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Abstract. Muon spin relaxation in the amorphous alloy $Ni_{35}Ti_{65}$ containing interstitial hydrogen has been measured in longitudinal geometry at zero applied magnetic field, at temperatures in the range 18–340 K. The relaxation profiles at all temperatures were consistent with time-dependent magnetic fields at the muon site and were analysed by means of a dynamic form of the Kubo–Toyabe relaxation theory. The correlation time of the local field at the muon below 80 K was found to be about 6 μs and to vary only slowly with temperature. Above $\sim$ 250 K, where the diffusion hopping rate of the hydrogen atoms is about 10^7–10^8 s^{-1}, as measured by nuclear magnetic relaxation methods, the activation energy of the correlation time is similar to the activation energy of the hydrogen diffusion. The relaxation of the muon spin at these temperatures is ascribed to the highly correlated motion of the muon and the hydrogen atoms. Some Monte Carlo calculations of the fluctuation rates of the local fields under such conditions are reported. According to the outcome of these calculations the experimental data are consistent with the muon having an intrinsically lower diffusion rate than the hydrogen atoms.

1. Introduction

Several measurements have been made of muon spin relaxation in amorphous alloys which contain hydrogen as an interstitial atom [1, 2], and in all cases the relaxation appears to be associated with the motion of either muons or hydrogen atoms. Typically, below $\sim$ 200 K the relaxation varies slowly with temperature, whereas at higher temperatures, where the hopping rate of the hydrogen atoms is 10^7–10^8 s^{-1}, the temperature dependence has an activation energy consistent with that of the hydrogen motion. This aspect of the experimental data is regarded as evidence that the motions of the muons and the hydrogen atoms are strongly correlated at the higher temperatures. In the present paper, measurements of muon spin relaxation in zero applied magnetic field on a hydrogen-loaded amorphous $Ni_{35}Ti_{65}$ alloy which confirm the general features of the previous experiments are reported. One result of these measurements, namely the value of the second moment of the local field distribution at the muon, has caused us to re-evaluate some earlier measurements made on the same alloy in transverse geometry and we are able to show that the correlation times calculated from the two experiments have a similar dependence on temperature.

An apparently paradoxical feature of these experiments is that the correlation time, τ, derived from the muon relaxation at high temperatures is generally greater than the average jump interval of the hydrogen atoms and a convincing explanation of the difference has not been given [3]. For uncorrelated motion, for example at low concentrations of hydrogen, $1/\tau = n_\mu + n_H$ where n_μ and n_H are the hopping rates of the muons and hydrogen atoms respectively and τ must be less than $1/n_H$. However, at the large hydrogen concentrations

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encountered in the experiments where the hydrogen atoms may block the motion of the muons, this equation no longer holds. Consequently, as an adjunct to the experimental work we have made calculations of muon correlation rates by means of Monte Carlo simulation methods, which demonstrate the effect of concentration. In the examples chosen, muons have either an intrinsically higher or lower mobility than hydrogen atoms. For highly mobile muons the simulation shows that $1/\tau$ approaches n_H at high concentration. On the other hand, when the muon is not very mobile $1/\tau$ is about $n_H/2$. We will show that the supposition that the muon has an intrinsically low mobility offers the most direct interpretation of the experimental data. However, since the calculations are based on a simplified version of the muon–hydrogen spin dipolar coupling and classical hopping we cannot claim that it offers a definitive explanation.

2. Experimental details

The $Ni_{35}Ti_{65}$ alloy was prepared by melting the constituent metals in an argon arc furnace and melt-spinning to produce a metallic glass ribbon. The experimental sample was charged with hydrogen by the electrolysis of dilute sulphuric acid, with a platinum anode and the specimen as the cathode, to a hydrogen to metal atom ratio of 1.5, as determined by subsequent desorption of the hydrogen. The experiment was conducted on the pulsed muon spectrometer, μSR, at the ISIS facility of the Rutherford Appleton Laboratory, UK with a sample made from high-purity (99.995%) Ni, known from our previous work [2] to be free of paramagnetic impurities which may give spurious muon depolarization rates. Sample temperatures were maintained in the range 20–340 K by a helium refrigerator and the time dependence of the muon polarization was measured in longitudinal geometry at zero applied magnetic field.

