

LA-UR-82-634

Conf. 820325-1

LA-UR--82-634

DE92 011982

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7409-ENG-24

TITLE MUON DIFFUSION IN NOBLE METALS

AUTHORS: M. E. Schillaci, C. Boekema, R. H. Heffner, R. L. Hutson, M. Leon, and C. E. Olson, Los Alamos National Laboratory; S. A. Dodds, Rice University; D. E. MacLaughlin, University of California, San Diego, CA; and P. M. Richards, Sandia National Labo.

SUBMITTED TO INTERNATIONAL SYMPOSIUM ON THE ELECTRONIC STRUCTURE AND PROPERTIES OF HYDROGEN, MARCH 4-6, 1982 RICHMOND, VA

MASTER

By acceptance of this article the publisher warrants that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, and to allow others to do so, for U.S. Government purposes. The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

Los Alamos Los Alamos National Laboratory Los Alamos, New Mexico 87545

## MUON DIFFUSION IN NOBLE METALS\*

M.E. Schillaci, C. Boekema, R.H. Jeffner, R.L. Hutson,  
M. Leon, C.E. Olsen  
Los Alamos National Laboratory, Los Alamos, NM 87545

S. A. Dodds  
Rice University, Houston, TX 77001

D. F. MacLaughlin  
University of California, Riverside, CA 92521

P. M. Richards  
Sandia National Laboratories, Albuquerque, NM 87185

## ABSTRACT

We have measured diffusion-induced muon depolarization in dilute AgCd and AgFe in the temperature range 200-700 K and have thereby determined the muon diffusion parameters in Ag. The diffusion parameters for  $\mu^+$  in Cu, Ag, and Au are compared with those of hydrogen. For Ag and Au, the  $\mu^+$  parameters are similar to those of hydrogen, whereas for Cu, the  $\mu^+$  parameters are much smaller. Lattice-activated tunneling and over-barrier hopping are investigated with computational models.

## INTRODUCTION

The similarity of the positive muon ( $\mu^+$ ) to a light proton (p) in a material makes the study of muon diffusion and trapping an ideal complement to the investigation of hydrogen in metals. Because the muon is so much lighter than the proton ( $m_\mu \approx m_p/9$ ) and because the muon diffusion can be studied at lower temperatures than hydrogen diffusion, the possibility of studying different diffusion mechanisms is enhanced.

Muon diffusion parameters can be derived from the temperature dependence of the muon depolarization rate,  $\Lambda_2$ , in a transverse magnetic field. The muon spin rotation (μSR) technique is described in several review articles.<sup>1</sup> In nonmagnetic metals, depolarization of the diffusing muon is caused by the inhomogeneous magnetic fields of the host nuclear dipole moments. As the muon hops more rapidly with increasing temperature, motional narrowing of the line width results--i.e.,  $\Lambda_2$  is reduced. Obviously, this method does not apply to metals having negligible nuclear moments, such as Ag and Au.

Another method for studying muon diffusion, reported earlier,<sup>2</sup> overcomes this limitation by doping the metal with small amounts of paramagnetic impurities and measuring  $\Lambda_2$  resulting from a muon's interaction with the electronic moments of the impurity ions. This mechanism is effective only if a muon moves to the vicinity of an impurity within its lifetime, and thus provides a measure of the  $\mu^+$  hopping time  $\tau_h$ . These measurements are also sensitive to the impurity ion spin dynamics and to the muon-ion interaction; the diffusion parameters thus determined, however, are not sensitive to the nature of the interaction, provided that it is reasonably short range.<sup>3</sup>

We report on measurements of muon diffusion in Ag, a continuation of our previous studies in Au.<sup>2</sup> Diffusion parameters for muons and hydrogen in Cu, Ag, and Au are then compared. A discussion of possible diffusion mechanisms and how they relate to the observed parameters follows.

