Muon spin relaxation in a superparamagnet: Field dynamics in $Cu_{98}Co_2$

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Zero-field muon-spin-relaxation techniques have been used in conjunction with magnetization measurements and numerical simulations to characterize the internal-field distributions and dynamics in a model superparamagnetic system, namely, dilute heterogeneous $Cu_{98}Co_2$. The internal dipolar field distribution in $Cu_{98}Co_2$ arising from the polydispersed, noninteracting magnetic Co clusters, is found to be Lorentzian in form, with a half width at half maximum which, for a given Co concentration, is independent of the details of distribution of cluster moments. Superparamagnetic relaxation of the clusters is shown to result in a root-exponential muon depolarization function with an associated relaxation rate that can be related directly to the mean fluctuation rate of the dipolar fields, but which is entirely independent of the width of the distribution of fluctuation rates. Analysis of the observed muon-spin-relaxation spectra of $Cu_{98}Co_2$ within the framework of the proposed model enables both the activation energies and intrinsic relaxation rates of the superparamagnetic Co clusters to be extracted. [S0163-1829(98)08741-4]

I. INTRODUCTION

Muon spin relaxation (μSR) is becoming an increasingly popular and powerful probe of the solid state. The unique sensitivity of an implanted muon to the static distributions and dynamic fluctuations of even the smallest atomic and nuclear magnetic fields within a sample can provide novel insights into a wide range of magnetic phenomena. In recent years μSR techniques have been applied with considerable success to the investigation of magnetic relaxation in systems as diverse as heavy fermions, spin-fluctuating itinerant magnets, and spin glasses.[1] In this paper we demonstrate that μSR can provide an equally effective tool for characterizing and quantifying the field distributions and cluster dynamics in archetypal superparamagnetic systems.

In this μSR study we shall focus upon dilute *Cu*-Co alloys as a model system. Indeed, heterogeneous *Cu*-Co has long been recognized as an almost ideal system with which to explore superparamagnetic phenomena.[2] Cobalt is virtually insoluble in copper,[3] while copper has less than 9% solubility in cobalt below 500 °C.[4] The growth of well-defined single-domain, ferromagnetic, Co-rich precipitates in an otherwise nonmagnetic Cu-rich matrix can thus be effected through careful sample preparation and subsequent annealing. At sufficiently dilute cobalt concentrations intercluster interactions are weak and the Co clusters, which are generally between a few nanometers and a few hundred nanometers in extent, behave as giant classical spins, fluctuating superparamagnetically at high temperatures. On cooling, the spin dynamics may ‘‘block’’ at a temperature that depends upon the anisotropy barriers associated with the individual clusters and also upon the effective time window of the experimental probe.

Although superparamagnetic *Cu*-Co has been the subject of numerous experimental studies over the last five decades, most investigations have been concerned either with the determination of cluster-size distributions or with the characterization of bulk magnetic properties. More recently interest has focused upon giant magnetoresistive effects in heterogeneous *Cu*-Co.[5] However, relatively little attention has been devoted to the study of cluster dynamics in the superparamagnetic state. This is, in part, a consequence of the range of characteristic frequencies associated with superparamagnetic cluster dynamics: the intrinsic relaxation rate of the Co clusters is of the order of 10^9 Hz, which is too slow to be studied by conventional inelastic neutron scattering, while far too fast for ac susceptibility methods. Other experimental techniques either have an insufficiently wide frequency window to allow the evolution of cluster dynamics to be followed over a wide range of temperatures, or alternatively require the application of an external magnetic field, thereby perturbing the superparamagnetic system. Zero-field μSR, on the other hand, has already been shown to be remarkably effective in studies of magnetic relaxation in spin-glass systems over an extremely broad frequency range ($10^4 < \upsilon < 10^{11}$ Hz),[6,7] a range directly comparable to that anticipated for superparamagnetic systems. We have taken advantage of this experimental window to view the superparamagnetic cluster spin dynamics indirectly via fluctuations of the associated dipolar fields.

In considering the application of μSR techniques to the problem of superparamagnetic relaxation we shall first characterize the internal-field distribution within dilute heterogeneous *Cu*-Co alloys using a combination of magnetization measurements and computational methods. We shall then introduce a muon depolarization function derived for the situation in which the muon senses the dynamic fields associated with fluctuating polydispersed Co clusters and, finally, apply this depolarization function to μSR spectra measured for dilute *Cu*-Co.

II. SAMPLE PREPARATION AND MAGNETIC CHARACTERIZATION

Polycrystalline ingots of $Cu_{98}Co_2$ were prepared in an argon arc furnace using metals of at least 99.99% purity. The ingots were severely cold rolled into disks of approximately 30 mm in diameter and 0.5 mm thick. The disks were sealed

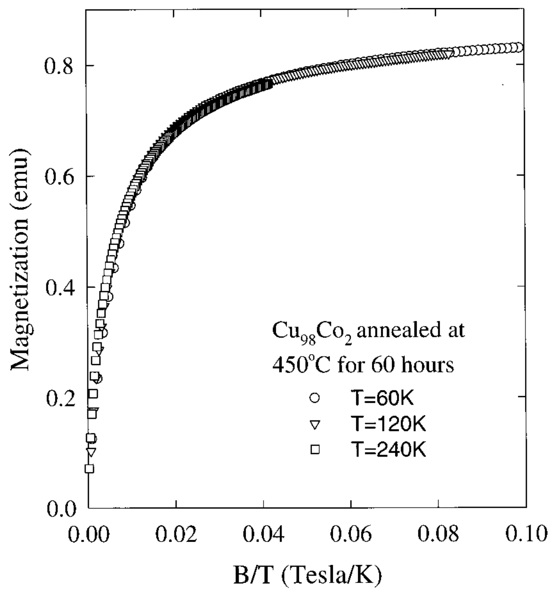

FIG. 1. The magnetic isotherms of the $Cu_{98}Co_2$ sample annealed for 60 h at 450 °C measured at 60, 120, and 240 K plotted as a function of the ratio of the applied field to temperature B/T. Note the superposition of the isotherms.

in quartz under an argon atmosphere, annealed for 24 h at 1000 °C and subsequently quenched into iced water. As cobalt is entirely soluble in Cu at 1000 °C the resulting quenched $Cu_{98}Co_2$ alloys have a metastable but homogeneous distribution of individual Co atoms. Co clusters were then nucleated and grown in a controlled manner by further annealing. The experiments described here were performed on a sample in the initial as-quenched state and upon two further samples annealed at 450 °C, one for 17 h and the other for 60 h.

