The Abdus Salam International Centre for Theoretical Physics

United Nations Educational, Scientific and Cultural Organization

IAEA International Atomic Energy Agency

1855-10

School and Workshop on Highly Frustrated Magnets and Strongly Correlated Systems: From Non-Perturbative Approaches to Experiments

30 July - 17 August, 2007

Muon Spin Relaxation in Frustrated Magnets

Amit Keren
Technion, Haifa, Israel

Muon Spin Relxation in Frustrated Magnets

Amit Keren

Technion

Abstract

A tutorial review of the major aspects of muon spin relaxation functions, with zero or longitudinal external field, and dynamic fluctuations is given. The emphasis is on frustrated magnets.

The muon spin spectroscopy technique operates in zero applied field. Therefore, standard perturbation methods to analyze the data, where the external field is considered large and the internal fields small, do not apply. Consequently, different methods are required to account for the muon relaxation function in zero and small external fields. Here, we lead the reader in three steps to a closed form equation which can be used to generate muon relaxation function under a variety of situations including the zero external field case, which is highly non-perturbative. In flight we explain the main feature of such functions. We then use this information to examine muon relaxation function in frustrated magnets.

I. RELAXATION FUNCTION IN A STATIC FIELD DISTRIBUTION

The fully polarized muon, after entering the sample, comes to rest in a magnetic environment. Since the mechanism which stops the muon is much stronger than any magnetic interaction, the muon maintains its polarization while losing its kinetic energy. Once the muon reaches its site, the muon spin starts to evolve in the local field $\mathbf{B}$. The muon polarization P_z along the $\hat{\mathbf{z}}$ direction is given by the double projection expression

$$P_z(\mathbf{B}, t) = \cos^2\theta + \sin^2\theta \cos(\gamma_\mu |\mathbf{B}| t) \tag{1}$$

where θ is the angle between the initial muon spin and the local field direction (see Fig. 1). This angle is related to the field values by

$$\begin{aligned} \cos^2\theta &= \tfrac{B_z^2}{\mathbf{B}^2} \\ \sin^2\theta &= \tfrac{B_x^2+B_y^2}{\mathbf{B}^2} \end{aligned}.$$

In a real sample, however, there will be a distribution of internal fields and the averaged polarization is written as

$$\overline{P}_z(t) = \int \rho(\mathbf{B}) \left[\frac{B_z^2}{\mathbf{B}^2} + \frac{B_x^2 + B_y^2}{\mathbf{B}^2} \cos(\gamma_\mu |\mathbf{B}| t) \right] d^3\mathbf{B} \tag{2}$$

where $\overline{P}_z(t)$ is the sample averaged polarization, and $\rho(\mathbf{B})$ is the field distribution which must obey

$$\int \rho(\mathbf{B}) d\mathbf{B}^3 = 1.$$

If the distribution of internal fields is only a function of $|\mathbf{B}|$ then we can write

$$\overline{P}_z(t) = \int \rho(|\mathbf{B}|) \left[\cos^2\theta + \sin^2\theta \cos(\gamma_\mu |\mathbf{B}| t) \right] B^2 dB d\Omega.$$

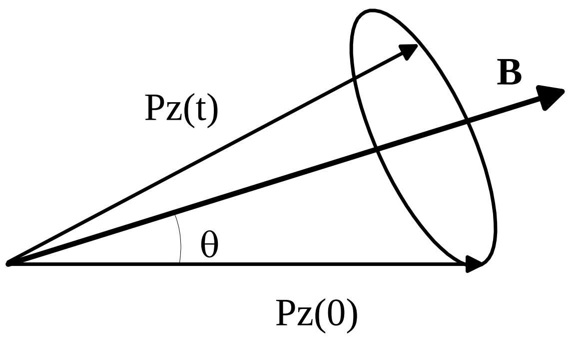

FIG. 1: Muon spin polarization rotating around a magnetic field in an arbitrary direction.

It is convenient to define

$$\rho'(|\mathbf{B}|) = 4\pi\rho(|\mathbf{B}|)$$

so that

$$\int \rho'(|\mathbf{B}|)\mathbf{B}^2 dB = 1.$$

and the angular dependence can be integrated out giving

$$\overline{P}_z(t) = \frac{1}{3} + \frac{2}{3}\int \rho'(|\mathbf{B}|)\cos(\gamma_\mu\,|\mathbf{B}|\,t)B^2 dB.$$

If, for example, the absolute value of the local field experienced by a muon is unique then

$$\rho'(|\mathbf{B}|) = \left(\frac{\gamma_\mu}{\omega_0}\right)^2 \delta(|\mathbf{B}| - \frac{\omega_0}{\gamma_\mu}).$$

This can happen in a powder of a ferromagnet or antiferromagnet (AFM) where there is only one muon site. In this case

$$P_z^{\omega_0}(t) = \frac{1}{3} + \frac{2}{3}\cos(\omega_0 t).$$

The time independent $1/3$ component represent muon with their initial polarization pointing effectively along the local field direction. These muons do not change their polarization.

