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Spin relaxation of a diffusively moving carrier in a random hyperfine field

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Relaxation, $\langle S_z(t)\rangle$, of the average spin of a carrier in a course of hops over sites hosting random hyperfine fields is studied theoretically. In low dimensions, $d = 1,2$, the decay of average spin with time is nonexponential at all times. The origin of the effect is that for $d = 1,2$ a typical random-walk trajectory exhibits numerous self-intersections. Multiple visits of the carrier to the same site accelerates the relaxation since the corresponding partial rotations of spin during these visits add up. Another consequence of self-intersections of the random-walk trajectories is that, in all dimensions, the average, $\langle S_z(t)\rangle$, becomes sensitive to a weak magnetic field directed along z. Our analytical predictions are complemented by the numerical simulations of $\langle S_z(t)\rangle$. The scenario of acceleration of spin relaxation due to returns applies also to the non-Markovian decoherence of a qubit surrounded by multiple fluctuators.

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Introduction. It is known for more than four decades that in crystalline semiconductor lacking inversion symmetry, the spin relaxation time is determined by spin-orbit coupling and is given by $\tau_s^{-1} = \langle \Omega_{\boldsymbol{k}}^2\rangle\tau$ [1], where $\Omega_{\boldsymbol{k}}$ is the precession frequency around the spin-orbit field, which depends on electron wave vector $\boldsymbol{k}$, and τ is the scattering time. Consider now the situation when the spin-orbit coupling is negligible, and the electron mobility is very low, so that the transport can be viewed as random inelastic hops of carriers between the sites. Then the spin relaxation is due to spin precession in random hyperfine fields of nuclei surrounding the sites, and the Dyakonov-Perel expression [1] takes the form $\tau_s^{-1} = b_0^2\tau$, where b_0 is the rms hyperfine field, and τ is the waiting time for a hop. The above situation is germane to the carbon-based organic semiconductors [2], which have low mobility and weak spin-orbit coupling. On the other hand, spin relaxation in these material is an issue of practical importance, since these materials are considered as promising candidates for spin-valve devices [3].

Naturally, for long τ_s, a typical partial rotation of spin, $\delta\varphi = b_0\tau$, during the waiting time is weak, $\delta\varphi \ll 1$. Assuming that all partial rotations are completely uncorrelated, the spin polarization, averaged over realizations of the hyperfine fields, falls off with the number of hops, N, as $\langle S_z(N)\rangle = S_z(0)\exp(-N\delta\varphi^2)$. This suggests that the evolution of $\langle S_z\rangle$ with time $t = N\tau$ is a simple exponent

$$\langle S_z(t)\rangle = S(0)\exp\left(-\frac{t}{\tau_s}\right). \tag{1}$$

The main message of the present Rapid Communication is that the random walk of a carrier over the sites induces the correlation in hyperfine fields "sensed" by the carrier spin. This correlation modifies the decay law, Eq. (1). The origin of correlation is the self-intersections of the random-walk trajectories (see Fig. 1). These self-intersections imply multiple visits of the carrier to the *same* site. Then the corresponding partial rotations *add up* which leads to acceleration of the spin relaxation. The effect is most dramatic if the carrier moves in one dimension. Then, in the course of N hops, the carrier visits $N^{1/2}$ sites, and the number of visits to a given site is also $N^{1/2}$. The N dependence of $\langle S_z\rangle$ can be found from the above derivation of Eq. (1) upon replacement $N \to N^{1/2}$ and $\delta\varphi \to N^{1/2}\delta\varphi$. This yields $\langle S_z(N)\rangle = S_z(0)\exp\left(-N^{3/2}\delta\varphi^2\right)$, and, correspondingly, the time dependence

$$\langle S_z(t)\rangle = S(0)\exp\left(-\frac{t^{3/2}}{\tau^{1/2}\tau_s}\right). \tag{2}$$

