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Anomalous temperature dependence in the depolarization rate of positive muons in pure niobium

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The temperature dependence of the depolarization rate for positive muons implanted into a pure niobium crystal has been measured in an external field of 102 Oe. Depolarization is observed at temperatures below 100 K, with a pronounced minimum occurring in the vicinity of 20 K. A study of the magnetic-field dependence at low temperature indicates that the muons occupy tetrahedral interstitial sites. The data are interpreted in terms of the self-trapping of the muon at low temperature, and trap-limited diffusion at temperatures above 65 K. Comparisons are made with the properties of hydrogen in niobium.

I. INTRODUCTION

With μ^+ particles one has the opportunity of extending studies of hydrogenic impurities in metals by a factor of approximately 9 in isotopic mass.[1] Since the niobium-hydrogen system has been extensively studied by a variety of techniques, we have investigated the diffusion of muons in a pure niobium crystal. Niobium is an advantageous material for study, as the isotopically pure ^{93}Nb has a large nuclear moment and relatively pure crystals are readily obtainable.

The diffusion of hydrogen in niobium shows significant deviation from a simple classical Arrhenius law at low temperatures.[2-4] Moreover, it has been found that hydrogen can be trapped at oxygen and nitrogen interstitial impurities,[5,6] and at vanadium and molybdenum substitutional impurities.[7,8] It has not been possible to observe isolated hydrogen impurities in pure niobium at low temperature due to H-H pair formation and hydride precipitation. In the present investigation we are interested in the nonclassical behavior at low temperatures for individually implanted muons. The present technique is especially sensitive to slow relaxation, i.e., where the mean time of stay of the muon at a particualr site is in the range 0.1–10 μ sec. Because of the transitory nature of the probe, we expect that slow diffusion would be observable since the muon would not be able to diffuse to trapping sites in its mean lifetime of 2.2 μsec. Theory predicts a self-trapping local lattice distortion will localize light interstitials, except for extraordinarily low temperatures, with many similarities to the electronic small polaron.[9] Previous studies indicate that the tetrahedral sites in bcc metals are favored for both muons and hydrogen.[10-13]

The muon spin rotation (μSR) technique[11] used here had earlier been applied to a study of copper by Gurevich, Grebinnik, and co-workers, who had observed an activated muon diffusion process.[14, 15] Recently, the method has been used to study a variety of metals.[16-21]

It is not expected that the diffusion of the muon will be affected by the creation of lattice damage by the introduction of the muon. Preliminary results for the computer simulations of Brice indicate that little damage is created in the final stopping region.[22]

II. EXPERIMENTAL PROCEDURE

The niobium crystal we studied is a disk 2.5 cm in diameter by 1.0 cm thick, cut in the (110) plane, which had originally been a part of the sample studied by Lankford *et al.*[12] A mosaic spread of less than 0.2° was measured with neutron diffraction.[23] The present crystal was given an oxygen anneal at 3×10^{-6} Torr and 2200 °C, to reduce the carbon impurities, and then outgassed at 4×10^{-10} Torr and 2300 °C. The dislocation density is typically 10^4 cm^{-2} for samples given this annealing treatment. The crystal contains 200 ppm Ta and about 10 ppm total of other impurities. The Mo and V impurity levels are less than 0.1 and 0.03 ppm, respectively. The Ta impurity is not effective as a trapping site for hydrogen,[7] which is consistent with the small lattice perturbation produced by substitutional Ta and with the equal valences of Ta and Nb.