In real systems, however, the local field experienced by different muons is rarely unique. It can vary from site to site as a result of nuclear moments, impurities, defects, or non-homogeneous freezing of the ionic moments. If, for example,

$$\rho'(|\mathbf{B}|) = \frac{\gamma_\mu}{\sqrt{2\pi}\Delta\mathbf{B}^2}\exp\left[-\gamma_\mu^2\left(|\mathbf{B}| - \frac{\omega_0}{\gamma_\mu^2}^2\right)/2\Delta^2\right]$$

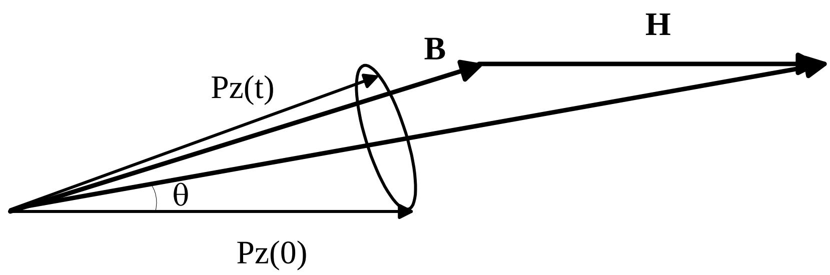

FIG. 2: Muon spin polarization rotating around the vector sum of an external magnetic field in the initial muon polarization direction, and an arbitrary internal field.

then

$$\overline{P}_z^{\Delta,\omega_0}(t) = \frac{1}{3} + \frac{2}{3}\exp\left(-\frac{\Delta^2 t^2}{2}\right)\cos(\omega_0 t).$$

When a longitudinal field (LF) is applied the situation becomes more complicated, and there is no closed form expression. However, some simplifications could be made to reduce the dimension of the integrals for the purpose of numerical calculations. For example, if the local field is completely random with a Gaussian distribution, and a field H is applied in the initial muon spin direction as in Fig. 2, then

$$\rho^{\mathrm{LF}}(\mathbf{B}) = \frac{\gamma_\mu^3}{(2\pi)^{3/2}\Delta^3}\exp\left(-\frac{\gamma_\mu^2[\mathbf{B}-H_{\mathrm{L}}\hat{\mathbf{z}}]^2}{2\Delta^2}\right). \tag{3}$$

In this case Eq. 2 could be simplified to [1]

$$\overline{P}_z^{\Delta}(\omega_{\mathrm{L}},t) = 1 - \frac{2\Delta^2}{(\omega_{\mathrm{L}})^2}\left[1-\exp(-\frac{1}{2}\Delta^2 t^2)\cos(\omega_{\mathrm{L}} t)\right] + \frac{2\Delta^4}{(\omega_{\mathrm{L}})^3}\int_0^t \exp(-\frac{1}{2}\Delta^2\tau^2)\sin(\omega_{\mathrm{L}}\tau)d\tau \tag{4}$$

where

$$\omega_{\mathrm{L}} = \gamma_\mu H.$$

This is known as the static-Gaussian-longitudinal-field Kubo-Toyabe (KT) function. Figure 3 shows $\overline{P}_z^{\Delta}(\omega_{\mathrm{L}},t)$ for a variety of ω_{L}. Interestingly, despite the fact that the external field is in the muon spin direction, wiggles are seen in the polarization, and their frequency is given by ω_{L}. When $\omega_{\mathrm{L}} \gg \Delta$ the muon does not relax any more. This is because the field at the muon site is nearly parallel to the initial muon spin direction. Finally, in the zero field case ($H_{\mathrm{L}} = 0$) Eq. 4 reduces to [1]

$$\overline{P}_z^{\mathrm{LF}}(0,t) = \frac{1}{3} + \frac{2}{3}(1-\Delta^2 t^2)\exp(-\frac{1}{2}\Delta^2 t^2). \tag{5}$$

