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Generalization of the Abragam relaxation function to a longitudinal field

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We obtain an analytical form for the muon relaxation function in a dynamical magnetic environment and an external longitudinal magnetic field using perturbation theory. The new relaxation function is compared with the semiclassical Kubo-Toyabe function and the result of Monte Carlo simulation. We also demonstrate the application of our result to the case of dilute spin glasses.

The muon spin relaxation (μSR) technique[1] has proven to be a very powerful tool for the investigation of magnetic phenomena. In this technique, polarized muons are implanted into a sample where their spin evolves in the local magnetic field until they decay. The decay positron is emitted preferentially along the final muon spin direction; by collecting several million positrons, we can reconstruct the time dependence of the muon spin polarization $[\mathbf{P}(t)]$ which, in turn, reflects the magnetic field at the interstitial muon site. In general this local field combines two contributions: the internal field and the externally applied field. When the internal field is random, for example, in the case of randomly oriented magnetic dipoles, measuring $\mathbf{P}(t)$ in the longitudinal field (LF) configuration reveals information on the internal field distribution and its dynamics. In the LF configuration the external field is applied parallel to the initial muon spin direction which is taken to be $\hat{\mathbf{z}}$, and the polarization $P_z(t)$ is measured by collecting positrons emitted parallel and antiparallel to $\hat{\mathbf{z}}$.

Due to a combination of static fields and dynamic fluctuations, $P_z(t)$ is expected to relax. In the case of purely static field distribution, relaxation occurs due to dephasing since different muons experience different magnetic fields and therefore precess with different frequencies. In this case not all the muons precess since some of them (1/3 of the ensemble) reside in sites where the local field points either parallel or antiparallel to their initial spin direction. These muons do not depolarize and in the absence of an external LF $P_z(t)$ recover to 1/3 at $t \to \infty$. The application of a LF only increases the relative number of muons experiencing a field parallel to their polarization and, as a result, the terminal value of the polarization increases. In the case of purely dynamic fluctuations, relaxation occurs due to the absorption of energy quanta by the muon spin Zeeman levels resulting in a spin flip. In this case the relaxation depends on the system's spectral density at the muon Larmor frequency. In real systems, the dominant source of relaxation often varies with temperature between the dynamic and the static mechanism. It is therefore important to find a model which incorporates sources of both dynamic and static relaxation as well as an external longitudinal field.

The semiclassical dynamical Kubo-Toyabe (DKT) model incorporates all of the above mentioned features and provides a practical relaxation function.[2,3] This function is obtained phenomenologically in two steps. First, the muon polarization function is evaluated according to an assumed static field distribution, typically Gaussian or Lorentzian. Next, the effect of dynamics is introduced via the strong collision model. In this model the polarization propagates in time according to the static relaxation process until a collision takes place. After the collision the relaxation resumes but with the initial polarization for the new interval taken as final polarization of the previous interval. This type of dynamical process (Markovian) leads to an exponential decay of the time-dependent field-field correlation function $\langle B_i(t)B_i(0)\rangle$. The Kubo-Toyabe formalism can be applied to any field distribution or collision rate and it yields the expected recovery at very slow dynamics. However, the DKT result is not analytical. Therefore, data analysis with the DKT model is somewhat difficult; it involves the storage of large multidimensional tables of relaxation functions. The lack of an analytical expression also limits our understanding of the polarization function.

In order to obtain an analytical expression for the relaxation function we apply the perturbation treatment developed by McMullen and Zaremba (MZ) in Ref. 4 to the same dynamical input of the DKT model. Our expression is therefore valid only in a limited range of parameters, as expected from a perturbation expansion. In order to illustrate the power of this approach we also discuss the polarization function in a spin glass.