In higher dimensions, $d = 2$ and $d = 3$, the number of self-crossings of an N-step random-walk trajectory is $\sim N$ and $\sim N^{1/2}$, respectively, i.e., each site is visited twice with probability ~ 1 for $d = 2$, and with probability $N^{-1/2}$ for $d = 3$. As a result, the change, $\langle \delta S_z(t)\rangle$, of the decay law, Eq. (1), due to accumulation of the partial rotations is of the order of $\langle S_z(t)\rangle$ for $d = 2$ and of the order of $(\tau/t)^{1/2}\langle S_z(t)\rangle$ for $d = 3$. But even in the latter case the correction to Eq. (1) can be important since it induces a sensitivity of $\langle S_z(t)\rangle$ to a *weak* external magnetic field directed along z. Recall that, without self-intersections, the B dependence of τ_s is given by the Hanle-type expression $\tau_s = \frac{1+B^2\tau^2}{b_0^2\tau}$, which applies for $B \gg b_0$ and predicts that sensitivity to B emerges at $B \sim \tau^{-1} \gg b_0$. We will demonstrate that, with self-crossings of the random-walk trajectories taken into account, the sensitivity to B develops at much smaller field $B \sim (\tau\tau_s^2)^{-1/3} \ll \tau^{-1}$ in one dimension and at $B \sim \tau_s^{-1} \ll \tau^{-1}$ for $d = 2$ and $d = 3$. Remarkably, the returns to the same site after a long time, t, give rise to the *oscillatory* correction $\propto \cos Bt$ to $\langle S_z(t)\rangle$, which is most pronounced for $d = 1$.

Diagrammatic expansion. To illustrate our main message, consider first a simplified situation, when the hyperfine field is located in the x,y plane. Moreover, we will assume that the randomness in the in-plane field, $b_\perp = (b_x,b_y)$, is exclusively due to randomness in the azimuthal angle ϕ, i.e., $b_x = b_0\cos\phi$, $b_y = b_0\sin\phi$ (see Fig. 1).

The spin operator satisfies the equation of motion $i\frac{d\widehat{S}}{dt} = [\widehat{S},\widehat{H}]$, with Hamiltonian $\widehat{H} = \widehat{S}\cdot\boldsymbol{b}(t)$. Excluding the in-plane components of the operator $\widehat{S}$, the equation of motion for S_z takes the form

$$S_z(t) = 1 - b_0^2\int_0^t dt_1\int_0^{t_1} dt_2\,\cos\left[\phi(t_1)-\phi(t_2)\right]S_z(t_2). \tag{3}$$

