μ^+ LOCALIZATION AND TUNNELLING IN Sc

F.N. GYGAX, A. AMATO, A. SCHENCK

Institut für Mittelernergiephysik der ETH Zürich, CH-5232 Villigen PSI, Switzerland

I.S. ANDERSON

Institut Laue Langevin, B.P. 156, F-38042 Grenoble Cédex 9, France

J.J. RUSH

Materials Science and Engineering Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA

G. SOLT

Paul Scherrer Institut (PSI), CH-5232 Villigen PSI, Switzerland

The angular and magnetic field dependence of the transverse μ SR relaxation rate in single crystal Sc shows unambiguously that, at low temperatures, the muon is localized along the $\hat{c}$ -axis joining interstitial tetrahedral sites. The best agreement with the measured results, covering a complete range of high and low field data, is obtained on assuming a muon state with fast tunnelling between two adjacent sites at positions $z/c = \pm 0.09$ above and below the $a-b$ plane. Both contributions from the muon-induced electric field gradient and that inherent to the crystal were considered.

We have performed μ SR measurements on single crystal samples of ScH_x , $x = 0, 0.05$ and 0.25 , aimed at determining both the muon site in the hcp lattice and the dynamics at temperatures below 300 K [1]. In what follows we discuss the results obtained for the pure Sc samples.

1. Muon Site

The μ SR relaxation rates have been measured as a function of the applied magnetic field in a transverse field geometry at various temperatures between 10 and 300 K. Scans were performed for different values of the angle θ between the hexagonal $\hat{c}$ -axis of the crystal and the direction of the externally applied field. All measurements show basically a levelling of the μ SR relaxation [2] below 50 K. Typical results at low temperature are presented in Fig. 1. Accordingly, between 10 and 50 K, where the

μ SR signals have clearly gaussian form, neither thermally activated nor other long range motion of the muon is indicated and we concentrate on the μ^+ site determination for that temperature domain.

Fig. 1. Selection of transverse field relaxation rates σ measured in a pure Sc single crystal as function of temperature (lower range). Data shown for two crystal orientations in different fields: + for $\theta = 0^\circ$ and 296 mT, o for $\theta = 0^\circ$ and 36 mT, x for $\theta = 45^\circ$ and 36 mT. The lines are meant to guide the eye.

The μ SR relaxation rates measured as function of the applied field B for $\theta = 0^\circ$ and $\theta = 45^\circ$ (Fig. 2) reach saturation values for $B \geq 0.3$ T. This indicates that the field distribution at the μ^+ site, originating from the

Fig. 2. Relaxation rate σ measured in Sc single crystals as function of the transverse magnetic field. The temperature was kept below 50 K and for most of the data was fixed at 12 ± 1 K. Data sets taken for the three crystal orientations $\theta = 0^\circ$ (diamonds), $\theta = 45^\circ$ (open circles) and $\theta = 90^\circ$ (x). A simple calculation of the second moment of the lattice dipolar field at the μ^+ (at a shifted T-site, assuming a radial EFG and 5.4% lattice expansion in a and b around the muon) yields the dashed line for $\theta = 0^\circ$, the solid line for $\theta = 45^\circ$ and the dotted line for $\theta = 90^\circ$.

magnetic nuclear Sc moments, has reached the Van Vleck limit. From the

measured $\sigma(B)$ dependences it is not obvious where the “low-field” limit is to be found exactly, however it is certainly below 10 mT. One sees that the $\sigma(\theta)$ dependences measured at 503 mT (Fig. 3a) and 7.8 mT (Fig. 3b) show two drastically different patterns.

Fig. 3. Transverse relaxation rate σ measured versus the crystal orientation θ , (a) in a field of 503 mT at 10 and 30 K, and (b) in a field of 7.8 mT at 30 K. The solid lines are calculated by assuming, like in Fig. 2, a shifted T-site for the μ^+ and a lattice expansion (see text).

We have performed calculations with increasing levels of sophistication to understand the measured second moment of the field distribution, M_2 , corresponding to the square of $\sigma(\theta, B)$, by assuming various sites for the μ^+ in the crystal lattice. Starting at the Van Vleck limit, i.e. with the simplest calculation, it is clear that an octahedral (O) site calculation gives an inadequate description of the angular pattern obtained experimentally (Fig. 4a). It should be noticed that in the Sc lattice the crystallographically equivalent “tetrahedral” interstitial sites split into two magnetically non-equivalent sub-sites. Thus the calculation of the effective second moment necessitates the proper average of M_2 over the sub-sites. For the muon at the center of a basic Sc tetrahedron (T), i.e. with $z/c = 0.1185$ in the undisturbed Sc lattice (c is the $\hat{c}$ -axis lattice constant and z the distance between the μ^+ and the basal lattice plane), the calculated $\sigma(\theta)$ function (Fig. 4a) reflects already the main trends in the data.