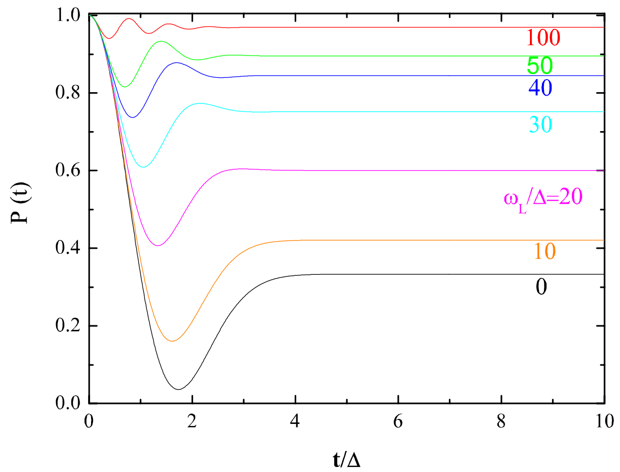

FIG. 3: Muon polarization function in a Gaussian internal field distribution and external field pointing in the initial muon spin direction. Different values of the external field H are shown.

This polarization function is known as the static-Gaussian-zero-field KT. At early time it has a Gaussian like behavior. It reaches a minimum on a time scale set by Δ after which it recovers and saturates again at $1/3$.

II. WHEN THE FIELD IS DYNAMIC

We start with the simplest solvable model for the muon polarization in an environment where the field dynamically fluctuates. Our solvable model considers a field at the muon site $\mathbf{B}$ exactly perpendicular to the $\hat{\mathbf{z}}$ direction, which flips with time between the up and down directions but maintains its absolute value. The $\hat{\mathbf{z}}$ direction is taken to be the initial muon spin direction and also the direction in which the polarization is measured. As a result the muon spin rotates with frequency $\pm\omega$. The polarization resulting from three such field flips is demonstrated in Fig. 4.

This can happen if a sample is an antiferromagnet (AFM) and the muon hops between different sites of opposite fields. We assume that the field fluctuation could be described by a flip rate probability per unit time ν. We further define ν_t as the total hop rate. If each

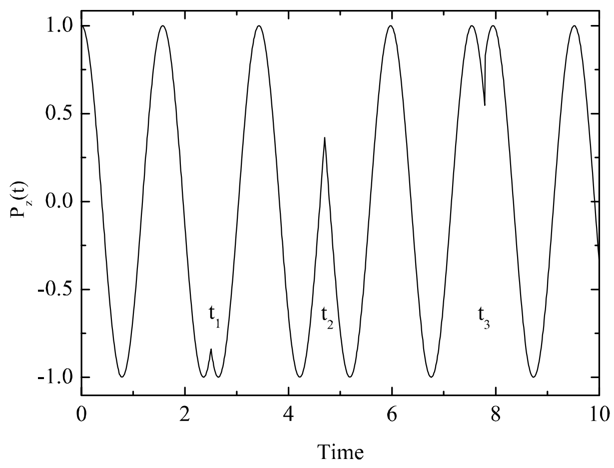

FIG. 4: A demonstration of the muon polarization in the $\widehat{\mathbf{z}}$ (initial) direction in the antiferromagnetic model in which the field is perpendicular to $\widehat{\mathbf{z}}$ but flips while maintaining its magnitude. In this figure three field flips are taking place.

time the muon hops it has the same chance as not of experiencing a field change then

$$\nu = \nu_t/2. \tag{6}$$

This assumption is called the strong collision approximation, and will only apply if the muon hops over long distances compared to the unit cell. For a discussion of the relaxation rate in this situation without the strong collision approximation (and arbitrary field distribution) see Ref. [2]. In our situation the field correlation function after the short interval $d\tau$ is given by

$$\langle B(d\tau)B(0)\rangle = B^2(1-\frac{\nu_t}{2}d\tau) - B^2\frac{\nu_t}{2}d\tau = B^2(1-\nu_t d\tau)$$

where $\langle\rangle$ stands for average. As can be proven by induction, after time τ

$$\langle B(\tau)B(0)\rangle = B^2\exp(-\nu_t\tau). \tag{7}$$

Thus the field correlation function decays exponentially with a correlation time of $1/\nu_t$.