Even at the lowest temperature at which the experiment was conducted, $\sim$ 18 K, the observed evolution of the muon spin polarization obtained from the asymmetry of the decay positrons did not have the characteristic shape associated with the relaxation function of static muons. According to the Kubo–Toyabe model [4, 5] this relaxation function when no magnetic field is applied has the form

$$G_Z(t) = \tfrac{1}{3}[1 + 2(1 - \Delta^2 t^2)\exp(-\tfrac{1}{2}\Delta^2 t^2)] \quad (1)$$

in which the polarization falls from its initial value to a minimum and then returns asymptotically to 1/3 of the initial value. Here Δ^2 is the zero-field second moment of the local field distribution given in frequency units. In quantum mechanical modifications of this model, for example that given by Celio [6], the polarization does not have the same asymptotic behaviour but returns from the minimum to a maximum value. The same general shape was observed in the experimental data at low temperatures. However, the theoretical maximum of the static function is close to 1/3 of the initial polarization whereas the maximum in the observed $G_Z(t)$, which was near 7 μs at low temperatures, fell well short of this. In fact, the difference in value between the observed maximum and the minimum near 5 μs was significantly less than its equivalent found in any of these models. We draw the conclusion that the muon was not stationary at any temperature in the experimental range and that the data are best explained by dynamic forms of the Kubo-Toyabe model which alloy for diffusion and have forms consistent with the observed $G_Z(t)$.

Analysis of the experimental data has been carried out with the aid of calculated versions of the relaxation functions based on the strong collision version of the dynamic Kubo–Toyabe model given by Hayano *et al* [5]. In strong collisions it is supposed that the correlation of the local field is lost when the muon makes a jump and the characteristic time, τ, associated with the correlation decay, is usually identified as the mean residence time of the muon at a given site having fixed surroundings. In the present work, the hydrogen atoms are also free to move and alter the dipole field at the muon, and it seems more correct to describe τ simply as the correlation time of the local field at the muon, irrespective of whether it depends on either the motion of the muon or the hydrogen atoms. Defining τ this way is in fact consistent with the earliest version of the Kubo–Toyabe model [4] and we note that the difference between the relaxation functions in this model and the Hayano version is not significant at small τ, where the effect of the hydrogen motion is greatest. This is also true of other modifications of the Kubo–Toyabe model which describe the relaxation of diffusing muons [7] and the principal differences which do occur are confined to the correlation times greater than Δ^{-1}, that is, at temperatures below about 150 K in the present experiment.

Time in the relaxation function is most conveniently given in units of Δ^{-1} and the parameter $\tau\Delta$ determines the time dependence and the functional form. At large $\tau\Delta$ and low temperature where the shape of the relaxation function changes considerably, it was necessary to adjust both Δ and τ to fit the data. This was done graphically in the first instance to find $\tau\Delta$, that is, the most appropriate shape for $G_Z(t)$, following by adjustment of Δ using a weighted mean square method. At temperatures below 60 K, the value of the second moment could be determined from the position of the minimum in the relaxation profile. At temperatures above 150 K, the diffusion has become fast enough to remove the minimum in $G_Z(t)$ and above 250 K, the relaxation is approximately exponential, which simplifies the fitting procedure. The experimentally derived value of Δ and the associated second moment, M_{2Z}, are shown in table 1 while the temperature variation of the correlation time of the local field derived from the relaxation data is given in figure 1. The estimated error in Δ is $\pm 5\%$ and in $\tau \pm 20\%$ at the lowest temperature, falling to $\pm 10\%$ at the highest temperature.

