Study of μ^+ Localization and Diffusion in Sc and ScH_{0.05}*

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The location and diffusion of positive muons in single crystal samples of Sc and ScH_{0.05} have been studied. Low temperature results clearly show that, as in the case of hydrogen, the muons occupy the tetrahedral sites in the hcp lattice and produce a slightly anisotropic local expansion of the surrounding metal atoms. At higher temperatures motional narrowing is observed due to the onset of μ^+ diffusion and the deduced correlation times follow an Arrhenius behaviour above 100 K. The corresponding activation energies are 0.025 eV for Sc and 0.054 eV for ScH_{0.05}. A significant deviation from Arrhenius behaviour was observed for the ScH_{0.05} sample below 100 K and may be related to muon trapping by H.

1. Introduction

The recent discovery of quantum effects associated with the diffusion of H in scandium and yttrium [1–3] has led us to study the behaviour of muons in these metals since such effects should be more readily observable for the muon whose mass is approximately nine times smaller than that of the proton. In the case of H two distinct kinds of motion have been observed. Long range diffusion involves hops between tetrahedral (T) sites via a neighbouring octahedral site and rather slow hopping rates are observed ($\sim 10^4 \text{ s}^{-1}$ at 300 K) with activation energies of the order of 0.57 eV [4]. On the other hand a fast localised motion between neighbouring T sites has been observed by neutron scattering with hopping rates exceeding 10^{10}

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Fig. 1. Transverse field relaxation rate σ measured in a pure scandium single crystal at 12 K as function of the applied magnetic field. Data sets were taken for the three crystal orientation $\theta = 0^\circ$ (triangles), $\theta = 43^\circ$ (open diamonds) and $\theta = -45^\circ$ (open circles). A simple calculation of the second moment of the lattice dipolar field at the μ^+ (at a T-site, assuming a radial EFG and isotropic local lattice expansion) yields the dotted line for $\theta = 0^\circ$ and the dashed line for $\theta = \pm 45^\circ$.

s^{-1} at 300 K [1]. This motion is attributed to protons moving in a rather symmetric potential whereas a similar but slower local motion observed in NMR experiments [5] seems to be correlated to protons involved in short range ordered pairs [6] and thus moving in asymmetric potentials. The temperature dependence of both localised motions does not follow an Arrhenius behaviour below 300 K apparently due to quantum effects.

In this paper we present results of muon spin rotation (μ SR) measurements in single crystal samples of Sc and $\text{ScH}_{0.05}$ aimed at determining both the muon site in the hcp lattice and the dynamics below 300 K. All measurements were made at the Paul Scherrer Institut, Switzerland.

2. Muon site

In pure Sc μ SR relaxation rates have been measured as function of the applied magnetic field in a transverse field geometry at a temperature of 12 K. Scans were performed for different values of the angle θ between the hexagonal c -axis of the crystal and the direction of the externally applied field (Fig. 1). It can be seen that (i) markedly field-dependent anisotropic gaussian relaxation rates σ are obtained, (ii) $\sigma(\theta)$ is symmetric about $\theta = 0^\circ$, (iii) for the 3 angles θ at which data were obtained the dependence $\sigma(B)$ reaches a saturation value for $B \geq 0.3$ T. From the latter observation it is

Fig. 2. Apparent hopping rate R_A displayed versus inverse temperature for muons in a ScH_{0.05} single crystal oriented with $\theta = 90^\circ$ relative to a transverse magnetic field of 36 mT. R_A is obtained from an Abragam formula fit to the μ SR data. An Arrhenius relation (dash-dotted line) holds for $T \geq 110$ K; it corresponds to an activation energy of 0.058 eV. From 110 K down to 95 K a surprising increase of R_A is detected.

concluded that the field distribution at the μ^+ site, originating from the magnetic nuclear Sc moments, corresponds to the high-field (Van Vleck) limit for B above 0.3 T.

Assuming that the positive muons are not diffusing at 12 K, the second moment, M_2 , of the field distribution can be easily calculated for the different interstitial μ^+ sites possible. Comparison between the calculated M_2 ($= \sigma^2$) and the measured relaxation $\sigma(0^\circ) = 0.3547(35) \mu s^{-1}$ and $\sigma(45^\circ) = 0.2029(12) \mu s^{-1}$, in the high field limit, indicates that only the tetrahedral interstitial sites have to be retained for the muons.

Better agreement between the calculations and the measurements was obtained by including a lattice expansion around the μ^+ , resulting in an increased distance of about 5% between μ^+ and its 4 nearest-neighbor Sc atoms. More precisely, calculations and measurement agree exactly if a slightly anisotropic local lattice expansion of 3% along the c -axis and a linear expansion of about 6% for all directions perpendicular to the crystal-line c -axis are assumed.

