Relaxation mechanism for electron spin in the impurity band of *n*-doped semiconductors

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We propose a mechanism to describe spin relaxation in *n*-doped III-V semiconductors close to the Mott metal-insulator transition. Taking into account the spin-orbit interaction induced spin admixture in the hydrogenic donor states, we build a tight-binding model for the spin-dependent impurity band. Since the hopping amplitudes with spin flip are considerably smaller than the spin-conserving counterparts, the resulting spin lifetime is very large. We estimate the spin lifetime from the diffusive accumulation of spin rotations associated with the electron hopping. Our result is larger but of the same order of magnitude than the experimental value. Therefore, the proposed mechanism has to be included when describing spin relaxation in the impurity band.

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I. INTRODUCTION

The renewed interest in spin relaxation in semiconductors[1] stems from the possible applications and fundamental science associated with the emerging field of spintronics.[2,3] The large measured values of the electron spin lifetime constitute a promise for the use of spin as a unit of quantum information and pose a considerable challenge for the identification of the appropriate mechanisms of spin relaxation. Interestingly, low-temperature experiments in various *n*-doped semiconductor bulk systems have found that the longest spin lifetimes occur close to the Mott metal-insulator transition (MIT) density.[4–8] Many aspects of the Mott transition have been thoroughly studied, making it a paradigm of condensed matter physics.[9] However, the connection between the spin and transport problems is only beginning to be explored.[10]

The comprehensive experimental and theoretical work of Dzhioev *et al.*[7] considered both sides of the MIT, which for GaAs occurs at $n_c \simeq 2\times 10^{16}$ cm^{-3}. In the deeply localized regime, with donor densities $n \leqslant 5\times 10^{15}$ cm^{-3}, the hyperfine interaction was shown to account for the measured spin lifetimes.[11] For higher densities, but still lower than n_c, the anisotropic exchange of localized spins was proposed as the dominant mechanism for spin relaxation.[12] Later calculations based on the same mechanism[13] initiated an ongoing controversy concerning the quantitative agreement between experiment and theory. The coupling between localized states in the impurity band and delocalized states in the conduction band, initially considered by Paget[14] for highly pure GaAs, has been recently applied[15] to densities slightly smaller than n_c, and the fitting parameter describing the strength of the exchange interaction is a factor of 3 larger than the theoretical estimation.

For densities above the transition, the well-known D'yakonov-Perel (DP) mechanism for conduction electrons was invoked.[7,16] This mechanism arises from the splitting of the conduction band due to spin-orbit interaction and yields a spin lifetime inversely proportional to the momentum relaxation time. This description applies to doping densities high enough so that mainly the conduction band is populated. [For GaAs, the hybridization of impurity and conduction bands occurs at a doping density $n_h \simeq 8\times 10^{16}$ cm^{-3} (Refs. 4, 17, and 18).] If one is interested in understanding the large lifetimes measured near the MIT, the DP mechanism is not applicable since, clearly, any mechanism invoking momentum relaxation via impurity scattering becomes meaningless in the impurity band. This difficulty lies at the origin of the lack of suitable theories of spin relaxation at low temperature near the MIT.[19]

In this work, we provide a theoretical frame and propose a spin-relaxation mechanism for the metallic-conduction regime of the impurity band. Our approach is to extend, by incorporating the spin-orbit interaction, the well-known model of Matsubara and Toyozawa (MT) of electron conduction at zero temperature.[17] The effect of the spin-orbit interaction is to introduce a spin admixture in the impurity states. We will refer to this as the impurity spin admixture (ISA). The ISA allows for spin-flip processes in electron-hopping events even in the absence of spin-dependent potentials. A tight-binding model built on the ISA states provides a theoretical framework to study spin dynamics and spin-dependent transport in the impurity band. Since we are interested in spin lifetimes, a first test of our model is to estimate the order of magnitude that the ISA mechanism predicts. We proceed by calculating the accumulated spin rotation angle along the diffusive evolution of the electron in the potential of the impurities. The time that it takes the spin to depart an angle of unity from its initial orientation is then taken as a qualitative measure of the spin lifetime.