Magnetization measurements were made on samples cut from the three $Cu_{98}Co_2$ disks over a temperature range of 3.8 to 300 K and in applied fields of up to 12 T, using an Oxford Instruments Vibrating Sample Magnetometer. The magnetization $M(B,T,\mu)$ of an ensemble of identical superparamagnetic clusters is expected to follow the classical Langevin form, i.e.,

$$M(B,T,\mu)=M_s\left\{\coth\left(\frac{\mu B}{k_B T}\right)-\frac{k_B T}{\mu B}\right\}, \tag{1}$$

where M_s is the saturation magnetization of the sample and μ is the magnetic moment of the superparamagnetic clusters. Although this expression does not precisely describe the measured magnetization of the annealed samples, due largely to the expected distribution of cluster moments, the measured magnetization is found to scale precisely with the ratio B/T over a wide range of temperatures, as illustrated in Fig. 1. Such scaling demonstrates that the annealed samples are ''good'' superparamagnets with very little intercluster exchange and also that the net magnetic moment of the clusters is essentially independent of temperature, at least below 240 K. In contrast the as-quenched sample exhibits a linear dependence of the magnetization with applied field at all temperatures, indicating that the magnetization of this sample is dominated by the magnetic response of individual Co atoms.

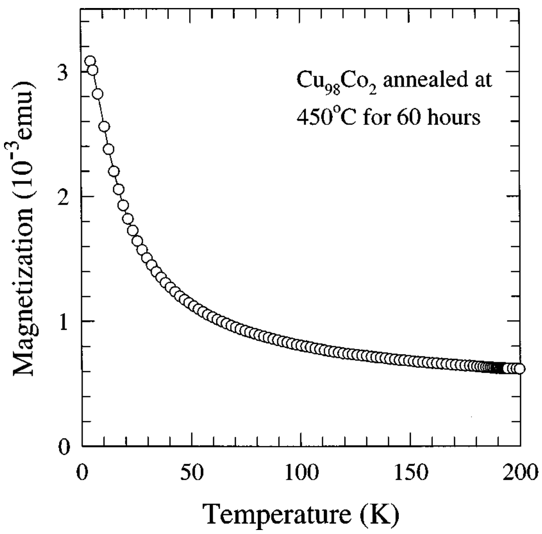

FIG. 2. The temperature dependence of the zero-field-cooled magnetization of the $Cu_{98}Co_2$ sample annealed for 60 h at 450 °C measured in a field of 10 mT. There is no evidence of superparamagnetic blocking over the temperature range of the measurements.

The zero-field-cooled magnetization, measured as a function of temperature in a field of 10 mT, provides little evidence of superparamagnetic blocking in any of the samples down to a temperature of 3.8 K (Fig. 2) and only a slight inflection in the temperature dependence of the magnetization is observed at the lowest temperatures. This implies that the anisotropy barriers associated with the superparamagnetic Co clusters, and hence the Co clusters themselves, are relatively small and, moreover, that the intercluster interactions are weak.

III. CLUSTER SIZE AND INTERNAL-FIELD DISTRIBUTIONS

In principle the magnetic moment μ of the Co clusters can be extracted from a fit of Eq. (1) to the measured magnetization of the annealed samples. In reality a distribution of Co cluster sizes, and hence of cluster moments, is expected. The magnetization should therefore be fitted using a Langevin function convoluted with an appropriate probability distribution of cluster moments. Rather than adopting the conventional route of assuming an arbitrary probability function for the cluster moment distribution, we have chosen an alternative approach that uses a modified reverse Monte Carlo (RMC) procedure to extract this distribution directly from the magnetic isotherms. Starting with an initially uniform distribution of cluster moments the RMC procedure repeatedly modifies the relative weighting across the distribution and convolutes the resulting distribution with the Langevin function. χ^2 is minimized at each stage until the best fit to the experimental magnetization curve is achieved. The resulting probability distributions for the two annealed samples are illustrated in Fig. 3.

The distribution of cluster moments is found to be closely log-normal in form for both the annealed samples, as can be seen in Fig. 3, where the lines through the data points generated by the *ab initio* RMC method represent the best fit of the modified log-normal distribution function

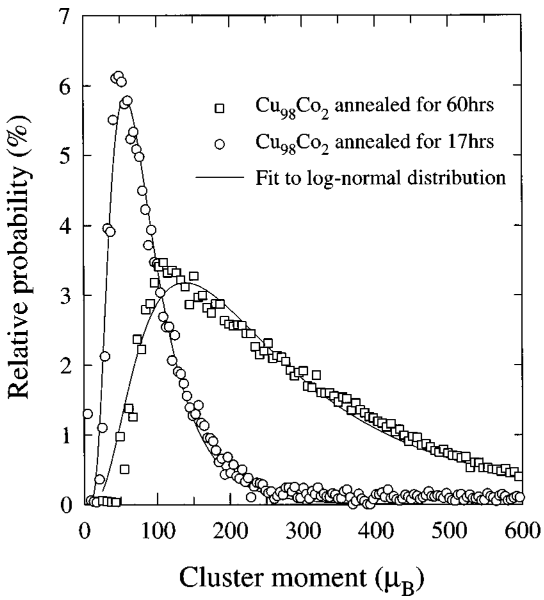

FIG. 3. The *ab initio* distribution of cluster moments obtained for both the annealed $Cu_{98}Co_2$ samples obtained by the modified reverse Monte Carlo method described in the text. The lines through the data represent the best fit to a log-normal distribution function.