3. The local field at the muon

The most obvious mechanism giving rise to the relaxation of the muon polarization is the dipolar coupling between muon and the nuclear spins of the hydrogen atoms. Confirmation that this is the principal mechanism is provided by comparing M_{2Z} with the second moment of the dipolar field at the hydrogen atoms themselves. This can be obtained from nuclear magnetic relaxation. The proton relaxation time, T_1 in $(Ni_{35}Ti_{65})H_{1.5}$, measured at the Larmor frequency $\omega = 2\pi \times 18$ MHz is known to be characteristic of diffusing spin dipoles and to have a minimum value of 20.4 ms at 375 K [8]. The relationship between the T_1 minimum and the second moment, M_2, depends to a moderate extent on the theoretical model chosen but, for low spin concentrations and cubic lattices, which have similar local atomic arrangements to those in metallic glasses, a good working relationship [9]

Table 1. The experimentally derived value of Δ and the associated second moment M_{2Z}

Δ (μs^{-1})	M_{2Z} (μs^{-2})	$M_{2Z}(H)$ (μs^{-2})
0.36	0.127	0.074

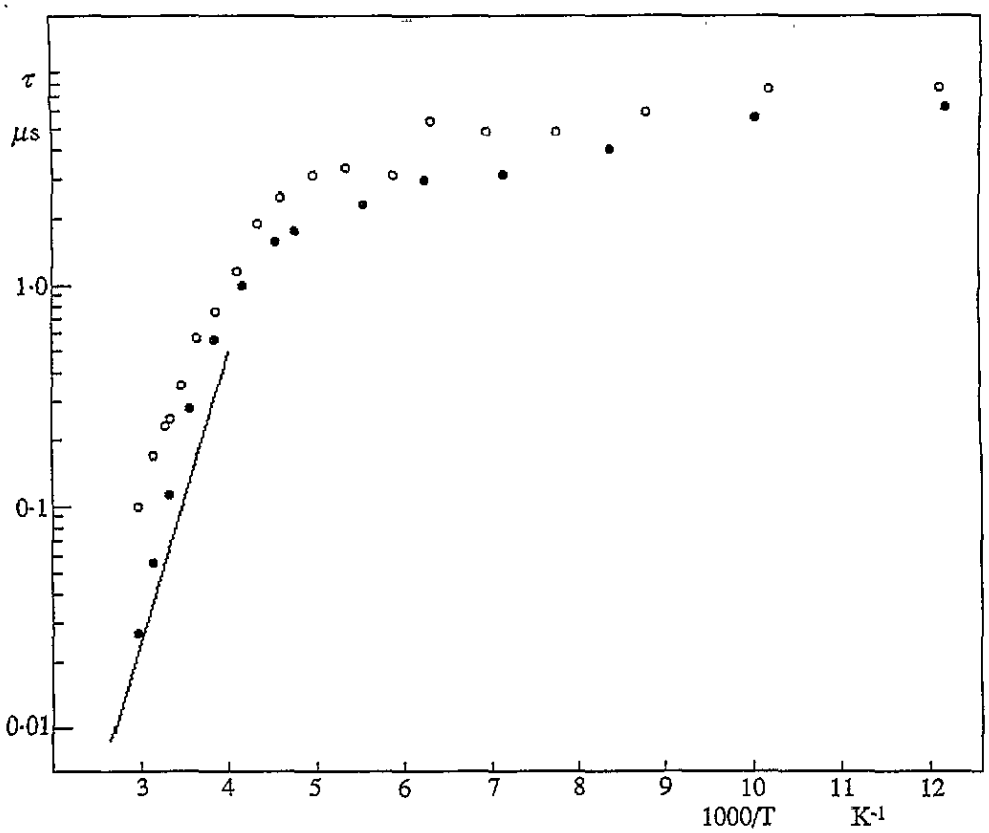

Figure 1. The correlation time, τ, of the local field at the muon calculated from its relaxation profile according to the dynamic form of the Kubo–Toyabe theory [5] and plotted as a function of reciprocal temperature. The closed points are from data measured in zero applied field in the present experiment and the open circles refer to data obtained in transverse geometry in an earlier experiment [14]. The solid line shows the interval between diffusion hops of the hydrogen atoms as calculated from nuclear magnetic relaxation [8]. As indicated in the text this line represents maximum, rather than probable, values for the jump interval. The scale on the horizontal axis has been chosen to give an adequate display of the differences between the various characteristic times above 250 K with the result that some experimental data is missing from the figure. Between ~ 80 K and 18 K the correlation time is a slowly varying function of temperature, never rising above 7 μs.

