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Spin precession in disordered systems: Anomalous relaxation due to heavy-tailed field distributions

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Abstract. – We investigate spin precession in the presence of randomly distributed magnetic moments which reorient by thermally activated transitions. Based on analytical calculations and scaling arguments, we find that the polarisation decay of a spin ensemble exhibits a rich behaviour characterised by stretched exponentials and power laws. The anomalous relaxation laws result from heavy-tailed local field distributions and are verified by computer simulations. The general problem is discussed in the context of μSR in random assemblies of superparamagnetic clusters.

Introduction. – The dynamics of disordered systems is often governed by spatial and temporal fluctuations that can be described by Gaussian statistics due to the central limit theorem (CLT). However, in an increasing number of systems it is found that anomalous broad Lévy distributions of the fluctuations play a key role [1]. Lévy distributions generalise the CLT if the fluctuations arise from a superposition of independent random contributions whose distributions exhibit no finite second moment [2]. Intriguing consequences for dynamical processes in many different scientific disciplines have been explored, mainly in the context of anomalous long-range transport phenomena (see, *e.g.*, [3]).

In this letter we investigate the effects of anomalous fluctuations on the local dynamics of spin systems. The precession dynamics of a spin probe in a local field $\boldsymbol{H}(t)$ is described by [4]

$$\frac{\mathrm{d}\boldsymbol{S}}{\mathrm{d}t} = \boldsymbol{S} \times \boldsymbol{H}. \tag{1}$$

Several experimental techniques, such as, *e.g.*, nuclear and electron magnetic resonance (NMR, ESR), muon spin relaxation (μSR), and βNMR, rely on eq. (1), which should be understood as the equation of motion of the quantum-mechanical expectation value. In the phenomenological approach by Bloch, the effects of fluctuating fields $\boldsymbol{H}(t)$ are taken into account by adding relaxation terms to eq. (1). These give rise to exponential decays of an initial spin polarisation with relaxation rates that can be expressed by spectral densities of the field fluctuations [5]. For non-exponential relaxations, however, the standard Bloch approach can no longer be straightforwardly applied. We may distinguish two particularly relevant cases: i) The fluctuations exhibit long-range temporal correlations, $\langle H(t)H(0)\rangle \sim t^{-\nu}$ with $\nu \le 1$ such that a characteristic correlation time does not exist, and ii) the field distribution $\psi(\boldsymbol{H})$ of local fields is anomalously broad, *e.g.* $H^2\psi(\boldsymbol{H}) \sim H^{-1-\eta}$ with $\eta < 2$, such that it exhibits no finite second moment. The first case is often referred to in the analysis of spin glass systems [6, 7].

The second case arises quite naturally when considering an ensemble of independent spins in a system of randomly distributed field sources. As an example of practical importance we refer in this work to μSR in random assemblies of superparamagnetic clusters [8, 9], whose magnetic moments reorient with a thermally activated rate ν. The corresponding dipolar fields decay as r^{-3} with distance from the spin probe. For testing our theoretical predictions, we more generally consider fields decaying as $r^{-\mu}$.

The aim of our work is as follows. We will show how broad local field distributions emerge in this conceptually simple system and how they give rise to rich anomalous spin relaxation scenarios, whose long-time relaxation behaviour is characterised by power laws and stretched exponentials with exponents depending on μ and d. We will make new predictions for μSR experiments in the systems referred to above. Due to the generality of the problem and the simple scaling arguments we invoke, our findings should also be of importance for related problems, where the precession dynamics plays a fundamental role.