The spin-relaxation mechanism based on the ISA that we study here is related to the Elliot-Yafet (EY) mechanism of spin relaxation of electrons in the conduction band.[20–22] The common characteristic between them is that both are allowed by the spin admixture of the relevant electronic states, which enables spin-flip transitions mediated by spin-independent interactions. On the other hand, they differ in the sense that while the EY spin-flip mechanism involves scattering of delocalized Bloch states, with well-defined lattice momentum $\mathbf{k}$, the ISA mechanism studied here is based on the fact that spin-flip transitions occur in hopping processes between lo-

calized impurity states. The EY relaxation rate is given by the momentum relaxation rate, but as we explained above, the latter does not have a meaning in our case.

The paper is organized as follows. In Sec. II, we briefly review the MT model of shallow impurities, while in Sec. III, we generalize it by introducing our tight-binding model with the impurity spin admixture and by calculating the matrix elements with spin flip. We calculate the spin-relaxation rate from the accumulated spin rotations in Sec. IV and provide concluding remarks in Sec. V. We present as appendixes the calculation of the spin rotation angle for arbitrary initial states and the alternative derivation of the spin-relaxation rate for initially delocalized states.

II. TIGHT-BINDING MODEL FOR SHALLOW IMPURITIES

The MT model consists of a tight-binding approximation built from the ground state of the doping impurities. For shallow donors, it is a standard approximation to restrict the expansion of the impurity ground state to conduction-band states,[23] and for an impurity located at the origin, we write it as

$$[\psi_{0,\sigma}](\mathbf{r}) = \sum_{\mathbf{k}} \phi(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} [u_{\mathbf{k},\sigma}](\mathbf{r}) \approx \phi(\mathbf{r}) [u_{\sigma}^{(0)}](\mathbf{r}). \quad (1)$$

The envelope function $\phi(\mathbf{r}) = (1/\pi a^3)^{1/2} \exp(-r/a)$, where a is the effective Bohr radius ($a \approx 100$ Å for GaAs), is hydrogenlike. We denote its Fourier transform as $\phi(\mathbf{k})$, while $[u_{\mathbf{k},\sigma}](\mathbf{r})$ represents the periodic part of the Bloch functions of the conduction-band states. Its dependence on $\mathbf{k}$, being much smoother than that of $\phi(\mathbf{k})$, leads to the last relation in Eq. (1), where $[u_{\sigma}^{(0)}] = [u_{\mathbf{k}=0,\sigma}]$. The spinors $[\psi_{0,\sigma}]$ and $[u_{\mathbf{k},\sigma}]$ are trivial since they are eigenstates of the operator S_z with eigenvalues $\sigma = \pm 1$. However, this will no longer be the case once we include the spin-orbit interaction. The Hamiltonian of the MT model can be simply written as

$$H_0 = \sum_{m \neq m',\sigma} t_{mm'}^{\sigma\sigma} c_{m'\sigma}^{\dagger} c_{m\sigma}, \quad (2)$$

where $c_{m'\sigma}^{\dagger}$ ($c_{m\sigma}$) represents the creation (annihilation) of an impurity eigenstate at the impurity site m' (m). The ground-state energy of an isolated impurity is taken as the origin of energies. The energy integral for the electronic transfer from site m to m' is given by

$$t_{mm'}^{\sigma\sigma} = \sum_{p \neq m} \langle \psi_{m'\sigma} | V_p | \psi_{m\sigma} \rangle. \quad (3)$$

The Coulomb-like potential produced by the impurity placed at $\mathbf{r}_p$ is $V_p(\mathbf{r}) = -e^2/\varepsilon|\mathbf{r} - \mathbf{r}_p|$. We denote the static dielectric constant (12.9 for GaAs) by ε and the electron charge by e. Due to the exponential decay of the wave functions, the dominant term in Eq. (3) is the two-center integral corresponding to $p = m'$,

$$\langle \psi_{m'\sigma} | V_{m'} | \psi_{m\sigma} \rangle = -V_0 \left(1 + \frac{r_{mm'}}{a}\right) \exp\left(-\frac{r_{mm'}}{a}\right), \quad (4)$$

with $V_0 = e^2/\varepsilon a$ and $r_{mm'}$ the distance between the two impurities.[24] For convenience, in Eq. (3), we switched from a spinor to a ket notation. The Hamiltonian [Eq. (2)] has been thoroughly studied using a variety of analytical and numerical techniques,[17,25–27] allowing a useful description of the impurity band and its electronic transport.