is $T_1(\text{min}) = \omega/0.84M_2$. This expression needs to be revised for larger concentrations but the necessary adjustment to the numerical factor according to Monte Carlo calculations is only about 4% [10]. For the present alloy the expression gives $M_2 = 6.56 \times 10^{-3}\ \mu s^{-2}$. If it is assumed that the muon occupies one of the hydrogen sites the second moment of the muon–hydrogen dipolar coupling can be calculated from this value by multiplying by the square of the ratio of the Larmor frequencies together with a number of factors which depend on the nature of the spin coupling. It is first necessary to introduce a factor of 4/9 [5], since, in contrast to the proton dipolar coupling, the muon–hydrogen interaction is between unlike spins and involves only the secular terms of the total dipolar coupling. Including this factor gives the transverse field muon second moment, $M_{2T}(H)$, equal in this case to $29.5 \times 10^{-3}\ \mu s^{-2}$. A second factor of 5/2 [5] is required to convert this high-field second moment to the zero-field moment arriving at the value of 0.074 μs^{-2} given in table 1.

The ratio of the experimental zero-field second moment and that calculated from the hydrogen dipolar coupling is 1.73. Similar ratios have been found in other metallic glasses [2] and it has been pointed out that they do not negate the inference that the local field at the muon arises solely from the nuclear spin dipoles of the hydrogen atoms. Since $M_2 \propto r^{-6}$, where r is the separation of the spins, this ratio can interpreted either as a decrease in the muon–hydrogen distances of $\sqrt[6]{1.73}$, that is about 10%, compared with the hydrogen separation, or as the partial occupation of sites between those filled by the hydrogen atoms. Provided muons do not obey the same rules of site occupancy as hydrogen atoms it can be argued that there is ample space available to them, since it is well known that site blocking occurs in hydrogen–metal alloys [11] which prevents a pair of hydrogen atoms occupying neighbouring interstitial sites in the metal matrix. The fraction of muons not obeying the same occupancy rules as hydrogen can only be small. A simple calculation assuming only first-neighbour interactions shows that just 1% of the muons need deviate from the site blocking rule to account for the difference in second moments if the 1% keep the same local coordination and are on average mid-way between hydrogen atoms.

It is also possible to show that the second moment is consistent with the spacing between hydrogen sites as determined by structural methods. The second moment, M_{2Z} for muons embedded in a simple cubic lattice of hydrogen nuclear spins, I, is

$$M_{2Z} = (2/3)I(I+1)\hbar^2\gamma_H^2\gamma_\mu^2 \sum_i r_i^{-6} \tag{2}$$

where γ_H and γ_μ are the gyromagnetic ratios of the hydrogen and muon respectively [12]. The sum $\sum_i$ is $8.5/a^6$ where a is the lattice parameter. When M_{2Z} has the experimental value, a turns out to be 2.37 Å and using the value derived from the nuclear second moment gives $a = 2.63$ Å. Neutron scattering methods [13] indicate that hydrogen in both crystalline and amorphous $NiTi_2$ occupies sites surrounded by either octahedral or tetrahedral arrangements of metal atoms, with the latter being preferentially occupied. The average spacing between hydrogen atoms in deuterated amorphous $NiTi_2$ has been identified as 2.56 Å for tetrahedral sites and ~ 3 Å for octahedral sites. The use of the cubic sum for comparison purposes is questionable, since in amorphous materials the atomic polyhedra are highly distorted and no two sites are equivalent. In addition, because of site blocking, the average coordination number of the hydrogen atoms is probably higher than for cubic lattices. The comparison is clearly approximate but, in spite of this, the similarity in the various values for the hydrogen spacing does lend support to the view that the depolarization rate is a consequence of the muon–hydrogen dipolar coupling and the muons and hydrogen atoms occupy similar interstitial sites. Calculations based on the interatomic spacings given in [13] show that the dipolar contribution from the nuclear spins on the metal atoms is more than an order of magnitude lower.

