh}}^2 = (1 - 2\overline{p_1^2}) \Delta^2 + (1 - 2\overline{p_2^2}) \Delta^2 + 4\Delta^2 \Delta_{1,2}^2.
$$

(19)

The corresponding expression for the interference contribution, $\Delta_{\text{int}}^2$, reads

$$
\Delta_{\text{int}}^2 = \frac{1}{2} \left[ (\overline{p_1} + 2\overline{p_2})(\overline{p_1} + 2\overline{p_2}) - 2(\overline{p_1} + 2\overline{p_2}) \Delta_{1} \right.
$$

$$
\left. - 2(\overline{p_1} + 2\overline{p_2}) \Delta_{1}^2 + 4\Delta_{1,2}^2 \right].
$$

(20)

Analytical expressions for $\overline{p_{1,2}}$ and $\Delta_{1,2}^2$ take a simple form in the limits of strong ($\Omega \tau \gg 1$) and weak ($\Omega \tau \ll 1$) magnetic fields:

$$
\overline{p_{1,2}} = \begin{cases} 
\frac{b_{i}^2}{\Delta_{1,2}^2} - \frac{b_{0}^2}{\Delta_{1,2}^2}, & \Omega_{1,2} \tau_{1,2} \ll 1 \\
\frac{1}{2} \int_{0}^{\infty} \frac{ds}{(1+s)^{\tau_{1,2}}} \exp \left[ -\frac{s b_i^2}{b_0^2} \right], & \Omega_{1,2} \tau_{1,2} \gg 1
\end{cases}
$$

(21)

$$
\Delta_{1,2}^2 = \begin{cases} 
\frac{b_{0}^2}{4}, & \Omega_{1,2} \tau_{1,2} \ll 1 \\
\frac{b_{0}^2}{4} + \frac{1}{2} \int_{0}^{\infty} \frac{s ds}{(1+s)^{\tau_{1,2}}} \exp \left[ -\frac{s b_i^2}{b_0^2} \right], & \Omega_{1,2} \tau_{1,2} \gg 1
\end{cases}
$$

(22)

Here $B$ is the external field directed along the z-axis. Eq. (21) describes the fall-off of the disorder-averaged spin-flip probability with $B$. For weak hyperfine field, $b_i \tau \ll 1$, the dependence $\overline{p_{1,2}}(B)$ evolves from small value, $b_{0}^2/2B^2$, to $b_{0}^2/2B^2$. In the opposite limit, $b_i \tau \gg 1$, the evolution starts from $\overline{p_{1,2}}(0) = 1/3$ and converges to $b_{0}^2/2B^2$ when $B$ exceeds $b_0$.

In the first case, we have $\Delta_{\text{incoh}}^2 \approx \overline{p_{1,2}^2}/2$, while $\Delta_{\text{incoh}}^2 \approx \Delta_{1}^2 + \Delta_{1}^2$, so that for the ratio $\Delta_{\text{incoh}}^2/\Delta_{\text{incoh}}^2$ we get $\tau_{1}^2/\tau_i^2 + (\tau_{1}^2 + \tau_i^2)$, i.e. the interference contribution is of the same order as $\Delta_{\text{incoh}}^2$. For strong hyperfine field, $\Delta_{1,2}$, and $\Delta_{\text{incoh}}^2$ do not depend on $\tau$. At $B = 0$, Eq. (22) yields $\Delta_{1,2}^2 = \frac{7}{45}$. Using this value, we get for the contributions to the variance: $\Delta_{\text{incoh}}^2 = \frac{7}{45} \left( \frac{2}{3} \right)^2$ and $\Delta_{\text{int}}^2 = \frac{4}{9} \left( \frac{2}{3} \right)^2$, i.e. the interference contribution is almost 20 times bigger than the incoherent contribution. Finally, consider the limit of strong hyperfine field and $B \gg b_0$. In this limit Eq. (22) yields $\Delta_{1,2}^2 = b_{0}^2/2B^2$, and we thus have:

$$
\Delta_{\text{int}}^2 = \frac{\Delta_{1,2}^2}{2} = \frac{b_0^2}{8B^4}, \quad \Delta_{\text{incoh}}^2 = 2\Delta_{1,2}^2 = \frac{b_0^2}{B^4} = 8\Delta_{\text{int}}^2.
$$

(23)

In summary, for all the domains of change of the dimensionless parameters $b_i \tau$ and $b_i/B$ the variance, $\Delta$, of the spin-flip probability is of the order of average $P_{st}$, and the interference contribution to $\Delta$ is comparable to $\Delta$ itself.