III. TIGHT-BINDING MODEL WITH IMPURITY SPIN ADMIXTURE

In order to extend the MT model to the spin case, we first generalize the shallow-donor wave functions to include the spin-orbit interaction. The spin-orbit effects coming from the orbital motion do not modify in an appreciable way the envelope functions $\phi(r)$. Therefore, the spin-orbit interaction affects mainly the spinor $[u_{\mathbf{k}}]$. As is well known, in zinc-blende semiconductors such as GaAs, the spin-orbit coupling leads to spin-mixed conduction-band states at finite wave vectors. Group theoretical arguments dictate the way in which the conduction and valence states are mixed by the spin-orbit interaction. Within the $\mathbf{k}\cdot\mathbf{p}$ approximation of Kane[28] (and using the notation of Ref. 29), the periodic part of the spin-mixed conduction-band states is given by

$$|\tilde{u}_{\mathbf{k}\sigma}\rangle = |u_{\sigma}^{(0)}\rangle + \mathbf{k} \cdot |\mathbf{u}_{\sigma}^{(1)}\rangle, \quad (5)$$

where

$$|\mathbf{u}_{\sigma}^{(1)}\rangle = \alpha_1 (|\mathbf{R}\sigma\rangle + \alpha_2 \mathbf{S} \times |\mathbf{R}\sigma\rangle). \quad (6)$$

The state $|u_{\sigma}^{(0)}\rangle$ is s-like since it describes the unperturbed wave function at the Γ point. The vector $|\mathbf{R}\rangle = (|X\rangle, |Y\rangle, |Z\rangle)$ represents the three p-like valence states. $\mathbf{S}$ is the angular momentum operator. The state $|\tilde{u}_{\mathbf{k}\sigma}\rangle$ is clearly not an eigenstate of S_z. However, we still characterize it with the label σ since the mixing is small, and $\langle \tilde{u}_{\mathbf{k}\sigma} | S_z | \tilde{u}_{\mathbf{k}\sigma} \rangle$ is much closer to $\sigma\hbar/2$ than to $-\sigma\hbar/2$. The weak spin mixing is governed by the small constants $\alpha_1 = i\hbar\{(3E_G + 2\Delta)/[6m^* E_G(E_G + \Delta)]\}^{1/2}$ and $\alpha_2 = 2\Delta/i\hbar(2\Delta + 3E_G)$. We denote the spin-orbit splitting of the valence bands by Δ, the conduction-band effective mass by m^*, and the band gap by E_G.

The mixing of Eq. (5) tells us that in the presence of a spin-orbit interaction, Eq. (1) takes the form

$$[\tilde{\psi}_{0\sigma}](\mathbf{r}) = \sum_{\mathbf{k}} \phi(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} \{[u_{\sigma}^{(0)}](\mathbf{r}) + \mathbf{k} \cdot [\mathbf{u}_{\sigma}^{(1)}](\mathbf{r})\}. \quad (7)$$

Using the hydrogenic character of $\phi(r)$, the ISA state centered at $\mathbf{r}_m$ reads

$$[\tilde{\psi}_{m\sigma}](\mathbf{r}) = \phi(\mathbf{r} - \mathbf{r}_m) \left\{ [u_{\sigma}^{(0)}](\mathbf{r}) + \frac{i}{a} \frac{(\mathbf{r} - \mathbf{r}_m)}{|\mathbf{r} - \mathbf{r}_m|} \cdot [\mathbf{u}_{\sigma}^{(1)}](\mathbf{r}) \right\}. \quad (8)$$

Electron hopping between ISA states in different impurity sites provides a mechanism for spin flip by connecting the σ and $\bar{\sigma} = -\sigma$ states. The Hamiltonian of the system now

contains a term without spin flip [described by the Hamiltonian H_0 of Eq. (2)] and a spin-flip term

$$H_1 = \sum_{m \neq m', \sigma} t_{mm'}^{\sigma\bar{\sigma}} c_{m'\bar{\sigma}}^{\dagger} c_{m\sigma}. \tag{9}$$

