

magnetization [S. Geller, H. J. Williams, R. C. Sherwood, and G. P. Espinosa, *J. Phys. Chem. Solids* **23**, 1525 (1962)]. Thus, if it is assumed that likewise  $\text{Fe}^{2+}$  ions in the dodecahedral sites do not contribute to the

magnetization and are only paramagnetic, variation of the applied magnetic field should cause a variation in both dichroism and absorption. However both remain constant.

## Diffusion and Trapping of Positive Muons in Al:Cu Alloys and in Deformed Al

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Depolarization of  $\mu^+$  particles implanted into quenched alloys of 0.042, 0.13, and 0.42 at.% Cu in Al shows peaks in the temperature dependence which are attributed to trapping of the muons by various metastable Cu impurity complexes, and provides evidence that positive muons can be used to study impurity correlations in metals.

Previous work has shown complex muon diffusion phenomena in metals.<sup>1-3</sup> This work is a study of one of the simplest systems of dilute impurities in metals which shows interesting muon depolarization dependence on temperature and therefore should be useful in understanding muon trapping and diffusion. For low concentrations of Cu in Al, though precipitation is not observed by transmission electron microscopy (TEM), there is indirect evidence from resistivity<sup>4</sup> and NMR<sup>5</sup> measurements as well as theoretical grounds<sup>6</sup> to suspect that Cu is not uniformly distributed.

To study this low-concentration region, we have measured the depolarization of implanted spin-polarized  $\mu^+$  particles (muon spin rotation<sup>7</sup>) in the temperature range 1.7 to 300 K. These experiments with muons provide a unique way of studying the thermal diffusion and transitory trapping processes for implanted particles, as well as the nature of impurities and other defects that act as traps. Over the temperature range studied, the weak depolarization that has been previously reported<sup>8</sup> for high-purity Al is explained by rapid diffusion of the muon.<sup>9</sup> Depolarization has previously been observed in Al:Cu.<sup>10</sup> The present data taken with a finer temperature mesh on quenched Al:Cu alloys reveal previously unobserved peaks in the temperature dependence, which can be explained on the basis of a model involving muon trapping at sites near Cu impurity complexes. So far trapping by impurities has not been treated theoretically. We presume that it arises from the local lattice distortion and electronic perturbation at sites near the impurity.

Alloys containing 0.042, 0.13, and 0.42 at.% Cu

were prepared from 99.99%-pure Al and Cu. Eight 0.3-cm $\times$ 7.5-cm $\times$ 10-cm plates fabricated from each ingot were combined to make muon-spin-rotation samples. Each plate was given a homogenizing treatment by annealing in air at 820 K followed by a quench (0.1 sec time constant) into agitated brine at 290 K. Prior to measurement, the 0.42% alloy was aged for 1.5 days at 390 K, the 0.13% alloy about 0.5 h at 300 K, and the 0.042% alloy 3 days at 300 K. Stereoscopic TEM measurements find no evidence of precipitation down to 200 nm and find an average dislocation density of  $10^9 \text{ cm}^{-2}$ .

In order to test the role of point defects and dislocations on muon trapping, we also studied a 99.999%-pure Al sample, deformed 50% in compression at approximately 77 K to a final thickness of 2 cm. Prior to the muon-spin-rotation measurements, the temperature of the sample was kept below 160 K so that vacancies created during deformation would not be lost. Subsequent TEM analysis at room temperature found dislocation cells about 1  $\mu\text{m}$  across, with a dislocation density of  $10^9$  to  $10^{10} \text{ cm}^{-2}$ .

