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A correlation length measured by zero-field muon spin relaxation in disordered magnets

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Abstract. Muon spin relaxation, as a local magnetic probe, is not normally expected to be able to provide measurements of any magnetic correlation length. When anomalously shallow static zero-field muon spin relaxation functions were observed for $CeNi_{1-x}Cu_xSn$ and a small number of other highly disordered magnetic systems, however, and a 'Gaussian-broadened Gaussian' relaxation function shown to fit them, it was suggested that an unusual short-range correlation among local moments in the material was acting. 'Range-correlated moment magnitude variation', in which ion-moment orientations are completely random, but moment magnitude varies slowly from place to place, is shown by Monte Carlo numerical simulation on a lattice of moments to generate the qualitative form of shallow static zero-field relaxation observed. Thus, the *shape* of the zero-field muon spin relaxation function is sensitive to a correlation length.

1. Introduction

Positive-muon spin relaxation (μSR) is a probe of local magnetic fields in materials (for introductory reviews, see [1]) that is particularly useful when carried out in zero external field (ZF). The magnetic moment of a μ^+ stopped at an interstitial site in a solid precesses in the local field at that site, and then emits a positron preferentially in the direction of the μ^+-moment at the instant of decay. Knowing the initial polarization of the muons and detecting the directions of the decay positrons, the experimenter can measure the time evolution of the polarization in the direction of initial orientation, called the (longitudinal) 'muon spin relaxation function', $G_z(t)$. In ferromagnetic or antiferromagnetic states, the well-ordered array of ion moments will often generate a unique field at the muon site, causing a single muon precession frequency observed as spontaneous oscillation of $G_z(t)$ (for reviews of μSR in magnetic materials, see [2]). In cases where the magnetic moments in the material are static (not fluctuating) yet not long-range ordered (as in frozen spin glasses and conductors containing only nuclear moments), the 'Kubo–Toyabe' relaxation functions [3] corresponding to either a Gaussian local field distribution

$$G_z^G(t) = \frac{1}{3} + \frac{2}{3}(1 - \Delta^2 t^2)\exp\left(-\frac{\Delta^2 t^2}{2}\right) \tag{1}$$

(where $\Delta = \gamma_\mu (B_x)_{rms}$, and γ_μ is the muon gyromagnetic ratio) or to a Lorentzian local field distribution

$$G_z^L(t) = \frac{1}{3} + \frac{2}{3}(1 - at)\exp(-at) \tag{2}$$

(where $a = \gamma_\mu (B_x)_{hwhm}$) have been sufficient to fit all data obtained until recently. In both of these functions, polarization drops from the initial value of unity to a single minimum, and

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then recovers to 1/3 at late times. This 1/3 asymptote is a characteristic feature of muon spin relaxation in static random local fields and zero external field. It is now known, however, that in a small number of materials, including Al–Mn–Si magnetic quasicrystals [4] and $CeNi_{1-x}Cu_xSn$ Kondo-lattice alloys [5], static ZF relaxation of initial polarization can lead to a minimum too shallow to be reproduced by either of the Kubo–Toyabe functions (equation (1) or equation (2)) above. In fact, the extreme case of monotonic decay to the 1/3 asymptote has been observed.

A prior publication [6] showed that the observed relaxation in $CeNi_{1-x}Cu_xSn$ alloys is fitted well by a relaxation function derived from a 'convolution broadening' of a Gaussian distribution. This means that instead of a single-Gaussian random local field distribution with a unique value of $(B_x)_{rms}$, a collection of distributions are used, corresponding to a collection of different muon sites, where the collection of values of $(B_x)_{rms}$ themselves form a Gaussian distribution of width W and most likely value B_0. This 'Gaussian-broadened Gaussian' static ZF Kubo–Toyabe relaxation function has the form

$$G_z^{GBG}(t) = \frac{1}{3} + \frac{2}{3}\left(\frac{1+R^2}{1+R^2+R^2\Delta_{eff}^2 t^2}\right)^{3/2}\left(1 - \frac{\Delta_{eff}^2 t^2}{1+R^2+R^2\Delta_{eff}^2 t^2}\right) \times \exp\left(\frac{-\Delta_{eff}^2 t^2}{2(1+R^2+R^2\Delta_{eff}^2 t^2)}\right) \quad (3)$$

where $\Delta_{eff}^2 = \gamma_\mu^2(B_0^2 + W^2)$ and $R = W/B_0$. Previously, however, no microscopic model of moments in a solid was presented that could produce such a field distribution.