Similar to the spin-conserving case [Eq. (3)], we have

$$t_{mm'}^{\sigma\bar{\sigma}} = \sum_{p \neq m} \langle \tilde{\psi}_{m'\bar{\sigma}} | V_p | \tilde{\psi}_{m\sigma} \rangle, \tag{10}$$

with the matrix elements

$$\langle \tilde{\psi}_{m'\bar{\sigma}} | V_p | \tilde{\psi}_{m\sigma} \rangle = C \int d^3 r \times \frac{(r - r_m)_-(z - z_{m'}) - (z - z_m)(r - r_{m'})_-}{|\mathbf{r} - \mathbf{r}_m||\mathbf{r} - \mathbf{r}_p||\mathbf{r} - \mathbf{r}_{m'}|} \times \exp[-(|\mathbf{r} - \mathbf{r}_m| + |\mathbf{r} - \mathbf{r}_{m'}|)/a]. \tag{11}$$

We have defined $C = V_0 |\alpha_1|^2 \alpha_3 / \pi a^4$, $r_\pm = x \pm iy$, and $\alpha_3 = 3\Delta(\Delta + 2E_G)/(2\Delta + 3E_G)^2$. In order to calculate the integral [Eq. (11)], we perform a rotation of coordinates from the crystallographic system (x, y, z) to a new one having the z axis along the line joining m and m'. Taking the origin at the middle point between these impurities, scaling all lengths with the distance $r_{mm'}/2$, and using dimensionless cylindrical coordinates $(\mathcal{Z}, \rho, \phi)$, we have

$$\langle \tilde{\psi}_{m'\bar{\sigma}} | V_p | \tilde{\psi}_{m\sigma} \rangle = \frac{C e^{i\varphi_{mm'}} r_{mm'}^2}{2} \int_{-\infty}^{+\infty} d\mathcal{Z} \int_0^{\infty} d\rho \int_0^{2\pi} d\phi \frac{\rho^2(\cos\phi + i \cos\theta_{mm'} \sin\phi)}{[\rho^2 + \rho_p^2 + (\mathcal{Z} - \mathcal{Z}_p)^2 - 2\rho\rho_p \cos(\phi - \phi_p)]^{1/2}} \times \frac{\exp\{-r_{mm'}[\sqrt{\rho^2 + (\mathcal{Z} - 1)^2} + \sqrt{\rho^2 + (\mathcal{Z} + 1)^2}]/2a\}}{\sqrt{\rho^2 + (\mathcal{Z} - 1)^2}\sqrt{\rho^2 + (\mathcal{Z} + 1)^2}}. \tag{12}$$

$\varphi_{mm'}$ and $\theta_{mm'}$ are the polar angles of the vector $\mathbf{r}_{mm'}$ in the original coordinate system. $(\mathcal{Z}_p, \rho_p, \phi_p)$ are the cylindrical coordinates of $\mathbf{r}_p$ in the new coordinate system.

As in the spin-conserving model, we start with the case where $p = m'$. The corresponding two-center integral [analogous to [Eq. (4)] is obtained by putting $\mathcal{Z}_p = 1$, $\rho_p = 0$ in Eq. (12). However, $\langle \tilde{\psi}_{m'\bar{\sigma}} | V_{m'} | \tilde{\psi}_{m\sigma} \rangle = 0$ due to the symmetry of the angular integral. This important fact is ultimately responsible for the large values of the spin lifetime in the regime of impurity-band conduction. The leading order effect will then be given by the three-center integrals corresponding to $p \neq m, m'$, which are, in general, very difficult to calculate in a closed form.[30] The angular integral can be performed in terms of elliptic functions, but since only the small arguments of the latter are relevant for the remaining integrals,[31] we can write

$$\langle \tilde{\psi}_{m'\bar{\sigma}} | V_p | \tilde{\psi}_{m\sigma} \rangle = \frac{C e^{i\varphi_{mm'}} \pi r_{mm'}^2}{4} \rho_p (\cos\phi_p + i \cos\theta_{mm'} \sin\phi_p) \times \int_{-\infty}^{+\infty} d\mathcal{Z} \int_0^{\infty} d\rho \left[\frac{\rho^2}{\rho^2 + \rho_p^2 + (\mathcal{Z} - \mathcal{Z}_p)^2} \right]^{3/2} \frac{\exp\{-r_{mm'}[\sqrt{\rho^2 + (\mathcal{Z} - 1)^2} + \sqrt{\rho^2 + (\mathcal{Z} + 1)^2}]/2a\}}{\sqrt{\rho^2 + (\mathcal{Z} - 1)^2}\sqrt{\rho^2 + (\mathcal{Z} + 1)^2}}. \tag{13}$$

