Spectral narrowing and spin echo for localized carriers with heavy-tailed Lévy distribution of hopping times

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We study analytically the free induction decay and the spin echo decay originating from the localized carriers moving between the sites which host random magnetic fields. Due to disorder in the site positions and energies, the on-site residence times, τ, are widely spread according to the Lévy distribution. The power-law tail $\propto \tau^{-1-\alpha}$ in the distribution of τ does not affect the conventional spectral narrowing for $\alpha > 2$, but leads to a dramatic acceleration of the free induction decay in the domain $2 > \alpha > 1$. The next abrupt acceleration of the decay takes place as α becomes smaller than 1. In the latter domain the decay does not follow a simple-exponent law. To capture the behavior of the average spin in this domain, we solve the evolution equation for the average spin using the approach different from the conventional approach based on the Laplace transform. Unlike the free induction decay, the tail in the distribution of the residence times leads to the slow decay of the spin echo. The echo is dominated by realizations of the carrier motion for which the number of sites, visited by the carrier, is minimal.

I. INTRODUCTION

A concept of spectral narrowing of the magnetic resonance lineshape was first formulated in the pioneering paper Ref. 1 and, subsequently, quantified in Ref. 2 more than sixty years ago. This concept pertains to diverse subfields of physics, such as vibrational molecular spectra in liquids[3], optical spectra of semiconductor quantum dots[4], and cold atoms[5]. It is also covered in the textbooks, see e.g. Refs. 6–9.

In application to free induction decay (FID) in nuclear magnetic resonance, the concept of spectral narrowing can be recapped as follows. In the presence of the time-dependent random magnetic field, $b(t)$, the decay of the FID signal is determined by the average

(a)

Dyakonov-Perel
(spectral narrowing)

(b)

With Lévy-type
distribution of
waiting time
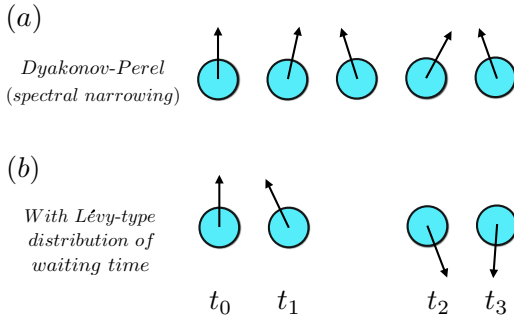
t_0 $\quad$ t_1 $\qquad$ t_2 $\quad$ t_3

FIG. 1: (Color online) The contrast between (a) Dyakonov-Perel spin relaxation with a single correlation time and (b) spin relaxation with broad distribution of the waiting times is illustrated schematically. Allowance for anomalously long waiting times accelerates the relaxation.

$\langle \exp\left[i \int_0^t dt' b(t') \right] \rangle$. The character of the decay depends on the relation between the typical magnitude, b_0, of $b(t)$ and the correlation time, τ. For long correlation time $b_0\tau \gg 1$, the decay is gaussian, $\propto \exp(-b_0^2 t^2)$, reflecting the gaussian distribution of the magnitudes of $b(t)$. In the opposite limit, $b_0\tau \ll 1$, the integrand rapidly changes sign, which is the origin of the spectral narrowing. If the time intervals between the subsequent sign changes are, δt_1, δt_2, δt_3, and so on, and we treat the values b_i as independent gaussian variables, then the average, $\langle \exp\left[i \int_0^t dt' b(t') \right] \rangle$, over the field realizations can be rewritten as $\exp\left[-b_0^2 \sum_{i=1}^n (\delta t_i)^2 \right]$. On the other hand, the number of the sign changes, n, is determined by the condition $\sum_{i=1}^n \delta t_i = t$. This leads to a simple exponential behavior, $\exp(-t/\tau_s)$, of the FID signal, where

$$\frac{1}{\tau_s} = b_0^2 \frac{\langle \delta t^2 \rangle_{\delta t_i}}{\langle \delta t \rangle_{\delta t_i}}. \tag{1}$$

If the random field is characterized by a *single* correlation time, τ, then the intervals δt_i obey the Poisson distribution

$$p_0(\delta t, \tau) = \frac{1}{\tau} \exp\left[- \delta t/\tau \right]. \tag{2}$$

Averaging with this distribution in Eq. (1) yields a well-known result, $\tau_s = 1/2b_0^2\tau$, for the decay rate. In the field of semiconductors this result is also known as the Dyakonov-Perel spin relaxation time[10].

A nontrivial situation emerges when the correlation times, τ, are broadly distributed. Then Eq. (1) takes the form

$$\frac{1}{\tau_s} = 2b_0^2 \frac{\langle \tau^2 \rangle}{\langle \tau \rangle}, \tag{3}$$

where the averaging is performed over the distribution, $F(\tau)$, of the correlation times. Such a situation is generic, e.g., for the dispersive transport in disordered semiconductors[11–17]. Broad distribution of the τ-values stems from the spread in the activation energies. Another example is a system with hopping transport, where the broad distribution of τ is the result of the spread in the hopping distances. Yet another entirely different area where the switching times are broadly distributed are various qubits, see e.g. the review Ref. 18. A qubit can be viewed as a spin. In real devices it is surrounded by two-level systems which switch in time in the telegraph-noise fashion. Such a switching can be viewed as a fluctuating magnetic field acting on spin.

In all the above cases $F(\tau)$ has a power-law tail: $F(\tau) \propto \tau^{-1-\alpha}$. Such a distribution, also known as the Lévy distribution, is normalizable for positive α. However, for $\alpha < 2$ the average $\langle \tau^2 \rangle$ diverges. Formally, this implies that τ_s turns to zero. On the physical level, this means that, on certain occasions, the spin spends enough time in some given field to exercise a full rotation, see Fig. 1. Although the portion of these occasions is small $\sim \int_{1/b_0}^{\infty} d\tau F(\tau) \sim (b_0 \tau)^{\alpha}$, they change the average spin dynamics dramatically. Theoretical study of this dynamics is the subject of the present paper. We find that, for $\alpha < 2$, the FID retains the form of a simple exponent, but the rate, τ_s^{-1}, shortens and becomes a strong function of the tail parameter, α. Our results can be summarized as

$$\frac{1}{\tau_s} = \begin{cases} D_1(\alpha) \, b_0^2 \tau, & \alpha > 2 \\ D_2(\alpha) \, b_0^{\alpha} \tau^{\alpha-1}, & 2 > \alpha > 1 \\ D_3(\alpha) \, b_0, & 1 > \alpha > 0, \end{cases} \quad (4)$$

where $D_1(\alpha)$, $D_2(\alpha)$, and $D_3(\alpha)$ are the dimensionless functions of the tail parameter. Change of the behavior of τ_s at $\alpha = 2$ is due to the formal divergence of $\langle \tau^2 \rangle$, while the change at $\alpha = 1$ is due to the formal divergence of $\langle \tau \rangle$, which enters into the denominator of Eq. (3). We also find that the crossovers at $\alpha = 2$ and $\alpha = 1$ take place within narrow intervals: $|\alpha - 2| \sim 1/|\ln b_0 \tau|$ and $|\alpha - 1| \sim 1/|\ln b_0 \tau|$.