Spin-polarized muons of momentum 96 MeV/*c* obtained at the Swiss Institute for Nuclear Research cyclotron, were implanted into the crystal with a transverse magnetic field applied. Free precession of the muon spin introduces an oscillatory factor in the muon decay spectrum as a result of the asymmetric positron emission ($\mu^+ \rightarrow e^+ + \bar{\nu}_\mu + \nu_e$). The oscillations are damped by virtue of the static local-field inhomogeneity created by the ^{93}Nb nuclear dipole moments.[11] The quantity of interest here is the damping rate, effectively a muon spin depolarization rate, which is determined by the sites occupied by the muons, electric-field gradient perturbations on the ^{93}Nb nuclei,[24] and the diffusion of the muons. Getting a quantitative display of our data necessarily depends upon choosing a model function to express the time dependence of the polarization amplitude $A(t)$. Lacking an *a priori* knowledge of the dynamics, we use the following time dependence[14,15]

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

In this model the parameter σ_s^2 is proportional to the second moment of the host nuclear dipole field and τ is an effective correlation time associated with the muon motion. Possible effects such as the diffusion to impurity or lattice defect traps are not taken into account explicitly in this analysis.[16,21,25] The form of Eq. (1) has the advantage of spanning the limiting forms of a Gaussian, $\exp(-\sigma_s^2 t^2)$, expected in the absence of diffusion, and exponential, $\exp(-2\sigma_s^2 \tau t)$, expected for rapid diffusion. Data were taken as a function of temperature with a transverse 102-Oe field applied in the [100] crystallographic direction. The fastest depolarization was found at our lowest temperature $T = 10$ K. We have made the assumption that τ^{-1} is sufficiently small at 10 K that $A(t) \approx \exp(-\sigma_s^2 t^2)$ and have used a Gaussian fit to determine $\sigma_s = 0.321 \pm 0.008$ μsec^{-1}. The implications of this assumption are discussed below. These data were fitted using Eq. (1) by treating τ^{-1} as an adjustable parameter that varies with temperature.

The results for our sample, designated Nb-2, are shown in Fig. 1, where we have plotted a depolarization rate that satisfies the relation $A(\Lambda^{-1}) = e^{-1}$. The qualitative shape of the temperature dependence is not changed when either an exponential or a Gaussian model function is substituted for $A(t)$. The temperature dependence of the correlation rate τ^{-1} is given in Fig. 2. For comparison, the previous results of Lankford *et al.*[12] (denoted by Nb-1), analyzed in the same fashion, are also given in Figs. 1 and 2. The Nb-1 sample has an interstitial impurity concentration of 170 ppm.

We find it most remarkable that the overall temperature dependence is not monotonic. It appears that although the muon motion is sensitive to temperature, slow jump processes are observed over an extended range of temperature, from 10 to 100 K. Our statistical analysis of the fits determined that the diffusion formula provides better fits than either an exponential or a Gaussian, with probabilities that range from 0.5 to 0.9. There is a good indication of a plateau at low temperatures, below 15 K. The minimum in Λ that occurs near 20 K is more pronounced in our purer sample; the depolarization rate at 20 K is smaller and the minimum region extends to a higher temperature. The depolarization rates at 10 K are about the same for the two samples. The depolarization

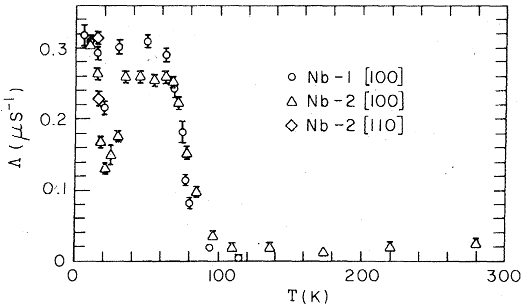

FIG. 1. Depolarization rate for muons implanted into niobium. Nb-2 refers to the present work and Nb-1 to the measurements of Lankford et al. (Ref 12.) The lowest temperature point for Nb-1 was corrected by a factor $\sqrt{2}$ as the measurement had been taken in zero field. The orientations of the external field of 102 Oe are given.