4. Comparison with earlier work

The value of the second moment found in the present experiment also has some bearing on the muon depolarization rates in this alloy measured earlier in transverse field geometry [14]. In the transverse experiment motional, narrowing was observed, particularly at temperatures above 250 K and, consequently, the polarization decay was analysed with the aid of the Abragam function [12]

$$G(t) = \exp[-M_{2T}\tau^2\{\exp(-t/\tau) - 1 + t/\tau\}] \tag{3}$$

which offers a way of tracing the evolution of the decay from its Gaussian form, characteristic of static dipolar coupling, at large τ to the motionally narrowed exponential form

$$G(t) = \exp(-\lambda t) \qquad \lambda = M_{2T}\tau \tag{4}$$

at small τ. In the earlier work, the experimental decay at the lowest temperature was taken to represent the static case which gave a magnitude for M_{2T} equal to 0.039 μs^{-2} in contrast to the value of 0.051 μs^{-2} found in the present work. The fact that experimental value of M_{2T} is lower clearly reinforces the conclusion that the muon is not static even at the lowest experimental temperature. The values of τ derived in the earlier paper can no longer be regarded as true. Consequently in figure 1, we give a new interpretation of earlier data based on the current value of M_{2T}. The new values of τ in the motionally narrowed region above 250 K have been obtained by multiplying the earlier data by the ratio of the new and old second moments and, in the region where the exponential approximation does not apply, τ has been obtained by fitting equation (3) directly to the experimental data using the present value of M_{2T}. It can be seen that the jump times derived from the transverse field measurements are consistently greater than those obtained in zero field by a factor of about 1.5. Apart from this the same general trend is observed in each data set.

5. The correlation time of the local field

In the motionally narrowed region above 250 K the temperature variation of τ is consistent with an activation energy 0.22 eV and 0.3 eV for the transverse field and zero-field data respectively. These values are similar to the activation energy of the hydrogen diffusion, which, as measured by nuclear magnetic relaxation [8], is 0.26 eV. This similarity is a frequently observed feature of this type of experiment [1–3] and it has generally been accepted as evidence that the motions of the muons and hydrogen atoms are highly correlated in the sense that the motion of the muon is blocked by the hydrogen atoms. The same conclusion, reinforced by the inference given above that the muons and hydrogen atoms occupy similar sites, can be reached in the present case also. A second feature, also found in other experiments [1–3], is that the muon correlation time is longer than the average interval between hops of the hydrogen atom. The solid line in figure 1 is the jump interval of the hydrogen atoms, τ_H, obtained from the assumption that $\omega\tau_H = 1$ at the temperature of the minimum in the nuclear magnetic relaxation time, T_1. It can be seen from the figure that the correlation time of the dipole field at the muons, as measured in zero external field at high temperature, is on average a factor of 1.6 greater than the hydrogen jump interval given by this line. The relation $\omega\tau_H = 1$, although often quoted, is only an approximation and in fact $1 > \omega\tau_H > \frac{1}{2}$ [10, 14] which means that the broken line essentially represents maximum possible, rather than likely, values for τ_H. A better estimate of the factor between the jump and correlation times is probably nearer 3 than 1.6.

If the same variation of the hydrogen jump rate continues to lower temperatures, below about 150 K, the hydrogen atoms may be considered as static in the correlation time of the muons. In this temperature range the apparent activation energy is very small and more characteristic of quantum mechanical tunnelling than classical hopping. The phonon-assisted tunnelling of small polarons [15, 16] is often used to describe the motion of muons in metals near this temperature. According to the original form [15] of this theory the hopping rate is

$$\tau^{-1}(T) = \nu_0 \exp[-E_a/kT] \tag{5}$$

with

$$\nu_0 = A_J T^{-1/2} = J^2[\pi/4\hbar^2 E_a k T]^{1/2} \qquad A_J = J^2[\pi/4\hbar^2 E_a k]^{1/2} \tag{5a}$$