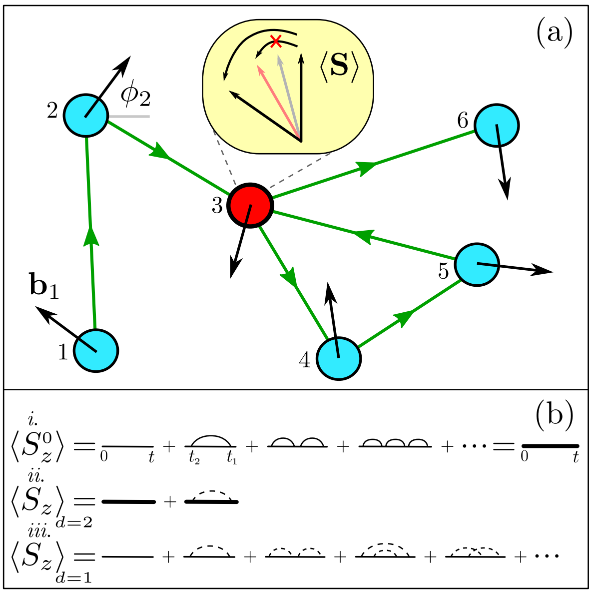

FIG. 1. (Color online) (a) In the course of diffusion $1 \to 2 \to 3 \to 4 \to 5 \to 3 \to 6$ over sites hosting random hyperfine fields (black arrows) a carrier visits site 3 *twice*. As a result, the partial spin rotation doubles (see enlargement). (b) (i) For a trajectory without self-intersections $\langle S_z(t)\rangle$ is given by a sequence of *nonintersecting* solid arcs encoding the correlator C_0. (ii) Graphical representation of Eq. (9) for the $d=2$ spin relaxation; self-intersections are captured by a single dashed arc encoding the correlator C_D. (iii) Spin relaxation for $d=1$ is described by diffusive diagrams *only*.

To find the time evolution of the average, $\langle S_z(t)\rangle$, it is necessary to iterate Eq. (3) as

$$S_z(t) = 1 - b_0^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \cos[\phi(t_1) - \phi(t_2)] + b_0^4 \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 \cos[\phi(t_1) - \phi(t_2)] \times \cos[\phi(t_3) - \phi(t_4)] - \cdots \quad (4)$$

and perform averaging over the random azimuthal angle, $\phi(t)$. Without self-intersections of the random-walk trajectories, this averaging is straightforward since the angles $\phi(t), \phi(t')$ are correlated only for $|t - t'| \lesssim \tau \ll t$, i.e.,

$$\langle \cos[\phi(t) - \phi(t')]\rangle = \exp[-|t - t'|/\tau] = C_0(t,t'). \quad (5)$$

The exponential character of C_0 expresses the Poisson distribution of the waiting times.

Each term of the expansion, Eq. (4), can be graphically expressed as a diagram (see Fig. 1). Because of the short-time decay of C_0, the arcs corresponding to C_0 terms are not allowed to cross. More precisely, each crossing of arcs gives rise to a small factor $\tau/t \ll 1$. On the other hand, averaging of each term with n nonintersecting arcs yields $(-1)^n (b_0\tau)^{2n}/n!$, and we restore Eq. (1).

As a consequence of self-intersections of the random-walk path, the difference $[\phi(t) - \phi(t')]$ can be small even if the moments t and t' are well separated in time. Quantitatively, this is captured by the diffusive contribution, C_D, to the correlator

$$\langle \cos[\phi(t) - \phi(t')]\rangle = \left[\frac{1}{2\pi D|t - t'|}\right]^{d/2} = C_D(t,t'), \quad (6)$$

where the diffusion coefficient D is $1/\tau$ assuming that the separation between neighboring sites is unity. In Eq. (6), self-intersections are accounted for in the continuous limit as a probability to return to the origin after moving diffusively for a time $t' - t$.

The correlator C_D should also be incorporated into the diagrammatic expansion; we denote it with dashed arcs (see Fig. 1). For example, the diagram involving only one dashed arc is given by

$$\lambda_d(t) = b_0^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \left[\frac{1}{2\pi D\,|t_1 - t_2|}\right]^{d/2}. \quad (7)$$

Evaluation of the double integral yields

$$\lambda_d(t) = \begin{cases} \frac{4}{3(2\pi)^{1/2}} \frac{t^{3/2}}{\sqrt{\tau}\tau_s}, & d = 1 \\ \frac{1}{2\pi\tau_s}\left[t \ln\left(\frac{t}{\tau}\right)\right], & d = 2 \\ \frac{2\tau^{1/2}}{(2\pi)^{3/2}\tau_s}\left(\frac{t}{\sqrt{\tau}} - 2\sqrt{t}\right), & d = 3, \end{cases} \quad (8)$$

where we have expressed b_0^2 in terms of τ_s and have taken into account that the diffusive description applies when $t_1 - t_2 \gtrsim \tau$. The double integral, Eq. (7), converges for $d = 1$ and the result, Eq. (8), confirms the qualitative argument given in the Introduction. Namely, the averaged expansion, Eq. (4), becomes a series in the dimensionless combination $b_0^2 t^{3/2}\tau^{1/2}$. Note also, that for $d = 1$ the diffusive contribution, Eq. (7), exceeds by $(t/\tau)^{1/2}$ the contribution coming from a single solid arc. This illustrates the fact that each site is visited many times in the course of a $d = 1$ random walk.