Directly, μSR 'senses' only the magnetic fields at the muon sites, and as such is a local probe that has not been expected to be sensitive to correlation lengths. In cases that require a non-trivial value of R in equation (3), however, that non-trivial value (or, equivalently, the shallowness of the minimum of polarization) is extra information not provided by the standard Gaussian or Lorentzian forms. One publication [4] had speculated that this kind of anomalous relaxation could be produced by random variation of the magnitude of the frozen moments, with a correlation range. The present paper presents the results of Monte Carlo simulations of the ZF muon spin relaxation functions generated by a microscopic model of such 'range-correlated moment magnitude variation' (RCMMV). This involves similar values of the magnitude of nearby magnetic moments that nonetheless have uncorrelated orientations. The results demonstrate that this type of correlation in a magnetically frozen (but disordered) state will generate the kind of shallow static ZF Kubo–Toyabe relaxation function that has been observed for the materials listed above. As the correlation length gets successively larger, the associated relaxation function more closely approaches monotonic relaxation to the 1/3 asymptote, as will be discussed. The extra information represented by the value of R is indeed the range of this correlation. In the model discussed here, this correlation is imposed, not generated by a plausible physical mechanism. The identification of candidate mechanisms is a subject for future research.

Throughout this discussion, it is necessary to understand that while the Lorentzian Kubo–Toyabe relaxation function has a shallower minimum than the Gaussian Kubo–Toyabe function, $G_z^L(t)$ does not fit any data of the type exemplified by the $CeNi_{1-x}Cu_xSn$ data. The Lorentzian case couples that shallower minimum with rapid initial relaxation. If forced to fit the initial relaxation observed in $CeNi_{1-x}Cu_xSn$ [5], the Lorentzian minimum occurs far later than in the data. If forced to fit the observed minimum, the initial decay of $G_z^L(t)$ is much faster than that of the data. The Gaussian-broadened Gaussian function places the minimum at essentially the same time, relative to the initial relaxation, as the standard Gaussian Kubo–Toyabe function, and does fit the data well. Since Gaussian-like behaviour is generally associated with dense-moment systems [2], a dense array of moments was specifically incorporated into the Monte Carlo model.

2. The microscopic Monte Carlo model

A Monte Carlo simulation of a microscopic model of moments on a lattice has been used before to simulate the field distribution at an interstitial muon site and the corresponding static ZF muon spin relaxation function [7]. For a cluster of a finite number of ions on a lattice, a vector magnetic moment is assigned to each magnetic ion, the assumed coupling of the moments to a muon at a particular site is used to calculate the effective field due to each ion at the muon site, and those individual-ion fields are vector summed to find the net local field at the muon for that particular moment configuration. For any particular initial muon polarization, the precession of the muon spin around the local field is calculated. Of particular interest is the projection of the polarization along its initial direction, as a function of time. This is the longitudinal relaxation function $G_z(t)$. For a single muon, this will oscillate at the Larmor frequency of the local field. If there is any disorder in the system, randomization characteristic of that disorder is included in the calculation, and the calculation is repeated many times. The many net local fields are collected into histograms representative of the local field probability distribution, and the projected polarization of many muons is averaged to generate the simulated static muon spin relaxation function.

One type of disorder that often occurs in samples studied with μSR is polycrystallinity. Most μSR experiments are performed on powders or polycrystalline ingots, where all possible orientations of the principal axes of the crystallites of the material, with respect to the initial muon polarization, occur. This averages over any anisotropy of the relaxation with respect to the crystal axes. All of the equations above assume ‘polycrystalline averaging’ (or perfect isotropy of the local field distribution). In a Monte Carlo program, this is achieved by randomizing the orientation of the lattice principal axes relative to the initial muon polarization, and this was done in every case in the simulations discussed below.

A previous publication [7] discussed Monte Carlo simulation of the static ZF relaxation functions that should be generated by non-dilute alloys, and that the path to the Lorentzian case in the dilute limit was not simple. That program used fixed-magnitude, random-direction moments in a cluster of lattice positions. Upon discovery of the anomalous, shallow-minima, static ZF relaxation functions, and with the expectation that some new type of disorder would be required in the explanation, a simple modification was made to that program which randomized the moment magnitude in addition to the orientation. The modification produced simulated field distributions and relaxation functions of the same qualitative form as those shown in the earlier paper. They evolved from the Gaussian Kubo–Toyabe function at high moment concentration, through two-minima forms (with two effective sites) at intermediate concentrations, to the Lorentzian limit at low concentrations, without producing anything that fitted the shallow relaxation functions of $CeNi_{1-x}Cu_xSn$ and magnetic Al–Mn–Si quasicrystals. *Completely random moment variation does not explain the anomalous relaxation that has been observed.*