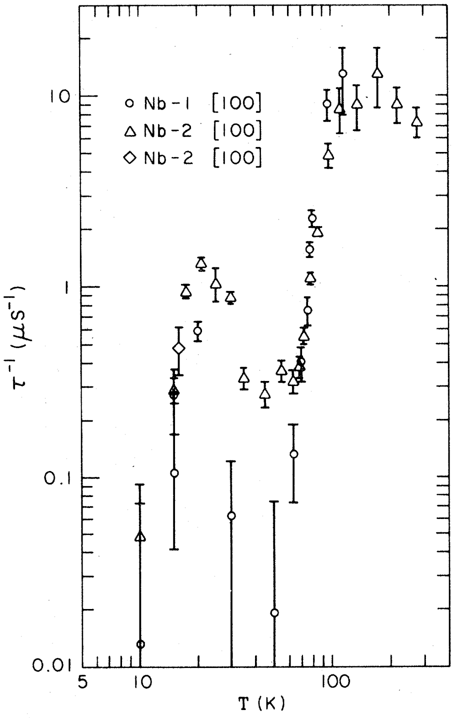

FIG. 2. Log-log plot of the temperature dependence of the correlation rates obtained in the analysis of the data according to Eq. (1) with $\sigma_s = 0.321\ \mu\text{sec}^{-1}$.

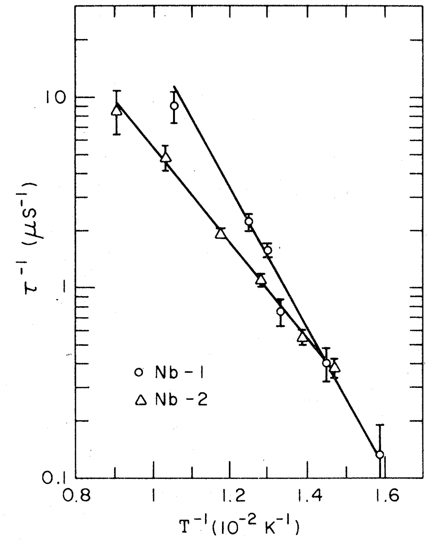

FIG. 3. Arrhenius plot of the temperature dependence of the correlation rate obtained from fits to data taken at 112 Oe. Nb-2, present work. Nb-1, Ref. 12. Least-squares fits are given by the lines.

rate exhibits a broad maximum region from 35 to 65 K, in which Λ is about 17% larger for the less pure Nb-1.

Before going on to discuss our interpretation of these results, it is important to mention the systematic effects of the uncertainty in the parameter σ_s. For a given set of data the parameter τ^{-1} is strongly correlated with σ_s, i.e., τ^{-1} increases with σ_s. This was investigated by testing another fitting procedure whereby both σ_s and τ^{-1} are treated as adjustable parameters. We find that the χ^2 has a broad shallow minimum in the $\sigma_s - \tau^{-1}$ plane. Making global fits over all the temperature points with a unique value for the parameter σ_s gives $0.353 \pm 0.012\ \mu\text{sec}^{-1}$ for Nb-1 and $0.394 \pm 0.012\ \mu\text{sec}^{-1}$ for Nb-2. The values of σ_s obtained in this manner are influenced by parasitic systematic errors which are difficult to estimate accurately. These include a small contribution to the μSR signal from muons stopping in the cryostat as well as accidental background events. That different σ_s values are obtained for the two sets of data is possibly indicative of this systematic uncertainty. The value $0.321\ \mu\text{sec}^{-1}$ obtained from the 10 K data and used in the analysis presented in Figs. 1–3 is judged to be a lower limit for σ_s. The systematic uncertainty in σ_s appears to be about 20%. A second independent consideration is that a more sophisticated formula than that given in Eq. (1) could possibly give a better fit to the data.[26]

The temperature region between 70 and 100 K is interesting in that a thermally activated process $\tau^{-1} = \nu_o \exp(-U/kT)$ provides a good representation of the temperature dependence of the τ^{-1} data. We show an Arrhenius plot of the data together with least-squares fits in Fig. 3. The results are $\nu_o = 10^{10.9 \pm 0.3}\ \text{sec}^{-1}$ and $U = 73 \pm 4$ meV for Nb-1 and $\nu_o = 10^{9.2 \pm 0.2}\ \text{sec}^{-1}$ and $U = 50 \pm 2$ meV for Nb-2. The indicated error bars are the statistical errors. The $\sigma_s - \tau^{-1}$ parameter correlation are such that both ν_o and U decrease with increasing σ_s. In the Nb-2 data a 20% increase in σ_s produces a 40% decrease in ν_o and a 12% decrease in U. We find larger effects for the Nb-1 data—a factor of 6 decrease in ν_o and a 21% decrease in U.