In two dimensions, the contribution $\lambda_2(t)$, to $\langle S_z(t)\rangle$ from a dashed arc exceeds logarithmically the contribution from one solid arc. On the other hand, this contribution contains a prefactor $(2\pi)^{-1}$. We will take advantage of the smallness of this prefactor and sum up *all* diagrams containing only zero or one dashed arc, as illustrated in Fig. 1(b). The most delicate ingredient of this procedure is that the insertion of solid arcs under a dashed arc amounts to the replacement $C_D(t_1,t_2) \to C_D(t_1,t_2)\exp[-\frac{(t_2-t_1)}{2\tau_s}]$. Physically, this means that between the two subsequent visits to the same site at time moments t_1 and t_2, the spin polarization is "forgotten" in the course of many short-time hops. The emergence of the nontrivial factor 1/2 in the exponent is demonstrated in the Supplemental Material [4] where we also show that the presence of a z component of the hyperfine field amounts to the replacement $C_D(t_1,t_2) \to C_D(t_1,t_2)\exp[-\frac{3(t_1-t_2)}{4\tau_s}]$.

For planar hyperfine fields the resulting expression for $\langle S_z(t)\rangle$, which is shown graphically in Fig. 1(b), takes the form

$$\langle S_z(t)\rangle = e^{-t/\tau_s} - \frac{g_2}{2\pi\tau_s} e^{-t/\tau_s} \int_0^t dt_1 \int_\tau^{t_1} dt_2 \frac{\exp\left[-\frac{(t_2-t_1)}{2\tau_s}\right]}{t_1 - t_2}, \quad (9)$$

where the numerical factor g_2 should be 1, but is retained intentionally for future comparison with numerics. The second term is responsible for the deviation from a simple exponential decay. This term can be easily reduced to a single integral, and we get

$$\langle S_z(t)\rangle = e^{-t/\tau_s}\left[1-\frac{g_2}{\pi}\int_{\tau/2\tau_s}^{t/2\tau_s}\frac{dw}{w}\left(\frac{t}{2\tau_s}-w\right)e^{w}\right]. \quad (10)$$

For small $t \ll \tau_s$, Eq. (10) yields the correction, $-\frac{g_2}{2\pi\tau_s}t\ln(t/\tau)$, to a simple exponent which reproduces Eq. (8). In the limit $t \gg \tau_s$ the correction takes the form $\frac{g_2}{\pi}(\frac{\exp(-t/2\tau_s)}{(t/2\tau_s)})$. In fact, this asymptote applies already at $t > \tau_s$. It decays slower than $\exp(-t/\tau_s)$, so that $\langle S_z(t)\rangle$ should exhibit a sign reversal followed by a minimum. Our numerics, see below, show that this minimum is very shallow.

Turning now to $d = 3$, we find that the first dominant term in Eq. (8) describes the contribution from short times and essentially renormalizes τ_s. The second subleading term comes from long diffusive trajectories. It yields a correction to $\langle S_z(t)\rangle$ which is small as $(\frac{t}{\tau_s})^{1/2}$ at $t \ll \tau_s$ and as $(\frac{\tau_s}{t})^{3/2}\exp(\frac{-t}{2\tau_s})$ at $t \gg \tau_s$. The importance of this correction is that it causes a sensitivity of $\langle S_z(t)\rangle$ to a weak external magnetic field, as we show below.

