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Muon methods for studying nanomagnetism

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Abstract. This paper describes the technique of muon spin relaxation and how it can be applied to the study of nanomagnetic materials. The low temperature case with static moments, high temperature superparamagnetism and the intermediate case close to the blocking temperature can all be studied. Simulation of the expected relaxation functions shows that the detailed form of the curve depends on the details of inter-particle interactions and anisotropies.

1. Introduction

The muon was first discovered in cosmic rays in the 1930s[1, 2]. Once it could be produced using proton accelerators its properties could be measured in detail[3]. High energy protons ($E > 280$ MeV, typically 500 to 1000 MeV) strike a nucleus of a light atom and produce pions by various reactions such as $p^+ + p^+ \rightarrow p^+ + n + \pi^+$. The positive and negative pions decay rapidly into muons by $\pi^+ \rightarrow \mu^+ + \nu_\mu$ with a lifetime of 26ns. Due to conservation of spin and the parity of the neutrino, the muons all have spin antiparallel to their momentum as seen in the rest frame of the pion. The muons also have a well defined momentum from this decay. We can therefore produce a 100% spin polarised muon beam.

Muons subsequently decay with a lifetime of 2.2 μs : $\mu^+ \rightarrow e^+ + \overline{\nu}_\mu + \nu_e$ and parity and spin conservation causes the positron to be emitted preferentially in the direction of the instantaneous muon spin. This polarisation is only 1/3 if all positron energies are included.

As a charged particle the muon interacts strongly with matter and will rapidly come to rest inside a bulk sample. This process takes place by electrostatic interactions and does not affect the spin polarisation.

Both positive and negative pions are produced, giving positive and negative charged muons, easily separated in the muon beam line. Most muon spin relaxation experiments use the positive variety. The muon has spin 1/2, magnetic moment 8.9 μ_N ($\gamma_\mu/2\pi = 135.5$ MHz/T) and mass $1/9\, m_p$, so it behaves like a light proton. When implanted in a material it will usually occupy an interstitial site and in metals the conduction electrons will screen its positive charge and give an exchange coupling to any local magnetic moment.

The principle of the muon spin relaxation experiment is to implant the spin-polarised muons in a material of interest, and measure the evolution of the spin as a function of time using the positron decays. It is therefore a local probe similar in theory to NMR or Mössbauer spectroscopy. In a muon spectrometer the sample position is surrounded by positron detectors to monitor the muon decay, and magnetic fields can be applied either parallel or perpendicular to the initial spin. Unlike NMR there is no reliance on the Boltzmann population of the spin states or to have a suitable spin-lattice relaxation rate, so measurements can be made at any temperature and in zero field if required.

2. Experiments

Some of the earliest muon experiments were performed on bulk ferromagnetic materials such as Fe, Ni [4] and Co [5] and followed the evolution of the internal field up to T_c. Muon behaviour in simple ferromagnets and antiferromagnets is now well understood. In a polycrystalline or multi-domain cubic material the muon polarisation is of the form

$$P(t) = \frac{1}{3}\exp(-t/T_1) + \frac{2}{3}\cos(\omega t)\exp(-t/T_2) \tag{1}$$

A review of a wide variety of muon studies of magnetism is given in [6].

Many experiments have been performed on dilute magnetic alloys and spin glasses [7] where the physical interaction of the muon spins to the magnetic moments is similar in principle to granular systems. The differences are that the time scale for fluctuations in nanomagnetic materials is often slower, and the interaction between the muon and its nearby magnetic moments is predominantly by dipolar rather than exchange interactions. Usually the non-magnetic matrix comprises the majority of the sample and gives the largest contribution to the observed muon signal - the muons interacting with dipolar magnetic fields.

Some work has been performed on nanomagnetic materials such as ball milled Ag-Fe [8, 9] and Co-Cu alloys where annealing causes phase separation [10] though the technique has not been widely applied at present.