Model. – To be specific, we consider the following model. We place a spin $\boldsymbol{S}$ at the origin of a d-dimensional system that contains randomly oriented point-like magnetic clusters with mean number density n at random positions (Poisson distribution of the cluster number in a volume ΔV with mean cluster number $n\Delta V$). A cluster with moment $\boldsymbol{m}$ and position $\boldsymbol{r}$ is assumed to induce a field contribution $\boldsymbol{h} = \boldsymbol{m}/r^{\mu}$ at the probe site. Each moment $\boldsymbol{m}$ changes its orientation to a set of possible other orientations with the rate ν. In particular we study two situations: In the first case, only the directions $\boldsymbol{m}$ and $-\boldsymbol{m}$ are possible (uniaxial case), while in the second case there are four additional orientations perpendicular to $\boldsymbol{m}$ corresponding to a cubic symmetry (multiaxial case). Initially, the spin is polarised in the z-direction, $\boldsymbol{S} = (0, 0, 1)$. The task is to solve eq. (1) for a given cluster configuration and a certain realization of the cluster reorientation process and to average this solution over all possible realizations. By finally averaging over all cluster configurations we obtain the spin polarisation $\langle S_z(t)\rangle$ at time t as measured in experiment. In the following we will discuss the relaxation behaviour for the generic situation $\mu > d/2$ [10].

Local field distribution, static case. – We start out by focusing on the time regime $t \ll \nu^{-1}$, where the field $\boldsymbol{H} = \sum_i \boldsymbol{h}_i$ can be viewed to be static, and the solution of eq. (1) reads $S_z(t) = (H_z^2/H^2) + [1 - (H_z^2/H^2)]\cos(Ht)$. The probability density $\psi_h(\boldsymbol{h})$ for the field $\boldsymbol{h} = \boldsymbol{m}/r^{\mu}$ of an individual cluster at distance r from the spin probe scales as $h^2\psi_h(\boldsymbol{h}) \sim r^{d-1}|\mathrm{d}r/\mathrm{d}h| \sim h^{-1-d/\mu}$. Hence, for $d/\mu < 2$ a second moment does not exist and the probability density $\psi(\boldsymbol{H})$ of the total local field $\boldsymbol{H}$ should, according to Lévy statistics [2], also scale as $H^2\psi(\boldsymbol{H}) \sim H^{-1-d/\mu}$ for large H. This was first discussed in [11] in the context of dilute dipolar or RKKY spin glasses with power law interactions. By an exact calculation we obtain

$$\psi(\boldsymbol{H}) = -\frac{1}{2\pi W^2 H}\,\mathrm{Re}\,L'_{\frac{d}{\mu},0}\left(\frac{H}{W}\right), \tag{2}$$

where $\mathrm{Re}\,L'_{\alpha,0}(u)$ denotes the real part of the derivative of the Lévy stable law $L_{\alpha,0}(u) = (2\pi)^{-1}\int \mathrm{d}k\,\exp[-iku - |u|^{\alpha}]$ to the index $(\alpha, 0)$. For $d = \mu$, *i.e.* in particular for dipolar fields in $d = 3$, one obtains a three-dimensional variant of the Lorentzian, $\psi(\boldsymbol{H}) = 1/\pi^2 W/(W^2 + H^2)^2$ [12]. The characteristic width $W = C_W m n^{\mu/d}$ is given by the field associated with the mean distance $n^{-1/d}$ of the clusters times a constant [13]. As expected, $4\pi H^2\psi(\boldsymbol{H}) \sim C_{\psi} W^{-1}(H/W)^{-1-d/\mu}$ for large H.