With the magnetic field along the z axis. Incorporating the constant, $\boldsymbol{B} = z_0 B$, and random, $b_z(t)$, components of the magnetic field amounts to the replacement

$$[\phi(t_1)-\phi(t_2)]\to\left[\phi(t_1)-\phi(t_2)+B(t_1-t_2)+\int_{t_1}^{t_2}dt' b_z(t')\right] \quad (11)$$

in Eq. (4). As discussed in the Introduction, the solid-arc diagrams describing the hops to nearest neighbors during the time intervals $\sim\tau$ develop the sensitivity to B only for strong $B \sim \tau^{-1}$. On the other hand, the dashed-arc diagrams are defined by much longer times, and are thus sensitive to much weaker B. Below, we demonstrate [4] that the B-dependent correction has the form $tB^{-1/2}$, $t\ \ln Bt$, and $tB^{1/2}$ for $d = 1$, 2, and 3, respectively. In addition, these corrections [4] *develop oscillations*.

Numerical results. We simulated the spin evolution numerically using the discrete version of the equation of motion

$$\begin{aligned}\boldsymbol{S}_i = {} & [\boldsymbol{S}_{i-1} - \boldsymbol{n}_i(\boldsymbol{n}_i\cdot\boldsymbol{S}_{i-1})]\cos b_0\tau \\ & + (\boldsymbol{n}_i\times\boldsymbol{S}_{i-1})\sin b_0\tau + \boldsymbol{n}_i(\boldsymbol{n}_i\cdot\boldsymbol{S}_{i-1}), \end{aligned}\quad (12)$$

so that the local hyperfine field had the same magnitude, b_0, on all sites, while the directions, $\boldsymbol{n}_i$, were defined by either a random azimuthal angle, ϕ_i, or by two spherical angles, ϕ_i and θ_i. The diffusive motion of a carrier was simulated by randomly choosing $\boldsymbol{n}_i$ at the next step from one of the nearest neighbors of $\boldsymbol{n}_i$ at the previous step.

Our numerical results are shown in Figs. 2 and 3. We started by verifying that, for a directed walk, when *all* $\boldsymbol{n}_i$ are uncorrelated, $\langle S_z(i)\rangle$ decays as a simple exponent. It is seen from Fig. 2 that, upon allowing self-intersections, the numerical curve $\langle S_z(t)\rangle$ drops below the result for uncorrelated $\boldsymbol{n}_i$ after several steps. For a spherical hyperfine field, $\ln\langle S_z(t)\rangle$ remains essentially linear at large t, but with bigger slope, i.e., the evolution of $\langle S_z(t)\rangle$ exhibits a crossover from one simple exponent at short times to another simple exponent at long times. By contrast, for planar hyperfine field, $\langle S_z(t)\rangle$