In the limit $r_{mm'}/a \gg 1$,[32] we can perform the integrals using the saddle-point approximation and obtain

$$\langle \tilde{\psi}_{m'\bar{\sigma}} | V_p | \tilde{\psi}_{m\sigma} \rangle = 4.2 C e^{i\varphi_{mm'}} (\cos\phi_p + i \cos\theta_{mm'} \sin\phi_p) \times \frac{r_{mm'}^{3/2} a^{1/2} \rho_p \exp(-r_{mm'}/a)}{[1 + (\rho_p^2 + \mathcal{Z}_p^2) r_{mm'}/a]^{3/2}}. \tag{14}$$

For a given impurity configuration, the matrix elements [Eq. (14)] determine the hopping integral [Eq. (10)] (with the terms $p = m, m'$ excluded) of the Hamiltonian H_1.[33] The system Hamiltonian $H_0 + H_1$ can be addressed numerically or by perturbation theory. In order to test the physical relevance of the proposed spin-flip mechanism, we will estimate the spin-relaxation time within some simplifying hypotheses that we discuss in what follows.

IV. SPIN-RELAXATION TIME: DIFFUSIVE APPROACH

Viewing the electron transport as a hop between impurity sites, we see that, since $|t_{mm'}^{\sigma\sigma}| \gg |t_{mm'}^{\sigma\bar{\sigma}}|$, there is a very small probability of spin-flip per hop, which may be translated into a mean spin-rotation angle $\gamma_{mm'}$. Assuming that these relative rotations are accumulated in a diffusive way, we can estimate the spin-relaxation time by

$$\frac{1}{\tau_s} = \frac{2}{3}\frac{\langle \gamma^2 \rangle}{\tau_c}, \tag{15}$$

where $\langle \gamma^2 \rangle$ is the ensemble average of $\gamma^2_{mm'}$ and τ_c is the mean hopping time. In Appendix A, we show that in the electron hop between impurities m and m', the spin typically rotates by an angle

$$\gamma_{mm'} = \sqrt{\frac{15}{2}}\frac{|t^{\sigma\bar{\sigma}}_{mm'}|}{|t^{\sigma\sigma}_{mm'}|}. \tag{16}$$

Given the form of Eq. (10) of the energy integrals, the typical rotation angle $\gamma^2_{mm'}$ involves a double sum over impurities p and p' ($\neq m,m'$), which can be approximated by its impurity average. Only the diagonal term ($p=p'$) survives the averaging, and we can write

$$\gamma^2_{mm'} \approx 27.7\left(\frac{C}{V_0}\right)^2 r^{3/2}_{mm'} a^{11/2} n_i (1+\cos^2\theta_{mm'}), \tag{17}$$

where n_i is the impurity density. The typical rotation angle between impurities m and m' increases with their distance $r_{mm'}$ as a power law. This dependence renders the impurity distribution crucial for the determination of the mean square rotation angle per hop. The distribution of doping impurities is known to be completely random and to lack hard-core repulsive effects on the scale of a [34]. Since the probability of jumping from a given impurity m to a second one m' is $|t^{\sigma\sigma}_{mm'}|^2/\Sigma_{m'}|t^{\sigma\sigma}_{mm'}|^2$, we obtain the impurity average of the typical rotation angle per hop as

$$\langle \gamma^2 \rangle \approx 1.5 \times 10^2 \left(\frac{C}{V_0}\right)^2 a^4 (n_i a^3). \tag{18}$$

The hopping time can be estimated from the perturbation theory by determining the characteristic time for the decay of the initial population by one-half, yielding

$$\frac{1}{\tau_c} = \frac{1}{\hbar}\sqrt{2\sum_{m'}|t^{\sigma\sigma}_{mm'}|^2}, \tag{19}$$

where an average over the initial position m is implicit. This estimation assumes orthogonality of the electronic orbitals at different impurity sites, which is actually not satisfied by the hydrogenic states. However, since the overlaps are very small, the nonorthogonality effects arising in the MT model are known to be small.[25,35]

From Eqs. (15), (18), and (19), we obtain

$$\frac{1}{\tau_s} \approx 6.8 \times 10^2 \frac{C^2 a^4}{V_0 \hbar}(n_i a^3)^{3/2}. \tag{20}$$

For GaAs, at the density of the MIT, Eq. (20) yields a spin-relaxation time of $\tau_s \approx 1400$ ns.