Of course τ^{-1} also increases with temperature in the region $T < 20$ K. Since we do not have sufficient data points in this region, we are unable to establish the form of the temperature dependence. Parameters characterizing the diffusion in this temperature region are discussed in Sec. III.

Information on the symmetry of the sites occupied by the muons can be inferred from data taken in strong external magnetic fields, which quench the electric-field gradient perturbation of the host nuclei.[24] Crystalline anisotropy in σ_s is expected to be most pronounced at strong fields, as has been seen experimentally by Camani *et al.*[27] in a study of muons in copper, and theoretically by

Hartmann.[24] A preliminary survey of the field dependence of the depolarization rate was carried out in the low-temperature region, $T = 15$ K, where τ^{-1} is small, for applied fields up to 5 kG and for the crystal orientations [100] and [110] parallel to the external field direction. Gaussian depolarization, with the model function $A(t) = \exp(-\sigma^2 t^2)$, gives a better fit to these data than does an exponential, as expected when the muon jump rate is small. Fitted values of σ are plotted in Fig. 4. Over this field region the depolarization rate decreases by about 15% for the external field applied along the [100] orientation and by about 37% for the [110] orientation. These changes are in the same direction as predicted by theory for the tetrahedral site assignment: 14% for the [100] and 22% for the [110] orientations, respectively. For the octahedral site, the theory predicts a 12% increase and a 33% decrease, respectively, for the [100] and [110] orientations.

Although this field dependence is qualitatively consistent with a tetrahedral site assignment, the depolarization rates we observe at high field are substantially smaller than the calculated values for muons occupying tetrahedral sites in an undistorted lattice. The calculated values of σ_s are 0.407 μsec^{-1} for a strong field in the [100] direction, and 0.350 μsec^{-1} for the [110] direction. The values shown in Fig. 4 for the Gaussian fit are 0.25 μsec^{-1} for [100] and 0.20 μsec^{-1} for [110]. Our temperature dependence results of Fig. 2 show that diffusion is not negligible at 15 K, where $\tau^{-1} = 0.3$ μsec^{-1}. Using the empirical formula of Eq. (1) with a fixed τ^{-1} and adjustable σ_s, we obtain corrected experimental values at 5 kG of 0.30 μsec^{-1} for the [100] and 0.24 μsec^{-1} for the [110]. The statistical precision obtained for the above values of σ_s is ± 0.01 μsec^{-1}. Therefore, diffusion does not account for all of the decrease and we find that a model assigning the muon to a single tetrahedral site would have to postulate a local outward displacement of the Nb neighbors by 11%–13%, in order to agree with the corrected measurements at 5 kG. By way of comparison, it was found that the lattice is dilated by 5.8% for hydrogen in niobium.[28]

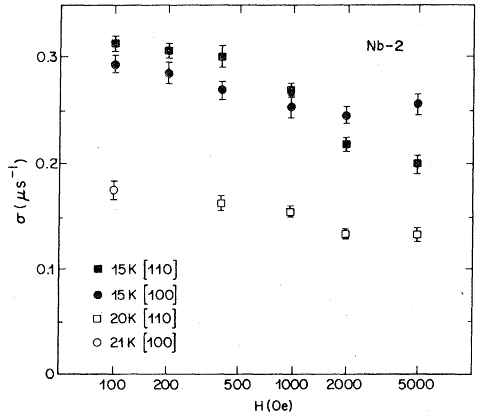

FIG. 4. Magnetic-field dependence of the depolarization rate, obtained from fits using a Gaussian function of time.