For n hops between sites of opposing fields, at times $t_1 < ... < t_n < t$, the polarization function g_n is given by

$$g(t_1,...,t_n,t) = Re\exp(i\sum_{j=1}^{n+1}[-1]^{j+1}\omega[t_j - t_{j-1}]) \tag{8}$$

where $t_{n+1} = t$. In this form it is clear that

$$g(t_1, ..., t_n, t) = Re \prod_{j=1}^{n+1} g_j(t_j - t_{j-1}) \tag{9}$$

where

$$g_j(t_j - t_{j-1}) = \exp([-1]^{j+1} i\omega[t_j - t_{j-1}]).$$

Next we calculate the probability that a field flip will occur at a time t_{i+1}, given that a previous change occurred at time t_i. For this we divide the time segment $t_{i+1} - t_i$ into m steps each dt long and take $m \to \infty$. This gives the probability

$$\lim_{m\to\infty} \left[1 - \nu \frac{t_{i+1} - t_i}{m}\right]^m \nu dt = e^{-\nu(t_{i+1}-t_i)} \nu dt.$$

Therefore, the probability for n field flips in the time segments $[t_1, t_1 + dt_1], ..., [t_n, t_n + dt_n]$ is

$$\prod_{i=1}^{n} \exp[-\nu(t_i - t_{i-1})] \nu dt_i = \nu^n \exp(-\nu t) \prod_{i=1}^{n} dt_i.$$

The averaged polarization is obtained by taking the sum over all possible numbers of field flips, weighted by their probability, and integrating over the times in which they can take place [1]. This leads to

$$P_z^{AFM}(t) = e^{-\nu t} g(t) + \nu e^{-\nu t} \int_0^t dt_1 g(t_1, t) + \nu^2 e^{-\nu t} \int_0^t dt_2 \int_0^{t_2} dt_1 g(t_1, t_2, t) + ... \tag{10}$$

where $g_n(t_1, ..., t_n, t)$ is given by 8. In the simple case presented here the series could be summed [3] and it leads to

$$P_z^{AFM}(t) = c_+ e^{z_+ t} + c_- e^{z_- t}, \tag{11}$$

where

$$c_\pm = \frac{1}{2} \pm \frac{\nu(\nu^2 - \omega^2)^{-\frac{1}{2}}}{2}$$

and

$$z_\pm = -\nu \pm (\nu^2 - \omega^2)^{\frac{1}{2}}.$$

Note that this result is correct regardless of the strong collision approximation of Eq. 6.

The polarization P_z^{AFM}, for selected values of ν/ω is shown in Fig. 5. Clearly, as the field flip rate increases the oscillations at frequency ω disappear. When $\nu/\omega \to \infty$ the muon behaves as if it experiences zero field, which is the average field. This is the μSR

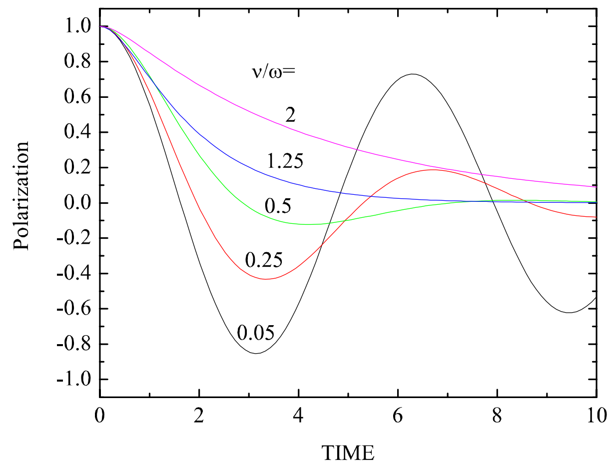

FIG. 5: The expected muon spin polarization in the antiferromagnetic model for various ratios of flip rates to oscillation frequency.

manifestation of motional narrowing. Another important aspect of this model is that when $\nu \gg \omega$ the polarization could be approximated by

$$P_z^{AFM}(t) = \exp\left(-\frac{\omega^2}{\nu_t}t\right). \tag{12}$$

This is usually refered to as Lorenzian relaxation. In contrast, when $\nu \lesssim \omega$ the relaxation is Gaussian like at early time. The value of $\omega^2 = \gamma_\mu^2 B^2$ could be estimated from the initial relaxation rate, as in the completely static case.