In conclusion of the Section, note that for $\tau \gg \tau_{1}$, the hops $1 \rightarrow 2$ between the sites do not affect the current. Except for anomalous configurations of hyperfine fields, when $\Omega_{2\perp}$ is much smaller than $\Omega_{1\perp}$, these hops also do not affect the spin memory. In the next Section we will demonstrate that multiple bounces of electron within a pair of sites, while not affecting the current, can significantly affect the spin memory. This effect, caused by interference, is most pronounced in the presence of an external magnetic field.

The partial spin-flip probabilities obviously fall off with magnetic field, $B$, which is parallel to the polarization in the injector. The result of the probabilistic approach, $P_{\text{incoh}}$, also falls off with $B$. As it is easy to see from Eq. (3), the probability $p_{st}$ is proportional to $1/B^2$ for $\Omega \tau \gg 1$. Concerning the magnitude of the interference term, Eq. (9), it can actually grow with $B$ if both partial probabilities, $p_{st}$, exceed 1/4. However, when they are both small, the magnitude of interference term also drops with $B$ as $1/B^2$. In the next Section we will demonstrate that electron bounces can transform the $1/B^2$ to a much weaker dependence.

III. EFFECT OF BOUNCING ON THE SPIN-FLIP PROBABILITY

Assume that $\tau_{2}$ is much bigger than $\tau_{1}$ and the activation energy for the back-hop $2 \rightarrow 1$ is small, Fig. 1b). In this case, as it was explained in the Introduction, while awaiting the hop $2 \rightarrow R$, the electron performs $m = \tau_{2}/\tau_{1} \gg 1$ hops $2 \rightarrow 1$ and back. This bouncing affects strongly the spin-rotation and enhances the interference contribution to $P_{st}$.

Note first that, within the probabilistic description, taking bounces into account is equivalent to modifying the partial probability $p_{st}^{(1)}$

$$
p_{st}^{(1)} = \frac{1}{2} - \frac{1}{2} (1 - 2p_{st})^m,
$$

(24)

where $m$ is odd. Eq. (24) expresses the fact that $p_{st}^{(1)}$ is the sum of probabilities to flip spin only once in the course of all bounces, only three times in the course of all bounces, and so on. Accumulation of the powers of
(1 − 2p_m) with m is natural since (1 − 2p_m) is the probability of spin preservation for one step. In reality, while bouncing, electron spin experiences an alternating magnetic field, which takes only two values. This favors the interference processes, and the result Eq. (24) should be compared to the result of treatment with interference taken into account. Within the latter treatment, the spin-flip amplitude is given by the non-diagonal elements of the matrix product \( U_m(t_m) \cdots U_2(t_2)U_1(t_1) \), where \( U_i(t_i) \) is the matrix Eq. (4) in which the fields corresponding to \( U_i \) and \( U_2 \) are \( \Omega_i \) and \( \Omega_2 \), respectively. The times, \( t_i \), are random, but have the same distribution.

To illuminate the importance of interference in course of bouncing, consider the following simple example. Suppose that \( m = 3 \) and that the external field is strong, i.e. \( \Omega_i \ll \Omega \). Assume as well, that the in-plane field components for all three steps are equal in magnitude and differ only in azimuthal orientations, \( \chi_i \). Then the non-diagonal matrix element of the product

\[
\left( \begin{array}{cc} u & -ie^{i\chi_3}v \\ -ie^{-i\chi_3}u & u \end{array} \right) \left( \begin{array}{cc} u & -ie^{i\chi_2}v \\ -ie^{-i\chi_2}u & u \end{array} \right) \left( \begin{array}{cc} u & -ie^{i\chi_1}v \\ -ie^{-i\chi_1}u & u \end{array} \right)
\]

takes a simple form

\[
\hat{A} = iu^2e^{-i(\chi_3−\chi_2+\chi_1)} − iv^2(e^{-i\chi_1} + e^{-i\chi_2} + e^{-i\chi_3}).
\]