As a next step, more features of the measured $\sigma(\theta)$ are reproduced when one assumes a muon shifted from the center of the T-site towards the $a - b$ basal plane of the tetrahedron, i.e. towards the next neighbour tetrahedral site. In fact taking $z/c = 0.05$ and a 5.4% expansion of a and b for the lattice around the μ^+ gives the more satisfactory solid lines in Fig. 3a. To fit the low field data of Fig. 3b (see the resulting solid line),

Fig. 4. (a): calculated relaxation rate $\sigma(\theta)$ in the Van Vleck limit. For the three cases the Sc lattice is undistorted. Dashed line for muons at O-sites, dotted line for μ^+ at the center of T-sites, solid line for μ^+ at shifted T-sites ($z/c = \pm 0.05$). (b): calculated relaxation rate $\sigma(\theta)$ at low magnetic fields. The muon is assumed at a shifted T-site ($z/c = \pm 0.05$), a and b of the lattice are expanded by 5.4% around the μ^+ . Dashed line for a purely axial EFG, parallel to the $\hat{c}$ -axis. With a purely radial EFG (i) dotted line at the low-field limit, (ii) solid line for $B = 7.8$ mT and $\nu_Q = 50$ kHz.

we assumed in addition a radial electric field gradient (EFG) around the μ^+ implying a quadrupolar frequency ν_Q of about 50 kHz for the closest Sc nuclei. (A purely axial EFG along the $\hat{c}$ -axis – Fig. 4b – is clearly not supported by the data at 7.8 mT.) At this point one might already be satisfied with this muon site determination reproducing quite well the data over a large range in applied field. However, it was necessary to assume a significant shift in the T-site position and an *ad hoc* lattice expansion. Furthermore, the calculations do not reproduce well the $\sigma(B)$ data at very low field (Fig. 2, solid line for $\theta = 45^\circ$ and dashed line for $\theta = 0^\circ$).

The agreement can be greatly improved by considering the fact that the large shift obtained for the assumed static muon position towards the $a - b$ plane can be taken as an indication for a state with the μ^+ tunnelling between adjacent T-sites *across* the plane. The calculations in this model show that the muon displacements from the two symmetrical T-“centers” is indeed smaller and that an agreement with the experiment can be obtained in this case even with an unrelaxed lattice. This can be qualitatively understood by simple considerations about the effect of μ -tunnelling on the second moment of the field distribution.

As a result the best possible assumption for the μ^+ dynamics is a rapid ($\nu \gg 1/\tau_\mu$, ν_0) tunnelling between adjacent T-sites, whose equilibrium positions are slightly shifted from the ideal T-centers towards each other,

with $z/c = \pm 0.09$ (here τ_μ is the μ^+ lifetime and ν_0 the reorientation frequency of the nuclei). Below the Van Vleck limit the effect of the predominantly radial EFG generated by the muon is prevalent. The strength of the quadrupolar coupling is found to be $\nu_Q \approx 67$ kHz for the neighbouring Sc nuclei. It contains a small contribution from an inherent crystalline axial EFG parallel to the $\hat{c}$ -axis ($\nu_Q^{\text{cr}} \approx 9$ kHz) which leads to the upturn of the calculated $\sigma(0^\circ, B)$ for B decreasing below 10 mT, in agreement with the data – see Fig. 5a. As can be seen from Fig. 5b,c the tunnelling model with the above parameters describes well the $\sigma(\theta)$ data measured at high and low field.

Fig. 5. Data compared to the calculated $\sigma(\theta, B)$ assuming μ^+ tunnelling between adjacent T-sites ($z/c = \pm 0.09$), $\nu_Q = 67$ kHz and $\nu_Q^{\text{cr}} = 9$ kHz. The Sc lattice is undistorted. Lines and data points in (a) identified as in Fig. 2. (b) for $B = 503$ mT and (c) for $B = 7.8$ mT.

2. Discussion

The field and orientation dependences of the muon relaxation rate at low temperature are well described by a model invoking a rapid tunnelling

between adjacent tetrahedral sites separated by $2z = 0.18c$. This separation is smaller than expected from the "ideal" position but is remarkably near to the value $2z = 0.194c$ found for D in Sc [3].

Furthermore, rapid local hopping ($\sim 10^{10} \text{ s}^{-1}$) of H between neighbouring T sites has been observed for H in Sc [4]. In this case the temperature dependence of the hopping rate is well described by a model in which the tunnelling rate is damped by coupling to conduction electrons. Considering the rather close separation of the adjacent T sites and the apparent shallow potential between them (as measured by inelastic neutron scattering [4]) the observation of tunnelling phenomena is not surprising, particularly for the light μ^+ .

Further measurements, particularly to study the interplay between muon and H in a scandium host, are under way.

Acknowledgement

We wish to thank R. Feyerherm (ETH Zurich) as well as V.G. Olshevsky and V.Yu. Pomjakushin (JINR Dubna) for their help in data taking.

References

- [1] F.N. Gygax, A. Amato, I.S. Anderson, J.J. Rush and A. Schenck, *Z. Phys. Chem.*, (1993), in press.
- [2] A careful examination indicates a very light drop of σ at the lowest temperature. This feature is not discussed here but will be treated in a future report.
- [3] C.K. Saw, B.J. Beaudry, and C. Stassis, *Phys. Rev. B* **27** (1983) 7013.
- [4] N.F. Berk, J.J. Rush, T.J. Udovic, and I.S. Anderson, *J. Less. Comm. Met.*, **172-174** (1991) 496.