The subject of this paper is a model that more successfully reproduces shallow static ZF relaxation functions. It was constructed to be a specific example of range-correlated moment magnitude variation, alluded to above. Retaining a lattice cluster and completely random moment orientations, moment magnitudes are held fixed in sub-clusters of moments, while they are varied randomly between sub-clusters. Forcing the sub-clusters to have a particular average size creates a correlation range. To obtain good statistical behaviour of the simulations using such sub-clusters, substantially larger overall cluster size is necessary: while the clusters of the previous Monte Carlo μSR paper [7] were spheres of up to 500 ions on fcc lattice sites, the work reported here used cubes of ions on simple-cubic lattice sites, with up to 126 ions on a side (containing, therefore, up to slightly more than two million ions). The use of cubic clusters, and a simple-cubic lattice, help reduce CPU time, as does the increase in computation speed available since that previous publication. The qualitative conclusions drawn should not depend on the particular lattice used, although fine details may well do so.

Within an overall cluster, the sub-clusters were chosen to be rectangular boxes, with each box's three side lengths chosen randomly between one ion thick and a maximum thickness (the 'cut-off', which controls the correlation range), subject to a simple packing constraint such that, in spite of the randomization, the overall cube is filled by the boxes, with no ions left out. For each box, a single value of the moment magnitude is chosen randomly between zero and unity. This rectangular-box configuration of correlated clusters is not physically likely, but is computationally simple. More realistic methods of generating correlations, usually involving some pairwise nearest-neighbour interaction, would require orders of magnitude more CPU time, which would be prohibitive. Furthermore, no physically reasonable candidate mechanisms for producing such correlations are known at this time. The simulations reported here provide a first glimpse of what RCMMV can do to static ZF relaxation functions.

More challenging, in terms of the microscopic physics, is the assumption of continuous moment variation between zero and a maximum. Ion magnetic moments in solids are usually single valued, and cases of static moment instability are usually discussed in terms of (usually only two) distinct values ('mixed valence'; see, e.g., [8]). A continuous range, or at least a large number of possible values, of moment magnitude seems necessary to generate shallow static ZF relaxation. Use of the models described here with only two possible moment values generates field distributions and relaxation functions clearly representative of two muon sites with two particular rms field values, and does not reproduce the shallow static ZF relaxation observed. The behaviour discussed here might well be generated by the size of the magnetic ion moment taking a distinct and unique value (not just on/off) for each and every different near-neighbour environment that occurs in the material. For a site-substitution alloy like $CeNi_{1-x}Cu_xSn$, this would result in a binomial distribution of moment sizes. Continuous ranges of moment values are a possibility in Kondo screening (see, e.g., [9]), but static spatial inhomogeneity with a continuous range of moment values may not yet have been considered in a Kondo-interaction calculation.

The correlation generated by the modelling should be monitored by a two-point correlation function, which, if it decays exponentially in the distance r between the two points, defines a correlation length. Any moment–moment correlation can only be defined at those discrete values of r that represent the separations of pairs of moments in the lattice cluster. For any such r, there will usually be a number $N(r)$ of pairs of moments, with magnitudes m_i and m_j, for which $r_{ij} = r$. The correlation function chosen was

$$C(r) \equiv \frac{1}{N(r)} \sum_{i,j} (1 - |m_i - m_j|)_{r_{ij}=r}. \tag{4}$$

At a particular r, the sum is over all pairs for which $r_{ij} = r$. In this notation,

$$N(r) \equiv \sum_{i,j} (1)_{r_{ij}=r}.$$

$C(r)$ is unity when all moments are the same size, and 2/3 when the magnitudes are completely random on the unit interval. Figure 1 shows $C(r)$ for a typical simulation run, with an overall cluster thickness of 44 ions and a sub-cluster thickness cut-off of 10 ions. All distances are stated as multiples of the distance between nearest-neighbour moments in the lattice