Here \( u^2 + v^2 = 1 \). For a sequential hopping all \( \chi_i \) are random. Then the average value of \( |\hat{A}|^2 \) is given by

\[
|\hat{A}|^2 = v^6 + 3u^4v^2.
\]

On the other hand, if the hops constitute a single bounce \( 1 \rightarrow 2 \rightarrow 1 \), we have \( \chi_1 = \chi_3 \), which leads to the following expression for average \( |\hat{A}|^2 \).

\[
|\hat{A}|^2 = v^6 + 5u^4v^2.
\]

The result Eq. (26) can be brought in correspondence with probabilistic description Eq. (24), if we identify \( |v|^2 \) with \( p_m \). The fact that Eq. (27) yields a bigger value for \( |\hat{A}|^2 \) is due to interference of the spin-flip amplitudes which arises as a result of visiting the site 1 twice. Multiple bouncing would amplify the role of interference. It is easier to capture this effect quantitatively by starting directly from the Schrödinger equation for electron spin in a time-dependent magnetic field.

In the next two subsections we will separately consider the effect of bouncing on the spin preservation in a zero and in strong external fields. We will demonstrate that in these two limits the effects of bouncing are opposite.

### A. Bouncing in a zero external field

The amplitudes \( a_i \) and \( a_2 \) for an electron to have an \( \uparrow \) and \( \downarrow \) projections of spin satisfy the system

\[
i\dot{a}_i(t) = \frac{i}{2} \left[ b_i(t) a_i(t) + b_i^*(t) a_i(t) \right],
\]

\[
i\dot{a}_2(t) = \frac{i}{2} \left[ b_i(t) a_i(t) − b_i^*(t) a_i(t) \right].
\]

Suppose that at time \( t = 0 \) electron spin is directed \( \uparrow \), so that \( a_2 = 0 \). A formal solution of the system Eq. (28) reads

\[
a_2(t) = -\frac{i}{2} \int_0^t dt' \left[ \frac{\epsilon}{2} \int_0^{t'} dt'' b_i(t'') \exp \left( -iBt'' \right) \right].
\]

If the net spin rotation during the time, \( \tau_i \), when electron waits for the hop \( 2 \rightarrow R \) is small, we can set \( a_i(t) = 1 \) and \( \exp[-\int_{t_0}^{t} \frac{\epsilon}{2} b_i(t')] = 1 \) in the integrand. This leads to the following result for the spin-flip probability

\[
P_{sf} = \left| b_{1\downarrow} (t_1 + t_3 + \cdots) + b_{2\downarrow} (t_2 + t_4 + \cdots) \right|^2,
\]

where \( t_1, t_3, \ldots \) are the time intervals spent by electron on the site 1, while \( t_2, t_4, \ldots \) are the time intervals spent by electron on the site 2; each time interval is \( \sim \tau_i \). It is an important consequence of bouncing that these time intervals add up, instead of averaging out, which would be the case for hopping over multiple sites. The big parameter \( \tau_i/\tau_1 \) allows us to replace these sums by \( \tau_i/2 \).

Then we get

\[
P_{sf} = \frac{\tau_i^2}{2} \left| \left| \frac{b_{1\downarrow} + b_{2\downarrow}}{2} \right|^2 = \frac{\tau_i^2}{2} \left| \Omega_1 \right|^2.
\]

The meaning of Eq. (31) is obvious: as a result of performing multiple “short” hops while awaiting the “long” hop electron spin “sees” the average hyperfine field, \( \left| \frac{\Omega_1}{2} \right| \). If the number of sites visited in course of waiting was big, the averaging of the corresponding hyperfine fields would lead to the suppression of \( P_{sf} \).

We assumed that the net spin rotation is small, \( b_i \tau_i \ll 1 \). However, the above derivation suggests that we could impose a much weaker requirement, \( b_i \tau_i \ll 1 \). This is because the effective averaging takes place over time \( \sim \tau_i \). If under the condition \( \Omega_i \tau_i \ll 1 \) the product \( b_i \tau_i \) is not small, then \( P_{sf} \) is given by the full Eq. (4) with \( \Omega \) replaced by the average of the vectors \( b_i \) and \( b_2 \).