Previous measurements of depolarization rates in Nb-1 at $T = 10$ K, also smaller than the prediction for single sites, were taken as evidence that the muon wave function is spread over a manifold of sites.[12] A tunneling-state model has been proposed as an explanation for a variety of anomalous data on hydrogen in niobium.[29] There is evidence that protons occupy the tetrahedral sites in niobium,[1] but it is not known whether tunneling states exist for isolated, as opposed to trapped, protons. Recent studies of oxygen- and nitrogen-doped samples have provided evidence for the tunneling of hydrogen which is trapped at sites adjacent to the impurities.[30,31] Thus, it is interesting to consider the T_4 model of Lankford *et al.*,[12] in which it is assumed that the muon rapidly tunnels among four tetrahedral sites forming a square ring arrangement in a {100} plane. We find that by taking a more modest 3% dilation of the lattice, this calculation gives depolarization rates of 0.30 μsec^{-1} for a strong field in the [100] direction, and 0.23 μsec^{-1} for [110], values that are in good agreement with the observed rates.

The fact that the muon occupies tetrahedral sites suggests a basic similarity to the properties of protons. Further measurements probing in detail the crystalline anisotropy at high field will need to be carried out at lower temperatures before the symmetry of the muon wave function and the local lattice distortion can be firmly established.

The small depolarization rates at 20 K shown in Fig. 4 extend to high field, so that the minimum anomaly found in Λ is apparently not an artifact of the magnitude of the applied field. We conclude that the muon becomes more highly mobile as temperatures increase towards 20 K.

III. DISCUSSION

It is clear that understanding the origin of the minimum in Λ near 20 K is central to interpreting this experiment. Recently, a similar minimum phenomenon was reported by Grebinnik *et al.* for a polycrystalline sample of pure Bi.[18] It was suggested that coherent diffusion of the muon in band-like states was observed at low temperature, where theory predicts that the diffusion rate of light interstitial particles decreases with increasing temperature, owing to phonon scattering.[9,32] A low-temperature plateau in the depolarization rate was identified with defect trapped states.

The interpretation we are proposing for the present experiment on niobium is different, and assumes that the muon diffusion rate increases monotonically with temperature in the range covered by the experiment. The low-temperature plateau in the depolarization rate near 10 K is attributed to the self-trapping of the muon on a set of tetrahedral interstitial sites, such as assumed in the T_4 model.[12] The minimum in Λ near 20 K is explained in terms of diffusion and impurity trapping. On the low-temperature side of the mimimum, the muon motion between rings occurs and the increase in diffusion rate with temperature causes Λ to decrease. In this temperature region the jump frequency is low enough so the muon remains untrapped during its lifetime. On the high-temperature side the time for diffusion to traps becomes small and the trapped muon lifetime large on the time scale of the experiment, and the depolarization rate increases with temperature.

The depolarization observed at temperatures between 35 and 65 K is attributed to muons trapped at impurities. The present work does not identify the impurities which are the effective traps. Thermal detrapping of muons is not observed until the temperature is raised to about 65 K. Above this temperature the muon jump rate increases and the depolarization rate decreases. The details of the muon's environment are probably not much different for either self-trapped or impurity trapped states, as the depolarization rate measured in the two regions $T < 15$ K and $35 < T < 65$ K differ by 20% or less. The most likely trapping sites are expected to have little effect on the depolarization rates as both natural ^{16}O and ^{12}C have zero nuclear moments, and the moment of ^{14}N is small. The contribution to the dipolar field near these impurities is therefore small.

We anticipate that further studies of the broad maximum region at high field, for various crystal orientations, will shed light on the nature of the trapped states and on possible ringlike tunneling states for the trapped and diffusing muons.