Another phenomenon which is strongly affected in the presence of multiple waiting time is spin echo[19]. The effect of the tail in $F(\tau)$ on the echo is opposite to the effect of $F(\tau)$ on the spin relaxation rate. The echo decays *slower* due to this tail. The average echo signal is determined by the realizations with longest waiting times.

The paper is organized as follows. In Sect. II we derive a closed equation for the FID averaged over the realizations of random fields. Asymptotic (in parameter $b_0\tau \ll 1$) solution of this equation is found in Sect. III, where we derive the result Eq. (4) and also find the crossover behaviors near $\alpha = 2$ and $\alpha = 1$. In Sect. IV we analyze the decay of the average echo signal. Concluding remarks are presented in Sect. V.

II. BASIC EQUATION FOR AVERAGE FID WITH MULTIPLE RELAXATION TIMES

As it was mentioned in the Introduction, the physical picture which we have in mind is the carrier motion between the sites either by hopping or by trapping-detrapping process[11–17]. The time-dependent magnetic field of a hyperfine origin[20], acting on the carrier spin, represents a sequence of steps $\boldsymbol{b}(t) = \sum_i \boldsymbol{b}_i [\Theta(t_{i+1} - t) - \Theta(t - t_i)]$, where $\Theta(x)$ is the step-function and the step durations, $(t_{i+1} - t_i)$, are distributed according to the Poisson distribution Eq. (2), in which τ_0 is distributed according to $F(\tau)$. Since the sites are separated in space, random fields, $\boldsymbol{b}_i$, at different sites are *completely uncorrelated*. We also assume that the motion is three-dimensional, so that the effect of occasional returns to the same site[21–23] are negligible.

Suppose that at time moment $t = 0$ a carrier occupies the site $i = 0$, and its spin is directed along the z-axis. After time t the carrier can either remain on the site $i = 0$ or hop on the neighboring site $i = 1$. The probability to stay is $p_0(t, \tau^{(0)})$, defined by Eq. (2), where $\tau^{(0)}$ is a waiting time for the hop $0 \rightarrow 1$. We assume that the external magnetic field is directed along x-axis, so that only the x-components of the fields $b^{(0)}$ and $b^{(1)}$ on the sites $i = 0$ and $i = 1$ are important. If the carrier stays on $i = 0$, then the z-projection of its spin after time t is equal to $\cos b^{(0)} t$. If the carrier hops after time $t_1 < t$, then this projection is equal to $\cos [b^{(0)} t_1 + b^{(1)} (t - t_1)]$. Taking into account that the moments t_1 are random, the value of $S_z(t)$ can be presented as a sum

$$S_z(t) = e^{-t/\tau^{(0)}} \cos b^{(0)} t \quad (5)$$

$$+ \int_0^t dt_1 \frac{d(1 - e^{-t_1/\tau^{(0)}})}{dt_1} \cos [b^{(0)} t_1 + b^{(1)}(t - t_1)].$$

The derivative in the integrand is the probability density of the hop. If there is a site $i = 2$ on which the carrier can hop from $i = 1$, the expression Eq. (5) gets modified. It acquires a third term describing the possibility of the hop $1 \rightarrow 2$, with corresponding waiting time $\tau^{(1)}$. If this hop takes place, S_z acquires the value $\cos [b^{(0)} t_1 + b^{(1)}(t_2 - t_1) + b^{(2)}(t - t_2)]$, where t_2 is a random moment of the hop $1 \rightarrow 2$ and $b^{(2)}$ is the random field on the site $i = 2$. Quantitatively, the form of the modified second term is the following

$$\int_0^t dt_1 \frac{d(1 - e^{-t_1/\tau^{(0)}})}{dt_1} \left\{ e^{-(t-t_1)/\tau^{(1)}} \cos [b^{(0)} t_1 + b^{(1)}(t - t_1)] + \right.$$

$$\left. \int_{t_1}^t dt_2 \frac{d(1 - e^{-(t_2-t_1)/\tau^{(1)}})}{dt_2} \cos [b^{(0)} t_1 + b^{(1)}(t_2 - t_1) + b^{(2)}(t - t_2)] \right\}. \quad (6)$$

Comparing Eqs. (5) and (6) one can establish a recurrent rule according to which the possibilities of 3, 4, and

so on hops are taken into account. For infinite number of possible hops Eq. (5) transforms into an infinite series. Averaging each term over the gaussian distribution of magnetic fields and realizations of waiting times generates a series for average spin projection, $\overline{S}_z(t)$. It can be verified that $\overline{S}_z(t)$ satisfies the equation

$$\overline{S}_z(t) = \left\langle e^{-t/\tau} \right\rangle e^{-b_0^2 t^2} + \int_0^t dt_1 A(t_1) \overline{S}_z(t - t_1), \quad (7)$$

where the function $A(t)$ is defined as

$$A(t) = \left\langle \frac{1}{\tau} e^{-t/\tau} \right\rangle e^{-b_0^2 t^2}, \quad (8)$$

and $\langle .... \rangle$ stands for averaging over the broadly distributed waiting times. For example, to verify that Eq. (7) yields the same result as averaged Eq. (6) for one hop it is sufficient to substitute the free term in Eq. (7) into the integrand.

The above closed equation describes the averaged spin relaxation. In the next Section we solve it in different domains of the tail parameter α.