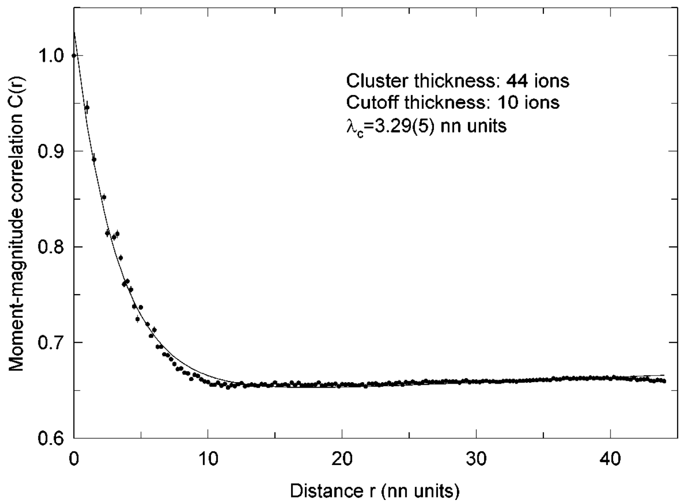

Figure 1. The moment magnitude correlation $C(r)$ (random $= 2/3$), as a function of distance r between the moments (in units of the distance between nearest-neighbour moments), for a typical Monte Carlo simulation of range-correlated magnetic moment variation (RCMMV, as described in the text), with a cubic cluster thickness of 44 ions, and a rectangular-box sub-cluster cut-off thickness of 10 ions. The solid curve is a fit of exponential decay to the baseline, with a deduced correlation length of 3.29(5).

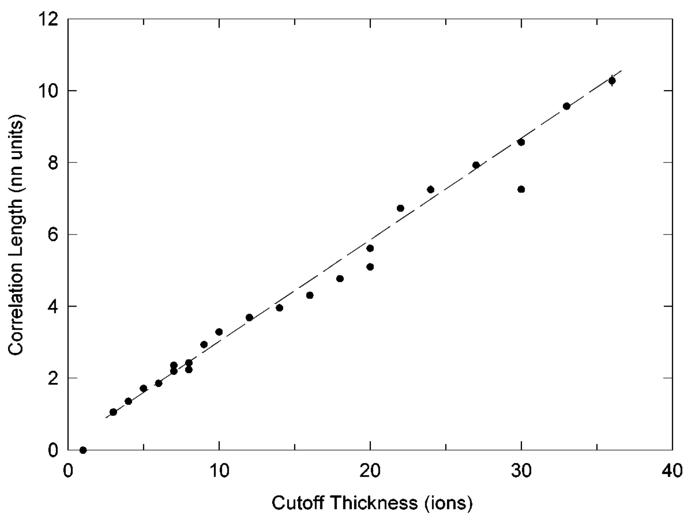

Figure 2. The RCMMV moment magnitude correlation length deduced from fits to $C(r)$ as a function of the sub-cluster cut-off thickness. The straight line (slope 0.29) is a guide for the eye.

(abbreviated as nn). It was found that for runs with cut-off greater than $\sim 1/4$ of the overall thickness, the correlation did not settle down near 2/3 for large r, so these were not used. Also shown in figure 1 is the fit of exponential decay to the baseline,

$$C(r) = A\mathrm{e}^{-\lambda_c r} + B \tag{5}$$

(where $A \simeq 1/3$ and $B \simeq 2/3$), with a deduced correlation length $\lambda_c = 3.29 \pm 0.05$ nn. Figure 2 shows the correlation length deduced from such fits as a function of the cut-off thickness. The correlation length is $\simeq$0.3 times the cut-off thickness. With overall clusters of up to two million ions, correlation lengths only up to about ten times the nn distance can be reasonably modelled.

For each overall cluster, once moment magnitudes were assigned as above, and moment directions randomly, numerous examples of the interstitial muon site (at the crystallographically unique unit-cell centre) were chosen at random from the interior volume at least five nn steps from the outside of the cluster. Modelling of the field distributions and static ZF relaxation functions was then performed as described previously [7]. The local field at each muon site was calculated in dipole coupling to all the moments in the cluster, and then added to the field distribution histograms. The component of muon polarization in its initial direction, as a function of time, was calculated in that local field and added to the relaxation-function histogram. For the larger cluster sizes, typically moments were assigned 600 times and more than 2000 muon sites were evaluated for each of those moment configurations.