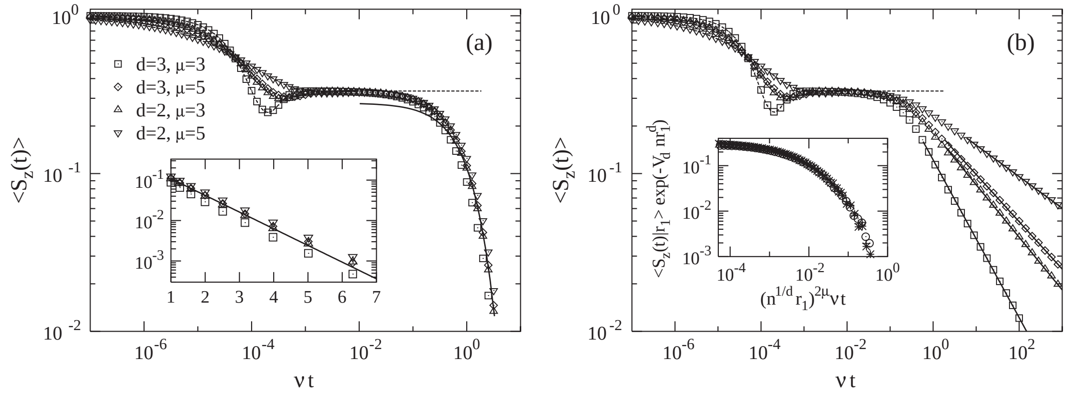

Fig. 1 – Spin polarisation $\langle S_z(t)\rangle$ as a function of νt in the slowly fluctuating case ($\nu/W = 10^{-4}$) for (a) multiaxial and (b) uniaxial cluster moments, and several μ and d. The symbols refer to the simulations and their assignment is the same in both figures. The dashed lines refer to the exact result (3), while the solid lines are fits according to the long-time behaviours (4), (7). The inset in (a) shows, on a semi-logarithmic scale, the exponential long-time relaxation of $\langle S_z(t)\rangle$ *vs.* νt that is almost independent of d and μ (the solid line is drawn as a guide for the eye). The inset in (b) demonstrates the scaling (6) for 4 different radii $r_1 \ll n^{-1/d}$, $r_1 = 1.0$ (+), 1.5 (×), 2.0 (∗), and 2.5 (○) in the case $\mu = d = 3$, $n = 0.01$.

Averaging $S_z(t)$ over $\psi(\boldsymbol{H})$ eventually yields

$$\langle S_z(t)\rangle = \frac{1}{3} + \frac{2}{3}\left[1 - \frac{d}{\mu}(Wt)^{d/\mu}\right]\exp\left[-(Wt)^{d/\mu}\right]. \tag{3}$$

For $d = \mu$, one recovers the Lorentzian Kubo-Toyabe function [14]. As shown in fig. 1 for different μ and d, the results from our simulations agree with eq. (3) for $\nu t \ll 1$. Laws of type (3) have been used in the literature to describe anomalous μSR line-shapes with $d/\mu \neq 1, 2$ that neither follow a Lorentzian ($d/\mu = 1$) nor Gaussian ($d/\mu = 2$) behaviour (see, *e.g.*, [15]). We note, however, that (3) is an exact result and should not be confused with an effective "power Kubo-Toyabe function" [16] that serves as a fitting function.

Case of slowly fluctuating cluster moments. – In the dynamic regime $t \gg \nu^{-1}$ we distinguish between the two cases of slowly or rapidly fluctuating cluster moments, where $\nu \ll W$ or $\nu \gg W$, respectively. In both cases we employ scaling arguments to derive the typical decay rates Γ of the spin polarisation. To tackle the problem of averaging over spatial cluster configurations, we consider subensembles of configurations that are specified by fixing the distances of the clusters closest to the spin probe. This concept is motivated by the hierarchy implied by the Lévy statistics, which for the field distribution (2) means that the n-th nearest cluster gives a contribution of order $n^{\mu/d}$ times smaller than the closest cluster (see, *e.g.*, [17]).

Let us begin with the case $\nu \ll W$ of slowly fluctuating cluster moments, where for the relevant cluster configurations the field $\boldsymbol{H}$ has a magnitude $H \gg \nu$ (other configurations have an exponentially small weight). In a time interval of order ν^{-1} then, the spin precesses many periods around the local field, whereby $S_z(t)$ oscillates around a mean value $\bar{S}_z(t)$. The changes of $\bar{S}_z(t)$ averaged over many realizations of the cluster dynamics determine the decay of spin polarisation.

In the multiaxial case, significant changes of $\boldsymbol{H}$, which occur in a time of order ν^{-1}, alter the axis of precession and $\bar{S}_z(t)$ relaxes with a rate proportional to ν. Hence we expect a simple exponential decay

$$\langle S_z(t) \rangle \sim \exp[-\,\mathrm{const}\,\nu t]\,, \tag{4}$$

which is confirmed by our simulations shown in fig. 1a.