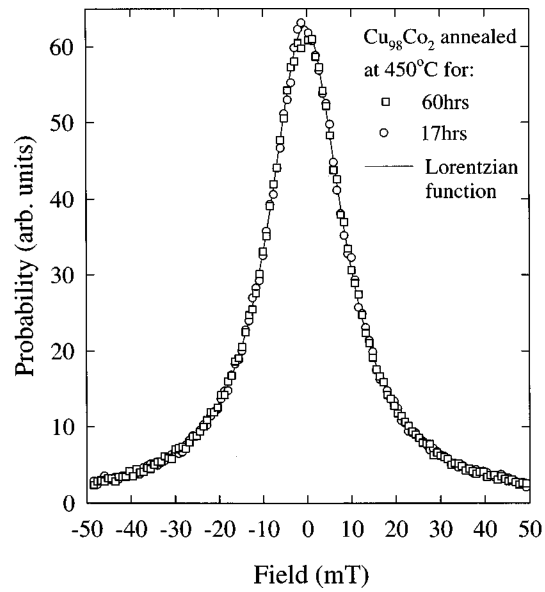

FIG. 4. The Monte Carlo simulated static *x*, *y*, and *z* components of the dipolar field profile at the muon site arising from the log-normal distributions of cluster moments illustrated in Fig. 2. The line through the data represents the best fit to a Lorentzian field profile with a HWHM of 10.1 mT. Note that the simulated field profile is identical for both the annealed $Cu_{98}Co_2$ samples.

$$F(\mu)=\frac{1}{\sqrt{2\pi}\sigma_m\mu}\exp\left[-\frac{1}{2\sigma_m^2}\ln^2\left(\frac{\mu}{\mu_m}\right)\right], \tag{2}$$

where μ_m and σ_m are the mean cluster moment and the standard deviation of the distribution, respectively. For the sample annealed for 17 h at 450 °C we find $\mu_m=77\mu_B$ and $\sigma_m=0.55$, while for the sample annealed for 60 h $\mu_m=237\mu_B$ and $\sigma_m=0.71$.

In a zero-field μSR experiment the muon senses the internal-field distribution within the sample. It is therefore useful to attempt to estimate this internal-field distribution directly from the magnetization measurements on the annealed $Cu_{98}Co_2$ samples. This has been achieved by adopting a second Monte Carlo procedure in which the magnitudes of the cluster moments are selected at random according to the log-normal distributions determined above for the respective samples. The associated clusters are then positioned at random within a cubic box $30\times30\times30$ nm^3. The process is repeated until the volume fraction of the box occupied by the Co clusters is consistent with a concentration of 2-at. % Co. The volume fraction is estimated by assuming that Co clusters adopt a face-centred-cubic structure with a lattice parameter close to that of pure Cu, and also by assuming that the magnetic moments of individual Co atoms are similar to those in bulk Co, i.e., $1.7\mu_B$ (although the latter assumption is perhaps contentious). Finally, the orientations of the individual cluster moments are selected at random and the *x*, *y*, and *z* components of the net dipolar field resulting from every Co cluster within the box are calculated at a fictitious muon site at the center of the box. This procedure is repeated 2×10^5 times, producing the field distributions shown in Fig. 4 for the two annealed $Cu_{98}Co_2$ samples.

The resulting *x*, *y*, and *z* components of the internal-field distributions are found to be precisely Lorentzian in form, as predicted for a system of dilute magnetic dipoles.[8] The solid line through the data in Fig. 4 represents a least-squares fit of a Lorentzian function to the field distributions, yielding a half width at half maximum (HWHM) of 10.1 mT. At first sight it may seem surprising that the two very different cluster moment distributions of Fig. 3 should produce identical Lorentzian field profiles. However, this is simply a consequence of the $1/r^3$ dependence of the dipolar fields arising from the clusters, and of the assumption that the cluster moment is directly proportional to the cluster volume: if the latter increases by a factor *n*, then clearly the average distance between the clusters increases by a factor $n^{1/3}$ in the dilute limit. For a given concentration of Co atoms the effect of increasing the magnitude of cluster moments is thus exactly compensated by the resulting decrease of the $1/r^3$ dipolar field term. The HWHM of the Lorentzian field distribution should thus remain constant and independent of the details of the cluster distribution for a given concentration of Co spins.