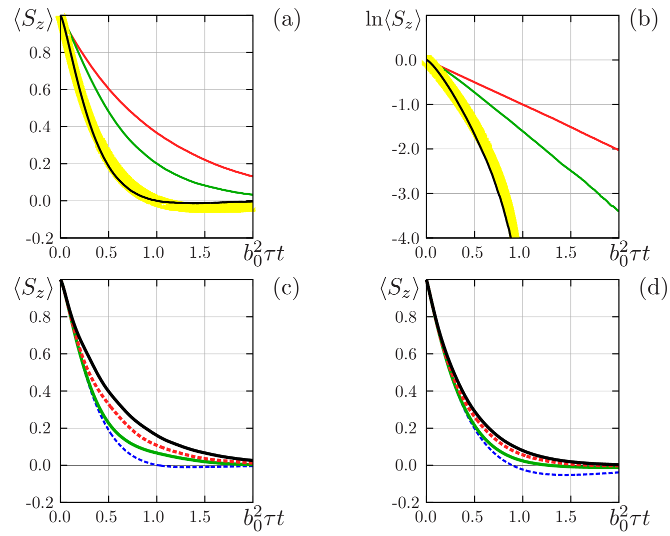

FIG. 2. (Color online) (a) $d = 2$ spin relaxation for uncorrelated (no self-crossings) hyperfine fields $\boldsymbol{b}_i$ (red), with self-intersections and spherically distributed $\boldsymbol{b}_i$ (green), and with self-intersections and planar $\boldsymbol{b}_i$ (black). (b) Same as (a) but in logarithmic scale. The decay $\langle S_z(t)\rangle$ is a simple exponent (red), shows crossover between two simple exponents (green), strongly nonexponential (black). The yellow line is plotted from Eq. (9) with $g_2 = 0.75$. (c) and (d) Weak external field $B \sim \tau_s^{-1}$ suppresses the effect of self-intersections. Numerical (c) and analytical (d) results illustrate how a simple-exponent decay is restored upon increasing $B\tau_s$. Results for $B = 0$ (blue), $B\tau_s = 2$ (green), $B\tau_s = 5$ (red), and $B\tau_s = 10$ (black) are shown.

is strongly nonlinear in the logarithm scale at all times. This completely nonexponential decay is very well described by Eq. (10) with $g_2 = 0.75$ instead of 1. As we argued above, self-intersections give rise to the sensitivity of the spin relaxation to magnetic field $B \sim \frac{1}{\tau_s} \ll \frac{1}{\tau}$. Evolution of the numerical curves with B is shown in Fig. 2. A significant slowing down of the relaxation starts from $B \sim \frac{5}{\tau_s}$. We have also plotted an analytical dependence of $\langle S_z(t)\rangle$ obtained by introducing $\cos Bt(x_1 - x_2)$ into the integrand of Eq. (9). Qualitatively, the numerical and analytical curves exhibit similar behavior.

Numerical results for random walk in one dimension are shown in Fig. 3. First, we established that these numerical results perfectly satisfy the scaling relation predicted from the qualitative reasoning. Namely, when plotted versus $t^{3/2}b_0^2\tau^{1/2}$, they all fall on a single curve. We also see that the empirical prediction, Eq. (2), does not apply. In fact, for purely planar hyperfine field, the numerical curve, $\langle S_z(t)\rangle$, drops to a *negative* value $\langle S_z\rangle \approx -0.16$ before approaching zero. As we explained above, only the dashed arcs are responsible for the spin relaxation for $d = 1$. Therefore, capturing the nontrivial decay of $\langle S_z\rangle$ analytically, requires summation of at least a part of dashed-arc diagrams to *all* orders.

In the Supplemental Material we present two variants of such summation. They essentially reduce to exponentiating of one-dashed-arc contribution, $\lambda_1(t)$, Eq. (7), and differ by the way the numerical factors in the diagrams with crossings are counted. Two ways of approximate counting yield $\langle S_z(t)\rangle = \exp[-\lambda_1(t)]$ and $\langle S_z(t)\rangle = 2\exp[-\frac{\lambda_1(t)}{2}] - 1$, which lie above