This value is larger but within an order of magnitude of the experimentally reported result of 200 ns (Refs. 6 and 7) at the MIT. We stress that our result does not depend on any adjustable parameter, but it relies on a few approximations. For example, a step of the calculation that could admit an alternative treatment is the impurity average of $\gamma^2_{mm'}$. If we assume that the hopping takes place only between nearest neighbors (separated by a typical distance $n_i^{-1/3}$), we obtain a value of 840 ns for τ_s at the MIT. In general, the order of magnitude agreement with the experimental value is not affected by the approximations. Therefore, the ISA mechanism needs to be included in the description of spin relaxation in the impurity band.

In materials with stronger spin-orbit interactions such as InSb and InAs, one obtains considerably smaller values of the spin lifetime. For the impurity density of 5×10^{14} cm^{-3} in InSb, our estimate yields a spin lifetime of 100 ns. This value is within an order of magnitude of the experimental result and close to the theoretical estimation of Ref. 29. The theoretical approach of this reference is justified only in the high-concentration limit since it describes single scattering of plane waves and treats electron-electron interactions through a corrective factor.

The approach presented in this section assumes a relatively narrow wave packet as the orbital part of the initial state. However, this is not an important restriction. In Appendix B, we show that under some simplifying hypotheses, assuming an initially delocalized state in the impurity band results in an equivalent expression for τ_s, up to a prefactor which is of order 1.

V. CONCLUSION

In this work, we have proposed a mechanism for spin relaxation in the regime where electron conduction occurs in the impurity band of doped semiconductors. The mechanism is based on the impurity spin admixture of the electronic ground state of the donors caused by the spin-orbit interaction. The impurity spin admixture states do not have a well-defined spin projection about a fixed spatial direction. Therefore, hopping between two of these states may connect different projections of the angular momentum.

Unlike the spin-conserving case, the matrix elements of the spin-flip hops are not dominated by a two-center integral, where the impurity potential corresponds to one of the extreme sites. We therefore have to consider three-center integrals, where the impurity potential is not centered around any of the two sites of the hop. Since the latter matrix elements are considerably reduced with respect to the former, the resulting spin lifetimes are very large.

Our calculation of spin-flip matrix elements yields a suitable model for studying electron and spin transfer in the regime of impurity concentrations just above that of the metal-insulator transition. Various treatments can be applied to our model Hamiltonian, including a calculation of the diffusive accumulation of spin rotation during the hopping process.

Our estimation of the spin lifetime results in values that are larger than the ones experimentally measured, but within the right order of magnitude. Therefore, the impurity spin admixture mechanism has to be taken into account in descriptions of the spin relaxation in the impurity band. Our model admits generalizations including other physical effects, such as doping compensation, electron-electron interaction, and a second electronic band, which may improve the agreement between theory and experiment.

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APPENDIX A: SPIN ROTATION ANGLE FROM SPIN-DEPENDENT HOPPING INTEGRALS

In this appendix we derive expression (16) for the typical spin rotation angle $\gamma_{mm'}$ used in Sec. IV and characterize the hopping process of an electron from site m to site m' of the effective tight-binding model. The spin rotation arises due to the presence of the spin-flip term H_1 of Eq. (9) in the Hamiltonian. Let us consider as initial state $|m\sigma\rangle$, describing an electron with spin up ($\sigma=+1$) on site m. If we apply the hopping matrix elements and perform a partial projection of final state on site m' without specifying the spin part of the final state, we get the spin part of the final state on site m' as

$$|s_{m'}\rangle = \langle m'|(H_0+H_1)|m\sigma\rangle = t_{mm'}^{\sigma\sigma}|\sigma\rangle + t_{mm'}^{\sigma\bar{\sigma}}|\bar{\sigma}\rangle, \quad \text{(A1)}$$

where $\bar{\sigma}=-1$. The corresponding spin orientation is given by the expectation value of the angular momentum

$$\langle \mathbf{S}_{m'}\rangle = \frac{\langle s_{m'}|\mathbf{S}|s_{m'}\rangle}{\langle s_{m'}|s_{m'}\rangle}. \quad \text{(A2)}$$