The relative contributions of the nearest neighbor distributed clusters to the dipolar field at the fictitious muon site have also been calculated as part of the above procedure. It is interesting to note that in all cases the nearest-neighbor cluster heavily dominates this field. Indeed, as Fig. 5 shows, almost 90% of the magnitude of the *x*, *y*, and *z* components of the internal field arises from the dipole moment associated with the nearest-neighbor cluster, with a further 8% contributed by the second-nearest neighbor. Just as the HWHM of the internal field distribution is independent of the cluster-size distribution, so is the relative contribution to the internal field at the muon site from successive nearest-neighbor clusters. Perhaps more surprisingly, the relative contributions from successive neighboring clusters to the normalized field at the muon site is also found to be independent of the concentration of the heterogeneous alloy, at least to concentra-

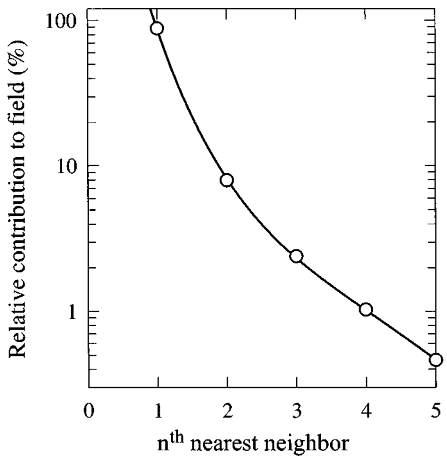

FIG. 5. Estimated relative contributions to the field at the muon site from nearest-neighbor Co clusters. The data shown here represent an average over 10^5 muon sites. The solid line is a guide to the eye.

tions of 10%. Figure 5 is therefore quite general for heterogeneous alloys in this relatively dilute limit.

The combination of magnetization measurements and Monte Carlo simulations used here has provided a comprehensive model of the internal-field distribution within the $Cu_{98}Co_2$ samples. It should now be possible to interpret the muon spin relaxation in $Cu_{98}Co_2$ within the framework of this model.

IV. μSR MEASUREMENTS

The μSR experiments were performed on the EMU spectrometer at the ISIS pulsed neutron and muon facility at the United Kingdom's Rutherford Appleton Laboratory.[9] μSR measurements were made in zero field on each of the three 30-mm-diam $Cu_{98}Co_2$ disks over a temperature range from 3.5 to 280 K.

ISIS produces high-intensity pulses of spin-polarized muons of approximately 70 ns FWHM at a repetition rate of 50 Hz. The resulting muons are implanted in the sample and subsequently decay with a half-life of $\tau = 2.19\ \mu$s emitting a positron preferentially in the direction of the muon spin at the instant of decay. The time histograms of positron counts, $N_F(t)$ and $N_B(t)$, collected in detectors placed in the forward F and backward B positions relative to the initial muon polarization thereby measure the time evolution of the muon polarization.

The zero-field muon-spin-relaxation function $G_z(t)$ is extracted from the two positron histograms, appropriately corrected for dead time, by taking the ratio

$$P(t) = a_0 G_z(t) = \frac{N_F(t) - \alpha N_B(t)}{N_F(t) + \alpha N_B(t)}. \tag{3}$$

Here a_0 is the initial asymmetry, typically taking a value between 0.21 and 0.25. α is a numerical factor, generally close to unity, which corrects for the relative F/B detector efficiencies and the asymmetry in positron attenuation resulting from absorption within the sample and ancillary sample environment. Experimentally the detector internormalization is achieved by applying a small (2-mT) transverse field to the sample and then adjusting α to ensure that the resulting asymmetry spectrum oscillates symmetrically about zero.

All of the measured spectra include a small, time- and temperature-independent background contribution a_{bg} to the asymmetry associated with those muons localizing in the (silver) sample holder. This background asymmetry was determined independently for our experimental configuration by replacing the sample with a hematite disk of identical size and applying a 2-mT transverse field. Hematite rapidly and completely depolarizes the implanted muons in the time frame of the measurement with the result that any remaining oscillating asymmetry can be attributed solely to those "background" muons that have localized outside the area of the sample. For the present configuration we obtain $a_{bg} = 0.021$.

It should be noted that the finite muon pulse width at the ISIS muon facility precludes the measurement of coherent internal fields greater than approximately 50 mT. This is simply a consequence of the experimental convolution of this pulse width with the time-dependent oscillatory positron signal associated with the coherent precession of the muons in the internal field. As a result the muon asymmetry associated with those muons localizing within the single-domain ferromagnetic Co clusters will be "lost" from the measurement.

A. μSR in quasistatic fields arising from Co clusters

Before proceeding to the more complex case of fluctuating Co clusters, we shall first examine the functional form of $G_z(t)$ for the situation in which the cluster spins are approximately *static* on the time scale of the muon probe. The computer simulations of the previous section have shown that the dipolar fields resulting from the distributed and randomly oriented Co cluster moments lead to a static internal field profile that is Lorentzian in form. In the static limit it is expected that $G_z(t)$ resulting from such a field profile is given by the static Lorentzian Kubo-Toyabe function:[10]

$$G_z^{\mathrm{LKT}}(t) = \tfrac{1}{3} + \tfrac{2}{3}(1 - at)\exp(-at), \tag{4}$$

where a/γ_μ is the half width at half maximum of the Lorentzian field distribution and $\gamma_\mu/2\pi$ (=135.54 MHz/T) is the gyromagnetic ratio of the muon. The expression $P(t) = a_1 G_z^{\mathrm{LKT}}(t) + a_{bg}$, where $a_{bg} = 0.021$ is the additional background asymmetry, has been fitted to the experimental μSR spectrum collected at 2.4 K from the $Cu_{98}Co_2$ sample annealed at 450 °C for 60 h. It can be seen in Fig. 6 that the static Lorentzian Kubo-Toyabe function adequately describes the spectrum at short times. The best fit of Eq. (4) gives a value of $a = 6.8\ \mu s^{-1}$, equivalent to a Lorentzian field distribution HWHM of $a/\gamma_\mu = 8$ mT. This value is a little lower than that of 10.1 mT provided by the computer simulations above. It is likely that the discrepancy arises from a small residual number of individual (nonclustered) Co spins within the sample. According to the most recent phase diagrams[4] 0.3-at. % Co is soluble in Cu at room temperature. Approximately 15% of the total amount of Co in the annealed $Cu_{98}Co_2$ samples might therefore be expected to occur as isolated atoms. The spins associated with such isolated atoms are likely to fluctuate at a sufficiently high rate that their contribution to the field at the muon site is subject to extreme motional narrowing. As the Lorentzian field