The results of the muon-spin-rotation measurements are shown in Fig. 1 and were analyzed in the following manner. The decay of the asymmetry in the muon-spin-rotation signal can be represented as  $\exp[-\gamma(t)]$ . The muon spin decay rate, which we define as  $d\gamma/dt$ , has an average value which is statistically weighted according to the radioactive decay law. Thus we can define a dimensionless depolarization parameter:

$$\alpha = \int_0^\infty \exp(-st)(d\gamma/dt) dt, \quad (1)$$

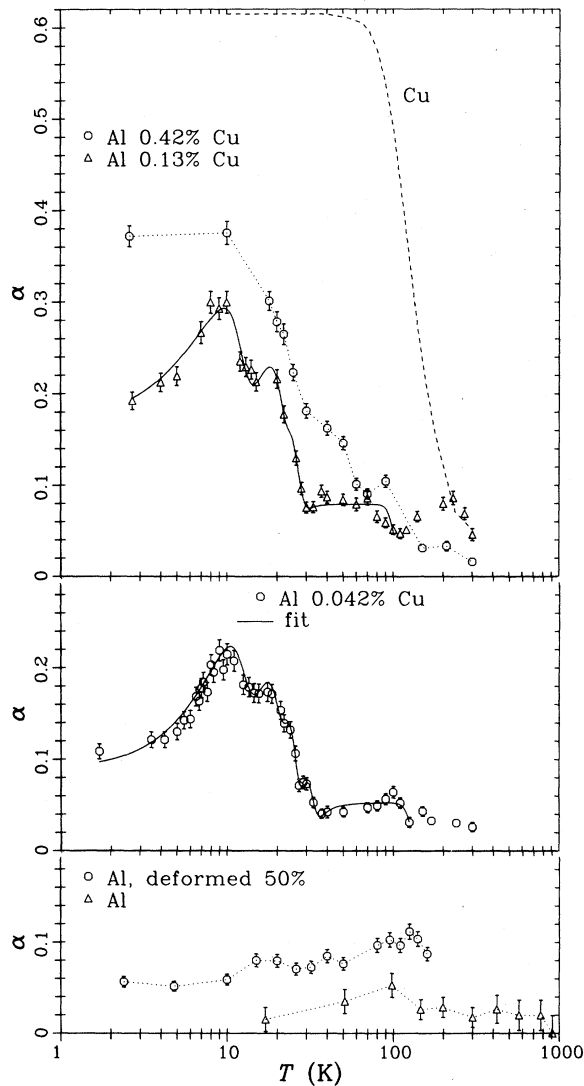


FIG. 1. Temperature dependence of the depolarization parameter  $\alpha$ , measured with external fields of 500 Oe for the 0.042% and 0.13% alloys and deformed Al, and 330 Oe for the 0.42% alloy. Data for Cu (Ref. 10, 30 Oe) and Al (Ref. 8, 75 Oe) are uncorrected for weak field perturbations (Ref. 11). The fits are Eq. (5) with the parameters in Table I.

where the muon lifetime  $s^{-1} = 2.2 \mu\text{sec}$ . We evaluate  $\alpha$  by fitting the experimental time histogram with a Gaussian function,  $\gamma(t) = \frac{1}{2}\alpha s^2 t^2$ , which we find gives a better fit than an exponential. We have chosen to represent the data in terms of the  $\alpha$  parameter because  $\alpha$  is also conveniently calculated from the theory, discussed below. For comparison, values of  $\alpha$  calculated from previous measurements on annealed polycrystalline samples of Al<sup>8</sup> and Cu<sup>12</sup> are also shown in Fig. 1.

The calculated value of  $\alpha$  is 0.44 for the muon localized at an octahedral interstitial site and 0.12 for a substitutional site, neglecting lattice relaxation and the finite extent of the muon wave function.<sup>9,11</sup>

Nonlinear screening calculations using the jellium model predict trapping of protons as well as muons at vacancies in Al.<sup>13,14</sup> We had assumed *a priori* that dislocations would also be capable of trapping the muons.

Our results for the deformed Al in the vicinity of 100 K are consistent with the trapping at open volume defects as opposed to trapping at interstitial sites by the strain fields surrounding defects.<sup>15</sup> A smaller fraction of the muons diffuse to traps at lower temperatures, indicating that the diffusion time increases with decreasing temperature.

The depolarization in the alloys at  $T < 50$  K is sufficiently large to be caused by the trapping of muons at interstitial sites. For the two more dilute alloys  $\alpha$  does not reach a constant maximum at low temperatures nor does it drop off smoothly with temperature, as it does for Cu (Fig. 1). In Cu the depolarization at low temperatures is the result of self-trapping at an octahedral interstitial site,<sup>16</sup> and the decrease at high temperatures is attributed to thermal diffusion.<sup>12</sup> The peaks observed in the alloy data at low temperatures increase in magnitude with increasing Cu concentration. This is expected for thermal diffusion of the  $\mu^+$  to traps whose population of Cu increases with impurity concentration.<sup>17</sup> What is surprising here is that complex structure is found above 10 K.