The uniaxial case is more subtle. To see this, we decompose the field $\boldsymbol{H}$ into the contribution $\boldsymbol{h}_1 = \boldsymbol{m}/r_1^\mu$ from the nearest cluster at distance r_1 and the contribution $\boldsymbol{H}_1$ from the other clusters, $\boldsymbol{H} = \boldsymbol{h}_1 + \boldsymbol{H}_1$. In the subensemble of all cluster configurations with given r_1, the variance of $\boldsymbol{H}_1$ is

$$\langle H_1^2 \mid r_1 \rangle = C_H h_1^2 \left(\frac{h_1}{W}\right)^{-d/\mu}. \tag{5}$$

For $r_1 \gg n^{-1/d}$, $h_1/W \ll 1$, and $\boldsymbol{H}_1$ dominates over $\boldsymbol{h}_1$. Hence one encounters the same physical situation as in the multiaxial case. For small $r_1 \ll n^{-1/d}$, however, $\boldsymbol{h}_1$ is dominant, so that changes $\boldsymbol{h}_1 \to -\boldsymbol{h}_1$ essentially revert the direction of precession and leave $\bar{S}_z(t)$ unchanged.

In this situation of small $r_1 \ll n^{-1/d}$ the presence of the contribution $\boldsymbol{H}_1$ causes the axis of the field $\boldsymbol{H}$ (irrespective of its direction) to wobble around the $\pm\boldsymbol{h}_1$-axis with the rate ν and an angular amplitude of order H_1/h_1. The wobbling motion together with the much faster precession leads to a diffusive type of motion of $\bar{S}_z(t)$ with a diffusion rate $\Gamma \sim (H_1/h_1)^2\nu$. As will be substantiated in the following, this leads to an extremely slow long-time decay of the polarisation dominated by those spins which are relatively close to one cluster (large h_1) and far away from any other clusters (small H_1).

To extract the asymptotic relaxation of the spin polarisation, we consider the subensemble of all cluster configurations with fixed distances r_1 and r_2 of the nearest and second-nearest cluster to the spin probe. In the configurations of this subensemble we can decompose $\boldsymbol{H}_1$ into $\boldsymbol{h}_2$ and $\boldsymbol{H}_2$, where $h_2 = m/r_2^\mu$ and $\langle H_2^2 \mid r_2\rangle$ satisfies (5) with h_1 replaced by h_2. Accordingly, for $r_1 < r_2 \lesssim n^{-1/d}$, $H_1^2 \sim m^2/r_2^{2\mu}$ and $\Gamma \equiv \Gamma(r_1, r_2) \propto (r_1/r_2)^{2\mu}\nu$, while for $r_2 \gtrsim n^{-1/d}$, $H_1^2 \sim W^{d/\mu}(m/r_2^\mu)^{2-d/\mu}$ and $\Gamma(r_1, r_2) \propto r_1^{2\mu} W^{d/\mu}(m/r_2^\mu)^{2-d/\mu}\nu$. Writing $\langle S_z(t) \mid r_1, r_2\rangle \sim \exp[-\Gamma(r_1, r_2)t]$ in the subensemble with given r_1 and r_2, we can average over the probability density $\phi_2(r_2 \mid r_1) = S_d n r_2^{d-1} \exp[-V_d n(r_2^d - r_1^d)]$ of r_2 $(r_1 \le r_2 < \infty)$ to obtain [18]

$$\langle S_z(t) \mid r_1 \rangle \sim \exp\left[V_d n r_1^d - \mathrm{const}\left[(n^{1/d} r_1)^{2\mu} \nu t\right]^{d/2\mu}\right] \tag{6}$$

for $\nu t \gg 1$ (and $r_1 \ll n^{-1/d}$). We have verified this prediction for various μ and d by our simulations. One example (for $\mu = d = 3$) is shown in the inset of fig. 1b.

Final averaging over the probability density $\phi_1(r_1) = S_d n r_1^{d-1} \exp[-V_d n r_1^d]$ of r_1 yields

$$\langle S_z(t) \rangle \sim (\nu t)^{-d/2\mu}\,. \tag{7}$$

This slow power law decay is in marked contrast to the exponential decay in the multiaxial case and it is verified in fig. 1b by our simulations.