where J is the tunnelling matrix and E_a is the adjustment between the muon energy levels on neighbouring sites. This expression may be fitted to the data in the range 80–170 K by choosing the values $E_a = 0.017$ eV and $A_J = 0.5\ \mu s^{-1}\ K^{1/2}$ but, because of the competing nature of the temperature dependences of the terms in equation (5), the fitted curve is concave upwards rather than concave downward as indicated by the data. The effect is particularly marked in the present case because of the low value of E_a. For reasons of presentation not all values of τ have been included in figure 1 and in fact τ is almost independent of temperature below 80 K. Possibly the discrepancy in the nature of the curves arises from the inappropriate use of small-polaron theory in disordered materials or from defects in its original form, which does not converge to classical behaviour at high temperature. Newer versions [17, 18] of the theory exist but, since these are more difficult to deal with in an experimental context and the possibility of obtaining a definitive identification the mechanism of the depolarization is not high, we have not proceeded any further.

In the temperature range where the muon and hydrogen motions are correlated, the degree of correlation and the relative activation energies and jump rates must depend on the concentration of the hydrogen atoms. In order to illustrate this point we have calculated, by means of Monte Carlo simulation, the correlation time of the local magnetic field at the muon in a simple model in which the hopping motion of the muon is blocked by the hydrogen atoms. When the concentration of the hydrogen is low enough to allow the muons to move independently, simple arguments based on the assumption that diffusion takes place by hopping to vacant sites lead to the relationship $\nu \equiv 1/\tau = n_\mu + n_H$, where n_μ and n_H are the hopping rates of the muons and hydrogen atoms respectively; that is, the local field at a muon ceases to be correlated with its former value when either the muon or a neighbouring hydrogen atom hops. If the hydrogen atoms and muons have the same hopping rate, ν has the value $\nu_1 = 2n_H$. On the other hand in the limiting cases of $n_\mu \gg n_H$ and $n_\mu \ll n_H$, ν becomes $\nu_\infty \approx n_\mu$ and $\nu_0 \approx n_H$, respectively. In addition $\nu_\infty/\nu_1 \approx \frac{1}{2}n_\mu/n_H$ and $\nu_0/\nu_1 \approx 1/2$. Of course when the two motions are independent the activation energies associated with n_μ and n_H are not necessarily identical. As the concentration of the hydrogen increases, blocking the motion of the muon, these simple relationships are no longer applicable. The analogous problem of nuclear magnetic relaxation rates in concentrated spin systems caused by random fluctuations of the dipolar coupling has been treated both theoretically [21] and by means of Monte Carlo simulation [10, 14]. An adaptation of the latter method would seem to be an attractive approach to the present problem except for the fact that the correlation functions which govern the muon depolarization are much less tractable than their nuclear magnetic equivalents. In order to circumvent this problem we have dealt instead with a simplified correlation function which, it turns out, appears to be able to demonstrate the effect of high concentration.

In the longitudinal geometry of the present experiment, the relaxation of the muon spin depends on the component of the local field in a plane transverse to the direction of the polarized beam and this in turn depends on the distribution and nuclear spin vectors of the neighbouring hydrogen atoms [5]. In order to reduce the number of averaging processes, we adopt the approximation that the local field at the muon can be replaced by its average over all (isotropic) directions of the hydrogen nuclear spin dipoles. Within the framework of the Kubo–Toyabe formulation this allows us to obtain the mean square local transverse field in a closed form [5], namely,

$$\langle H_T^2 \rangle = I(I+1)\gamma^2 \sum_i r_i^{-6}(5 - 3\cos\theta_i)/3 \tag{6}$$

where r_i, θ_i, ϕ_i are the polar coordinates of the nuclear spins, I. It is then possible to calculate the correlation function of this quantity by Monte Carlo simulation in much the same way as the correlation functions of the spin dipolar coupling. The details relating to the hopping diffusion of the particles in the Monte Carlo method used in present work to do this have been given elsewhere [14]. The simulation assumes classical hopping but accounts for the atom–vacancy correlation effects which occur at high concentration. The principal modification made was that one particle, the muon, was given a different hopping rate from the others and the correlation function of its local field at several different hydrogen concentrations was evaluated as the hopping progressed. Three examples of muon jump rates were chosen, corresponding to $n_\mu = 10n_H$, n_H and $0.1n_H$, respectively.