III. SOLUTION OF EQUATION FOR $\overline{S}_z(t)$

For concreteness, we choose the distribution function of the waiting times in the form

$$F(\tau) = \frac{C_\alpha \tau_0^\alpha}{(\tau_0^2 + \tau^2)^{\frac{1+\alpha}{2}}}, \quad (9)$$

where the τ_0 parameter is the timescale on which the distribution of waiting times crosses over to the Levy distribution.

For $\tau \gg \tau_0$, we have $F(\tau) \propto \tau^{-1-\alpha}$, and the coefficient C_α which insures the normalization is given by

$$C_\alpha = \frac{2\Gamma(\frac{1+\alpha}{2})}{\pi^{1/2}\Gamma(\frac{\alpha}{2})}. \quad (10)$$

As we demonstrate in the Appendix, for the multiple trapping model[17] the form Eq. (9) describes accurately not only the tail but the entire body of the distribution. Moreover, we show in the Appendix that Eq. (9) applies for general distribution of energies of the sites. While the *typical* time, τ_0, is short, $\tau_0 \ll b_0^{-1}$, the distribution has a long tail $F(\tau) \propto \tau^{-1-\alpha}$.

We start the analysis of the basic equation Eq. (7) by noticing that at times $t \gg \tau_0$ the first term is small. Indeed, averaging with the help of Eq. (9), yields

$$\left\langle e^{-t/\tau} \right\rangle \sim \left(\frac{\tau_0}{t} \right)^\alpha. \quad (11)$$

A crucial step of the analysis is making use of the fact that spin relaxation takes place over a large number of hops. This allows one to expand $\overline{S}_z$ in the integrand

$$\overline{S}_z(t - t_1) \approx \overline{S}_z(t) - t_1 \frac{d\overline{S}_z}{dt}(t). \quad (12)$$

Later in the text we come back and check that the results obtained justify the above step by virtue of smallness of the parameter $b_0\tau_0$. Upon the expansion, Eq. (7) can be easily solved yielding

$$\overline{S}_z(t) = \exp\left[-\int_0^t dt_1 \Phi(t_1) \right], \quad (13)$$

where the function $\Phi(t)$ is defined as

$$\Phi(t) = \frac{1 - \int_0^t dt_2 A(t_2)}{\int_0^t dt_2 t_2 A(t_2)}. \quad (14)$$

For $\alpha > 1$ the characteristic time, τ_s, for the spin dynamics is much longer than b_0^{-1} (we will check this assumption later). On the other hand, even for wide distribution of the waiting times, the function $A(t)$ falls off dramatically for $t \gtrsim b_0^{-1}$. This allows one to extend the upper limits in the integrals in Eq. (14) to ∞. Then Eq. (13) reduces to a simple exponential decay

$$\overline{S}_z(t) = \exp\left(-\frac{t}{\tau_s} \right), \quad (15)$$

with the decay rate given by

$$\frac{1}{\tau_s} = \Phi(\infty) = \frac{\int_0^\infty dt \left\langle \frac{1}{\tau} e^{-t/\tau} \right\rangle \left(1 - e^{-b_0^2 t^2} \right)}{\int_0^\infty dt\, t \left\langle \frac{1}{\tau} e^{-t/\tau} \right\rangle e^{-b_0^2 t^2}}. \quad (16)$$

If the averages in the numerator and denominator decayed rapidly at $t \gtrsim \tau_0$, we would be allowed, by virtue of the condition $b_0\tau_0 \ll 1$, to replace $\exp\left(-b_0^2 t^2 \right)$ by 1 in the denominator and expand $\left[1 - \exp\left(-b_0^2 t^2 \right) \right]$ in the numerator. After that the integrations over time can be carried out explicitly, retrieving the standard expression Eq. (3) for the Dyakonov-Perel relaxation time. Calculating $\langle \tau^2 \rangle$ and $\langle \tau \rangle$ with the help of the distribution Eq. (9) we find

$$\frac{1}{\tau_s} = \frac{\pi^{1/2}(\alpha - 1)\Gamma(\frac{\alpha-2}{2})}{2\Gamma(\frac{\alpha+1}{2})} b_0^2 \tau_0. \quad (17)$$

This expression is valid if $\langle \tau^2 \rangle$ is finite, which corresponds to $\alpha > 2$. The prefactor in this expression specifies the function $D_1(\alpha)$ in Eq. (4). The function $D_1(\alpha)$ falls off monotonically with α. At $\alpha \gg 1$ it behaves as $\left(\frac{2\pi}{\alpha} \right)^{1/2}$. Such a decay of $D_1(\alpha)$ might seem surprising, since for a rapidly decaying tail one would expect τ_s to saturate. In fact, this saturation does take place. Indeed, for $\alpha \gg 1$, the distribution Eq. (9) assumes the form $F(\tau) \approx \left(\frac{2\alpha}{\pi\tau_0^2} \right)^{1/2} \exp\left[-\alpha\tau^2/2\tau_0^2 \right]$, so that the typical time is $\sim \tau_0/\alpha^{1/2}$.

In the domain $1 < \alpha < 2$ the value of $\langle \tau^2 \rangle$ diverges while $\langle \tau \rangle$ remains finite. The latter still allows one to set $b_0 = 0$ in the denominator of Eq. (16), but the numerator cannot be expanded anymore. The explicit expression for the numerator in Eq. (16) reads

$$\int_0^\infty dt \left\langle \frac{1}{\tau} e^{-t/\tau} \right\rangle \left(1 - e^{-b_0^2 t^2}\right) = \qquad (18)$$

$$\int_0^\infty d\tau \int_0^\infty dt \frac{C_\alpha \tau_0^\alpha}{(\tau_0^2 + \tau^2)^{\frac{1+\alpha}{2}} \tau} e^{-t/\tau} \left(1 - e^{-b_0^2 t^2}\right)$$

Upon introducing the new variables $z = t/\tau$ and $w = b_0 z \tau$, the integral in the right-hand side takes the form

$$C_\alpha (b_0 \tau_0)^\alpha \int_0^\infty dz\, z^\alpha e^{-z} \int_0^\infty dw \frac{1 - e^{-w^2}}{[(b_0 \tau_0 z)^2 + w^2]^{\frac{\alpha+1}{2}}}. \qquad (19)$$

Since the characteristic z in Eq. (19) is ~ 1, we can neglect $(b_0 \tau_0 z)^2$ in the denominator, after which the double integral factorizes, yielding

$$C_\alpha (b_0 \tau_0)^\alpha \Gamma(\alpha) \Gamma\left(\frac{2-\alpha}{2}\right). \qquad (20)$$

Then the corresponding expression for the relaxation time in the domain $1 < \alpha < 2$ acquires the form

$$\frac{1}{\tau_s} = (\alpha - 1)\Gamma(\alpha)\Gamma\left(\frac{2-\alpha}{2}\right)\frac{(b_0 \tau_0)^\alpha}{\tau_0}, \qquad (21)$$

where the prefactor specifies the function $D_2(\alpha)$ in Eq. (4).