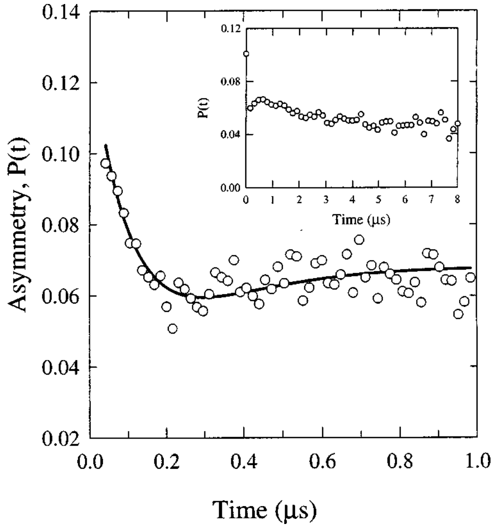

FIG. 6. The zero-field muon depolarization spectrum measured at 2.4 K for the $Cu_{98}Co_2$ sample annealed at 450 °C for 60 h. The line through the data represents the best fit to a static Lorentzian Kubo-Toyabe function. The inset shows the spectrum extending to longer times. The slight decay in the tail of the Lorentzian Kubo-Toyabe function indicates the persistence of fluctuating dipolar field even at these low temperatures.

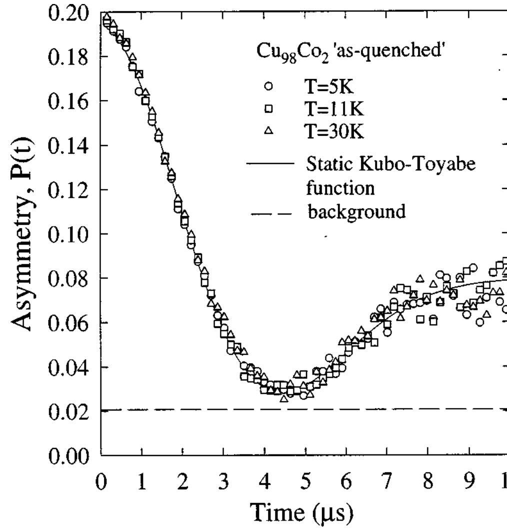

FIG. 7. The zero-field muon depolarization spectrum measured for the ''as-quenched'' $Cu_{98}Co_2$ sample at several temperatures. The solid line represents a least-squares fit of a static Kubo-Toyabe function with a depolarization rate $\sigma = 0.36\ \mu s^{-1}$, close to that of pure Cu.

width at the muon site is directly proportional to the concentration of *static* magnetic impurity spins within the sample the field-distribution width obtained from the simulations can be corrected to account for the dynamic isolated Co spins. This correction leads to a revised estimate of 8.5 mT for the HWHM of the Lorentzian field distribution, in reasonable agreement with the value determined directly from the muon spectrum. The agreement between the measured and estimated internal-field distribution width provides some justification for the assumptions that the muon occupies entirely random positions within the Cu-Co matrix, and that the moment at each Co site is comparable to that in bulk Co.

Although the static Lorentzian Kubo-Toyabe function provides a reasonable description of the muon depolarization in the low-temperature spectra obtained from the $Cu_{98}Co_2$ sample annealed for 60 h, a slight decay of the $\frac{1}{3}$ tail of the muon spectrum at longer times confirms that the Co clusters are not entirely static (i.e., blocked) within the time window of the muon probe, even at 2.4 K. These effects are far more pronounced for the sample annealed for only 17 h, while the isolated Co spins in the as-quenched sample remain wholly dynamic, with the associated field distribution fully motionally narrowed, even down to the lowest temperatures.

B. Contribution from nuclear dipole fields

In the above analysis only those dipolar magnetic fields associated with Co clusters have been considered. Such fields are sufficiently strong to decouple the muon from the much smaller fields associated with the nuclear moments at the Co and Cu sites. If, on the other hand, the cluster moments are rapidly fluctuating the resulting dipolar field at the muon site is motionally narrowed and the muon senses only the static nuclear dipole fields. This is observed in the as-quenched $Cu_{98}Co_2$ sample, as can be seen in Fig. 7 where a static Gaussian Kubo-Toyabe function[11]

$$G_z^{\mathrm{KT}}(t) = \tfrac{1}{3} + \tfrac{2}{3}(1 - \sigma^2 t^2)\exp(-\tfrac{1}{2}\sigma^2 t^2) \quad (5)$$

has been fitted to the muon depolarization spectra measured over a range of temperatures from 3.5 to 30 K using the relationship $P(t) = a_1 G_z^{\mathrm{KT}} + a_{\mathrm{bg}}$. σ^2/γ_μ^2 is the second moment of the Gaussian field distribution at the muon site arising from the Co and Cu nuclear moments. The fit of Eq. (5) to the data gives $\sigma = 0.36\ \mu s^{-1}$, close to the value expected for pure Cu.[12] This, once again, indicates that the muon occupies random positions within the Cu-Co matrix. Previous studies of the nuclear dipole fields in pure Cu (Ref. 12) indicate that the muon occupies an octahedral site in the face-centered-cubic Cu lattice and is almost stationary below 70 K. The absence of any significant decay of the $\frac{1}{3}$ tail of the Kubo-Toyabe function in Fig. 6 indicates that the muon is similarly stationary in as-quenched $Cu_{98}Co_2$ to temperatures of at least 30 K.