For niobium we believe there is good reason for expecting that the muon diffusion rate is large enough in the temperature region between 35 and 65 K to allow the μ^+ particles to diffuse to residual impurities. We refer to previous studies of the magnetic bcc metals, Fe and Cr, where evidence was found that muons move rapidly at low temperature.[10,33] In Fe and Cr the local dipolar field is electronic in origin, owing to the ferromagnetic, or antiferromagnetic state, and is three orders of magnitude larger than the nuclear dipolar fields in niobium. Thus, the measurements of the depolarization rate are sensitive to correspondingly more rapid jump rates. A damped μSR signal was observed at 23 K in Fe by Nishida *et al.*[10] and at 77 K in Cr by Kossler *et al.*[33] It has been argued that the muon jumps between tetrahedral interstitial sites in Fe.[11] The correlation rate indicated by the data on pure Fe is on the order of 3×10^9 sec^{-1} at 23 K. Also, the absence of observable μSR signals at liquid-helium temperatures was taken as evidence that slow jump processes occur at the lowest temperatures measured. Measurements in less pure Fe samples have found a temperature interval where the μSR signal disappears, and has been explained in terms of fast depolarization of muons trapped at impurities.[34]

Because of the similarities among Nb, Fe, and Cr as bcc metals, we expect that the jump rate for muons in these metals to be of the same order of magnitude. In order for the muons to diffuse to the impurities in 2.2 μsec, the muon lifetime, the jump rate would have to be on the order of 5×10^9 sec^{-1}, for trap concentrations of 10 ppm. This jump rate, which is reasonable to expect, is about equal to the jump rate for hydrogen, measured at higher temperatures and extrapolated to 100 K.

The decrease in Λ for temperatures above 65 K is attributed to trap-limited diffusion of the muon. The small depolarization rates observed at temperatures above 100 K are associated with comparatively short times of stay at impurities. The temperature dependence of this process should be described by the quantum tunneling process of Flynn and Stoneham which has a high-temperature asymptotic behavior of the form[9]

$$\tau^{-1} = (\pi/4\hbar^2 E_a kT)^{\frac{1}{2}}\,|J|^2 \exp(-E_a/kT), \qquad (2)$$

where $|J|$ is the tunneling energy in the unrelaxed lattice and E_a the lattice activation energy. Deviations from Eq. (2) are expected to occur for $T \lesssim \Theta_D$, where Θ_D is the Debye temperature. Such deviations from the Arrhenius temperature dependence have been observed for hydrogen relaxations in Nb and Ta.[30,35] While the data presented in Fig. 3 appear to fit Eq. (2), they extend over a limited temperature range, so that it is not possible to detect curvature. It is therefore not possible to establish whether the data are obtained in a temperature range suitable for the high-temperature asymptotic limit. Values of ν_0 are found in the Arrhenius fits which are up to four orders of magnitude smaller than for hydrogen. This is in the direction predicted by quantum theory but may also result from the use of the Arrhenius relation in a region of positive curvature. To check this possibility, we have used $\Theta_D = 275$ K obtained from specific-heat measurements[36] in evaluating the integral expressions for the jump rate calculated for the Debye model by Stoneham.[35] The τ^{-1} rate near 85 K can then be fitted with $E_a = 66$ meV and $|J| = 0.22 \pm 0.05$ meV. The $|J|$ value is much lower than the theoretically expected value, which is about 0.1 eV.

We assume trap-limited diffusion takes place, where E_a is the enthalpy for escape from impurities and is given by the sum of the impurity binding and migration enthalpies. The activation enthalpy is enhanced at a trapping site by the interaction of the lattice dilation around the muon with that around the impurity. Thus, trapping causes E_a to be larger than the migration enthalpy of diffusion between interstitial sites. Experiments on the trapping of hydrogen by oxygen and nitrogen impurities find binding enthalpies of about 0.1 eV,[6] larger than the migration enthalpy: 0.061 eV, measured at low temperature.[2] Richter *et al.* recently reported an activation enthalpy of 166 meV for the detrapping of hydrogen from nitrogen impurities in niobium, for neutron diffraction measurements taken at temperatures above 250 K.[37] Substantially larger binding and migration enthalpies are observed for deuterium, 0.13 and 0.12 eV, respectively.[3,6] Thus, the muon results fit the trends of finding migration enthalpies that increase with the mass of the interstitial.