A. Vicinity of $\alpha = 2$

We see that at the demarkation value $\alpha = 2$ both functions $D_1(\alpha)$ and $D_2(\alpha)$ diverge, so that the expressions Eq. (17) and Eq. (21) yield $\tau_s \to 0$. This suggests that the crossover domain should be treated more carefully. Namely, we rewrite the integral over w in Eq. (19) as

$$\int_0^\infty dw \frac{1 - e^{-w^2}}{[(b_0 \tau_0 z)^2 + w^2]^{\frac{\alpha+1}{2}}} = \frac{\Gamma(\frac{2-\alpha}{2})}{\alpha} \qquad (22)$$

$$+ \int_0^\infty dw \left[\frac{1}{[(b_0 \tau_0 z)^2 + w^2]^{\frac{\alpha+1}{2}}} - \frac{1}{w^{\alpha+1}}\right]\left(1 - e^{-w^2}\right).$$

The integral in the right-hand side converges at small $w \sim b_0 \tau_0 z$, which allows one to expand $(1 - e^{-w^2})$. Upon introducing the variable $v = (\frac{w}{b_0 \tau_0 z})^2$, this integral takes the form

$$\frac{(b_0 \tau_0 z)^{2-\alpha}}{2} \int_0^\infty dv \left[\frac{v^{1/2}}{(1+v)^{\frac{\alpha+1}{2}}} - \frac{1}{v^{\frac{\alpha}{2}}}\right]. \qquad (23)$$

Now we rewrite the integral $\int_0^\infty \frac{dv}{v^{\frac{\alpha}{2}}}$ as a sum of integrals from 0 to 1 and from 1 to ∞. The integral from 1 to ∞ is then combined with the first integral in Eq. (23) in which domain of integration is shifted by 1. This yields

$$\frac{(b_0 \tau_0 z)^{2-\alpha}}{2}\left[-\int_0^1 dv \frac{1}{v^{\frac{\alpha}{2}}} + \int_1^\infty dq \frac{(q+1)^{1/2} - q^{1/2}}{q^{\frac{\alpha+1}{2}}}\right]. \qquad (24)$$

Note that the second integral in the square brackets remains finite at $\alpha = 2$, while the first integral diverges. Keeping only the diverging part and combining it with $\frac{1}{\alpha}\Gamma\left(\frac{2-\alpha}{2}\right) \approx \frac{1}{2-\alpha}$, we establish the behavior of the spin relaxation rate Eq. (21) near $\alpha = 2$

$$\frac{1}{\tau_s} = 2b_0^2 \tau_0 |\ln b_0 \tau_0| \Upsilon\left((\alpha - 2)|\ln b_0 \tau_0|\right). \qquad (25)$$

where the crossover function $\Upsilon(z)$ is defined as

$$\Upsilon(z) = \frac{1 - e^{-z}}{z}. \qquad (26)$$

Thus the expressions Eq. (17) and Eq. (21) are valid outside the interval $|\alpha - 2| \sim |\ln b_0 \tau_0|^{-1}$, which is parametrically narrow.

B. Vicinity of $\alpha = 1$

We see that Eq. (21) yields $\tau_s \to \infty$ as α approaches 1 from the above. This is the result of the divergence of $\langle \tau \rangle$ in this limit. To regularize the behavior of Eq. (21), we need to calculate the denominator in Eq. (16) more accurately. We start from the explicit form of this denominator

$$\int_0^\infty dt\, t A(t) = \int_0^\infty d\tau \int_0^\infty dt \frac{C_\alpha \tau_0^\alpha}{(\tau_0^2 + \tau^2)^{\frac{1+\alpha}{2}} \tau} t e^{-t/\tau} e^{-b_0^2 t^2} \qquad (27)$$

The same change of variables $z = t/\tau$ and $w = b_0 z \tau$, allows to cast the integral in the the form

$$C_\alpha \tau_0 (b_0 \tau_0)^{\alpha-1} \int_0^\infty dz\, z^\alpha e^{-z} \int_0^\infty dw \frac{w e^{-w^2}}{[(b_0 \tau_0 z)^2 + w^2]^{\frac{\alpha+1}{2}}}. \qquad (28)$$

Formally, the singular behavior of this integral at $\alpha = 1$ follows from the fact that at $\alpha = 1$ integration over w yields logarithm if we neglect a small parameter $(b_0 \tau_0)^2$ in the denominator. To capture this behavior, we rewrite the integral over w using the integration by parts

$$\int_0^\infty dw \frac{w e^{-w^2}}{[(b_0 \tau_0 z)^2 + w^2]^{\frac{\alpha+1}{2}}} = \qquad (29)$$

$$\frac{1}{\alpha - 1}\left[(b_0 \tau_0 z)^{1-\alpha} - \int_0^\infty dw \frac{2w e^{-w^2}}{[(b_0 \tau_0 z)^2 + w^2]^{\frac{\alpha-1}{2}}}\right].$$

Now we can safely neglect $(b_0\tau_0)^2$ in the denominator and perform the integration over z, which yields

$$\frac{C_\alpha \tau_0 (b_0\tau_0)^{\alpha-1}}{\alpha-1}\left[\Gamma(2\alpha-1)(b_0\tau_0)^{1-\alpha} - \Gamma(\alpha)\Gamma(\frac{3-\alpha}{2})\right]. \tag{30}$$