These correlation functions were found to depart from the simple exponential form, making it impossible to define a single correlation time. For $n_\mu = n_H$ at the lowest concentration, 0.1, the time dependence could be described by the sum of two exponentials, one of which had a decay rate consistent with $\nu_1 = 2n_H$, as indicated above. The rate associated with the tail of the decay is about an order of magnitude less than this. At higher concentrations there is an increasing disparity between the decay rates and $2n_H$. However, it turns out that the correlation functions have the same functional shape, and scale linearly with time. In the absence of a better method we have taken this scaling factor as an indication of the way in which the muon correlation time changes with concentration. It can be seen from the inset of figure 2 that the scaling factor, normalized to unity at the 0.1 concentration changes by about 2 over the whole concentration range, that is $\nu_1 \equiv 2n_H$ at dilute concentration, falling to $\nu_1 \equiv n_H$ at high concentration. Both the departure from exponential and this change in correlation time have analogues in the spin correlation functions in nuclear magnetic relaxation [10, 14]. A similar scaling is also found in the other correlation functions, so it is possible to evaluate the ratios ν_{10}/ν_1 and $\nu_{0.1}/\nu_1$ for the jump rates $n_\mu = 10n_H$ and $n_\mu = 0.1n_H$ in the same way. Their variation with concentration is given in figure 2, where it can be seen that at low concentration the ratios are consistent with ν_∞/ν_1 and ν_0/ν_1 given above. At large hydrogen concentrations, muons with intrinsically high diffusivity clearly become less mobile as their diffusion paths are blocked by the hydrogen atoms. It is a reasonable guess that in the single-vacancy limit of high concentration ν_∞/ν_1 will converge to unity since the motion of the vacancy is controlled by the hydrogen atoms. However, it has not been possible to demonstrate this convergence even though the simulation was conducted with only $\frac{1}{2}\%$ of the hydrogen sites vacant. It therefore seems from these results that blocking the motion of an intrinsically highly mobile muon cannot explain the experimental difference between the muon correlation time and the hydrogen jump interval.

In contrast, muons with an intrinsically low diffusivity turn out to be relatively less affected and for them $\nu_{0.1}/\nu_1$ does not rise above 0.56, even at the highest concentration. In this case there is no reason for convergence to $\nu_{0.1}/\nu_1 = 1$ and furthermore, since here $\nu_1 \equiv n_H$, the muon correlation time, τ, is given by $\tau/\tau_H = 1/0.56 = 1.8$, where $\tau_H = 1/n_H$. It thus takes about two hops of the hydrogen atoms to remove the correlation of the local dipolar field. In view of the fact that the dipolar coupling has a very short range, this number is likely to be sensitive to the coordination of the near-neighbour hydrogen sites. The site coordination number in the model is 6 but in amorphous alloys it may be higher since, as indicated above, a form of site blocking exists in metals in which hydrogen atoms tend not to occupy neighbouring sites. This raises the possibility that in a realistic model more hydrogen atoms would need to move in order to reduce the correlation which in turn increases the factor between the correlation time and jump interval.

This description of the muon–hydrogen correlations at high concentration may be of

