Diffusion of Positive Muons in Vanadium

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At low temperatures the muon jump rate is nearly linear in temperature for vanadium. This result provides evidence of one-phonon–assisted tunneling in the presence of impurity-induced lattice strains. A local maximum is observed in the depolarization rate near 80 K, which can be explained in a model of diffusion to impurity traps at higher temperatures.

Interest in the diffusion of positive muons in metals stems in part from (1) the theoretically predicted qualitative similarity to the diffusion of hydrogen in metals,[1,2] (2) the fact that muon spin rotation measurements are characteristically carried out with isolated μ^+ particles, and (3) the fact that at low temperatures the diffusion rates may become sufficiently small that the μ^+ particle may diffuse without being trapped at impurities during its 2.2-μs lifetime after implantation. Certain deviations from classical behavior have been predicted for the diffusion of light interstitial impurities at low temperatures.[1-3] The bcc metals have been particularly interesting cases where rapid diffusion of hydrogen is observed.[3]

Both with the isotopes of hydrogen and with muons it is important to obtain a better understanding of the role other impurities play in the diffusion of the light interstitials. Small-polaron theories have been presented which predict that light interstitials diffuse by phonon-assisted tunneling processes.[1,2] Kagan and Klinger have suggested that in practical situations the random lattice strains caused by impurities and lattice imperfections would interfere with possible coherent tunneling at low temperatures, tending to localize the light interstitial.[2] At low temperatures the one-phonon-assisted tunneling model would therefore apply, which predicts a linear temperature dependence of the diffusive jump rate.[1,4] This is in good agreement with our data presented here on polycrystalline vanadium containing 0.4 at.% impurities.

Petzinger has recently proposed a model that takes into account both the diffusion of the muon and its trapping by impurities.[5] The trapping can take place either at the impurity sites themselves or in the lattice strain fields surrounding the impurities. This model finds that the muon spin depolarization rate, which is a measure of the localization of the muon, can have a nonmonotonic dependence on temperature and even exhibit a number of peaks. Maxima in the depolarization rate are produced by a competition between the diffusion-limited trapping rate, which increases with temperature, and the thermally activated release from traps.

The technique of Gurevich *et al.*[6] and Grebinnik *et al.*,[7] which takes advantage of the inhomogeneous local magnetic field produced by the host nuclei, has been applied here. In muon-spin rotation measurements the asymmetry in the decay of polarized muons ($\mu^+ \rightarrow e^+ + \bar{\nu}_\mu + \nu_e$) results in an oscillatory time dependence for the positron emission in the presence of an external transverse magnetic field. The damping of the oscillation amplitude $A(t)$ is approximated by the following model function[8]:

$$A(t) = \exp[-2\sigma^2\tau^2(e^{-t/\tau} - 1 + t/\tau)], \qquad (1)$$

where theoretically σ^2 is proportional to the second moment of the static nuclear field distribution and τ is a site correlation time that models the muon jump processes. This formulation of motional averaging is based on an exponentially decaying probability $\exp(-t/\tau)$ that a muon stays at a given site. This is probably correct for diffusion between traps, but for diffusion between interstitial sites the mean time of stay may be a factor of 2 smaller than the site correlation time.[9] The jump path for the muon is not provided by the muon-spin rotation measurements.

The calculation of σ from second moments is a straightforward application of the work of Van Vleck,[10] although Hartmann has recently shown that the quantization axes of the host nuclei are perturbed by the electric field gradients in the vicinity in the muon.[11] Anisotropy in the dipolar field therefore makes σ dependent upon the magnitude of the transverse external field.