Finally, the behavior of τ_s in the vicinity of $\alpha=1$ can be expressed in terms of the crossover function $\Upsilon(z)$ defined by Eq. (26)

$$\frac{1}{\tau_s} = \frac{(\alpha-1)\Gamma(\alpha)\Gamma\left(\frac{2-\alpha}{2}\right)}{\Gamma(2\alpha-1) - \Gamma(\alpha)\Gamma(\frac{3-\alpha}{2})(b_0\tau_0)^{\alpha-1}}\frac{(b_0\tau_0)^\alpha}{\tau_0} \tag{31}$$

$$\approx \frac{(\alpha-1)\pi^{1/2}}{1-(b_0\tau_0)^{\alpha-1}}\frac{(b_0\tau_0)^\alpha}{\tau_0} = \frac{\pi^{1/2}b_0}{|\ln b_0\tau_0|}\frac{1}{\Upsilon\big((1-\alpha)|\ln b_0\tau_0|\big)}.$$

Unlike Eq. (25), the crossover function appears in the denominator. Directly at $\alpha=1$ Eq. (31) yields

$$\frac{1}{\tau_s} = \pi^{1/2}\frac{b_0}{|\ln b_0\tau_0|}. \tag{32}$$

The fact that for all α greater or equal 1 the value τ_s^{-1} is smaller than b_0 justifies the ansatz: $\overline{S}_z(t-t_1) = \overline{S}_z(t) - t_1 d\overline{S}_z/dt$ and the extension of the upper limit in Eq. (14) to ∞.

C. $\alpha < 1$

For $\alpha < 1$ we cannot extend the limits of integration in Eq. (14) to infinity. Instead, we rewrite the expression Eq. (8) for $A(t)$ as follows

$$A(t) = \left\langle\frac{1}{\tau}e^{-t/\tau}\right\rangle\left(e^{-b_0^2t^2} - 1\right) + \left\langle\frac{1}{\tau}e^{-t/\tau}\right\rangle. \tag{33}$$

The integration over t in the second term can be carried out explicitly. Then Eq. (14) acquires the form

$$\Phi(t) = \frac{\left\langle e^{-t/\tau}\right\rangle + \int\limits_0^t dt_2 \left\langle\frac{1}{\tau}e^{-t_2/\tau}\right\rangle\left(1 - e^{-b_0^2t_2^2}\right)}{\int\limits_0^t dt_2 t_2 \left\langle\frac{1}{\tau}e^{-t_2/\tau}\right\rangle e^{-b_0^2t_2^2}}. \tag{34}$$

In calculating both averages we can use the power-law tail of the distribution Eq. (9)

$$\left\langle\frac{1}{\tau}e^{-t/\tau}\right\rangle \approx C_\alpha \frac{\tau_0^\alpha}{t^{\alpha+1}}\Gamma(\alpha+1), \quad \left\langle e^{-t/\tau}\right\rangle \approx C_\alpha\frac{\tau_0^\alpha}{t^\alpha}\Gamma(\alpha). \tag{35}$$

This expressions apply for $t \gg \tau_0$. Substituting them into Eq. (34) and then Eq. (35) into Eq. (13), we arrive at the final result for $\overline{S}_z(t)$

$$\overline{S}_z(t) = \exp\left[-\int\limits_{b_0\tau_0}^{b_0t} dx \frac{\frac{1}{x^\alpha} + \alpha\int\limits_0^x dx_1 x_1^{-\alpha-1}(1-e^{-x_1^2})}{\alpha\int\limits_0^x dx_1 x_1^{-\alpha} e^{-x_1^2}}\right]. \tag{36}$$
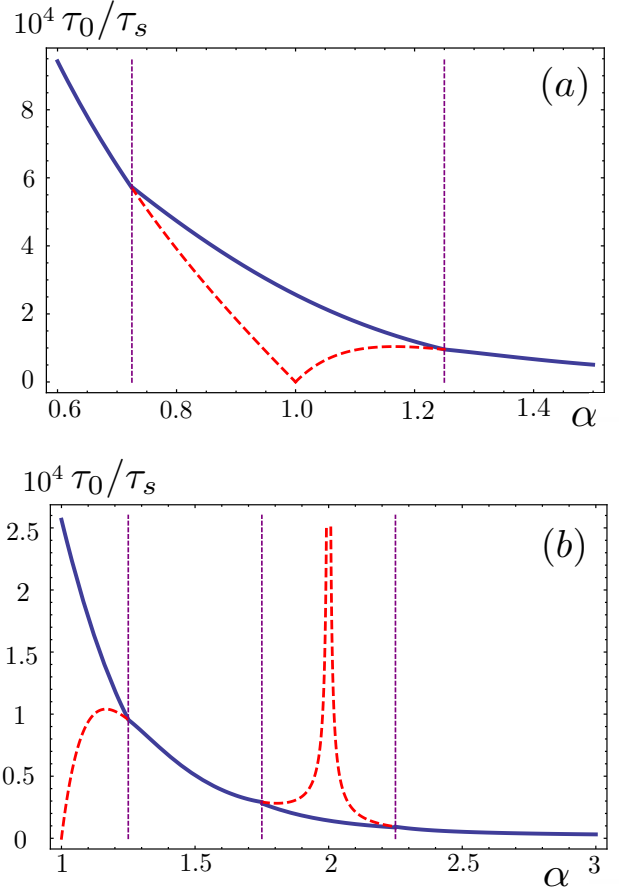
FIG. 2: (Color online) (a) Acceleration of the relaxation rate with decreasing the tail-parameter, α, is illustrated in the domain $0.6 < \alpha < 1.5$. Dashed red curves are plotted from Eq. (39) (for $\alpha < 1$), and Eq. (21) (for $1 < \alpha < 1.5$). The curves exhibit a dip near $\alpha=1$. They are connected by the crossover expression Eq. (31). (b) Domain $1 < \alpha < 3$. Dashed red curves are plotted from Eq. (21) (for $1 < \alpha < 2$), and from Eq. (17) (for $\alpha > 2$). The curves exhibit a divergence near $\alpha=2$. They are connected by the crossover expression Eq. (25). All the curves are plotted for $b_0\tau_0 = 10^{-3}$.

To analyze Eq. (36) in the domain $\tau_0 < t < b_0^{-1}$, we note that for small x the integrand in the exponent behaves as $\frac{1-\alpha}{\alpha x}$ which leads to the power-law decay of $\overline{S}_z(t)$

$$\overline{S}_z(t)\Big|_{t\ll b_0^{-1}} \approx \exp\left[-\frac{1-\alpha}{\alpha}\ln(t/\tau_0)\right] = \left(\frac{\tau_0}{t}\right)^{\frac{1-\alpha}{\alpha}}. \tag{37}$$

This behavior should be compared to the first term in Eq. (7), which we neglected. The power-law Eq. (37) dominates at $\alpha > 0.6$.