MZ wrote the polarization $P_z(t)$ as a perturbation series

$$P_z(t) = \langle\sigma_z(t)\rangle = \langle\sigma_z(t)\rangle_{(0)} + \langle\sigma_z(t)\rangle_{(1)} + \langle\sigma_z(t)\rangle_{(2)} + \cdots, \quad (1)$$

where

$$\langle\sigma_z(t)\rangle_{(0)} = P_z(0),$$
$$\langle\sigma_z(t)\rangle_{(1)} = 0,$$
$$\langle\sigma_z(t)\rangle_{(2)} = P_z(0)\frac{-\gamma_m^2}{4}\int_0^t d\tau(t-\tau) \times[e^{i\omega_L\tau}\Phi_{+-}(\tau) + e^{-i\omega_L\tau}\Phi_{-+}(\tau)],$$

and where

$$\Phi_{ij}(t'-t'') = \langle B_i(t')B_j(t'') + B_j(t'')B_i(t')\rangle_0.$$

In these equations $B_i(t)$ denotes the ith spatial component of the time-dependent fluctuating field; $B_\pm = B_x \pm iB_y$; γ_m is the muon gyromagnetic ratio; $\omega_L = \gamma_m B_L$ is the Larmor precession frequency in the LF B_L; and $\langle\rangle_0$ is the thermal average with respect to the states of the system in the absence of the muon.[4] This expansion is derived from basic principles (a magnetic Hamiltonian) and no assumptions are made regarding the field distribution. The only assumption needed to complete the calculation concerns the field-field correlation function $\Phi_{ij}(\tau)$. We assume the same time dependence of $\Phi_{ij}(\tau)$ as in the DKT model, namely,

$$\Phi_{ij}(\tau) = 4(\Delta^2/\gamma_m^2)\exp(-\nu\tau), \quad (2)$$

where Δ/γ_m is the second moment of the instantaneous field distribution, and ν is the inverse correlation time. Equation (1) is an expansion in products of the internal magnetic field, which is on the scale of Δ, and time, which is on the scale of $1/\nu$. Therefore it is an expansion in Δ/ν and is expected to be a good approximation for $\Delta/\nu < 1$. We also expect Eq. (1) to give an accurate account for the relaxation at $\nu t < 1$ since the nth term in Eq. (1) involves n integrations in time, and its contribution is proportional to the volume of integration $(\nu t)^n$. The requirements on the parameter ν prevent us from taking the limits $\nu \to 0$ and $t \to \infty$ simultaneously. Therefore we cannot discuss the static fluctuation case at $t \to \infty$ and we do not expect to find the 1/3 recovery in this theory. If we now write the polarization as

$$P_z(t) = P_z(0)\exp\left[-\Gamma(t)t\right], \quad (3)$$

expand it in powers of Γ, and compare it with Eq. (1) after the integration, we find that

$$\Gamma(t)t = 2\Delta^2 \frac{\{[\omega_L^2+\nu^2]\nu t + [\omega_L^2-\nu^2][1-e^{-\nu t}\cos(\omega_L t)] - 2\nu\omega_L e^{-\nu t}\sin(\omega_L t)\}}{(\omega_L^2+\nu^2)^2}, \quad (4)$$

which is our desired analytic result (AR). We can see from this formulation that, in general, the polarization relaxes; however, some oscillations of frequency ω_L exist near $\nu t \to 0$.

We now examine the behavior of Eq. (4) in three limits: (I) zero field ($\omega_L \to 0$), (II) the fast fluctuation regime ($\nu t \to \infty$ and $\nu > \Delta$), and (III) early times ($\nu t \to 0$ and $\omega_L t \to 0$).

(I) In the zero field limit we find that Eq. (4) simplifies to

$$\Gamma(t)t = \frac{2\Delta^2}{\nu^2}(e^{-\nu t} - 1 + \nu t).$$

This relaxation form is equivalent to the well known Abragam relaxation function for the transverse field configuration,[5] apart from an overall factor of 2. This factor of 2 is expected since in the longitudinal configuration both $\hat{x}$ and $\hat{y}$ fluctuations contribute to the relaxation, whereas in the transverse configuration only fluctuations in the $\hat{z}$ direction cause relaxation.