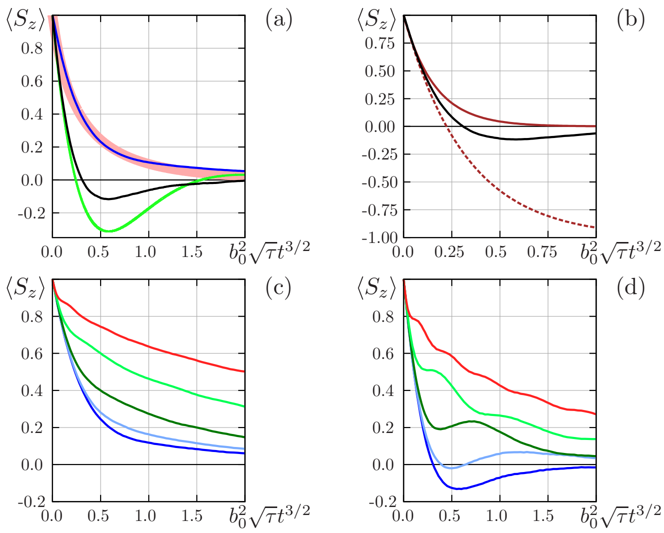

FIG. 3. (Color online) For $d = 1$ random walk the decay, $\langle S_z(t)\rangle$, is a universal function of $b_0^2\sqrt{\tau}t^{3/2}$. (a) Numerical results for a planar hyperfine field (black) exhibit spin reversal at intermediate time. For a spherically distributed $\boldsymbol{b}_i$ (blue) the decay is monotonic but nonexponential and is accurately captured by the solution of the self-consistent equation, Eq. (13) (pink). The green curve is the numerical solution of Eq. (13) for planar hyperfine field. (b) The black curve is the same as in (a), while brown and dashed brown show the result of partial summation of the diffusive diagrams (see text). Weak external field slows down the decay of $\langle S_z(t)\rangle$ for both spherical (c) and planar $\boldsymbol{b}_i$ (d). Numerical results are shown for the following values of $\frac{B}{b_0^{4/3}\tau^{1/3}}$: 0 (dark blue), 1 (light blue), 2 (dark green), 4 (light green), and 8 (red).

and below the numerical results (see Fig. 3). An alternative approach is to sum only the contributions from nonoverlapping diffusive diagrams, Fig. 1. It leads [4] to a self-consistent equation

$$\frac{d\langle S_z\rangle}{du} = -\Lambda \int_0^u du_1 \langle S_z(u_1)\rangle \frac{\exp\left[-\left(u^{2/3}-u_1^{2/3}\right)^{3/2}\right]}{u^{1/3}u_1^{1/3}\left(u^{2/3}-u_1^{2/3}\right)^{1/2}}, \quad (13)$$

where $u = b_0^2\tau^{1/2}t^{3/2}$ and $\Lambda = \frac{4}{9(2\pi)^{1/2}}$. For planar fields the numerator in Eq. (13) is one. The numerical solutions of Eq. (13) are shown in Fig. 3. We see that for the spherical case the solution closely reproduces the simulated decay of $\langle S_z(t)\rangle$. For the planar case, the depth of the minimum in $\langle S_z(t)\rangle$ predicted by the self-consistent equation is -0.28 instead of -0.16. In addition, Fig. 3 indicates that the oscillations in $\langle S_z(t)\rangle$ develop as the magnetic field is switched on, namely, at $B \approx b_0^{4/3}\tau^{1/3}$.

Discussion. In modern spintronics [5] the spin-relaxation time, τ_s, is a key parameter which determines whether or not a given material is suitable for applications. In the past decade, the values τ_s are routinely inferred from Hanle curves in an external magnetic field, i.e., from the behavior, $(1 + B^2\tau_s^2)^{-1}$, of the nonlocal resistance [6–9]. In this regard, our finding of *accelerated relaxation* can be reformulated as follows: memory effects broaden the Hanle curve and affect its shape transforming the Lorentzian closer to Gaussian. Note, that hyperfine fields, being the origin of spin relaxation, are crucial for the memory effects considered above. With spin-orbit fields, the memory effects [10–12] play the opposite role, i.e., they tend to *preserve* spin.

Our main finding can also be reformulated in the language qubit decoherence in a random environment (see, e.g., the recent review, Ref. [13]). A two-level system constituting a qubit naturally maps on a spin in a magnetic field. A common model of the environment [13] is an ensemble of two-level systems surrounding a qubit, which "modulate" this magnetic field at the random time moments of their switching. As a result, the spin senses the random field which assumes discrete values, much like the spin of an electron hopping over the sites. The main finding of the present Rapid Communication—that returns in the course of diffusion accelerate the relaxation—also applies to the qubit decoherence. Indeed, in the language of the environment, a random-walk return corresponds to all two-level systems coming to their initial state. At short enough times, before the magnetic field of the environment "explores" all of its possible values, the statistics of the returns is quite similar to the statistics of random walks.

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