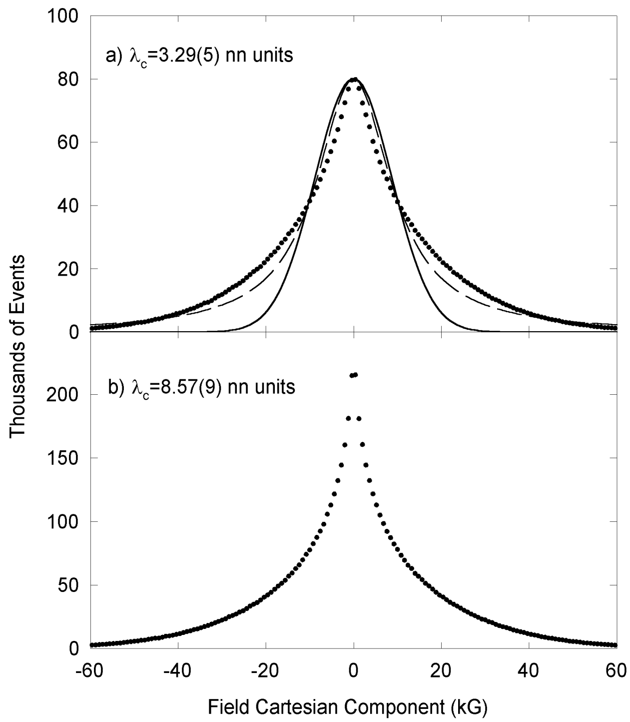

Figure 3. Monte Carlo RCMMV local field Cartesian-component distribution histograms: (a) for an overall thickness of 44 ions and a correlation cut-off thickness of 10 ions; the solid (dashed) curve shows the Gaussian (Lorentzian) distribution with the same maximum and fwhm; (b) for an overall thickness of 126 ions and a correlation cut-off of 30 ions.

3. Results

The Gaussian and Lorentzian Kubo–Toyabe relaxation functions are named after the distributions of one Cartesian component of the magnetic field, $P(B_x)$, that generate them. As previously discussed, however [7], particularly in the case of polycrystalline averaging, the distribution of the magnetic field magnitude $P(|\boldsymbol{B}|)$ is more directly related to the static ZF

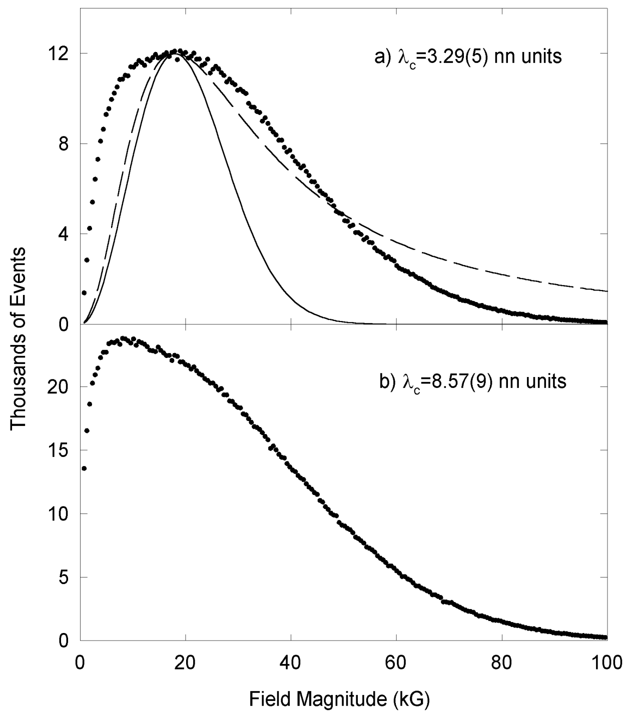

Figure 4. Monte Carlo RCMMV local field magnitude distribution histograms: (a) for an overall thickness of 44 ions and a correlation cut-off thickness of 10 ions; the solid (dashed) curve shows the Maxwellian (Lorentzian [7, 10]) magnitude distribution with same maximum value and position; (b) for an overall thickness of 126 ions and a correlation cut-off of 30 ions.

relaxation function, by

$$G_z(t) = \frac{1}{3} + \frac{2}{3}\int_0^\infty P(|\boldsymbol{B}|)\cos(\gamma_\mu|\boldsymbol{B}|t)\,\mathrm{d}|\boldsymbol{B}|. \qquad (6)$$