In the long-time limit $b_0t \gg 1$ the integral in Eq. (36) is determined by large x. For large x the integrand saturates at the value $\frac{(1-\alpha)\Gamma(\frac{2-\alpha}{2})}{\alpha\Gamma(\frac{3-\alpha}{2})}$, so that the resulting ex-
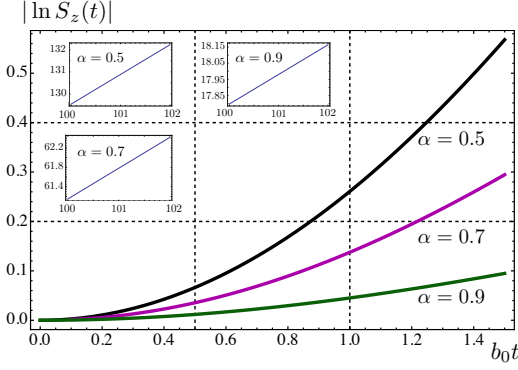
FIG. 3: (Color online) The time evolution of $|\ln \overline{S}_z(t)|$ is plotted from Eq. (36) for $b_0\tau_0 = 10^{-3}$ and the values of the tail-parameter: $\alpha = 0.9$ (green), $\alpha = 0.7$ (purple), and $\alpha = 0.5$ (black). The smaller is α, the later the curves converge to the straight lines. This is illustrated by the insets showing $|\ln \overline{S}_z(t)|$ calculated for very long times $b_0 t \approx 100$, when the slopes are saturated at the values predicted by Eq. (38). Between $b_0 t \approx 1.3$ and $b_0 t \approx 100$ the slopes of $\alpha = 0.9$, $\alpha = 0.7$ and $\alpha = 0.5$ curves increase by the factors of 1.8, 2.03, and 2.2, respectively. The domain where the first term in Eq. (7) dominates the decay corresponds to $b_0 t \lesssim 10^{-3}$, and is not represented in the figure.

pression for $\overline{S}_z(t)$ reads

$$\overline{S}_z(t)\Big|_{t \gg b_0^{-1}} \approx \left(b_0\tau_0\right)^{\frac{1-\alpha}{\alpha}} \exp\left[-\frac{(1-\alpha)\Gamma(\frac{2-\alpha}{2})}{\alpha\Gamma(\frac{3-\alpha}{2})} b_0 t\right]. \tag{38}$$

Since we neglected the first term in Eq. (7), the result Eq. (38) does not capture the initial stage of the decay, which is dominated by this first term. The decay in the entire time domain is described by Eq. (38) when α is so close to 1 that the prefactor in Eq. (38) is not small. Then Eq. (38) captures the spin decay which is a simple exponent with

$$\frac{1}{\tau_s} \approx (1-\alpha)\pi^{1/2} b_0. \tag{39}$$

Comparing to Eq. (4), we conclude that the function $D_3(\alpha)$ has the form $(1-\alpha)\pi^{1/2}$ near $\alpha = 1$. Note, that this expression is consistent with Eq. (31) for $(\alpha - 1) \gg |\ln(b_0\tau_0)|^{-1}$.

The overall behavior of the relaxation rate with α is illustrated in Fig. 2. In Fig. 3 we plot the time evolution of $\ln \overline{S}_z(t)$ using the general expression Eq. (36). We see that, the smaller is α, the later the curves converge to the straight lines corresponding to a simple exponential behavior. Three insets in Fig. 3 show $\ln \overline{S}_z(t)$ at very long times, $b_0 t \approx 100$, after this convergence. It can be seen that between $b_0 t \approx 1.3$ and $b_0 t \approx 100$ the slopes of $\alpha = 0.9$, $\alpha = 0.7$ and $\alpha = 0.5$ curves increase by 1.8, 2.03, and 2.2, respectively.

IV. SPIN ECHO DECAY WITH A LÉVY-TYPE WAITING TIMES DISTRIBUTION

It is well known[19] that the motion narrowing strongly affects the decay of the spin echo, which is formally defined as $\overline{S}_E(T) = \left\langle \exp\left[i\left(\int_0^{\frac{T}{2}} dt b(t) - \int_{\frac{T}{2}}^T dt b(t)\right)\right]\right\rangle$. If the random magnetic field is characterized by a single correlation time τ, the decay of the spin echo would follow the FID signal, i.e. $\overline{S}_E(T) = \overline{S}(T)$. The situation is very different for the broad distribution of τ. Indeed, the shortening of the FID time, τ_s, for distribution Eq. (9) with power-law tail was due to the possibility for a carrier to occasionally sit on a given site, i, during the time, τ_i, much longer than a typical time, τ_0. These sites give anomalously strong contribution, $\left\langle \exp\left[i\int_{t_i}^{t_i+\tau_i} dt b_i(t)\right]\right\rangle$, to the decay. The same physics suggests that, for distribution of τ with power-law tail, the echo signal will decay *slowly* with T. This is because the contributions from anomalously long residence times are eliminated in the echo signal. To quantify this statement, consider a situation when a carrier populates a certain site, i, at time $T_1 < T/2$ and leaves it at time $T_2 > T/2$. Then the contribution from this site to the unaveraged echo signal is given by

$$\exp\left\{ib_i\left[\left(\frac{T}{2}-T_1\right)-\left(T_2-\frac{T}{2}\right)\right]\right\} = \exp\left[ib_i\left(T-T_1-T_2\right)\right]. \tag{40}$$

Assume now, that the carrier makes many hops before arrival to the site i and many hops after departure from the site i. Then the probabilities to preserve spin during the time intervals $(0, T_1)$ and (T_2, T) are given by $\exp\left[-T_1/\tau_s\right]$ and $\exp\left[-(T - T_2)/\tau_s\right]$, respectively. As a result, the contribution to $\overline{S}_E(T)$ from this realization of the random fields reads

$$\int_0^{\frac{T}{2}} \frac{dT_1 e^{-\frac{T_1}{\tau_s}}}{\tau_s} \int_{\frac{T}{2}}^T \frac{dT_2 e^{-\frac{T-T_2}{\tau_s}}}{\tau_s} \left\langle p_0(T_2-T_1, \tau)\right\rangle e^{-b_0^2(T-T_1-T_2)^2}. \tag{41}$$

To analyze this expression, it is convenient to introduce a new variable, $T_3 = T - T_2$. Then it takes the form

$$\int_0^{\frac{T}{2}} \frac{dT_1 e^{-\frac{T_1}{\tau_s}}}{\tau_s} \int_0^{\frac{T}{2}} \frac{dT_3 e^{-\frac{T_3}{\tau_s}}}{\tau_s} \left\langle p_0(T-T_3-T_1, \tau)\right\rangle e^{-b_0^2(T_3-T_1)^2}. \tag{42}$$

The average in Eq. (42) is equal to $C_\alpha\Gamma(\alpha)\tau_0^\alpha/\left(T - T_3 - T_1\right)^\alpha$, see Eq. (35).