III. ALL INGREDIENTS COMBINED

In the presence of a dynamic and longitudinal field, numerical methods must be applied. For this purpose it is useful to write the infinite series of Eq. 10 in a compact form. We will now show that this series obeys the equation

$$P_z(\nu,t) = e^{-\nu t}g(t) + \nu\int_0^t dt' P_z(\nu,t-t')e^{-\nu t'}g(t') \tag{13}$$

known as the Volterra equation of the second kind. This equation could be solved numerically [4]. To verify the equivalence between Eq. 13 and Eq. 10, we first note that the first term on the right hand side (r.h.s) of Eq. 13 is the same as the first term of Eq. 10. Next we

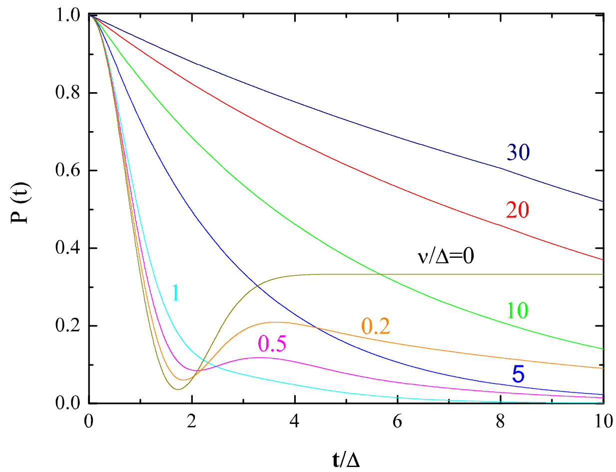

FIG. 6: Expected muon relaxation in a dynamic field with Gaussian instantaneous distribution and no external field. Different values of fluctuation rates are shown.

generate a sister equation to Eq. 13 by substituting $t' \to t''$ followed by $t \to t - t'$ in this equation. Finally, we replace $P_z(\nu, t-t')$ under the integral in Eq. 13 by the sister equation. This leads to

$$P_z(\nu, t) = e^{-\nu t} g(t) + \nu e^{-\nu t} \int_0^t dt' g(t-t') g(t') + \ldots$$

if $g(t_1, t) = g(t - t_1) g(t_1 - 0)$ (strong collisions), so that now both the first and the second terms on the r.h.s of Eq. 13 and Eq. 10 agree. Repeating this operation on Eq. 13 will regenerate Eq. 10.

The Volterra equation gives a good description of the dynamics only when the strong collision approximation, manifested in Eq. 9, is valid. The input to the Volterra equation is the static function $g(t)$. Therefore, there are three ways of using Eq. 13 to obtain dynamic information. The first one is in simple cases where $g(t)$ is known analytically as was done by Brewer *et al.* [5] for F-μ-F bond. The second one is when $g(t)$ must be obtained numerically as in the cases of Gaussian [1] or Lorenzian [6] field distribution with external longitudinal field. The third way is to measure $g(t)$ by cooling the system to low enough temperatures that dynamic fluctuations are no longer present, and to use the measured $g(t)$ in the Volterra equation [7].

Now we are in position to take one of our polarizations generated by static field distrib-

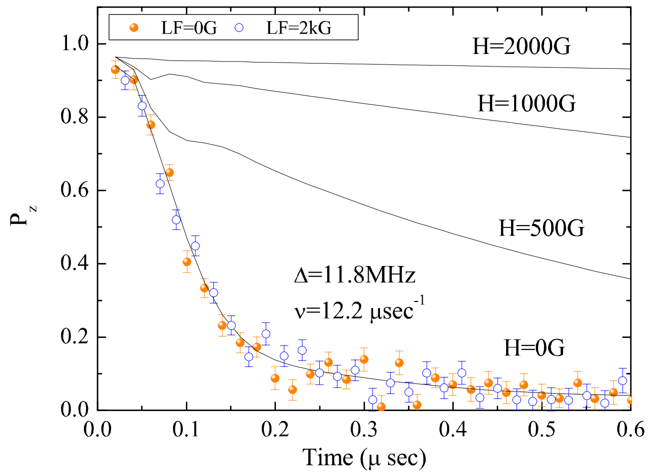

FIG. 7: Solid lines: Expected muon relaxation in a combination of internal field fluctuations with Gaussian instantaneous distribution, and longitudinal external field H. Symboles: Relaxation data from Ref. [9] in two different values of H. The disagreement between data and model indicates unusual behavior.

ution, say Eq. 4, use it as an input in the Voltera equation, and see how it behaves if the magnetic field fluctuates between different values. For example, if we take Eq. 5 as an input to Eq. 13 we obtain the polarizations shown in Fig. 6.