As the muon is a light particle it may diffuse through the sample lattice at higher temperatures, and possibly trap at defect sites and grain boundaries. This may start at temperatures as low as 50K for some pure metals or well above room temperature for other materials. This has to be taken into account once the length scale of the diffusion within the muon's lifetime becomes comparable with the microstructure of the material.

The muon can also interact with the nuclear moments of nearby atoms. The nuclei in the non-magnetic matrix will usually have a slower relaxation rate T_1 than the muon due to their lower moments, so they will give an approximately static random field at the muon site leading to a Gaussian relaxation in a transverse field measurement. In a magnetically ordered or strongly paramagnetic phase the varying relaxation rate of the nuclei also changes the observed signal [11]. An ideal model system would have components with low or zero nuclear moment such as Ag or Fe. In other cases such as the Co-Cu alloys the contribution to the muon relaxation due to the nuclei must be taken into account.

3. Simulations

Computer simulations based on the Monte-Carlo technique can be used to follow the spin flips of individual grains, and to calculate any desired physical property such as a magnetic hysteresis loop. The same simulation can be used to generate the muon response to the static or fluctuating moments of the grains. Similar procedures have been used for the muon response in magnetic alloys [12].

3.1. Static magnetisation

Below the blocking temperature the magnetic grains have static moments whose values and orientations depend on the magnetic history of the sample in addition to near-neighbour and bulk demagnetisation interactions. It is therefore possible to calculate the local fields experienced by the muon at any possible stopping site. At low temperature where muon diffusion is negligible, the exact stopping sites at interstitial positions in the lattice can usually be approximated by a uniform distribution, possibly weighted to take account of the different densities of the grains and the non-magnetic matrix.

An ideal dilute system with random moments can be shown to give a Lorentzian distribution of internal field components such as $P(B_x) = \frac{B_0}{\pi(B_x^2 + B_0^2)}$ [13] where the average field B_0 depends on

the concentration of magnetic particles and their magnetic moments. This gives an exponential relaxation in a transverse external field and the "Lorentzian Kubo-Toyabe" form in zero field:

$$P(t) = \frac{1}{3} + \frac{2}{3}(1 - \lambda t)\exp(-\lambda t) \qquad (2)$$

In applied longitudinal fields the relaxation is suppressed with the final polarisation increasing from 1/3 to 1. In general the observation of a zero field relaxation of rate σ which is suppressed in a field $B \sim \sigma/\gamma_\mu$ is an indication of static magnetism, either electronic or nuclear.

In many real materials, the non-magnetic matrix contains nuclei with large moments, such as H in organic compounds. The field contribution would be Gaussian in form and would add to the field from the nanomagnets. We can usually assume that the spin relaxation rate of the nuclei is much lower than for the muon, due to their lower moments, so we still have a static field. For small nuclear fields the effect is to increase the mean field in the lowest field regions furthest from particles, so making the "dip" lower and earlier in time. High nuclear fields can largely dominate the field at the muon, giving almost the Gaussian Kubo-Toyabe with a small Lorentzian contribution at short times. A possible function which empirically fits the simulated results is:

$$P(t) = \frac{1}{3} + \frac{2}{3}(1 - \lambda t - \sigma^2 t^2)\exp(-\lambda t - \sigma^2 t^2/2) \qquad (3)$$

where the magnetic relaxation rate λ and the nuclear relaxation σ are independently specified. Another possibility is the stretched Kubo-Toyabe [14]

$$P(t) = \frac{1}{3} + \frac{2}{3}(1 - (\sigma t)^\beta)\exp(-(\sigma t)^\beta/\beta) \qquad (4)$$

where the exponent β takes values between 1 (negligible nuclear fields) and 2 (strong nuclear fields with weak particle magnetism).

It has been shown that on average 90% of the local field at a muon site is due to one nearest grain [10]. However this approximation breaks down for those muons roughly equidistant from two or more grains, whose lower internal fields give rise to the "tail" of the muon spin relaxation curve.