The most sound consequence of Eq. (42) is that the echo signal survives at times much longer than τ_s. Indeed, characteristics T_1, T_3 in Eq. (42) are $\sim \tau_s$. For $T \gg \tau_s$ the upper limits in the integrals can be extended to infinity, while the average can be replaced by

$C_\alpha \Gamma(\alpha) \tau_0^\alpha / T^\alpha$. Then we get

$$C_\alpha \Gamma(\alpha) \left(\frac{\tau_0}{T}\right)^\alpha \int\limits_0^\infty dx \; e^{-x} \int\limits_0^\infty dy \; e^{-y} \exp\left[-b_0^2 \tau_s^2 (x-y)^2\right], \tag{43}$$

where we have introduces the dimensionless variables $x = T_1/\tau_s$ and $y = T_3/\tau_s$. Within a numerical constant the double integral is equal to $(b_0 \tau_s)^{-1}$. Thus, we conclude that the echo signal falls off as a power law:

$$\overline{S}_E(T) \sim (b_0 \tau_s)^{-1} \left(\frac{\tau_0}{T}\right)^\alpha. \tag{44}$$

It is seen from Eq. (44) that $\overline{S}_E(T)$ contains a small parameter $(b_0 \tau_s)^{-1}$. At this point, we note that due to a long tail in the waiting time distribution, there is a non-exponential probability that during time, T, the carrier does not hop at all. The contribution to echo signal from such realizations does not contain random magnetic field and falls with T in the same way as Eq. (44). Thus, the final result for the echo signal reads

$$\overline{S}_E(T) = \left\langle p_0(T, \tau) \right\rangle = C_\alpha \Gamma(\alpha) \left(\frac{\tau_0}{T}\right)^\alpha. \tag{45}$$

Comparing the contributions Eq. (44) and Eq. (45), we see that the latter contribution dominates. We thus conclude that the leading contribution to the echo signal comes from sparse realizations when the carrier remains on the same site. It is worth emphasizing that the contribution of such realizations to the average spin is negligible. Certainly the dramatic difference between FID and the echo at times *longer than the spin relaxation time* is the consequence of the power-law tail in $F(\tau)$.

V. CONCLUDING REMARKS

1. As a quantitative measure of acceleration of the relaxation rate, caused by the tail in the distribution, $F(\tau)$, one can consider a ratio of the times for the values of the tail-parameter $\alpha = 1$ and $\alpha = 2$. From Eqs. (25) and (32) one finds

$$\frac{\tau_s(\alpha = 2)}{\tau_s(\alpha = 1)} = \frac{\pi^{1/2}}{2 b_0 \tau_0 |\ln(b_0 \tau_0)|^2}. \tag{46}$$

Both values of τ_s are determined by the tail. Parametrical, in $b_0 \tau_0 \ll 1$, difference between the two values is due to the fact that for $\alpha = 1$ the portion of sites on which the carrier spin exercises a full rotation is $\sim (b_0 \tau_0)$ for $\alpha = 1$ and $\sim (b_0 \tau_0)^2$ for $\alpha = 2$.

2. In replacing the expression Eq. (13) by $\exp(-t/\tau_s)$, we argued that this replacement is valid for $t \gtrsim b_0^{-1}$. This means that in Eq. (13) we chose the lower limit $t_1 \sim b_0^{-1}$. Uncertainty in this lower limit leads to uncertainty

in the prefactor in Eq. (15) $\sim \exp\left(1/b_0\tau_s\right)$, which can be neglected since the product $b_0\tau_s$ is big. Yet another contribution to the prefactor comes from $\int\limits_{\tau_0}^{1/b_0} dt_1 \Phi(t_1)$. To estimate this contribution, we note that for $t \lesssim b_0^{-1}$

$$\Phi(t)\Big|_{t<b_0^{-1}} \approx \frac{1 - \int\limits_0^t dt_2 \left\langle \frac{1}{\tau} e^{-t_2/\tau} \right\rangle}{\int\limits_0^t dt_2 t_2 \left\langle \frac{1}{\tau} e^{-t_2/\tau} \right\rangle} = \frac{\left\langle e^{-t/\tau} \right\rangle}{\left\langle \tau - (t+\tau)e^{-t/\tau} \right\rangle}. \tag{47}$$

For $t \gtrsim \tau_0$ we can neglect the second term in the denominator and perform integration over time, yielding

$$\int\limits_0^t dt_1 \Phi(t_1)\Big|_{t<b_0^{-1}} = \frac{\left\langle \tau(1 - e^{-t/\tau}) \right\rangle}{\langle \tau \rangle}. \tag{48}$$

Thus, the contribution to the prefactor from small times does not exceed 1. In fact, the contribution Eq. (48) comes from neglecting the first term, $\left\langle \exp(-\frac{t}{\tau} - b_0^2 t^2) \right\rangle$, in the basic equation Eq. (7). We effectively replaced the first by the initial condition: $\overline{S}_z(t \sim \tau_0) = 1$. More accurate calculation, based on the Laplace transform, suggests that for $\alpha > 1$ the true prefactor is 1.