In order to provide a basis for comparison with our data, several hypothetical cases were considered. For a muon localized at either substitutional, tetrahedral, or octahedral sites in a rigid vanadium lattice, in the Zeeman limit of large external field, σ is 0.178, 0.399, and 0.429 μs^{-1}, respectively. Rapid tunneling in a multiplet of interstitial sites was proposed for hydrogen in niobium by Birnbaum and Flynn[12] to explain a variety of anomalous data. We have accordingly considered an analogous model where the muon wavefunction is spread over four tetrahedral interstitial sites in $\{100\}$ crystallite planes, finding that $\sigma = 0.27$ μs^{-1}. The weak-external-field limit was calculated by rotating the quantization axes of the ^{51}V nuclei near the muon from the direction of the external field towards the muon's mean position. For the occupancy of single sites the calculated σ is increased by a factor of $\sqrt{\frac{5}{3}}$. For the four-tetrahedral-site configuration we obtain $\sigma = 0.0370$ μs^{-1}.

Polarized muons were obtained from decays of pions in flight at the Space Radiation Effects Laboratory synchrocyclotron. The target was a 407-g annealed polycrystalline vanadium sample, containing the following major impurities (in ppm): Al (1300), O (1300), Fe (600), Si (250), C (370), and N (6). Transverse external fields of 37, 58, and 1786 Oe were used in the temperature range 5.5 to 300 K. The data at each field were fitted using the model function of Eq. (1) with a single adjustable parameter for σ and a different parameter for τ^{-1} at each temperature point. The fit at 58 Oe gives $\sigma = 0.37 \pm 0.01$ μs^{-1}, and at 1786 Oe, $\sigma = 0.261 \pm 0.007$ μs^{-1}. The calculation for σ based on the four-site tunneling model is consistent with these results. We recognize that it is also possible to obtain comparable values from a model in which the muon occupies single tetrahedral sites and the ^{51}V neighbors are displaced outwards by about 12%. Theoretically, local lattice relaxation is not expected to be so large.[13]

We have plotted the temperature dependence of a depolarization rate Λ defined through $A(\Lambda^{-1}) = e^{-1}$ in Fig. 1. The depolarization rate of 0.209 μs^{-1} is observed at $T = 10$ K in the 1786-Oe field and is equal, within statistical error, to the 0.208-μs^{-1} depolarization rate measured in a 400 Oe field by Hartmann *et al.*[14] It appears that the Zeeman limit applies to our high-field data.[11] There is a local maximum in Λ in the vicinity of 80 K, where the character of the diffusion proc-

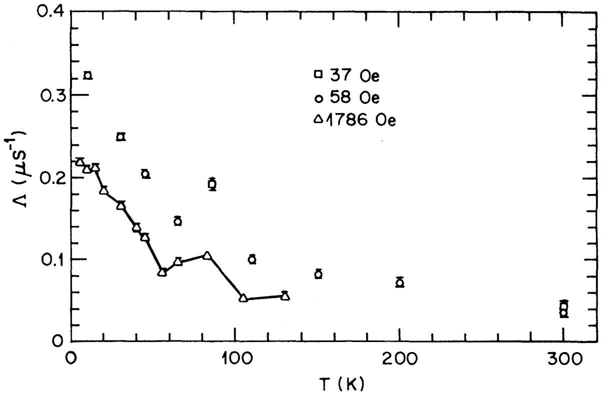

FIG. 1. Depolarization rate vs temperature for three values of external field.

τ^{-1} values are much smaller than the jump rates observed for hydrogen. This is explained by impurity trapping at high temperatures, where τ is close to being the mean time of stay at impurities.[5] We note that for a jump rate on the order of 3×10^7 s^{-1}, the muon may diffuse a distance equal to the mean distance between the impurities in 2.2 μs. We therefore associate the strucess apparently changes.

The temperature dependence for the correlation rate τ^{-1} is shown in Fig. 2. For $T \lesssim 50$ K it is linear, where $\tau^{-1} = (2.4 \pm 0.8)\times 10^4$ s^{-1} K^{-1} T and the normalized $\chi^2 = 2.1$. The theoretical expression for multiphonon processes, asymptotically approaching $\tau^{-1} \propto T^7$ at low temperatures,[1] does not fit these data, where we find $\chi^2 \sim 10^{11}$. At these temperatures the jump rates for hydrogen (extrapolated[15]) and τ^{-1} for the muon data are of the same order of magnitude. We believe it unlikely that the low-temperature behavior is an effect of trapping and detrapping from impurities, since we expect the binding enthalpy to impurities to be on the order of 0.1 eV, as it is for hydrogen.