Case of rapidly fluctuating cluster moments. – Next we discuss the case $\nu \gg W$ of rapidly fluctuating cluster moments. The field $\boldsymbol{H}$ in the relevant cluster configurations now has a magnitude $H \ll \nu$ and the spin rotates only by a small angle in a time interval of order ν^{-1}. This means that the concept of a mean value $\bar{S}_z(t)$ is not useful any longer, since the phase of the precession matters. Reorientations of $\boldsymbol{h}_1$ are effective for the spin relaxation both in the presence of uniaxial and multiaxial cluster moments.

The small angular changes of the spin lead again to a diffusive type of motion of $S_z(t)$. In time ν^{-1} the angular change is of order H/ν and the corresponding diffusion rate $\Gamma \sim (H/\nu)^2\nu$.

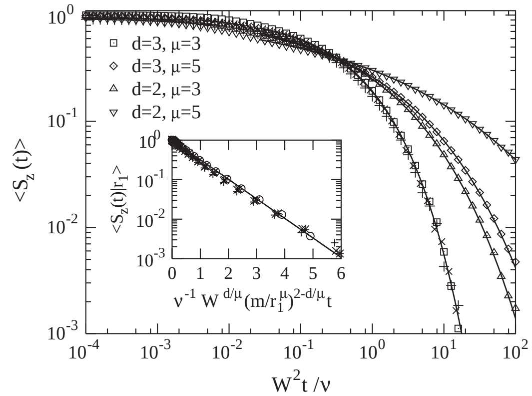

Fig. 2 – Spin polarisation $\langle S_z(t)\rangle$ as a function of W^2t/ν in the case of rapidly fluctuating cluster moments ($\nu/W = 10$ (□), 50 (+), and 100 (×) for $d = \mu = 3$, and $\nu/W = 10$ for the three other combinations of d and μ). Data points refer to the simulations and the solid lines are fits according to eq. (8). The inset shows the exponential decay of $\langle S_z(t) \mid r_1\rangle$ and the scaling as discussed in the text for 4 different radii $r_1 \gtrsim n^{-1/d}$, $r_1 = 6.5$ (+), 7.0 (×), 7.5 (∗), and 8.0 (○) in the case $\mu = d = 3$, and $n = 0.01$ (the solid line is drawn as a guide for the eye).

Decomposing the field $\boldsymbol{H} = \boldsymbol{h}_1+\boldsymbol{H}_1$ as before, and taking into account the dominant contributions, we thus find $\Gamma \equiv \Gamma(r_1) \propto \nu^{-1}m^2/r_1^{2\mu}$ for $r_1 \lesssim n^{-1/d}$ and $\Gamma(r_1) \propto \nu^{-1}W^{d/\mu}(m/r_1^{\mu})^{2-d/\mu}$ for $r_1 \gtrsim n^{-1/d}$ (cf. eq. (5)). We then write $\langle S_z(t) \mid r_1\rangle \sim \exp[-\Gamma(r_1)t]$ for $\nu t \gg 1$ and $r_1 \gg (m/\nu)^{1/\mu}$ (for $r_1 \ll (m/\nu)^{1/\mu}$, $h_1 \gg \nu$, *i.e.* one encounters a situation corresponding to the case of slowly fluctuating clusters moments, which is subdominant here except for uniaxial clusters at very long times, see below). This exponential decay of $\langle S_z(t) \mid r_1\rangle$ is demonstrated in the inset of fig. 2 for $d = \mu = 3$ in the regime $r_1 > n^{-1/d}$. By averaging over r_1 we finally obtain

$$\big\langle S_z(t)\big\rangle \sim \exp\Big[-\text{const}\,\big(\nu^{-1}W^2t\big)^{d/2\mu}\Big]. \tag{8}$$

To perform the average we have used a saddle point approximation, where analogous comments apply as given in [18]. It is interesting to note that similar stretched exponential relaxation laws were derived in the case of NMR by averaging over a distribution of Bloembergen-Purcell-Pound–type relaxation rates [19]. Figure 2 confirms both the scaling with (W^2t/ν) and the stretched exponential decay for the same μ and d values as in fig. 1. In the uniaxial case the stretched exponential decay (8) will, at long times, be masked by the much slower power law decay (7) that stems from the rare configurations with $h_1 = m/r_1^{\mu} \gg \nu$.