The increase in depolarization rate at $T \lesssim 20$ K has been ascribed to motion of untrapped muons. The temperature dependence of τ^{-1} should be described by the low-temperature asymptotic form of the Flynn-Stoneham theory

$$\tau^{-1} = 4.5 \times 10^3 \pi |J|^2 E_a^2 \times [(2T/\Theta_D)^7/\hbar(k\Theta_D)^3] \exp(-5E_a/k\Theta_D). \quad (3)$$

The data are not sufficient to test the T^7 behavior, and the parameters E_a and $|J|$ obtained above are not expected to apply to the motion of untrapped μ^+ at $T \lesssim 20$ K. However, we can get an approximate value of $E_a = 16 \pm 1$ meV, as E_a is determined by a slowly varying logarithmic function of the magnitude of $|J|$ and the τ^{-1} rate at $T \sim 20$ K.

Apart from the weaker depolarization near 20 K shown by the Nb-2 data, which we have associated with a smaller trapping rate at impurities, the depolarization near 50 K is also weaker. Since the depolarization rate remains nearly constant over a factor of 2 in temperature, it appears that in this temperature range trapping takes place quickly on the time scale of the experiment. Further work on crystals with controlled amounts of impurities would be required in order to establish any significance to the differences observed between the Nb-1 and Nb-2 data at temperatures near 50 K and higher.

Note added in proof. A μSR investigation of nitrogen-doped niobium with a description in terms of a two-state trapping model has been given by M. Borghini, O. Hartmann, E. Karlsson, K. W. Kehr, T. O. Niinikoski, L. O. Norlin, K. Pernestål, D. Richter, J. C. Soulié, and E. Walker, CERN (to be published). The results of the present experiment on niobium have been fitted with a theory for muon diffusion in the presence of traps by K. G. Petzinger and R. L. Munjal, Bull. Am. Phys. Soc. 23, 360 (1978), and independently by E. Zaremba and T. McMullen, *ibid.* 23, 361 (1978).

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[1] A recent review has been given by J. Völkl and G. Alefeld, in *Diffusion in Solids: Recent Developments*, edited by A. Nowick and J. Burton (Academic, New York, 1975).

[2] G. Schaumann, J. Völkl, and G. Alefeld, Phys. Status Solidi 42, 401 (1970).

[3] H. Wipf and G. Alefeld, Phys. Status Solidi A 23, 175 (1974).

[4] D. Richter, B. Alefeld, A. Heidemann, and N. Wakabayash, J. Phys. F 7, 569 (1977).

[5] C. Baker and H. K. Birnbaum, Acta Metall. 21, 865 (1973).

[6] R. F. Mattas and H. K. Birnbaum, Acta Metall. 23, 973 (1975).

[7] T. Matsumoto, Y. Sasaki, and M. Hihara, J. Phys. Chem. Solids 36, 215 (1975).

[8] T. Matsumoto, J. Phys. Soc. Jpn. 42, 1583 (1977).

[9] C. P. Flynn and A. M. Stoneham, Phys. Rev. B 1, 3966 (1970).

[10] N. Nishida, R. S. Hayano, K. Nagamine, T. Yamazaki, J. H. Brewer, D. M. Garner, D. G. Fleming, T. Takeuchi, and Y. Ishikawa, Solid State Commun. 22, 235 (1977).

[11] A. Schenck, *Nuclear and Particle Physics at Intermediate Energies*, edited by J. B. Warren (Plenum, New York), pp. 159–297.

[12] W. F. Lankford, H. K. Birnbaum, A. T. Fiory, R. P. Minnich, K. G. Lynn, C. E. Stronach, L. H. Bieman,

W. J. Kossler, and J. Lindemuth, Hyperfine Interactions 4, 833 (1978).

[13] H. D. Carstanjen and R. Sizmann, Phys. Lett. A 40, 93 (1972).

[14] I. I. Gurevich, A. E. Mel´eshko, I. A. Muratova, B. A. Nikol´sky, V. S. Roganov, V. I. Selivanov, and V. C. Sokolov, Phys. Lett. A 40, 143 (1972).