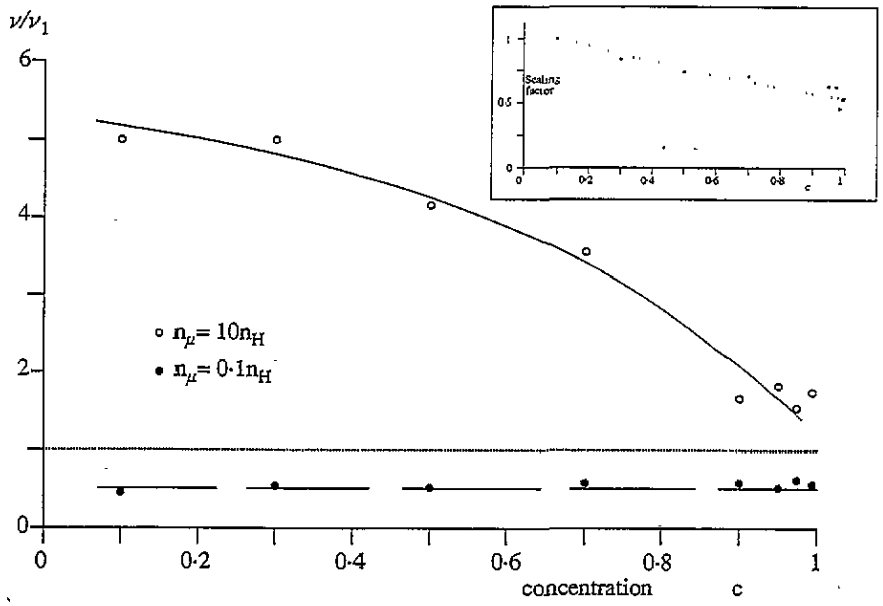

Figure 2. Relative decay rates, ν, of the correlation function of the local field at the muon from Monte Carlo calculations based on the simple model of muon–hydrogen correlated diffusion described in the text. In the inset the scaling factor of the correlation decays obtained when the jump rate of the muon, n_μ, equals the hydrogen jump rate, n_H, is shown as a function of the hydrogen atom/site ratio, c. The scaling factor, which, as indicated in the text, is a measure of the relative decay rate as a function of hydrogen concentration, has been normalized to unity at $c = 0.1$. Note that the decay rate of the correlation function is reduced by a factor of about 2 as the atom/site ratio is increased to near unity. The main figure shows decay rates, ν, for n_μ equal to $10n_H$ and $n_H/10$ relative to the decay rate, ν_1, for $n_\mu = n_H$. Only the decay of the $10n_H$ curve shows a marked change from its value at very small hydrogen concentration when the atom/site ratio is increased.

limited validity because of its appeal to classical hopping and the use of a simplified form of the dipolar coupling. Given that these limitations are not significant, the simulation appears to offer a direct explanation of the experimental observation that $\tau > 1/n_H$ provided it can be assumed that the intrinsic mobility of the muon is less than that of the hydrogen atoms above 250 K. Whether this assumption is a reasonable one, given the masses hydrogen atoms and muon, is open to question. However, it is reinforced by the results of experiments which make comparison between hydrided and hydrogen-free alloys. Unfortunately, such experiments depend on the presence of large nuclear spin moments on the metal atoms and the number of candidates among amorphous alloys is limited. However, the persistence of long muon correlation times to high temperatures has been observed in amorphous CuZr [19] and CuTi alloys [20]. In the CuTi alloys at low temperatures, the correlation time is essentially the same whether hydrogen is present or not and also similar to the present observations. However, at temperatures around 300 K the correlation time in the hydrided sample falls below that in the hydrogen free alloy by a factor of about 5. On the other hand, supporting evidence from crystalline alloys is more ambiguous. In some cases there is little qualitative difference between the crystalline and amorphous states [1] and in others, including crystalline CuTi alloys, the motion of the muon is more rapid when no hydrogen is present [3, 20]. We cannot offer an explanation of why the muon should have an intrinsically

lower mobility than the hydrogen atoms and, as far as we are aware, there is none in the current literature in spite of the accumulation of experimental data which could be explained by this proposition.

6. Summary

In the present experiment the muon depolarization rates have distinct similarities with earlier measurements [1, 2] on hydrided amorphous alloys, and in particular the activation energy of the correlation time at temperatures above 200 K is similar to the activation energy of the hydrogen diffusion. The simplest explanation of the data is that the correlation times found at low temperatures are those pertaining to a static array of metal and hydrogen atoms and possibly the local tunnelling of the muons. As the low activation energy would imply, this intrinsically low diffusion rate of the muons apparently persists to higher temperatures and, in view of the results of the Monte Carlo calculation of the correlation functions, we conclude that the correlation time at the higher temperatures is reduced by the motion of the hydrogen atoms rather than any significant change in the relatively slow motion of the muons.

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