Finally, in Fig. 7 we present the most complicated relaxation function combining Gaussian field distribution, fluctuations, and longitudinal field. We have chosen a special value of the parameters Δ, ν and H, for reasons that will become clear soon. It is interesting to mention that for the case $\nu \gtrsim \Delta$ there is an approximate expression for this relaxation function, which is given by

$$\overline{P}_z(t) = \exp(-\Gamma(t)t)$$

where

$$\Gamma(t)t = \frac{2\Delta^2}{(\omega_{\rm L}^2+\nu^2)^2}\left\{[\omega_{\rm L}^2+\nu^2]\nu t + [\omega_{\rm L}^2-\nu^2][1-e^{-\nu t}\cos(\omega_{\rm L} t)] - 2\nu\omega_{\rm L} e^{-\nu t}\sin(\omega_{\rm L} t)\right\}.$$

It approximate the exact function, even at $H = 0$, much better than is expected [8].

IV. MUON RELAXATION IN FRUSTRATED MAGNETS

At the very beginning of the research in the field of frustrated magnets it was noticed that the muon relaxation function is unusual. The symbols in Fig. 7 show the polarization at a temperature of 100 mK in the kagome system SCGO in zero field and a longitudinal field of 2 kG [9]. First, no oscillations are found, so the internal field is random with either static or dynamic nature. Second, the relaxation at early time is Gaussian, with a time scale of 0.1 μsec, so Δ must be on the order of 10 MHz. Third, there is no recovery so there must be some dynamic as in Fig. 6. But it must be that $\nu \sim \Delta$. If ν had been larger, the initial relaxation would have been Lorenzian (See Eq. 12 and Fig. 6). If ν had been much slower, the polarization would have recovered. Therefore, we conclude that $\nu \sim \Delta$ ~10 MHz. In these circumstances a field of 2 kG which is equivalent to $\omega_L = 170$ MHz should have "decoupled" the relaxation. This is not happening. The solid lines in Fig. 7 represent the expected decoupling which is very different from the observed one. Until today only one model has been proposed to explain this problem [9], which received the name sporadic adynamic (SD) [10].

In this model, the system is not magnetic most of the time, and magnetic with dynamic fluctuations only a fraction f of the time. In zero field it is clear that such a case will lead to $\overline{P}_z^{\text{sd}}(0,t) = \overline{P}_z^{\Delta,\nu}(0,ft)$. However, even when the H is applied, the polarization changes only when the internal field is on. Therefore even in this case $P_z^{\text{sd}}(\omega_L,t) = \overline{P}_z^{\Delta,\nu}(\omega_L,ft)$. Since ω_L and t always enter the relaxation function as a product we must have $\overline{P}_z^{\text{sd}}(\omega_L,t) = \overline{P}_z^{f\Delta,f\nu}(f\omega_L,t)$. What we actually estimated from the data to be 10 MHz is $f\Delta$, and $f\nu$. When the field is on Δ and ν are much higher than 10 MHz. In other words, the effect of the longitudinal field is reduced by factor f. This is the reason we do not see decoupling. This model is very successful in explaining μSR data.

The problem with this model is that the muon relaxation is temperature independent when the Gaussian relaxation is observed. Therefore, the system is in its ground state. In the ground state the system can not have time evolution, namely, the field cannot turn on and off. Also, NMR measurements when available tell a different story.

---

[1] R. S. Hayano, Y. J. Uemura, J. Imazato, N. Nishida, T. Yamazaki, and R. Kubo, Phys. Rev. B **20**, 850 (1979).

[2] J. R. Klauder and P. W. Anderson, Phys. Rev. **125**, 912 (1962).

[3] A. Keren, L. P. Le, G. M. Luke, B. J. Sternlied, W. D. Wu, Y. J. Uemura, S. Tajima, S. Uchida, Phys. Rev. B **48**, 12926 (1993).

[4] W. H. Press, B. P. Flannery, A. A. Teukolsky, and W. T. Vetterling, *Numerical Recipes* (Cambridge University Press, Cambridge, 1989).

[5] J. H. Brewer, D.R. Harshman, R. Keitel, S. R. Kreitzman, G. M. Luke, D. R. Noakes, and R. E. Turner, Hyperfine Interactins **32**, 677 (1986).

[6] Y. J. Uemura T. Yamazaki D. R. Harshman M. Senab, and E. J. Ansaldo, Phys. Rev. B **31** (1985).

[7] A. Keren, J. Phys.: Condens. Matter **16**, 1 (2004).

[8] A. Keren, Phys. Rev. B **50**, 10039 (1994).

[9] Y. J. Uemura *et al.*, Phys. Rev. Lett. **73**, 3306 (1994).

[10] D. Bono, *et al.* Phys. Rev. Lett. **93**, 187201 (2004).