(II) In the fast fluctuation limit we find

$$\Gamma(t)t = \frac{2\Delta^2\nu}{\omega_L^2+\nu^2}t \quad (5)$$

and the relaxation has an exponential shape. The relaxation rate Γ is simply given by the Fourier transform of Eq. (2) evaluated at ω_L. This demonstrates how the relaxation rate, in the fast fluctuation limit, depends on the spectral density at the Larmor frequency. The same result is obtained by the DKT theory in this limit.[3] Since the muon lifetime (2.2 μsec) restricts the value of t to less than 10 μsec, this limit is valid only for fluctuations $\nu \gg 0.1$ μsec^{-1}.

(III) In the early time limit we find

$$\Gamma(t)t = \Delta^2 t^2 + O(t^3), \quad (6)$$

producing a Gaussian relaxation. We see that the relaxation at the early time is independent of both the external longitudinal field and the fluctuation rate; it depends on the second moment of the field distribution. In this case the DKT theory and the AR also agree.[3] A typical experiment limits the time range to $t > 10$ nsec; therefore the early time limit materializes only if $\nu \ll 0.1$ nsec^{-1} and $\omega_L \ll 1$ T (independent of Δ).

When $\nu \simeq \Delta$ we expect some differences between the AR and the DKT model. To distinguish the two functions we provide Figs. 1(a) and 1(b) in which $P_z(t)$ is obtained with the AR and the DKT model, respectively, for $\nu = \Delta$ and several longitudinal fields. The DKT model in this figure is obtained from the tables currently used at the μSR facility in TRIUMF for data analysis when a Gaussian field distribution is assumed. In Fig. 1(c) we also show the polarization function obtained by directly integrating the equation

$$\frac{d\mathbf{P}(t)}{dt} = \gamma_m\left[\mathbf{P}(t) \times (\mathbf{B}(t) + B_L\hat{\mathbf{z}})\right],$$

where

$$\mathbf{P}(0) = \hat{\mathbf{z}}.$$

The field $\mathbf{B}(t)$ is determined by Monte Carlo (MC) simulation in a cyclic order with two steps. In the first step a field $\mathbf{B}$ is selected randomly with a Gaussian distribution of width Δ^2/γ_m^2; in the second step a time t is selected randomly with probability distribution $\nu\exp(-\nu t)$. The

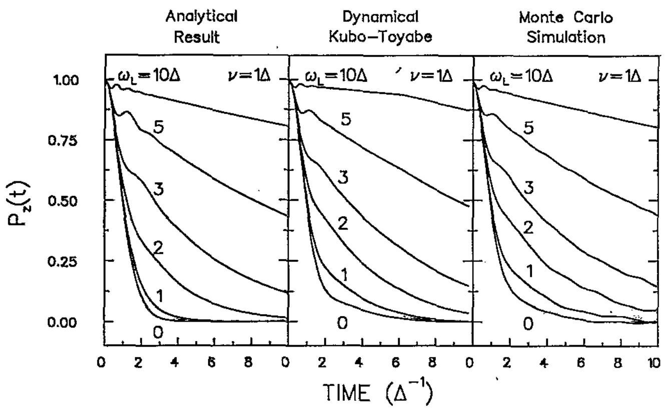

FIG. 1. Muon relaxation function in the longitudinal field configuration obtained in three different models: (a) the perturbation expansion (analytical result), (b) the dynamical Kubo-Toyabe model with a Gaussian field distribution, and (c) Monte Carlo simulations described in the text. The longitudinal field is given by $H_L = \omega_L/\gamma_m$.