### B. Bouncing in a strong external field

In a strong external field, \( B \gg b_i \), the net spin rotation is small both for weak, \( b_i \tau_i \ll 1 \), and for strong, \( b_i \tau_i \gg 1 \), hyperfine fields. In the limit \( B \tau_i \gg 1 \), when electron spin rotates many times around the external field while waiting for the hop \( 2 \rightarrow R \), the waiting time drops out of \( P_{sf} \), see Eq. (3). Effect of bouncing on \( P_{sf} \) can be studied perturbatively with respect to hyperfine field. In a zeroth order we have, \( a_i(t) = \exp \left( -\frac{i\epsilon}{2} t \right) \), \( a_2 = 0 \). In the first order the expression for \( a_2(t) \) takes the form

\[
a_2(t) = -\frac{i}{2} \int_0^t dt' \left[ \frac{\epsilon}{2} \int_0^{t'} dt'' b_i(t'') \exp \left( -iBt'' \right) \right].
\]
It is convenient to subtract $\bar{b}_\perp$ from $b_\perp(t')$ in the integrand and rewrite Eq. (32) as

$$a_2(t) = -\tau_\perp \frac{1}{2B} \exp(-iBt) + \tilde{a}_2(t),$$

where $\tilde{a}_2(t)$ is determined by Eq. (32) in which $b_\perp(t')$ in the integrand is replaced by the difference $(b_\perp(t') - \bar{b}_\perp)$. The term $\tilde{a}_2(t)$ captures the effect of bouncing. Next, it is convenient to divide the domain of integration in Eq. (32) into $N = Bt/2\pi$ intervals $2B \pi n < t' < 2B \pi (n + 1)$, and reduce the integration to a single interval $0 < t' < 2B \pi$. This yields

$$\tilde{a}_2(t) = -\frac{i}{2} \sum_{n=0}^{N} 2\pi/B \int_0^{2\pi/B} dt' \exp(-iBt') \left[ b_\perp(t' + 2\pi/B n) - \bar{b}_\perp \right].$$

In the domain $1/2 < B < 1$ the right-hand side of Eq. (34) is a sum of $N$ statistically-equivalent and independent terms, each being zero on average. In each term the integrand changes sign $2\pi/B \tau_1$ times with magnitude $\Delta b_\perp = \bar{b}_\perp$ - $b_\perp$. This allows us to estimate $|\tilde{a}_2(t)|^2$ as follows

$$|\tilde{a}_2(t)|^2 \sim |\Delta b_\perp|^2 \tau_1^{1/2} \left[ \frac{2\pi}{B \tau_1} \right]^{1/2} \sim |\Delta b_\perp|^2 \tau_1.$$ (35)

The factor $N^{1/2}$ accounts for the fact that all $N$ terms in the sum (34) are random. The factor $\left( \frac{2\pi}{B \tau_1} \right)$ accounts for the fact that each term is the sum of $2\pi/B \tau_1$ random contributions.

We see that magnetic field has dropped out of the “bouncing” estimate for $|\tilde{a}_2(t)|^2$. It dominates over the “regular” part, given by the first term in Eq. (33), for $t \tau_1 \gg 1/B^2$. Since the characteristic $t$ is $\sim \tau_2$, this defines a characteristic field

$$B_c = \frac{1}{(\tau_1 \tau_2)^{1/2}}.$$ (36)

At $B \sim B_c$ the spin-flip probability crosses over from $P_{sf} \sim \bar{b}_\perp^2 / B^2$ to a plateau value

$$P_{sf} \sim \bar{b}_\perp^2 \tau_1 \tau_2.$$ (37)

In deriving Eq. (37) we assumed that many bounces took place during the period, $2\pi/B$, of the in-plane spin rotation. This assumption is justified since $B_c \tau_1 \sim (\tau_1 / \tau_2)^{1/2} \ll 1$. As magnetic field increases above $1/\tau_1$, the spin will execute many in-plane rotations in course of every bounce. Then the integral in the expression for $\tilde{a}_2$ can be viewed as a sum of $\tau_1/\tau_1$ random contributions each of being of the order of $\Delta b_\perp / B$. Then we can again estimate $\tilde{a}_2$, and subsequently, $P_{sf}(B)$, from the variance. The result reads

$$P_{sf}(B) \bigg|_{B \gg 1/\tau_1} \sim \frac{|\Delta b_\perp|^2}{B^2} \left( \frac{\tau_2}{\tau_1} \right).$$ (38)

Note that the bouncing-related spin-flip probability, Eq. (38), exceeds the result $P_{sf} \sim \bar{b}_\perp^2 / B^2$ in the absence of bouncing by a large factor $\bar{b}_\perp / \tau_1$, which is the number of bounces.