This leads to

$$\langle \mathbf{S}_{m'}\rangle = \frac{\hbar/2}{|t_{mm'}^{\sigma\sigma}|^2+|t_{mm'}^{\sigma\bar{\sigma}}|^2}\begin{pmatrix} 2\,\mathrm{Re}[t_{mm'}^{\sigma\sigma}t_{mm'}^{\sigma\bar{\sigma}*}] \\ 2\,\mathrm{Im}[t_{mm'}^{\sigma\sigma}t_{mm'}^{\sigma\bar{\sigma}*}] \\ |t_{mm'}^{\sigma\sigma}|^2-|t_{mm'}^{\sigma\bar{\sigma}}|^2 \end{pmatrix}, \quad \text{(A3)}$$

such that the angle with respect to the initial orientation (0,0,1) is given by

$$\phi_{mm'}^{(+1)} = \arccos\left(\frac{|t_{mm'}^{\sigma\sigma}|^2-|t_{mm'}^{\sigma\bar{\sigma}}|^2}{|t_{mm'}^{\sigma\sigma}|^2+|t_{mm'}^{\sigma\bar{\sigma}}|^2}\right). \quad \text{(A4)}$$

For our case of spin-flip hopping matrix elements, which are much smaller than the elements which conserve the spin, we can expand the above exact result for small rotation angles ($|t_{mm'}^{\sigma\bar{\sigma}}|^2 \ll |t_{mm'}^{\sigma\sigma}|^2$), yielding

$$\phi_{mm'}^{(+1)} = \frac{2|t_{mm'}^{\sigma\bar{\sigma}}|}{|t_{mm'}^{\sigma\sigma}|}. \quad \text{(A5)}$$

It is straightforward to calculate the spin rotation angle for a general spin state $|s_m\rangle=\cos\,\xi|\sigma\rangle+e^{i\eta}\sin\,\xi|\bar{\sigma}\rangle$ on the initial site m. The rotation angle $\phi_{mm'}^{(\xi,\eta)}$ is now given by

$$\cos(\phi_{mm'}^{(\xi,\eta)}) = 1 - \frac{2|t_{mm'}^{\sigma\bar{\sigma}}|^2[1+\sin^2(2\xi)\cos^2(\eta)]}{|t_{mm'}^{\sigma\sigma}|^2+|t_{mm'}^{\sigma\bar{\sigma}}|^2+2\sin(2\xi)\cos(\eta)\mathrm{Re}[(t_{mm'}^{\sigma\sigma})^* t_{mm'}^{\sigma\bar{\sigma}}]}. \quad \text{(A6)}$$

As in the special case above, in the limit of small rotation angle, the above expression simplifies, and we have

$$\phi_{mm'}^{(\xi,\eta)} = 2\sqrt{1+\sin^2(2\xi)\cos^2(\eta)}\frac{|t_{mm'}^{\sigma\bar{\sigma}}|}{|t_{mm'}^{\sigma\sigma}|}. \quad \text{(A7)}$$

If we perform a root mean square average over the initial conditions (parametrized by the angles ξ and η), we get

$$\sqrt{\langle\phi_{mm'}^2\rangle} = \frac{\sqrt{5}|t_{mm'}^{\sigma\bar{\sigma}}|}{|t_{mm'}^{\sigma\sigma}|}, \quad \text{(A8)}$$

which has an additional prefactor of $\sqrt{5}/2$ with respect to Eq. (A5). Since the calculated angle $\phi_{mm'}$ with respect to the initial spin orientation contains the two components of $\gamma_{mm'}$ that are relevant for spin relaxation, one has

$$\langle\gamma_{mm'}^2\rangle = \frac{3}{2}\langle\phi_{mm'}^2\rangle, \quad \text{(A9)}$$

and we finally obtain the expression given in Eq. (16) of Sec. IV.