## RESULTS

Measurements were made on polycrystalline samples of Ag doped with either the S-state ion  $Gd^{3+}$  or the crystal-field-split ion  $Er^{3+}$  and were carried out at the Clinton P. Anderson Meson Physics Facility (LAMPF) at Los Alamos. The reader is referred to our earlier work<sup>2</sup> for the details of the experiment. Figure 1 shows the data for  $\Lambda_2$  as a function of temperature, T, for AgGd and AgEr, taken in a transverse applied magnetic field of 80 Oe. In each case, the higher concentration data are scaled by the concentration ratio, showing that the concentration dependence of  $\Lambda_2$  is linear. The observed peaks are clearly identified with muon-impurity ion interactions by comparison with data taken in the pure host, where  $\Lambda_2(T)$  is negligible. The data begin to rise out of the background ( $T \sim 200$  K) at the temperature at which the muons are hopping fast enough to reach a magnetic impurity within a muon lifetime. Since the rising portions of the data are almost identical, muon diffusion in the host is not affected by the nature of the impurity.

The curves shown in Fig. 1 are model fits to the data. A description of the model used has been given elsewhere<sup>2,3</sup>; we give only a brief outline here. The muon is assumed to hop between adjacent octahedral interstitial sites which are arranged in shells surrounding an impurity out to a radius corresponding to the mean volume per impurity. The muon-ion interaction is assumed to have two components--dipolar and contact, which apply only to sites adjoining the impurity. The spin-lattice relaxation rate is assumed to have the Korringa form. The muon hopping rate is assumed to have an Arrhenius form,  $\tau_h^{-1} = \nu_0 \exp(-\epsilon/T)$ . Thus there are four adjustable parameters; (1) muon-ion contact interaction strength; (2) impurity ion spin-lattice relaxation coefficient; (3) the activation energy  $\epsilon$ ; and (4) the pre-exponential factor  $\nu_0$ .

While the muon-ion interaction and the impurity spin dynamics are certainly interesting topics, we shall discuss only the diffusion parameters here. Any ambiguity in the determination of  $\nu_0$  and  $\epsilon$  is substantially reduced by the fact that in the rising portion of the data,  $\epsilon$  and  $\nu_0$  are not very sensitive to the values of the other two parameters. The muon diffusion parameters extracted from the model fits to the Ag data are listed in Table I along with those for Au and Cu. Also included for comparison in Table I are the hydrogen diffusion parameters for all three metals. We also list the Debye temperature,  $\Theta_D$ , and the temperature range studied,  $\Delta T$ , for each case.

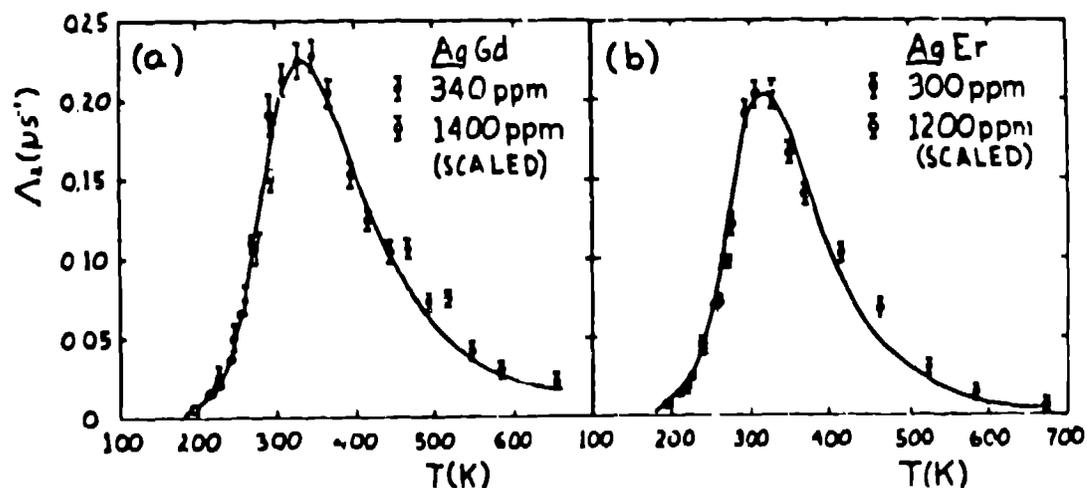


Fig. 1. Temperature dependence of muon depolarization rate in (a) AgGd and (b) AgEr at 80 Oe transverse applied field. The higher-concentration data has been scaled by the concentration ratios. Curves are model fits as discussed in text.