A motional-narrowing theory for  $\gamma(t)$ , related to that used for nuclear magnetic resonance, has been proposed<sup>17,18</sup> to explain muon diffusion and trapping in terms of correlation functions:

$$\gamma(t) = \sum_i \sigma_i^2 \int_0^t dt' \int_0^{t'} dt'' F_i(t'') G_i(t' - t''). \quad (2)$$

The sum extends over all interstitial sites in the sample, each having a local frequency distribution of second moment  $\sigma_i^2$  from nuclear dipolar fields.  $F_i(t)$  is the probability that the muon occupies site  $i$  at time  $t$  and  $G_i(t)$  is the site autocorrelation function. Equation (2) can be rewritten to represent equivalent unperturbed sites in the host and  $n$  types of trapping sites. The result is similar to Eq. (2), where we now define  $f_0$  as the probability that *any* host site is occupied, and  $f_m$  for  $m = 1, \dots, n$  as the probability that a trap of type  $m$  of concentration  $c_m$  is occupied. A

good approximation<sup>18</sup> to the autocorrelation function for any site of type  $m$  is  $g_m = \exp(-p_m t)$ . When the traps are assumed to be dilute, the  $f_m$  are then solutions to the following rate equations:

$$\frac{df_0}{dt} = -p_0 c f_0 + \sum_{m=1}^n p_m f_m [f_0(0) = 1 - c], \quad (3)$$

$$\frac{df_m}{dt} = p_0 c f_0 - p_m f_m [f_m(0) = c_m; m \neq 0], \quad (4)$$

where  $c = \sum_m c_m$ . The renormalized jump rate between host sites is  $p_0$ . The  $p_m$  for  $m > 0$  are detrapping rates given as  $p_m = p_0 \exp(-H_m/kT)$ , with  $H_m$  the binding enthalpy to the trapping site. Actual lattice structure is not taken into account explicitly in this model.

Since our definition of  $\alpha$  is identical to the Laplace transform, the solution for  $\alpha$  can be expressed as

$$\alpha = \sum_{m=0}^n \sigma_m^2 \bar{f}_m(s) / (s + p_m), \quad (5)$$

where  $\bar{f}_m(s)$  is the transform of  $f_m(t)$ . Thus one can solve the theoretical Eq. (2) quite easily as one does not need to solve the integral convolution of this equation in the time domain. Since the depolarization in pure Al is small,  $p \gg s$  and the  $m=0$  term is negligible. The model predicts a peak in the temperature dependence of  $\alpha$  when  $p_0$  increases monotonically with temperature and the trapping times and detrapping lifetimes are comparable to the muon lifetime, i.e., when the conditions  $p_0 c_m \approx s$  are satisfied. Thus, the peak appearing at the lowest temperature is associated with traps present in the highest concentration, assumed to be a single Cu impurity. The additional peak structure observed at higher temperatures is due to a lower concentration of traps with higher binding enthalpies, assumed to be di- and tri-Cu clusters.

Equation (5) was used to fit the data on the alloys for  $T \leq 100$  K using a nonlinear least-squares procedure with  $n=5$ . Because of a lack of a quantitative theoretical treatment, we have used an empirical formula for the temperature dependence of  $p_0$ :

$$p_0 = a \exp(-H_0/kT) + b + dT^2, \quad (6)$$

where the three terms are to represent the following. At high temperatures the Flynn-Stoneham model<sup>19</sup> predicts an Arrhenius dependence and at low temperatures a constant that is proportional to the coherent tunneling rate plus a one-phonon-assisted tunneling term that is linear in temperature and dependent upon strain broadening. We

find that the  $dT^2$  term fits the data better than a linear term. The fitted parameters are given in Table I. For the highest-concentration alloy (0.42%) the total number of traps is sufficiently high that the contribution of the individual traps is more difficult to observe and the data therefore were not fitted.