field is kept constant during the time t after which we return to the first step. The final polarization function is obtained by averaging over an ensemble of separate muons. The simulations shown in Fig. 1(c) were made using 10 000 fictitious muons and $\nu = \Delta$. Comparing the three theories in Fig. 2 we observe that the DKT model resembles the simulation more closely in a low LF, while the analytical result better describes the simulation in a high LF. We also observe that the AR relaxes somewhat faster then the DKT function and is harder to decouple with a longitudinal magnetic field. Otherwise, the three methods yield very similar results even for such a large value of the expansion parameter Δ/ν; in fact, they are probably experimentally indistinguishable when we consider the typical resolution of the μSR technique. We note that oscillations near $\nu t \to 0$ are present in all three methods, and that the early time behavior is universally independent of ω_L.

In order to demonstrate the application of the AR to a real system, we use the data obtained by Luck *et al.* in a LF-μSR experiment on copper (Cu).[6] In Cu, the local field originates from random nuclear moments, and dynamical fluctuations occur due to muon diffusion, which mainly take place in two temperature regions: At high temperatures ($T > 100$ K) the diffusion is generated by thermal excitation; at very low temperatures ($T < 1$ K) the diffusion is dominated by quantum tunneling.[7] In the intermediate range, the muon, and therefore the local field, is static. In Figs. 3(a) and 3(b) we show the μSR data in Cu at 45 K (static region) and 150 K (dynamic region), respectively. The solid lines represent a fit of the AR. At 45 K we find a clear recovery of the polarization at $t \to \infty$, and, as expected, the terminal value of the polarization increases with increasing LF. At this temperature the condition $\Delta/\nu < 1$ is not valid, and we can apply the AR only to the polarization at the early time ($t < 1$ μsec). On the other end, when the condition $\Delta/\nu < 1$ is valid, as in the case shown in Fig. 3(b), no recovery is observed and the AR fits the data in the entire time range. In the fits shown in Fig. 3(b), ω_L is determined by the external field (within experimental certainty), and Δ and ν are global parameters. As is evident in the figure the agreement in this instance between theory and experiment is satisfactory.

Having discovered the analytical form for the relaxation function, Eq. (4), we can now apply it to spin glasses. In a spin glass, such as CuMn or AuFe, the magnetic impurities are located randomly in the sample.[8] The random distance between the muon and the impurities results in a distribution of second moments. Therefore we treat the parameter Δ as a statistical variable. As an example, let us consider the case of a dilute spin glass at

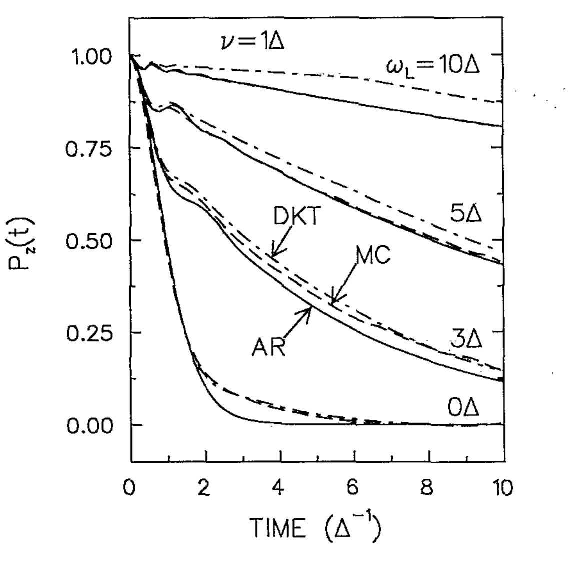

FIG. 2. The analytic result (AR) obtained by the perturbation expansion compared with the dynamical Kubo-Toyabe (DKT) relaxation function with a Gaussian field distribution, and Monte Carlo (MC) simulation for several longitudinal fields.