C. μSR in dynamic fields arising from fluctuating cobalt clusters

So far we have considered the two extreme cases of muon spin relaxation in fields associated with dilute and almost static magnetic clusters, and in a system in which the dipolar fields arising from rapidly fluctuating atomic or cluster moments are effectively motionally narrowed. We shall now focus on the more interesting intermediate regime in which the relatively slow dynamics of the superparamagnetic Co clusters fall within the frequency window of the muon probe. In the absence of any existing theoretical model of muon spin relaxation in the presence of fluctuating superparamagnetic clusters, we have decided to adopt the methodology and formalism proposed by Uemura *et al.*[13] to describe μSR in conventional dilute spin glasses above the glass temperature. Within this spin-glass model the fluctuation of atomic spins leads to a field at the muon site that is modulated in time. The dynamic range of this field modulation is different for each muon site, depending upon the local configuration of surrounding spins. Uemura *et al.* approximated the variable dynamic range by an atomic Gaussian field distribution of width Δ/γ_μ, i.e.,

$$P(H_i)=\frac{\gamma_\mu}{\sqrt{2\pi}\Delta}\exp\left[-\frac{\gamma_\mu^2H_i^2}{2\Delta^2}\right],\quad i=x,y,z. \tag{6}$$

The probability of finding a muon at a site with a width Δ is then calculated to be

$$\rho(\Delta)=\sqrt{2\pi}\frac{a}{\Delta^2}\exp\left[-\frac{a^2}{2\Delta^2}\right], \tag{7}$$

ensuring that the field distribution averaged over all muon sites maintains a Lorentzian profile with HWHM of a/γ_μ as expected for a system of dilute spins.

Assuming that all spins fluctuate at the same rate ν, resulting in a Markovian modulation of the field H such that

$$\frac{\langle H(t)H(0)\rangle}{\langle H(0)^2\rangle}=\exp(-\nu t), \tag{8}$$

the muon relaxation function for a single muon site, in the fast fluctuation limit ($\nu/a>20$), becomes

$$G_z(t,\Delta,\nu)=\exp(-2\Delta^2t/\nu). \tag{9}$$

Finally, spatially averaging over all muon sites, leads to the dynamic spin-glass depolarization function

$$G_z^{\mathrm{sg}}(t)=\int_0^\infty G_z(t,\Delta,\nu)\rho(\Delta)d\Delta=\exp[-(4a^2t/\nu)^{1/2}]. \tag{10}$$

This simple ''root exponential'' muon-spin-relaxation function has been shown to account successfully for the observed zero-field muon response in several dilute spin-glass systems.[13]

At first sight it appears to be a relatively straightforward exercise to extend this analysis from a system of dilute individual spins to one of dilute magnetic clusters. In both cases the internal-field distribution is Lorentzian in form, and the concept of a Gaussian-distributed variable dynamic range of field modulation is not unreasonable. However, in the case of a real superparamagnet, such as the annealed $Cu_{98}Co_2$ samples discussed here, the observed wide distribution of cluster sizes is expected to lead to a similarly wide range of cluster-spin fluctuation rates ν and hence to a stretched exponential or Kohlrausch-like field autocorrelation function[14] rather than the simple exponential form given by Eq. (8). It turns out, in fact, that such a distribution of fluctuation rates has remarkably little effect on the functional form of the muon spin relaxation given by Eq. (10). This can be demonstrated by considering the implications of the results of our numerical simulations of the internal-field distributions within superparamagnet $Cu_{98}Co_2$.

The Monte Carlo simulations show that, for a given concentration of clusters, the Lorentzian field profile at the muon site is entirely independent of the cluster-size distribution. Consequently $\rho(\Delta)$ of Eq. (7) must also be independent of the cluster-size distribution. Moreover, we have found that

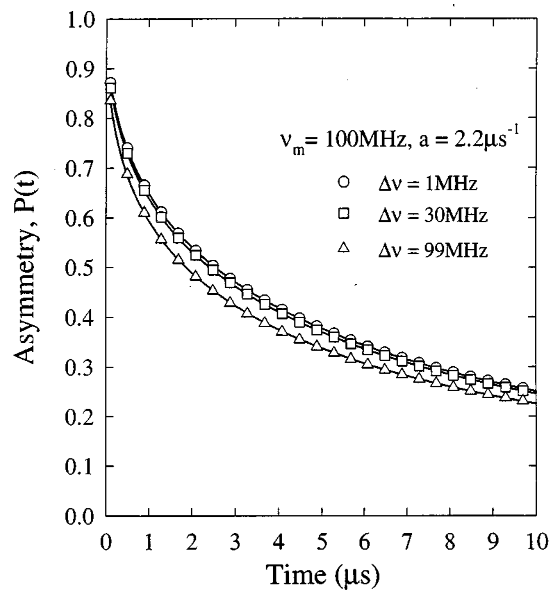

FIG. 8. Simulated zero-field muon depolarization spectra for a system of dilute dipolar spins with an increasingly wide range of fluctuation frequencies. The spectra have been calculated using numerical integration to solve Eq. (12) for several widths of the frequency distribution $\Delta\nu$ assuming a mean fluctuation rate of $\nu_m=100$ MHz and a Lorentzian field parameter width of $a=2.2\ \mu\mathrm{s}^{-1}$. The lines through the simulated spectra are fits to a stretched exponential function $\exp[-(\lambda t)^\beta]$. For (a) $\Delta\nu=1$ MHz the fit provides $\lambda=0.194\ \mu\mathrm{s}^{-1}$ and $\beta=0.5$, for (b) $\Delta\nu=30$ MHz we find $\lambda=0.197\ \mu\mathrm{s}^{-1}$ and $\beta=0.5$ while (c) for $\Delta\nu=99$ MHz we find $\lambda=0.238\ \mu\mathrm{s}^{-1}$ and $\beta=0.46$. These values compare with $\lambda=0.194\ \mu\mathrm{s}^{-1}$ and $\beta=0.5$ expected for an ideal δ-function distribution with $\Delta\nu=0$.

the field at the muon site and therefore, by implication and to a first approximation, the dynamic field range Δ is determined principally by the nearest-neighbor cluster. These two properties of a dilute heterogeneous dipolar system indicate that cluster size itself is unimportant in defining Δ, and also that Δ and the fluctuation frequency can be considered to be independent quantities. The muon relaxation function associated with a site at which the dynamic range is Δ, averaged over an arbitrary distribution of cluster fluctuation frequencies, can therefore be written as

$$G_z(t,\Delta,\nu)=\int_0^\infty f(\nu)\exp(-2\Delta^2t/\nu)d\nu, \tag{11}$$

where $f(\nu)$ is the normalized distribution of fluctuation frequencies.