Thus, unlike the case $B = 0$, the bouncing causes the growth of the spin-flip probability. The probability Eq. (38) for strong fields matches $P_{sf}$ for intermediate fields, Eq. (37), at $B \sim \frac{1}{\tau_1}$.

In conclusion of the subsection we summarize the results for $P_{sf}$ in different domains of magnetic field

$$P_{sf}(B) = \begin{cases} \bar{b}_\perp^2 \tau_2^2, & 0 < B < \frac{1}{\tau_1}, \\ \frac{|\Delta b_\perp|^2}{B^2} \left( \frac{\tau_2}{\tau_1} \right), & \frac{1}{\tau_1} < B < \frac{1}{\tau_2}, \\ \frac{|\Delta b_\perp|^2}{B^2} \tau_1, & B > \frac{1}{\tau_2}. \end{cases}$$ (39)

The evolution of the spin-flip probability with magnetic field is illustrated in Fig. 4.

IV. DISCUSSION

- Conventional treatments of spin relaxation neglect interference effects. This happens at the stage when the exact equation for the density matrix is solved using the “tau-approximation”, see e.g. the review Ref. 19. Concerning the effect of bouncing considered in the present paper, there is an analog of bouncing in spin relaxation caused by spin-orbit coupling. In course of the orbital electron motion in a strong magnetic field, it keeps returning to the origin after undergoing the same sequence of scattering events. This “memory” results in shortening of the spin relaxation time. 19 Similarly, Eq. (24), where bouncing is treated probabilistically, predicts

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FIG. 4: (Color online) Schematic illustration of the enhancement of the spin-flip probability due to multiple bounces. In the absence of bouncing, the plateau (1) at small external fields crosses over to the $1/B^2$ behavior (green dashed line) at $B \sim 1/\tau_2$, where $\tau_2$ is the waiting time for the hop $2 \to R$. When the waiting time, $\tau_1$, for the hops $1 \to 2$ and $2 \to 1$ is much shorter than $\tau_2$, the spin-flip probability decreases (2), develops a second plateau (3) at $B \sim (\tau_1 \tau_2)^{-1/2}$, see Eq. (39), and crosses over (4) to $1/B^2$ behavior (blue dashed line) at $B \sim 1/\tau_1$. 

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that $P_a$ approaches to 1/2 faster as the number, $m$, of bounces grows. We emphasize that the quantum treatment of bouncing leads to the opposite result.

- Absence of the Hanle effect reported in Refs. [14] and [15] can be interpreted as independence of $P_{sf}$ on the magnitude of the external field. In this regard, we note that the partial $p_{sf}$ values given by Eq. [3] increase monotonically with increasing magnetic field, for any field orientation. However, in Sect. II we demonstrated that, when the partial probabilities $p_{sf}$ are in the vicinity of 1/2, the dependence of the net two-step probability, $P_{sf}$, on these partial probabilities is non-monotonic. Moreover, the derivative of $P_{sf}$ with respect to the magnetic field passes through zero. This indicates that, for a range of parameters where $P_{sf}$ is near its maximum, there is no sensitivity to the magnitude of the applied field. Note however, that, since this behavior is a consequence of interference, it does not survive averaging over hyperfine field distributions.