The significance of these results is that qualitatively our model describes the data at low temperatures. Quantitatively, the binding enthalpies giving rise to the trapping at low temperatures are quite small. The concentration of traps producing the peak at 10 K is consistent with individual Cu impurity traps. The structure seen at  $T > 10$  K is attributed to appreciable concentrations of microclusters of two or more Cu. We note that if the distribution of impurities were random, the fraction of interstitial sites with more than one Cu nearest neighbor would be about two to three orders of magnitude smaller than the fraction with one neighbor. Our larger fractions indicate that clustering is energetically more favorable even at these low Cu concentrations.

The depth of the  $m=5$  trap requires a different interpretation. On the basis of our findings for the deformed Al and our TEM results, we suspect that dislocations are responsible for the structure observed at temperatures near 100 K. This is also consistent with our small  $\sigma_m$  parameter. Diffusion along these dislocations explains the smaller depolarization at 300 K.

The results of this Letter are the first evidence demonstrating that positive muons provide a unique way of studying impurity correlations in

TABLE I. Results of fitting  $\alpha$ , given by Eq. (5), to the data. Because of correlations between parameters, values are determined to order-of-magnitude accuracy, except for the binding enthalpies where shown. The parameters in Eq. (6) are  $a = 2.6 \times 10^{14} \text{ sec}^{-1}$ ,  $b = 2.1 \times 10^8 \text{ sec}^{-1}$ ,  $d = 7.8 \times 10^6 \text{ sec}^{-1} \text{ K}^{-2}$ , and  $H_0 = 22 \text{ meV}$ .

Alloy (%)	$m$	$\sigma_m$ ( $10^5 \text{ sec}^{-1}$ )	$c_m$ (ppm)	$H_m$ (meV)
0.042	1	3	410	9(5)
	2	3	180	16(7)
	3	3	48	22(15)
	4	3	4	36(20)
	5	1	4	200(60)
0.13	1	3	910	9
	2	3	510	18
	3	3	110	25
	4	3	40	28
	5	1.3	180	160

metals. Extension of this work to other impurities in Al is currently in progress.

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<sup>11</sup>The Zeeman energy is expected to exceed electric quadrupole splittings for <sup>27</sup>Al near the  $\mu^+$  and Cu (Ref. 5) impurities. Electric field gradients and the location of the muon can be determined from the magnetic field dependence for single crystals. [See P. Jena *et al.*, Phys. Rev. Lett. **40**, 264 (1978); and O. Hartmann, Phys. Rev. Lett. **38**, 832 (1977).] In weak fields  $\alpha$  may be a factor 5/3 larger.

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## Undamped Lattice Vibrations in Systems with Two Incommensurate Periodicities

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Despite the fact that the Frenkel-Kontorova model, with chain of atoms and sinusoidal potential incommensurate, lacks translational symmetry, the structure factor  $S(q, \omega)$  for this model is shown to exhibit always phonon peaks of negligible width.

There has recently been an upsurge in interest in lattices with two incommensurate periodicities. Examples of such systems are charge-density waves incommensurate with the underlying lattice,<sup>1</sup> some superionic conductors in which the mobile ions order in a structure incommensurate with the rest of the lattice,<sup>2</sup> and some thin films which are incommensurate with the substrate.<sup>3</sup> Such systems have no translational symmetry because there exists no nonzero translation under which both periodicities are invariant. Two of us have also shown<sup>4</sup> along with Aubry<sup>5</sup> that a model for such systems, known as the Frenkel-Kontorova model (which consists of a chain of atoms connected by springs, interacting with a sinu-

soidal potential), exhibits a sharp transition at a critical value of the strength of the sinusoidal potential such that below this critical strength the ground state of an infinite chain incommensurate with the sinusoidal potential is continuously degenerate.<sup>6</sup> This implies the existence of a zero-frequency sliding mode and the possibility of free sliding of one periodicity with respect to the other.

We will show that the dynamical structure factor  $S(q, \omega)$  (i.e., the imaginary part of the phonon Green's function for the chain) exhibits sharp phonon peaks of exceedingly small width, as for a system with translational symmetry. Furthermore, because of the existence of a continuously