To illustrate the effect, we use a simple simulation where each spin is free to orient itself parallel to the local field from its neighbours (or the nearest easy axis to such a field). We can also model the ideal case with random magnetisation, or the "remanent magnetised" case with all moments parallel to a previously applied field. In the latter case the demagnetising field from the bulk magnetisation should be taken into account - muon samples are rarely spherical, but usually a flat plate perpendicular to the muon beam and field.

Figure 1 shows the simulated muon relaxation functions in zero applied field for the same particle distribution and various interactions. Although there is an apparent change of overall relaxation rate, the curves actually coincide at short times. The form of the relaxation curve varies, with the minimum polarisation having a different value.

These curves can be fitted with a stretched Kubo-Toyabe function as in equation 4 where the exponent β has the value 1.0 for the randomly oriented moments, 0.96 for Ising and 0.92 for the Heisenberg particle moments. This deviation from the ideal Lorentzian Kubo-Toyabe form is in the opposite direction to the effect of nuclear fields.

In many cases the magnetic particles are not dilute. Often we can still consider the fields in the non-magnetic matrix as being due to ideal point dipoles at the centre of each particle, giving the same form of field distribution, except that the high field end of the distribution is now cut off at the surface of the particle. There is now a significant fraction of muons stopping inside the particles, often with a well defined local field. The effect is to remove the faster relaxing part of the Lorentzian Kubo-Toyabe function and replace it with an oscillating polarisation, which may
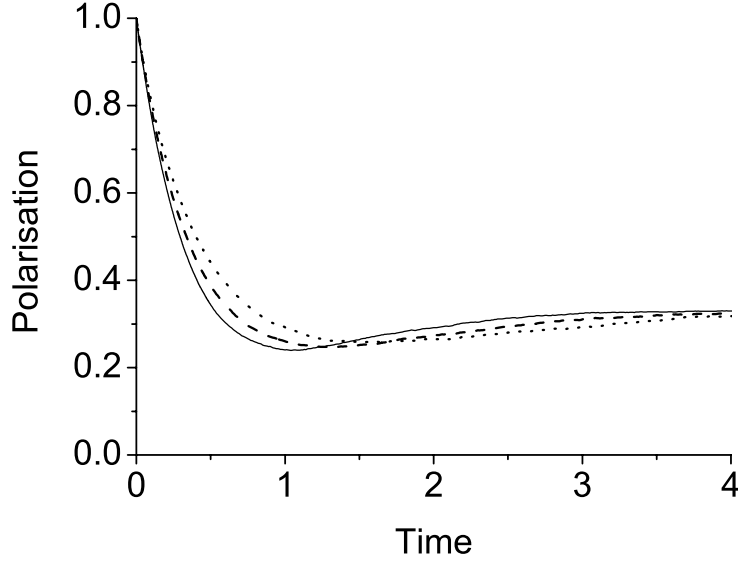
Figure 1. Simulated zero field muon relaxation for static particle magnetisation. —— random moments, giving the Lorentzian Kubo-Toyabe form, - - - - "Ising" moments relaxed along individual easy axes, ······ "Heisenberg" moments relaxed by rotation.

not be detected. The remaining polarisation may be approximately Gaussian in form, with a "missing fraction" from inside the particles.

Effects such as particle shape (needles or plates) and material texture will have to be considered to model the exact behaviour.

3.2. Fluctuating magnetisation

Well above the blocking temperature for any particle, each muon experiences a fluctuating local field with some characteristic fluctuation rate ν and RMS field value B_i. This gives a single effective "field power density" at that site, and therefore leads to a simple exponential relaxation for any one muon. This relaxation rate is independent of longitudinal field up to fields of order $\gamma_\mu B_{ext} = \nu$ and decreases as $1/B^2$ for higher fields. If this form of field dependence is observed it can be used to measure the fluctuation rate directly. If the relaxation rate is constant over the accessible magnetic field range, this implies that $\nu > \gamma_\mu B_{max}$ and its variation with temperature can still be used to infer the spin fluctuation rate.