- The fact that electron flips its spin as it travels between the electrodes constitutes an additional source of shot noise [20] and, thus, affects the Fano factor. The above calculation of $P_{sf}$ is insufficient to find the Fano factor of “two-site” transport with spin flip. The reason for this is that the transport of charge is incoherent while the spin-transport is fully coherent. Qualitatively, the complexity of description of noise follows from the fact that the Fano factor must depend on both $P_{sf}$ and the magnetizations $P_1$, $P_2$ of the electrodes. The latter conclusion can be inferred from the reasoning presented in Ref. [21]. Suppose that magnetizations of the electrodes are anti-parallel, and $P_{sf}$ is small. Then, no matter what is the actual mechanism of transport, the Fano factor should be 1, which is the Poissonian value. This is because, in order to be transferred between the electrodes, electron must flip the spin. For small $P_{sf}$ it is waiting time for the spin-flip which is the bottle-neck for transport, since it is much longer than the waiting time for all hops. All we can say is that, if one electrode changes from oppositely polarized to non-polarized, the Fano factor changes from 1 to $\frac{b_{\sigma}^2 \tau_0^2}{\tau_0^2 + b_{\sigma}^2 \tau_0^2}$, which is the Fano factor for spin-independent transport through the same sites. Rigorous evaluation of the Fano factor with magnetization of electrodes taken into account, requires solving the equation for time evolution of the full density matrix.

- As was mentioned in the Introduction, in diffusive transport, spin-memory loss is incorporated via the “survival” probability, $\exp(-L/l_s)$, which we replaced by $1 - 2P_{sf}$. The probabilistic description, on the other hand, predicts that $P_{sf}$ falls off exponentially with the number of steps, $N$, see Eq. [17], or equivalently with time, but not with length.

The exponential dependence $\exp(-L/l_s)$ is recovered upon the transformation

$$P_{sf}(L) = \int dN P_{sf}(N) \exp \left(-\frac{L^2}{N r^2}\right), \quad (40)$$

where $r$ is the length of a diffusion step. This yields

$$l_s = \frac{r}{\sqrt{\ln(1 - 2p_{sf})}}. \quad (41)$$

Here we would like to emphasize that the concept of spin diffusion length does not apply for multi-step transport [11,12]. The reason for this is twofold. Unlike diffusion, the relationship between $L$ and $r$ in multistep transport is $r = \sqrt{L/a}$, and $N = L/r = \sqrt{L/a}$, where $a$ is the under-barrier decay length [20]. Secondly, also unlike diffusion, the waiting time for the next step, which is the time for spin precession, is also a function of $L$ and $N$, specifically, $\tau = \tau_0 \exp(2L/Na)$. As a result we get

$$P_{sf} = \frac{1}{2} \frac{b_{\sigma}^2 \tau_0^2 \exp \left(\sqrt{\frac{L}{a}}\right)}{1 + b_{\sigma}^2 \tau_0^2 \exp \left(\sqrt{\frac{L}{a}}\right)}. \quad (42)$$

We see from Eq. (42) that for multistep transport the spin-memory falls off with thickness of the active layer, $L$, slower than for diffusive transport. Anomalous dependence of TMR on the device thickness was reported in Ref. [21]. However Eq. (42) does not explain the established facts that TMR vanished with increasing bias and temperature [21,23].

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Appendix A

For spin-independent unidirectional transport the current between the electrodes can be viewed as a sequence of cycles

$$I(T) = \delta(T - T_1) + \delta(T - T_1 - T_2) + \delta(T - T_1 - T_2 - T_3) + \cdots, \quad (A1)$$

where $T_i$ is a random waiting time for the next electron to be transferred between $L$ and $R$. Suppose now that the left electrode is polarized $\uparrow$, while the right electrode
is polarized ↓. Then the electron transfer requires a spin-flip, and Eq. (A1) should be modified as

\[ I_{↑↓}(T) = P_{a}(T)\delta(T-T_1) + P_{a}(T)\delta(T-T_1-T_2) \]
\[ + P_{a}(T)\delta(T-T_1-T_2-T_3) + \cdots, \quad (A2) \]

where \( P_{a}(T) \) is a quantum-mechanical probability that after a composite process with duration \( T \) the electron flips its spin. For the situation considered in this paper the composite process is an inelastic two-hop tunneling. To calculate the average current one should take the limit of large \( T \) and average over the compound waiting times, \( T_i \), with distribution function \( F(T) \). This averaging is convenient to perform using the integral representation of the \( \delta \)-function. Then the sum Eq. (A2) turns into a geometrical progression, the summation of which yields

\[ \langle I_{↑↓}(T) \rangle = \int_{0}^{2\pi} \frac{d\alpha}{2\pi} e^{i\alpha T} \frac{P_{a}(T') \exp(-i\alpha T')}{1 - \exp(-i\alpha T')} . \quad (A3) \]