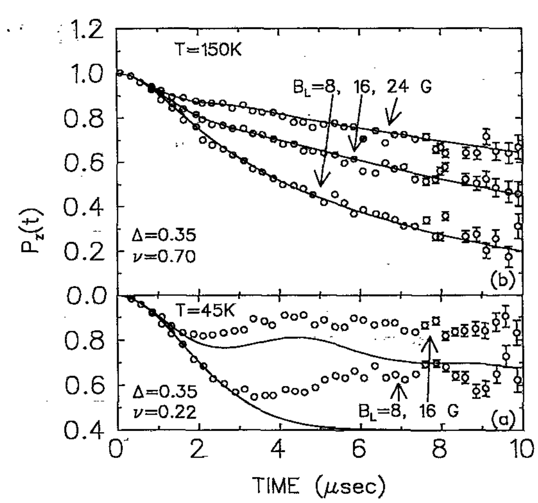

FIG. 3. Longitudinal field μSR in copper at $T = 45$ K (a) where the muon is slowly diffusing ($\nu < \Delta$) and Eq. (4) is applicable only at early time, and at $T = 150$ K (b) where the muon diffuses quickly ($\nu > \Delta$) and Eq. (4) fits the data well.

temperatures above the freezing temperature T_g. In this case, Uemura *et al.*[8] demonstrated that the distribution of Δ can be approximated by

$$\rho(\Delta) = \sqrt{2/\pi}\frac{a}{\Delta^2}\exp\left(-\frac{a^2}{2\Delta^2}\right). \quad (7)$$

The spin glass relaxation function is obtained by averaging Eq. (3) with respect to Δ. In order to maintain the condition $\Delta < \nu$, the parameter a must satisfy $a < \nu$. The resulting Uemura function is

$$P_z^{\rm SG}(t) = \int_0^\infty \exp[-\Gamma(t)t]\rho(\Delta)d\Delta$$
$$= \exp\left(-\sqrt{2a^2\gamma(t)t}\right), \quad (8)$$

where

$$\gamma(t) \equiv \Gamma(t)/\Delta^2.$$

Using $\Gamma(t)$ from Eq. (5) we see that, in the fast limit, the relaxation has a root-exponential shape. $\Gamma(t)$ given by Eq. (6) leads to an exponential relaxation in the early time limit. In Fig. 4 we show the polarization function obtained with Eq. (8) for $\nu = a$. From the figure we observe that, in the spin glass case, the LF is less effective in recovering the polarization than in the case of a field distribution with a unique width. This slow recovery is a consequence of the high field tail resulting from the convolution of the field distribution and Eq. (7). We also note, in Fig. 4, the exponential relaxation at the early times and the presence of oscillations with frequency ω_L near $\nu t \to 0$.

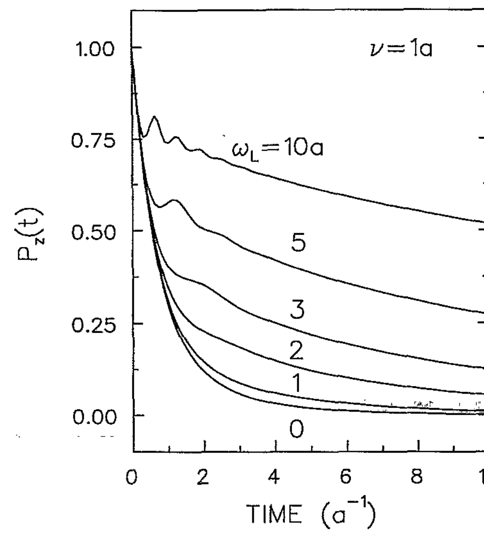

FIG. 4. Muon relaxation function in a spin glass in the longitudinal field configuration as given by Eq. (8). The longitudinal field as given by $H_L = \omega_L/\gamma_m$.

In conclusion, we have found a new analytical relaxation function which incorporates static and dynamic field fluctuations in the longitudinal field configuration. We made no assumptions regarding the instantaneous field distribution. The validity of this function is limited to either $\nu > \Delta$ or the early time.

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