Therefore both the component and magnitude distributions were monitored by the Monte Carlo program. Examples are shown in figure 3 and figure 4, respectively. For comparison, curves in figure 3 show the Gaussian and Lorentzian distributions with the same fwhm as the simulation: the RCMMV model generates distributions more sharply peaked at low field than either of the standard ones. The curves in figure 4 show Maxwellian and Lorentzian [7, 10] magnitude distributions, and the model clearly generates distributions with more probability of the occurrence of low fields, relative to the most likely field. Figure 5 shows a sequence of simulated static ZF relaxation functions, for the correlation lengths shown. The simulation shown for no correlation (every moment magnitude and direction random) is almost indistinguishable from the standard Gaussian Kubo–Toyabe function. The model, as described, naturally keeps the average moment (magnitude) per unit volume constant, independently of the correlation length, and this results in a constant rms local field Cartesian component $(B_x)_{rms}$. For moments in the interval (0, 1 μ_B) at 1 Å spacing, as shown for figures 3 and 4, $(B_x)_{rms} = 19.8 \pm 0.2$ kG. As a further result, it can be seen in figure 5 that the initial drop of polarization from 1.0 to ∼0.6 is also essentially independent of the correlation, and that the time at which minimum polarization occurs is not strongly dependent on the correlation, but that the value of minimum polarization achieved increases with λ_c. Figure 6 shows the minimum polarization of the Monte Carlo relaxation functions, plotted as a function of $1/\lambda_c$.

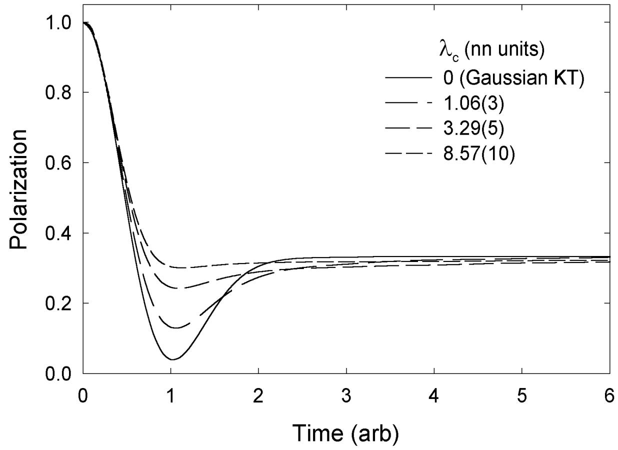

Figure 5. Monte Carlo RCMMV static ZF muon spin relaxation functions for the moment magnitude correlation lengths indicated.

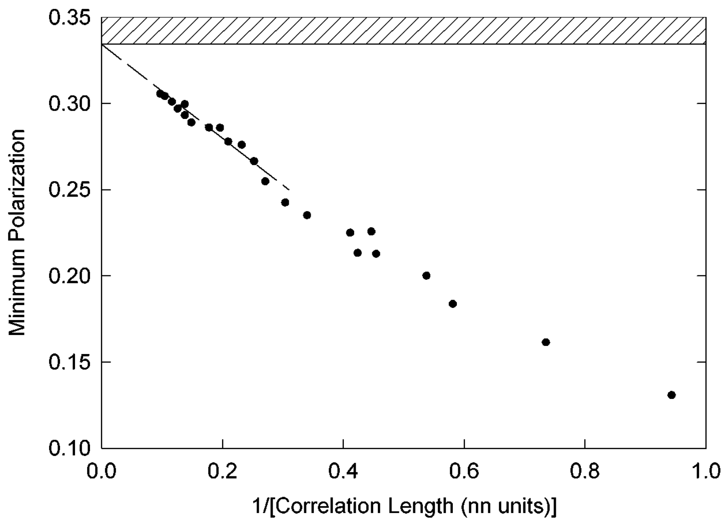

Figure 6. The minimum polarization achieved by the Monte Carlo RCMMV static ZF muon spin relaxation functions, as a function of the reciprocal of the moment magnitude correlation length λ_c. The horizontal line represents the 1/3 asymptote, above which the minimum polarization cannot rise. The dashed line is a guide for the eye.

Comparison of the simulated relaxation with the Gaussian-broadened Gaussian analytic form (equation (3)) can be made by treating each Monte Carlo spectrum as data and fitting $G_z^{GBG}(t)$ to it, as shown in figure 7. The fits are reasonable, but not perfect. Figure 8 shows that the deduced GBG ratio parameter R appears to have a simple straight-line relationship with the reciprocal of the correlation length, $1/\lambda_c$.

The RCMMV simulated relaxation cannot be properly least-squares fitted to experimental data, because only discrete values of the cut-off (and hence, the correlation length) are available. Figure 9 shows a comparison of simulated RCMMV for $\lambda_c = 2.94$ nn and low-temperature ZF μSR in $CeCu_{0.2}Ni_{0.8}Sn$ [5, 6]. The agreement between the two is fairly good.

