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THE POSITIVE MUON AND THE POSITRON AS PROBES OF DEFECTS

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The positive muon and the positron are each being used nowadays to investigate defects in condensed matter. A brief summary of the experimental methods employed with each particle is given in this paper. Similarities and differences between the behavior of the two leptons when implanted in condensed matter are pointed out, and by means of a comparison between muon and positron data in Al it is shown that the combination of muon and positron experiments can serve as a useful new probe of defects in solids.

INTRODUCTION

In this paper an up-to-date comparison of the positive muon and the positron as probes in the study of defects in metals will be made. Even though both of these particles are positively charged their properties in metals are significantly different owing mainly to mass and decay rate differences. It is hoped this parallel examination will provide some insight into new experiments which could resolve some of the questions remaining in both fields.

Both the positron and the positive muon are members of the lepton family with spin $1/2$. The muon has a mass 206.769 times that of the positron; a condition which creates distinct differences in the fundamental dynamics of the motion of these two particles in condensed matter. The positron, being stable in vacuum, annihilates in most metallic systems in times on the order of 0.2 nsec, whereas the positive muon decays in approximately 2.2 μ sec. These factors of $\sim 10^4$ in the mean lifetime and $\sim 10^2$ in the mass result in dissimilarities between the defect sensitivities of the particles.

The parity violating decay of a polarized muon¹ into a positron and two neutrinos allows one to determine the diffusive nature of the muon motion, i.e. to differentiate trapped muons from those diffusing in the perfect lattice. Since the positron is emitted preferentially in the direction of the muon's spin, one can measure the average polarization of an ensemble of implanted muons as a function of time. The observed spin depolarization is the result of inhomogeneous local magnetic fields which the muons sample as a result of diffusion.² The positron, usually obtained from radioactive sources, is implanted into the sample and is studied by using the decay products, principally two photons occurring from the annihilation of an electron and positron whose masses are converted into electromagnetic energy. By measuring the decay rate of the positron, the two-photon angular distribution, or the Doppler shift of the annihilation photons one can determine whether or not the positron is localized at a defect.³ A muon or positron which reaches a site where the mean escape time is large compared to its respective lifetime will be called trapped. Both of the particles are implanted into a metal and reach thermal energies^{2,3} in times much shorter than their mean lifetimes.

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Due to differences in the measurement techniques, mass and lifetimes, these two probes complement each other rather than duplicating the information supplied by their decay products. The present activity in both fields is mainly aimed at attempting to understand low temperature data where intrinsic and extrinsic defects play an important role in localizing these particles. The particle-impurity, or particle-defect potential and the trapping rates are not well known. At high sample temperatures the positron has provided considerable new information on the vacancy formation enthalpies of different metallic systems³ in contrast to the muon which has thus far provided little information concerning equilibrium defects in this temperature region.⁴

Owing to space limitations I will specifically discuss both low and high temperature data for only one metal, Al. Aluminum was chosen for this comparison because a significant amount of experimental work has been performed with positrons and muons. Well-annealed high-purity Al provides a good host to study defects in as the muon has a long depolarization time extending from 30 mK⁵ to 900K⁴ indicating that self trapping does not occur. Since Al is a simple "sp"-electron metal, it is also amenable to rather precise theoretical treatment. Aluminum should also provide a baseline for other more complicated metals.

POSITRON MEASUREMENTS

Positron measurements can in principle be made in all metallic systems although, in Group I metals, for example, thermally induced vacancies do not show signs of positron trapping.³ There are three standard positron measurements: 1) lifetime, a measurement of the time a positron resides in a metal before annihilation; 2) angular correlation, the angular distribution of the annihilation photons; 3) Doppler broadening, the Doppler shift in the energy of the emitted photons. The latter two measurements yield information on the electron momenta distribution at the site of the positron during annihilation. The lifetime is a measure of the expectation value of the electron density for all momenta over the extent of the positron wavefunction in the metal. Like the muon, the Coulomb interactions repel the positron from the ion cores; therefore, annihilation mainly occurs with conduction electrons. Angular correlation measurements are becoming widely used in extracting Fermi surface information.³