The modified form of the final depolarization function of Eq. (10) is therefore

$$G_z(t)=\int_{\Delta=0}^\infty\int_{\nu=0}^\infty\rho(\Delta)f(\nu)\exp(-2\Delta^2t/\nu)d\nu\ d\Delta. \tag{12}$$

In practice $f(\nu)$ is difficult to establish for a real superparamagnet without prior and precise knowledge of the origin of cluster anisotropy. For large clusters the terms associated with volume anisotropy might be expected to dominate cluster dynamics, in which case $f(\nu)$ can be determined from the distribution of anisotropy energies $f(E_a)$ and hence cluster volumes, via the Arrhenius relation $\nu=\nu_0 \exp(-E_a/kT)$. In general, however, this is not possible as surface anisotropy, magnetocrystalline anisotropy, and shape anisotropy also contribute to E_a. Nevertheless, we can demonstrate the effects of a broad distribution of cluster fluctuation frequencies by considering a simple top-hat distribution centered around ν_m with a width of $\pm\Delta\nu$ and thence numerically evaluating the double integral of Eq. (12). As an illustration we have performed this calculation for a Lorentzian field distribution with a parameter a of 2.2 μs^{-1} and a mean fluctuation frequency of $\nu_m=100$ MHz, for a number of widths of the frequency distribution in the range $0\leqslant\Delta\nu/\nu_m\leqslant 1$. The resulting muon-spin-relaxation functions $G_z(t)$ are shown in Fig. 8, where the solid lines represent a fit of a stretched exponential function $\exp[-(\lambda t)^{\beta}]$ to the numerically evaluated $G_z(t)$. For $\Delta\nu/\nu_m=0$, $f(\nu)$ is a delta function and $G_z(t)$ is defined by Eq. (10), with $\lambda=4a^2/\nu=0.194\ \mu s^{-1}$. The fit of the stretched exponential function to $G_z(t)$ for $\Delta\nu/\nu_m=0$ correspondingly gives $\lambda=0.194\ \mu s^{-1}$ and $\beta=0.5$, as expected. However, it is interesting to note that even for the situation where $\Delta\nu/\nu_m=0.99$, $G_z(t)$ remains closely root exponential in form, with $\lambda=0.238\ \mu s^{-1}$ and $\beta=0.46$. Clearly the root exponential form of $G_z(t)$ given by Eq. (10) continues to provide an adequate description of the muon-spin-relaxation function despite an increasingly wide distribution of fluctuation frequencies. It should also be noted that the relaxation rate λ extracted from the fit of the stretched exponential function to $G_z(t)$ remains within a few percent of that associated with the mean fluctuation rate ν_m of the system, while β similarly remains within a few percent of 0.5.

We have therefore arrived at a reasonable model function with which to describe the muon spin relaxation in an ensemble of dilute superparamagnetic clusters in which both cluster sizes and cluster fluctuation frequencies are broadly distributed. As the nuclear fields at the muon site, associated with Cu and Co nuclear moments, offer an independent channel of muon spin relaxation to that resulting from the dynamic dipolar fields associated with the magnetic clusters, the final superparamagnetic muon-spin-relaxation function can therefore be written as the product of the two relaxation functions, namely,

$$P(t)=a_1\{\tfrac{1}{3}+\tfrac{2}{3}(1-\sigma^2t^2)\exp(-\tfrac{1}{2}\sigma^2t^2)\}\times\exp[-(\lambda t)^{1/2}]+a_{\rm bg}, \tag{13}$$

where

$$\lambda=4a^2/\nu_m\,. \tag{14}$$

The measured muon-spin-relaxation spectra of the $Cu_{98}Co_2$ sample annealed for 60 h at 450 °C are shown in Fig. 9. The solid lines represent least-squares fits of Eq. (13) to the spectra. In the fitting procedure σ has been fixed at the value of 0.36 μs^{-1} determined from the as-quenched $Cu_{98}Co_2$ sample leaving the magnetic relaxation rate λ as the only free parameter. Equation (13) clearly provides an excellent description of the relaxation spectra. Equally good fits are obtained for the spectra collected from the sample annealed for 17 h at 450 °C.

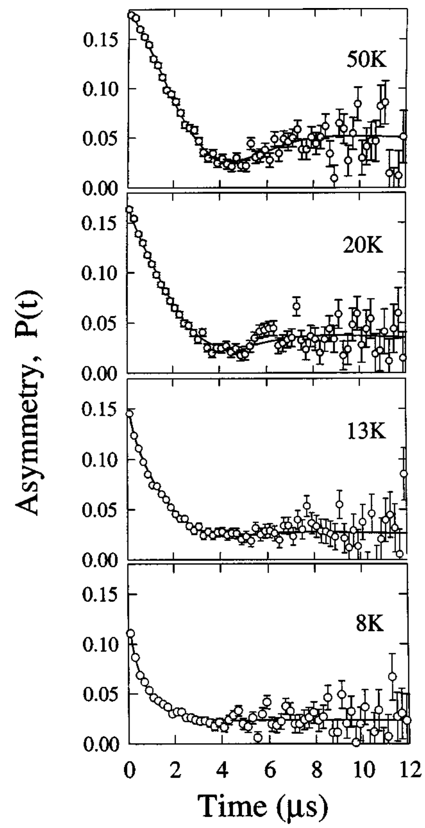

FIG. 9. The zero-field muon depolarization spectra measured for the $Cu_{98}Co_2$ sample annealed at 450 °C for 60 h. The solid lines represent fits of Eq. (13) to the data with only the muon depolarization rate λ as a free parameter.