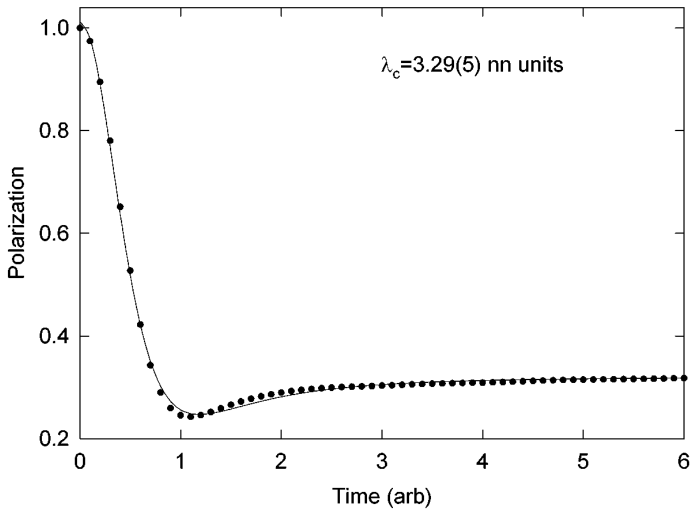

Figure 7. The Monte Carlo RCMMV static ZF muon spin relaxation function for a cluster overall thickness of 44 ions and a correlation cut-off thickness of 10 ions (circles), with a fit of a Gaussian-broadened Gaussian relaxation function (curve). The deduced GBG ratio parameter is $R = 0.543(6)$.

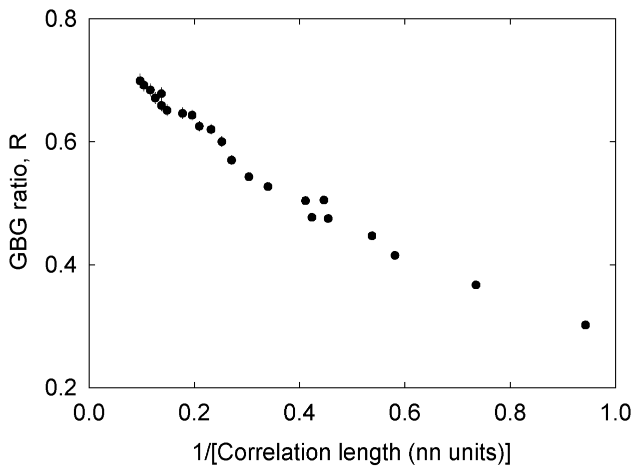

Figure 8. The GBG ratio parameter R, deduced from fits to Monte Carlo RCMMV static ZF μSR spectra, as a function of $1/\lambda_c$.

4. Discussion

The Monte Carlo model is crude in several respects, one of which is the forced total lack of correlation beyond the sub-cluster thickness cut-off parameter value. Thus (figure 1) it can only approximate the exponential decay of correlation with distance to be expected in a real situation. Nonetheless it shows that RCMMV can generate arbitrarily shallow static ZF relaxation functions. There is at this time no other microscopic model known to do so.

The field distributions generated by the RCMMV Monte Carlo model have dramatically enhanced probabilities of low fields relative to those of the average (or rms) fields of the

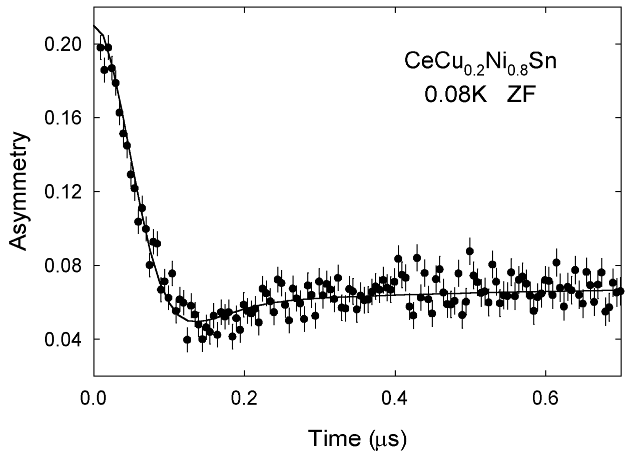

Figure 9. Comparison of the Monte Carlo RCMMV relaxation function having correlation length $\lambda_c = 2.94$ nn to the low-temperature ZF μSR observed in $CeCu_{0.2}Ni_{0.8}Sn$.