For the moment we have not succeeded in explaining exactly the observed $\sigma(B, \theta)$ for field values below the Van Vleck limit. By assuming electrical field gradients (EFG) coupled to the Sc nuclear quadrupole moments and either oriented parallel to the c -axis or pointing radially to the muon location, one can reasonably well reproduce the $\sigma(B)$ behaviour for the $\theta = \pm 45^\circ$ orientations. However, for $\theta = 0^\circ$ the dip of $\sigma(B)$ observed for B below the Van Vleck limit seems difficult to understand.

Fig. 3. Apparent hopping rate R_A displayed versus inverse temperature for muons in a pure Sc crystal oriented with $\theta = 0^\circ$ relative to a transverse magnetic field of 36 mT. R_A is obtained from an Abragam formula fit of the μ SR data. An Arrhenius relation (solid line) holds for $T \geq 80$ K; it corresponds to an activation energy of 0.029 eV. In contrast to the situation in $\text{ScH}_{0.05}$, R_A decreases continuously over the temperature range measured.

3. μ^+ and/or H diffusion

The muon depolarization rates were measured in transverse fields of 36 and 296 mT in the temperature range 12–340 K for various values of θ . Motional narrowing, due to an averaging of the field distribution sampled by the muon, was observed for Sc above 50 K and for $\text{ScH}_{0.05}$ above 70 K. The data were interpreted in terms of the commonly used Abragam approximation [7] to extract the diffusional correlation time τ_c which reflects the μ^+ hopping rate and the motion of associated protons if present. As can be seen from Figs. 2 and 3 both samples exhibit a temperature regime where the correlation time is well described by an Arrhenius behaviour. For the pure Sc sample this regime extends down to 80 K, however in the case of $\text{ScH}_{0.05}$ a clear departure from Arrhenius behaviour may be observed below 100 K where the apparent μ^+ hopping rate first increases significantly and then decreases with decreasing temperature. The activation energies derived from the data in the Arrhenius regime, 0.025 eV for Sc and 0.054 eV for $\text{ScH}_{0.05}$, are low compared to the value found for the long range diffusion of H in scandium but interestingly enough the value for $\text{ScH}_{0.05}$ is similar to binding energies (~ 0.06 eV) found for H pairs in Sc [8].

The observed hopping rates reach a value of $\sim 10^7 \text{ s}^{-1}$ at 300 K which is faster than the rate for long range diffusion of H but slower than the

localised H hopping observed by both neutron scattering and NMR. Thus it seems likely that the present correlation times reflect the long range motion of μ^+ which is faster than that of H.

Although further measurements are required to elucidate the significant increase in apparent hopping rate observed in $\text{ScH}_{0.05}$ at 100 K it is plausible that such a behaviour may be explained by trapping of the μ^+ by H. It is worthwhile remembering that muons are implanted in the sample in non-equilibrium conditions and have an average lifetime of 2.197 μs . Thus for a H trap to be effective the diffusion rate must be fast enough for the muon to reach the trap within this lifetime. Hence the low temperature rate in the $\text{ScH}_{0.05}$ sample may be representative of free μ^+ diffusion which increases with temperature to a point where the traps become effective and reduce the apparent hopping rate. At higher temperatures the μ^+ may escape from the trap so that the measured rate then reflects the escape rate.

The apparent hopping rates gradually lose reliability below 80 K because of their increasing sensitivity to the assumed low temperature σ entered into the Abragam formula. These plateau values, assumed to correspond to static muons, are not precisely defined and it is therefore not possible to draw valid conclusions without additional studies.

References

1. I. S. Anderson, N. F. Berk, J. J. Rush, T. J. Udovic, R. G. Barnes, A. Magerl and D. Richter, *Phys. Rev. Lett.* **65** (1990) 1439.
2. G. Cannelli, R. Cantelli, F. Cordero, F. Trequattrini, I. S. Anderson and J. J. Rush, *Phys. Rev. Lett.* **67** (1991) 2682.
3. R. G. Leisure, R. B. Schwarz, A. Migliori, D. R. Torgeson, I. Svare and I. S. Anderson, *Z. Phys. Chem.* **179** (1993) 359.
4. I. S. Anderson, D. K. Ross and J. E. Bonnet, *Z. Phys. Chem. Neue Folge* **164** (1989) 923.
5. R. G. Barnes, *J. Less-Common Met.* **172–174** (1991) 509.
6. O. Blaschko, *J. Less-Common Met.* **172–174** (1991) 237.
7. A. Abragam, *The Principles of Nuclear Magnetism*, Clarendon, Oxford, 1961.
8. N. F. Berk, J. J. Rush, T. J. Udovic and I. S. Anderson, *J. Less-Common Met.* **172–174** (1991) 496.