Discussion. – In summary, we have shown that spin precession in the presence of randomly distributed and fluctuating field sources leads to an anomalous relaxation of an initially polarised spin probe, which is characterised by stretched exponentials (eqs. (3), (8)) or power laws (eq. (7)). The deviations from a simple exponential decay are caused by Lévy-type local field distributions (eq. (2)). These render a treatment in terms of Gaussian processes impossible, but allowed us to perform an analysis based on subensembles of cluster configurations that are defined with respect to the most dominant contributions to the local field, *i.e.* the field sources closest to the spin probe.

It is important to stress that a simple mean-field–type description of the relaxation process would fail, as was already pointed out by Uemura *et al.* [20] in the case $\mu = d = 3$. In such a mean-field description one might employ a "strong collision approximation" [21], where the field $\boldsymbol{H}$ at the probe site is drawn anew from (2) with the rate ν (thereby neglecting the fluctuations in the spatial cluster configurations). By scaling arguments similar to those outlined above, one can show that this approach leads, for $t \gg \nu^{-1}$, to an exponential relaxation $\langle S_z(t)\rangle \sim \exp[-\Gamma_{\rm mf} t]$ both in the cases of slowly and rapidly fluctuating cluster moments and irrespective of whether the clusters possess only one easy axis or more. For $\nu \ll W$, one obtains $\Gamma_{\rm mf} \propto \nu$, while for $\nu \gg W$, $\Gamma_{\rm mf} \propto \nu(W/\nu)^{d/\mu}$ [22].

Our approach can be readily applied to experiments [8,9]. For example, for the system in [8] the width $W = C'_W nm$ [13] follows from the experimental known parameters n and m without any additional modelling [23]. Moreover, our work allows us to make new predictions. For example, one can test for dimensionality effects in the anomalous relaxation laws by depositing superparamagnetic cluster onto a surface (or by embedding them in separated layers within a bulk material). Both the static and the dynamic situation will be accessible by tuning W via the cluster concentration n, and by tuning $\nu = \nu_0 \exp[-E_{\rm aniso}/T]$ via the temperature T. For example, for the multiaxial iron nanoclusters used in [9], one finds for the attempt frequency $\nu_0 \simeq 10^9\,{\rm s}^{-1}$ and for the anisotropy energy $E_{\rm aniso} \simeq 51\,{\rm K}$. In that experiment $W \simeq 7\times 10^5\,{\rm s}^{-1}$ (for 0.1% volume fraction iron in a silver matrix), implying a crossover from the static to the dynamic situation around a temperature $T_x = E_{\rm aniso}/\ln(\nu_0/W) \simeq 7\,{\rm K}$.

We restricted our treatment here to point clusters with unique moment m and neglected interactions between the moments. As long as the cluster sizes are much smaller than the mean distance $n^{-1/d}$, crossover effects to a Debye-like relaxation behaviour typical for Gaussian processes should be of minor importance. A broad distribution of cluster sizes, however, may require a refined analysis in the dynamic regime by defining the subensembles with respect to both the nearest-cluster distance and the cluster size (in the static regime the results remain unchanged except that m has to be replaced by its average value). One can easily generalise our approach to situations where an additional external field is present. The μSR experiment then corresponds closely to a measurement of the transverse relaxation in NMR probes.

Interactions between the cluster moments at high temperatures T can be accounted for by a temperature-dependent width $W = W(T)$ in (2) (for an approximate calculation in $\mu = d = 3$, see [24]). At low temperatures T, by contrast, the cluster dynamics cannot be described any longer by a Poisson process with rate ν. For dipolar systems in $d = 2, 3$, this occurs for $T \lesssim 2m^2 n^{d/3}$ [25, 26]; *e.g.*, in the materials studied in [8, 9] this would be below 1 K. In this low-temperature regime the problem becomes more difficult and the relaxation laws (7), (8) may no longer hold true. A non-Poissonian cluster dynamics is often encountered in spin glasses [6, 7], orientational glasses [27] and related systems [28].

Having mentioned these limits of our findings, we hope that our work will stimulate further research on the challenging problem of spin precession in disordered systems. Our scaling methods should give deeper insight into the spin relaxation in disordered systems and may be extended to describe μSR or related dynamical probes also in other complex systems.

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