In the region $T > 50$ K, on the other hand, the ture in Λ and τ^{-1} observed for $T > 50$ K with impurity trapping. In the absence of trapping, positive curvature in τ^{-1} vs T is expected at higher temperatures,[1] where multiphonon processes begin to contribute.

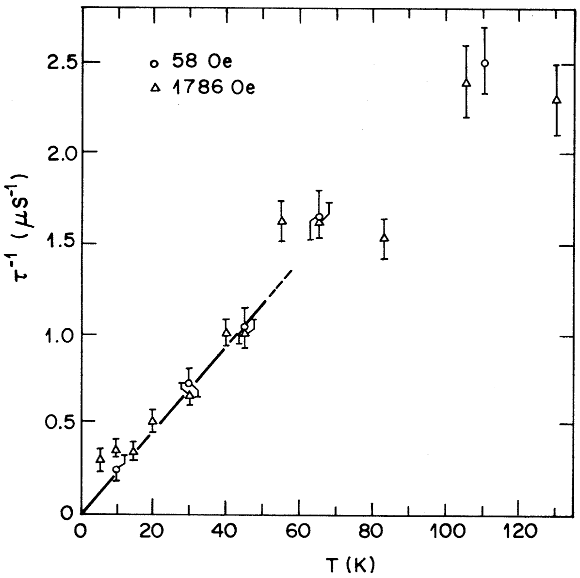

FIG. 2. The correlation rate parameter τ^{-1}, derived by least-squares fit with the diffusion expression, Eq. (1).

The effects of static strain fields around interstitial impurities have been considered theoretically by Kagan and Klinger.[2] A mean energy difference ΔE between neighboring interstitial sites may be estimated: $\Delta E = bc^{4/3}(\Delta V/V)_\mu(\Delta V/V)_{\text{imp}}$, where b is a deformation-potential coefficient,[2] $b = M\omega_D^2a^2/\pi^2 = 12$ eV, and c is the impurity concentration. M is the ^{51}V mass, a the lattice constant, and ω_D the Debye frequency. The fractional volume of solution, 0.4 for hydrogen, was used for $(\Delta V/V)_\mu$ and the value 0.9 for $(\Delta V/V)_{\text{imp}}$.[1] For our sample, we have $\Delta E \sim 0.001$ eV.

The one-phonon process[1,4] dominates at low temperatures if the condition $kT > \Delta E > J$ is satisfied, where J is a tunneling energy for transitions between neighboring states in the limit of $\Delta E = 0$. The jumps occur between sites where the muon localization energies differ by a mean amount ΔE. A characteristic of this theory is that the transition rate for $kT \gg \Delta E$ becomes independent of ΔE and linear in temperature. If we take into account the geometrical factors of the theory only approximately, the relaxation rate is given by[4]

$$\tau^{-1} \approx J^2b^2kT/3\pi\rho\hbar^4v_s^5, \qquad (2)$$

where ρ is the mass density and v_s the sound velocity. Fitting this expression to the low-temperature data gives $J/\hbar \sim 10^9$ s^{-1}. Thus indeed $kT > \Delta E > J$ is satisfied.

In conclusion, we have found that the motion of muons in vanadium at low temperatures can be represented by a phonon-assisted tunneling mechanism. These results provide the first evidence of one-phonon–assisted tunneling by light interstitial particles in metals. At higher temperatures we observe the predicted effect of the diffusion of the muons to impurity traps. Clearly, further work on the effect of impurities and studies of single-crystal specimens would be very interesting.

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