For a mono-disperse sample where all fluctuation rates are equal, the Lorentzian field distribution from the dipolar fields gives rise to a root exponential form $P(t) = \exp -(\lambda t)^{0.5}$ for the overall muon spin relaxation. With a distribution of grain moments or rates the power law may vary from 0.5, usually to a lower value.

3.3. Intermediate cases

Close to the blocking temperature, or when the sample has a large distribution of particle sizes, the muons experience internal fields varying on the same timescale as their precession frequencies and relaxation rates. To correctly simulate this case we must follow the detailed time dependence of each spin.

We can model this case by taking a small volume of sample containing a few hundred simulated magnetic particles, with initial moment directions. Choose random muon sites within this volume and calculate the local fields at each site, taking care with boundary effects. Next
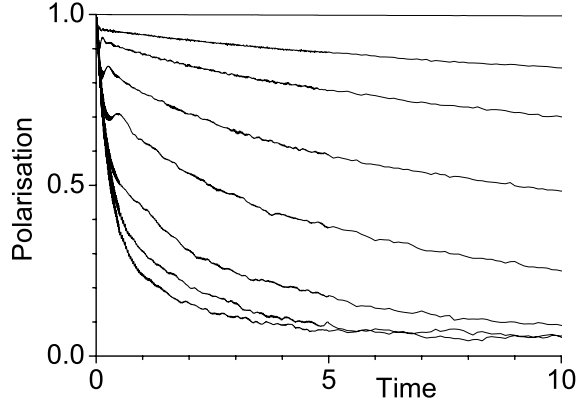
Figure 2. Simulated muon relaxation in a slowly fluctuating nanomagnet for various magnetic fields from zero (lowest curve) to high longitudinal field (top).
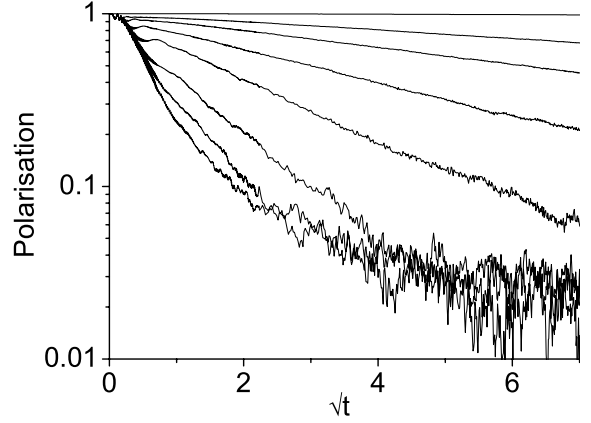

Figure 3. The data from figure 2 plotted as $\log(P(t))$ versus $\sqrt{t}$ to show the root exponential nature.

allow the muon spins to precess and measure the total polarisation for time steps from t=0 to some maximum, typically 20 μs. At randomly chosen times one or more particle moments will flip, and the local fields at the muons are adjusted appropriately. We then repeat the whole process with new particles and muon sites until sufficient statistical accuracy is obtained. Some typical results are shown in figure 2.

In zero field those muons close to one grain, with relaxation rate greater than that grain's spin flip rate, contribute to a Lorentzian Kubo-Toyabe relaxation form, while those further away with lower fields give a root exponential form. In an applied field the relaxation rate of the root exponential part decreases as the Larmor frequency of the muon becomes larger than the fluctuation rate, as seen in figure 3. Because the relaxation rate is lower, more of the muons have relatively higher fluctuation rates and so the fraction of the polarisation showing a root exponential form increases. The minimum in the Kubo-Toyabe portion is not observed in the zero field case but the corresponding "wiggles" in applied field are visible, because their period becomes shorter than the relaxation time.

4. Conclusions

Muon spin relaxation can give much information on the local properties of nanomagnetic materials. Simulation of the magnetic dynamics and calculation of the corresponding muon relaxation functions should confirm the model and give values for the interactions and particle anisotropies.

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