standard field distributions. As illustrated in figure 3, $P(B_x)$ becomes progressively more sharply peaked around $B_x = 0$ as the correlation length increases. In figure 4, meanwhile, the most likely field magnitude shifts to progressively lower values relative to the high-field tail of $P(|\boldsymbol{B}|)$. A strictly phenomenological form which roughly fits the magnitude distributions is

$$P_z^{phen}(B) = \frac{N}{\mathcal{W}}\left(\frac{B}{\mathcal{W}}\right)^p \exp\left(\frac{-B^2}{2\mathcal{W}^2}\right) \tag{7}$$

where $\mathcal{W}$ is a width parameter. The power p is close to 2 for $\lambda_c = 0$ (where the simulation magnitude distribution is almost indistinguishable for the Maxwellian case, and $\mathcal{W} = (B_x)_{rms}$), but decreases as λ_c increases. A graph of p as a function of $1/\lambda_c$ suggests that in the limit of long correlation length, $p \to 0$; that is, the magnitude distribution would be approximately Gaussian (with $\mathcal{W} = |\boldsymbol{B}|_{rms}$). No such closed-form approximation has yet been found for $P(B_x)$.

Note that the relaxation function $G_z(t)$, as determined from equation (3), is (a constant plus) the cosine Fourier transform of $P(|\boldsymbol{B}|)$. From this, the (extrapolated) change of the magnitude distribution from possessing a peak at non-zero field to possessing a peak only at $|\boldsymbol{B}| = 0$, as $\lambda_c \to \infty$, implies a change in G_z from underdamped (possessing a local minimum) to overdamped (showing monotonic relaxation to the asymptote) in that limit. Similarly, the extrapolation of the minimum polarization on figure 6 suggests that the local minimum disappears into the 1/3 asymptote for infinite λ_c. It thus appears that the complete range of shallow minima, down to vanishingly small, is spanned by this model.

The extrapolation of the Gaussian-broadened Gaussian ratio R, in figure 8, however, suggests a limit value for R of slightly less than 0.8, which still implies a small local minimum of polarization. The fits of G_z^{GBG} to the Monte Carlo relaxation functions are not perfect, and, to fit the Monte Carlo initial relaxation better, overestimate the depth of the minimum for the larger-λ_c cases. This discrepancy may be due to the crudeness of the Monte Carlo model (but that idea can only be tested by developing models that are more realistic). It would be nice to be able to use figure 8 as a calibration curve for conversion of measured GBG ratio values to underlying correlation lengths, but the imperfect match of $G_z^{GBG}(t)$ to the RCMMV Monte Carlo relaxation functions limits confidence in this. Direct comparison of individual simulations to observed data is useful, but is not as impartial as least-squares fitting is: there is a risk of systematic personal bias in the deduced parameter values. For the data of figure 9, since the simulation for $\lambda_c = 3.29$ nn looks almost as good as the 2.94 nn case shown, it is reasonable to conclude that $\lambda_c = 3.1 \pm 0.3$ nn.

Care must be exercised in thinking about the statistics of the model in the large-correlation-length limit, '$\lambda_c \to \infty$'. In this limit, $C(r)$ becomes unity, and it would seem that all moments are the same size, which is a case well known to generate a relaxation function well approximated by the Gaussian Kubo–Toyabe function, not the monotonic relaxation to 1/3 asserted above. The difference is that for any finite r, no matter how large, as many muons will find sites surrounded by very small moments as will land at sites with moments near the maximum value. For all finite r, the moments are uniformly spread over the unit interval. This is only a formal problem. Physically, a correlation length $\lambda_c \sim 30$ nn units (typically less than 200 Å in real materials) would generate a static relaxation function with a minimum so shallow that it would be well approximated by monotonic relaxation, within the statistical accuracy of a normal μSR spectrum, and thus would appear to be at the limit, yet with sample dimensions of at least tenths of millimetres, there will still be room for millions of such correlated regions with vastly different moment magnitudes.

5. Conclusions

The observed anomalous form (with the characteristic shallow minimum) of static ZF muon spin relaxation in disordered magnetic materials such as $CeNi_{1-x}Cu_xSn$, which was previously found to conform to the phenomenological 'Gaussian-broadened Gaussian' relaxation function, has been shown here, by Monte Carlo simulation, to be associated with a subtle short-range partial-ordering effect: range-correlated moment magnitude variation (RCMMV). The shallowness of the ZF relaxation, relative to standard Gaussian Kubo–Toyabe relaxation, is in a one-to-one relationship with the moment magnitude correlation length, and so μSR, as a local probe, in these circumstances provides a measure of a correlation length. It is a strange and heretofore unexpected correlation, however, of static spatial inhomogeneity of the moment magnitudes: the moments not just 'on' or 'off', but ranging over enough distinct values to be approximated by a continuous range from zero to a maximum.

Acknowledgments

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