3. Formal solution of Eq. (7) can be expressed in the form of the inverse Laplace transform, see e.g. Ref. 24

$$\overline{S}_z(t) = \frac{1}{2\pi i} \int\limits_{\gamma - i\infty}^{\gamma + i\infty} ds \; e^{st} \frac{S_z^{(0)}(s)}{1 - K(s)}, \tag{49}$$

where the functions $S_z^{(0)}(s)$ and $K(s)$ are defined as

$$S_z^{(0)}(s) = \int\limits_0^\infty dt \left\langle e^{-t/\tau} \right\rangle e^{-st - b_0^2 t^2}, \tag{50}$$

$$K(s) = \int\limits_0^\infty dt \left\langle \frac{1}{\tau} e^{-t/\tau} \right\rangle e^{-st - b_0^2 t^2}$$

The decay of $\overline{S}_z(t)$ is defined by the poles, $s = s_0$, for which $K(s_0) = 1$. For $\alpha > 1$, one can retain only the smallest s_0 and find it by expanding $K(s)$ at small s. This readily yields $s_0 = -\Phi(\infty) = -\tau_s^{-1}$, i.e. the same expression Eq. (16) for the decay rate as was found in Sect. II from the different approach. The justification for expanding $K(s)$ is that the exponent $\exp\left(-b_0^2 t^2\right)$ ensures the convergence of the integral Eq. (50) at $t \sim b_0^{-1}$ when $\exp\left(-s_0 t\right)$ is close to 1. Thus, for $\alpha > 1$, the results of the two approaches to solving Eq. (7) coincide. Moreover, the solution Eq. (49) takes into account the first term in Eq. (7), which we have neglected. The prefactor in front
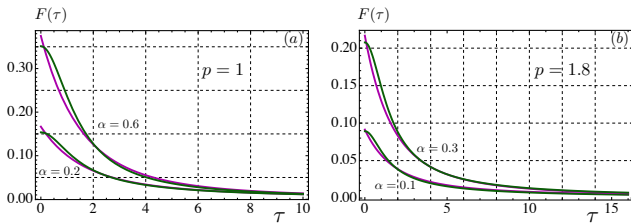
FIG. 4: (Color online) Distribution function, $F(\tau)$, of the waiting times in the multiple-trapping model is shown with purple lines for the densities of tail states of the form $g(\varepsilon) \propto \exp\left[-\left(|\varepsilon|/\mathcal{T}_0\right)^p\right]$ with $p = 1$ (a) and $p = 1.8$ (b). Green lines are the interpolations of $F(\tau)$ with the form Eq. (9) of the main text.

of $\exp\left(-t/\tau_s\right)$ calculated from Eq. (49) is given by

$$-\frac{S_z^{(0)}(s_0)}{K'(s_0)} \approx \frac{\int\limits_0^\infty dt \left\langle e^{-t/\tau} \right\rangle e^{-b_0^2 t^2}}{\int\limits_0^\infty dt\; t \left\langle \frac{1}{\tau} e^{-t/\tau} \right\rangle e^{-b_0^2 t^2}}. \qquad (51)$$

It appears that we can neglect the exponent $\exp\left(-b_0^2 t^2\right)$ in the integrands in the numerator and the denominator. This is because both integrals converge for $\alpha > 1$ and are equal to $\langle\tau\rangle$, which is finite for $\alpha > 1$. Thus the true prefactor is equal to 1, as was mentioned above.

4. For $\alpha < 1$ the formal solution Eq. (49) becomes useless. This is because the pole s_0 cannot be found analytically, and, moreover, many poles (corresponding to $s \sim b_0$) contribute to $\overline{S}_z(t)$. This also follows from our solution Eq. (37) and from Fig. 3. It is seen that $\overline{S}_z(t)$ follows a simple exponential behavior only for large times, $b_0 t \gtrsim 1$.

5. In the paper Ref. 16 the effect of the power-law tail in $F(\tau)$ on the decay of $\overline{S}_z(t)$ was analyzed. The analysis relied on the solution of Eq. (7) in the form of Eq. (49). The authors did not analyze the behavior of τ_s in different domains of the tail-parameter, α. They rather realized that retaining a single pole becomes inadequate for $\alpha < 1$, and resorted to the numerics. Our results Eqs. (17), (21) and Eqs. (25), (31) for the crossover domains are fully analytical. Obtaining these results was facilitated by exploiting the small parameter $b_0\tau_0 \ll 1$ for $\alpha > 1$ and by solving Eq. (7) using an alternative approach for $\alpha < 1$.

Appendix A: Applicability of the waiting times distribution Eq. (9) to the multiple trapping model

In the multiple trapping model[11–17], the waiting time is determined by activation of electron from a localized state in the tail to the conduction band. If the energy position of the localized state is $-\varepsilon$, then the activation rate is equal to $\Gamma(\varepsilon) = \Gamma_0 \exp[-\varepsilon/\mathcal{T}]$, where $\mathcal{T}$ is the temperature. Actual waiting times, t_i, are random. While the average waiting time is Γ^{-1}, the distribution of the waiting times for a given ε is given by the Poisson distribution

$$f_\varepsilon(\tau) = \Gamma(\varepsilon) \exp\left[-\Gamma(\varepsilon)\tau\right]. \qquad (A1)$$

The remaining task is to average $f_\varepsilon(\tau)$ over ε with the weight determined by the density of the tail states, $g(\varepsilon)$. In the multiple trapping model the form of $g(\varepsilon)$ is a simple exponent $g(\varepsilon) \propto \exp\left[-\varepsilon/\mathcal{T}_0\right]$. The final expression for the waiting times distribution reads

$$F(\tau) \propto \int\limits_{-\infty}^0 d\varepsilon\, g(\varepsilon) f_\varepsilon(\tau) \propto \frac{\alpha}{\tau^{\alpha+1}} \int\limits_0^{\Gamma_0\tau} dx\; x^\alpha e^{-x}, \qquad (A2)$$

where $\alpha = \mathcal{T}/\mathcal{T}_0$. For large waiting times $\tau \gg \Gamma_0^{-1}$, we have $F(\tau) \propto \tau^{-(\alpha+1)}$. At $\tau \to 0$ the power-law divergence is cut off. The character of cutoff is not precisely the one given by Eq. (9), but they match very closely, as illustrated in the Figure 4. In organic semiconductors the density of the tail states is better approximated by a stretched-exponential form $g(\varepsilon) \propto \exp\left[-\left(|\varepsilon|/\mathcal{T}_0\right)^p\right]$, with p close to 2, see Refs. 25–27. Repeating the above steps for this $g(\varepsilon)$ we found that $F(\tau)$ can still be closely approximated with Eq. (9), see Figure 4.

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