In the limit \( T \to \infty \) one can set \( \alpha = 0 \) in the numerator and expand the denominator to the lowest order. After that the integration over \( \alpha \) can be easily performed leading to the natural result

\[ \langle I_{↑↓} \rangle = \frac{\langle P_{a} \rangle}{(T)} . \quad (A4) \]

where \( \langle P_{a} \rangle \) is defined as

\[ \langle P_{a} \rangle = \int P_{a}(T)F(T)dT. \quad (A5) \]

For a particular case of a two-hop transport we have \( T = t_1 + t_2 \), where \( t_1 \) and \( t_2 \) are distributed with \( f_{t_1,t_2}(t) = \frac{1}{\tau_{1,2}} \exp(-t/\tau_{1,2}) \). Then Eq. (A5) assumes the form

\[ \langle P_{a} \rangle = \int_{0}^{\infty} dt_1 \int_{0}^{\infty} dt_2 P_{a}(t_1,t_2)f_{t_1}(t_1)f_{t_2}(t_2). \quad (A6) \]

This is exactly the quantity calculated in Sect. II. From Eq. (A4) we conclude that for calculation of average current one should multiply this quantity by \( 1/\langle T \rangle \), which is the current between unpolarized electrodes.

From the same reasoning we confirm that opposite directions of polarization of the electrodes the current is equal to \( I_{↑↓} = (1 - \langle P_{a} \rangle)/\langle T \rangle \). Therefore the expression for TMR with completely polarized electrodes takes the form

\[ \text{TMR} = \frac{I_{↑↓} - I_{↑↑}}{I_{↑↓} + I_{↑↑}} = 1 - 2P_{a}. \quad (A7) \]

For partial polarization of electrodes with concentrations \( N_{↑}, N_{↓} \) of ↑ and ↓ electrons in the left electrode and \( n_{↑}, n_{↓} \) in the right electrode, the general expressions for \( I_{↑↓} \) and \( I_{↑↑} \) can be presented as

\[ I_{↑↓} = \Gamma_{↑↓}(N_{↑}n_{↓} + N_{↓}n_{↑}) + \Gamma_{↑↓}(N_{↑}n_{↓} + N_{↓}n_{↑}), \quad (A8) \]

\[ I_{↑↑} = \Gamma_{↑↑}(N_{↑}n_{↓} + N_{↓}n_{↑}) + \Gamma_{↑↑}(N_{↑}n_{↓} + N_{↓}n_{↑}), \quad (A9) \]

where \( \Gamma_{↑↓} \) and \( \Gamma_{↑↑} \) are the rates for the transfer processes from ↑ to ↑ and from ↑ to ↓. These rates are the characteristics of the active layer and do not depend on the polarizations of the electrodes. Naturally, we have \( \Gamma_{↑↓} = \Gamma_{↓↑} \) and \( \Gamma_{↑↑} = \Gamma_{↓↓} \). The expression Eq. (2) follows from Eqs. (A8) and (A9) in two steps. We relate the concentration via the degrees of polarization as

\[ P_{1} = \frac{N_{↑} - N_{↓}}{N_{↑} + N_{↓}}, \quad P_{2} = \frac{n_{↑} - n_{↓}}{n_{↑} + n_{↓}}, \quad (A10) \]

yielding

\[ \text{TMR} = \frac{2P_{1}P_{2}(\Gamma_{↑↓} - \Gamma_{↑↑})}{(\Gamma_{↑↑} + \Gamma_{↑↓}) - P_{1}P_{2}(\Gamma_{↑↑} - \Gamma_{↑↓})}. \quad (A11) \]

Finally, we relate the rates \( \Gamma_{↑↓}, \Gamma_{↑↑} \) via \( P_{a} \) as

\[ \frac{\Gamma_{↑↓} - \Gamma_{↑↑}}{\Gamma_{↑↑} + \Gamma_{↑↓}} = 1 - 2P_{a}, \quad (A12) \]

and arrive to Eq. (2).
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