Table I. Diffusion parameters for muons and proton in noble metals

$\mu^+$ in:	$\nu_0$ ( $s^{-1}$ )	$\tau$ (K)	$\Delta T$ (K)	$Q_D$ (K)
Cu <sup>a</sup>	$10^{7.46 \pm 0.04}$	$551 \pm 15$	80-250	343
Ag <sup>b</sup>	$10^{13.5 \pm 0.5}$	$3200 \pm 200$	200-700	225
Au <sup>c</sup>	$10^{13.5 \pm 0.5}$	$1350 \pm 100$	85-230	165
$p$ in:				
Cu <sup>d</sup>	$10^{14}$	$4640 \pm 30$	720-1200	343
Ag <sup>e</sup>	$10^{13.8}$	3620	950-1120	225
Au <sup>f</sup>	$10^{12.6}$	2840	770-1260	165

<sup>a</sup>Ref. 4; <sup>b</sup>this work; <sup>c</sup>Ref. 2; <sup>d</sup>Ref. 5; <sup>e</sup>Ref. 6; <sup>f</sup>Ref. 7

## DISCUSSION

The obvious observation to be made from Table I is that the diffusion parameters for muons in Ag and Au are similar to the hydrogen parameters, while both  $\nu_0$  and  $\tau$  for  $\mu^+$  in Cu are very much smaller. This suggests significant incoherent tunneling for  $\mu^+$  in Cu,<sup>4</sup> but all of the other parameters are consistent with over-barrier hopping. Indeed, Teichler<sup>8</sup> has shown that the tunneling model gives good agreement with the  $\mu^+$  data for Cu. We examine the remaining cases in the context of the over-barrier hopping model.

We have calculated the potential of a point unit charge as a function of position within the unit cell of each metal following the calculation of Teichler,<sup>9</sup> to which the reader is referred for details. Electron screening densities around the point charge were obtained from the self-consistently calculated curves of Jena and Singwi.<sup>10</sup> The host ion-conduction electron pseudopotentials of Gubanov and Nikulin<sup>11</sup> were used, but their energy parameter,  $\epsilon$ , was varied as described below. Two general features of all of the potentials studied are: (1) only the octahedral (O) well is deep enough for the  $\mu^+$  ground state--the tetrahedral (T) well is too shallow; and (2) the O-O barrier is higher than the O-T barrier. Experimental evidence indicates that  $\mu^+$  is localized at the O site in Cu<sup>12</sup> and Au,<sup>13</sup> so we assume it is in Ag also.

The pseudopotential energy parameter,  $\alpha$ , was varied in order to adjust the difference,  $\Delta_\mu$  (or  $\Delta_p$ ), between the O-T barrier height and the  $\mu^+$  ( $p$ ) ground state. The over-barrier activation energy is then given by  $\epsilon_\mu = \Delta_\mu + B_\mu$ , where  $B_\mu$  ( $B_p$ ) is the  $\mu^+$  ( $p$ ) self-trapping energy. In the harmonic approximation, the ratio would be given by  $B_\mu/B_p = \sqrt{m_p/m_\mu} \approx 3$ ; however, we shall use the ratio calculated by Teichler<sup>9</sup>-viz.,  $B_\mu/B_p = 90/40 = 2.25$ . The two unknown quantities,  $\alpha$  and  $B_p$ , are determined from the two measured activation energies,  $\epsilon_p$  and  $\epsilon_\mu$ . In Table II, we list the values of  $\alpha$ ,  $B_p$ , and  $B_\mu$  together with the value,  $\alpha_0$ , which yields the correct host atom nearest-neighbor separation in lowest order perturbation theory within the pseudopotential scheme.<sup>14</sup> We note that the  $\alpha$ 's are within  $\sim 5^\circ$  of the  $\alpha_0$ 's. The self-trapping energies required to fit the data in Au and Ag are 2 to 4 times greater than those calculated<sup>9</sup> for Cu. To actually calculate  $B_\mu$  ( $B_p$ ) is difficult because the result depends very sensitively on both the host-host potential and the host- $\mu^+$  ( $p$ ) potential. Better potentials are clearly needed for this task.