The relaxation rate λ for both annealed samples is found to follow simple Arrhenius behavior, as witnessed by the linear temperature dependence of $T\ln(\lambda)$ for the two samples (Fig. 10). This result implies, through Eq. (14), that the mean fluctuation rate ν_m of the internal dipolar fields at

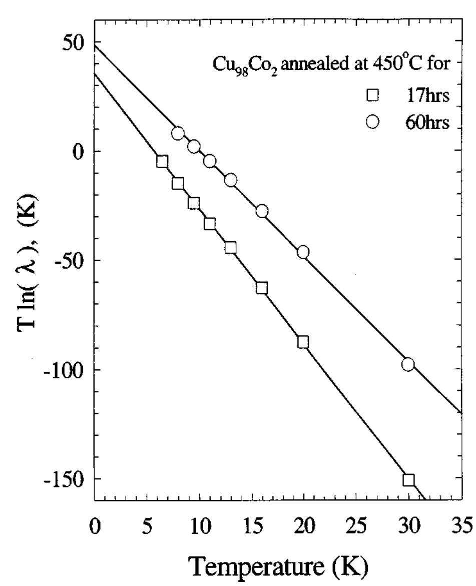

FIG. 10. The temperature dependence of the muon depolarization rate λ for the two annealed $Cu_{98}Co_2$ samples shown as an Arrhenius plot in the form $T\ln(\lambda)$ vs T.

the muon site arising from the ensemble of dynamic superparamagnetic clusters is also Arrhenius-like, i.e.,

$$\nu_m = \nu_0 \exp(-E_a/kT), \tag{15}$$

where ν_0 is the intrinsic fluctuation frequency of the fields and E_a is a mean activation energy. As we have shown that the field at a muon site is dominated by the contribution from the nearest-neighbor cluster, it is not unreasonable for us to associate ν_m with the mean fluctuation rate of the magnetic clusters themselves. Similarly ν_0 and E_a may be assumed to be closely related to the characteristic fluctuation frequency and anisotropy energy of the clusters.

From the linearized Arrenhius plots of Fig. 10 we can estimate cluster activation, or anisotropy, energies of 35.4 and 48.4 K for the $Cu_{98}Co_2$ samples annealed at 450 °C for 17 and 60 h, respectively. Correspondingly, using the value of $a = 6.8\ \mu s^{-1}$ determined from the fit of the Lorentzian Kubo-Toyabe function to the low-temperature quasistatic muon-spin-relaxation spectrum shown in Fig. 6, we obtain entirely reasonable intrinsic cluster fluctuation frequencies ν_0 of 0.7×10^9 Hz and 2.7×10^9 Hz for the two annealed samples.

Taking into consideration the Co cluster moment distributions determined by the magnetization measurements and RMC calculations discussed in Sec. II, it is apparent that anisotropy energies of the superparamagnetic clusters in the annealed $Cu_{98}Co_2$ samples scale neither with cluster volume nor surface area. It is therefore unlikely that either simple volume anisotropy or surface pinning play a dominant role in defining the intrinsic anisotropy barriers of the Co clusters within these dilute samples.

V. CONCLUSIONS

In the preceding sections we have been able to establish a theoretical framework within which μSR spectra obtained from an archetypal superparamagnetic system, namely, $Cu_{98}Co_2$, can be understood and interpreted. We have shown that for a dilute, heterogeneous, superparamagnetic system the zero-field muon depolarization function is essentially root exponential in form. Perhaps surprisingly, the associated muon-spin-relaxation rate is found to be directly related to the mean of the distribution of fluctuation frequencies of the internal dipolar fields arising from the superparamagnetic clusters, while remaining virtually independent of the width of this distribution. This behavior is closely similar to that observed in dilute spin-glass systems by Uemura *et al.*,[3] upon whose theoretical model we have based our analysis. However, the observed behavior is in marked contrast to that reported more recently by Campbell *et al.*[7] for concentrated spin-glass systems. While Uemura *et al.* report a root exponential muon-spin-depolarization function at all temperatures above the glass temperature, Campbell *et al.* describe a stretched exponential form for the depolarization function in which the exponent β decreases from unity at high temperatures to $\frac{1}{3}$ at the glass temperature, reflecting directly a theoretically predicted increasing width of the distribution of field fluctuation rates with decreasing temperature.[14] In the case of the superparamagnet system discussed here, the independence of the muon relaxation rate on the width of the distribution of the field fluctuation rates, and consequently the observation of a root- rather than stretched-exponential depolarization function is shown by our magnetization measurements and Monte Carlo simulations to be a natural consequence of the properties of a system of dilute, polydispersed, noninteracting dipoles.

We are currently extending our measurements and modeling techniques from the relatively simple case of dilute, noninteracting superparamagnetic systems discussed here to more concentrated systems in which both intercluster interactions and blocking are significant. We are also using transverse-field muon-spin-rotation methods to determine the effects of applied external fields on the internal-field distributions and dynamics in these superparamagnetic Cu-Co alloys. The results of these studies will be reported shortly.

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