[15] V. G. Grebinnik, I. I. Gurevich, V. A. Zhukov, A. P. Manych, E. A. Mel´eshko, I. A. Muratova, B. A. Nikol´sky, V. I. Selivanov, and V. A. Suetin, Zh. Eksp. Teor. Fiz. 68, 1548 (1975) [Sov. Phys.-JETP 41, 777 (1976)].

[16] W. B. Gauster, A. T. Fiory, K. G. Lynn, W. J. Kossler, D. M. Parkin, C. E. Stronach, and W. F. Lankford, J. Nucl. Mater. 69/70, 147 (1977).

[17] O. Hartmann, E. Karlsson, K. Pernestål, M. Borghini, T. O. Niinikoski, and L. O. Norlin, Phys. Lett. A 61, 141 (1977).

[18] V. G Grebinnik, I. I. Gurevich, V. A. Zhukov, A. I. Klimov, V. N. Mayorov, A. P. Manych, Y. V. Mel´nikov, B. A. Nikol´sky, A. V. Piragov, A. N. Ponomarev, V. I. Selivanov, and V. A. Suetin, Pis´ma Zh. Eksp. Teor. Fiz. 25, 322 (1977) [JETP Lett. 25, 298 (1977)].

[19] R. H. Heffner, W. B. Gauster, D. M. Parkin, C. Y. Huang, R. L. Hutson, M. Leon, M. E. Schillaci, M. L. Simmons, W. Trifthäuser, Hyperfine Interactions 4, 838 (1978).

[20] O. Hartmann, E. Karlsson, L. O. Norlin, K. Pernestål, M. Borghini, and T. Niinikoski, Hyperfine Interactions 4, 824 (1978).

[21] W. B. Gauster, R. H. Heffner, C. Y. Huang, R. L. Hutson, M. Leon, D. M. Parkin, M. E. Schillaci, W. Trifthäuser, and W. R. Wampler, Solid State Commun. 24, 619 (1977).

[22] D. Brice (private communication).

[23] The neutron scan was kindly carried out by S. Shapiro.

[24] O. Hartmann, Phys. Rev. Lett. 38, 832 (1977).

[25] A. Seeger, Phys. Lett. A 53, 324 (1975).

[26] V. G. Baryshevsky and S. A. Kuten´, Fiz. Tverd. Tela 18, 2873 (1976) [Sov. Phys. Solid State 18, 1677 (1976)].

[27] M. Camani, F. N. Gygax, W. Rüegg, A. Schenck, and H. Schilling, Phys. Rev. Lett. 38, 836 (1977).

[28] H. Pfeiffer and H. Peisl, Phys. Lett. A 60, 363 (1977).

[29] H. K. Birnbaum and C. P. Flynn, Phys. Rev. Lett. 37, 25 (1976).

[30] C. G. Chen and H. K. Birnbaum, Phys. Status Solidi A 36, 687 (1976).

[31] G. Pfeiffer and H. Wipf, J. Phys. F 6, 167 (1976).

[32] Yu. Kagan and M. I. Klinger, J. Phys. C 7, 2791 (1974).

[33] W. J. Kossler, A. T. Fiory, D. E. Murnick, C. E. Stronach, and W. F. Lankford, Hyperfine Interactions 3, 287 (1977).

[34] H. Graf, W. Kündig, B. D. Patterson, W. Reichart, P. Roggwiller, M. Camani, F. N. Gygax, W. Rüegg, A. Schenck, and H. Schilling, Helv. Phys. Acta 49, 730 (1976).

[35] A. Stoneham, J. Phys. F 2, 417 (1972).

[36] H. A. Leupold and H. A. Boorse, Phys. Rev. 134, A1322 (1964).

[37] D. Richter, T. Springer, and K. W. Kehr, *Proceedings of the Second International Congress on Hydrogen in Metals*, (Plenum, Paris, 1977), paper 2B5.