If tunneling were the dominant mechanism rather than over-barrier hopping, a much smaller pre-exponential factor,  $\nu_{01}$ , would result unless the transfer integral,  $J$ , is large.<sup>15</sup> For  $\mu^+$  ( $p$ ) tunneling, we calculate  $J$  values for Ag and Au that are similar to that for Cu--on the order of 10 eV. Since only  $\mu^+$  in Cu exhibits a small  $\nu_{01}$ , we conclude that incoherent tunneling is not important for  $\mu^+$  in Ag and Au.

Table II. Self-trapping energies and pseudopotential parameters determined from the diffusion data.

	Cu	Ag	Au
$\alpha_0$ -eV	14.22	12.19	12.23
$\alpha$ -eV	13.25	11.73 $\pm$ .03	11.72 $\pm$ .02
$B_\mu$ -meV	90 <sup>d</sup>	344 $\pm$ 26	177 $\pm$ 13
$B_p$ -meV	40 <sup>d</sup>	153 $\pm$ 12	79 $\pm$ 6

<sup>d</sup>These are computed values from ref. 9.

## SUMMARY

We have presented muon diffusion parameters in Ag determined from measurements of the muon depolarization rate versus temperature in samples doped with dilute magnetic impurities. Comparison of the diffusion parameters for muons and hydrogen in the noble metals leads to the conclusion that only for  $\mu^+$  in Cu is incoherent tunneling a dominant mechanism. Model studies of over-barrier hopping indicate that this mechanism can explain the remaining data if the self-trapping energies for Au and Ag are 2 to 4 times greater than those calculated<sup>9</sup> for Cu.

## REFERENCES

1. See, for example, A. Seeger in *Hydrogen in Metals I*, edited by G. Alefeld and J. Völkl (Springer-Verlag, Berlin, 1978).
2. J. A. Brown, R. H. Heffner, R. L. Hutson, S. Kohn, M. Leon, C. F. Olson, M. F. Schillaci, S. A. Dodds, T. L. Estle, D. A. Vanderwater, P. M. Richards, and O. D. McMasters, *Phys. Rev. Lett.* **47**, 261 (1981).
3. M. F. Schillaci, R. L. Hutson, R. H. Heffner, M. Leon, S. A. Dodds, and T. L. Estle, *Hypt. Int.* **8**, 663 (1981).
4. V. G. Grebinnik, I. I. Gurevich, V. A. Zhukov, A. P. Manych, F. A. Meleshko, I. A. Muratova, E. A. Nikoskil, V. I. Solivanov, and V. A. Suetin, *Zh. Eksp. Teor. Fiz.* **68**, 1548 (1975) [*Sov. Phys. JETP* **41**, 777 (1969)].
5. L. Katz, M. Guinan, and E. J. Korep, *Phys. Rev.* **B4**, 330 (1971).
6. H. Katsuta and R. E. McLellan, *Scripta Metallurgica* **13**, 65 (1979).
7. K. Fichtenauer and D. Liebscher, *Z. Naturforsch.* **17A**, 355 (1962).
8. H. Teichler, *Phys. Lett.* **64A**, 78 (1977).
9. H. Teichler, *Phys. Lett.* **7A**, 313 (1978).
10. P. Jena and K. S. Singwi, *Phys. Rev.* **B17**, 3518 (1978).
11. A. I. Gubanov and V. K. Nikulin, *Fiz. Tver. Tel.* **7**, 2701 (1965), [*Sov. Phys. Solid State* **7**, 2184 (1966)].
12. M. Camani, F. N. Gygi, W. Büegg, A. Schenck and H. Schilling, *Phys. Rev. Lett.* **39**, 836 (1977).
13. K. Maier, G. Flik, D. Herlach, G. Jünemann, A. Seeger, and H.-D. Carstanjen, *Phys. Lett. A* (in press).
14. N. W. Ashcroft and D. C. Langreth, *Phys. Rev.* **159**, 500 (1967).
15. D. Emin, M. I. Baskes, and W. D. Wilson, *Phys. Rev. Lett.* **42**, 791 (1979).

\*Work supported by the U. S. Department of Energy and National Science Foundation.