APPENDIX B: SPIN-RELAXATION TIME: GOLDEN RULE APPROACH FOR EXTENDED INITIAL STATES

In this appendix we use the Fermi golden rule to calculate the spin decay rate of a spin-polarized extended initial state $|i\sigma\rangle$. Such a state will be a superposition of the eigenstates of H_0 polarized with spin σ, which we denote as $|\alpha\sigma\rangle$. The presence of the spin-flip term H_1 of Eq. (9) leads to a mixing of the spin states. Fermi's golden rule gives the spin decay rate,

$$\frac{1}{\tau_s} = \frac{2\pi}{\hbar}\rho_{\bar{\sigma}}|M|^2, \quad \text{(B1)}$$

of $|i\sigma\rangle$ into the spin-reversed states $|\alpha\bar{\sigma}\rangle$ with $\bar{\sigma}=-\sigma$, where $\rho_{\bar{\sigma}}$ is the density of states with spin $\bar{\sigma}$ and $|M|^2$ is the typical value for the absolute square of the matrix elements $M_\alpha=\langle\alpha\bar{\sigma}|H_1|i\sigma\rangle$ of the spin-flip term H_1 between the initial state with spin σ and spin-reversed states $|\alpha\bar{\sigma}\rangle$ having the same energy. With the projections $i_m=\langle m\sigma|i\sigma\rangle$ and $\alpha_m=\langle m\bar{\sigma}|\alpha\bar{\sigma}\rangle$ onto on-site basis states $|m\sigma\rangle$ and $|m\bar{\sigma}\rangle$, respectively, we can write

$$|M_\alpha|^2 = \sum_{m,m',m'',m'''} t_{mm'}^{\sigma\bar{\sigma}}(t_{m''m'''}^{\sigma\bar{\sigma}})^* i_m\alpha_{m'}^* i_{m''}^*\alpha_{m'''}. \quad \text{(B2)}$$

For generic wave functions of a disordered system on the metallic side of the metal-insulator transition, one can expect the phases to be fluctuating with the site index m. The four-fold sum will therefore be dominated by the diagonal terms with $m=m''$ and $m'=m'''$, and one gets

$$|M_\alpha|^2 \approx \sum_{m,m'} |t^{\sigma\bar{\sigma}}_{mm'}|^2 |i_m|^2 |\alpha_{m'}|^2. \tag{B3}$$

In the metallic regime, the wave functions are delocalized over all impurity sites. We assume an approximately homogeneous density $|\alpha_m|^2=|i_m|^2=1/N_i$, where N_i is the number of impurity sites. Neglecting the spin-flip matrix elements between distant sites m and m' as compared to the matrix elements $t^{\sigma\bar{\sigma}}_{\text{n.n.}}$ between m and its Z nearest neighbors, we obtain

$$|M|^2 \simeq \frac{Z}{N_i} |t^{\sigma\bar{\sigma}}_{\text{n.n.}}|^2. \tag{B4}$$

With the estimate for the density of states $\rho_{\bar{\sigma}} \simeq N_i/W$, where $W \simeq Z|t^{\sigma\sigma}_{\text{n.n.}}|$ is the width of the impurity band, we finally get from Eq. (B1) the spin decay rate,

$$\frac{1}{\tau_s} \simeq \frac{2\pi}{\hbar} \frac{|t^{\sigma\bar{\sigma}}_{\text{n.n.}}|^2}{|t^{\sigma\sigma}_{\text{n.n.}}|}. \tag{B5}$$

From the diffusive approach presented in Sec. IV, one can determine the spin lifetime using Eqs. (15), (16), and (19). Assuming that the hopping processes between nearest neighbors dominate as compared to hopping elements between more distant sites, one obtains the estimate

$$\frac{1}{\tau_s} \simeq \frac{5\sqrt{2Z}}{\hbar} \frac{|t^{\sigma\bar{\sigma}}_{\text{n.n.}}|^2}{|t^{\sigma\sigma}_{\text{n.n.}}|}, \tag{B6}$$

which agrees with the result of the Fermi golden rule approach Eq. (B5) up to a numerical factor of order 1.

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