If a defect produces a local decrease in the ion core density, the probability of finding the positron is increased in this region. Owing to change in the lattice potential around an open volume defect both the electron density and momentum are modified relative to the perfect lattice. These defects therefore produce a unique signature both in the decay rate and the angular distribution of the annihilation photons. The most useful information has been obtained from thermally induced monovacancies from which a formation enthalpy can be extracted;³ however, non-equilibrium defects can also be studied by this technique. Owing to its increased sensitivity to these open volume defects⁶ the positron has also been found to be very useful in the study of small vacancy clusters and voids produced by neutron and electron irradiation. The positron sensitivity ranges from atomic defect concentrations of 0.1 to 100 ppm.³ The latter concentration is where "saturation trapping" occurs, i.e. where >99% of the positrons annihilate in the trapped state.

It should be mentioned that positron trapping by monovacancies in metals indicates that the capture rate, and not the diffusion to the traps, controls the trapping rate,⁷ κ , in contrast to muon trapping. Experimentally this has an inherent advantage in that κ becomes temperature independent. This has been shown theoretically using an analogy of positron trapping to low-energy neutron capture by nuclei⁸ and by the application of the Fermi golden rule.⁷ For larger defects this temperature independence should and does appear to break down at low temperatures.⁹

While the positron is localized at a defect it should fundamentally be possible to extract the electron momentum profile associated with this defect. Although a significant theoretical attempt has been made in this area,¹⁰ the difficulties of a fully self-consistent calculation of both the positron and electron wave-functions in a defect has not been attempted. Very few quantitative comparisons between theory and experiment have been made either for angular correlation or for Doppler-broadening measurements even for the relatively simple case of a monovacancy. In measuring positron lifetimes associated with different defects it is difficult to experimentally resolve the individual components owing to parasitic fitting problems in deducing different exponentially decaying functions of unknown intensities. It is therefore impractical to resolve more than two traps or make an unambiguous assessment of the relative concentrations of each type of trap. Even with the above difficulties, a significant amount of research has been performed on more complicated types of defects such as voids, dislocations, stacking faults and grain boundaries.³

The low temperature properties of the positron are beginning to be examined more carefully and there appears to be some sensitivity to impurities or possibly those impurities associated with defects.¹¹ Recently, positron-lifetime experiments have measured the interaction of monovacancies and interstitial carbon impurities in electron irradiated α -iron.¹² The results show that asymmetric carbon-vacancy pairs form at annealing temperatures of 220K. Debate persists on the effect of these types of shallow traps as the capture rate may not be sufficiently high to allow localization of the positron in these regions at low temperatures.¹³ This area of positron research is presently very active.

Owing to the development of high flux monoenergetic positron beams in ultra-high vacuum,¹⁴ experiments designed to study well-characterized surfaces and those defects associated with the surface region are now in progress. The interaction of the positron with surfaces or surface defects is at present the least understood area in positron studies.

MUON MEASUREMENTS

For completeness the different measurement techniques for muons will be briefly mentioned, although for a more in depth appraisal the reader is referred to the review papers in the present conference proceedings or those previously published.^{3,16} The muon, which is characterized as a radiative magnetic probe, can be obtained in highly spin polarized beams. Owing to the emission of the positron preferentially in the direction of the muon's spin one can observe the time evolution of its spin under static and dynamic local magnetic fields. Three basic measurements can be made: 1) Muon Spin Rotation where the muon precesses about an applied transverse magnetic field thus allowing the measurement of the transverse relaxation function (depolarization rate); 2) Muon Spin Relaxation which yields the longitudinal relaxation function by measurement of the forward/backward decay asymmetry; 3) Muon Spin Resonance which is detected by applying an r.f. field when measuring the forward/backward decay asymmetry thereby yielding the Larmor frequency and both of the above relaxation functions.¹⁶ In all these measurements the time is determined between the muon's arrival at the sample and the appearance of its decay positron. The transverse relaxation function has been most widely used to date in defect studies although, as will be discussed, longitudinal relaxation functions will be helpful in unraveling present dilemmas which exist in the field of impurity or defect trapping.

In the past few years it has become painfully obvious that sample characterization is extremely important in muon studies. For example, only 42 ppm Mn⁵ was found to significantly effect the depolarization rates in Al measured at low sample temperatures (<40K). This characterization is necessary if one wants to determine

the intrinsic diffusive properties of the muon in the host sample under various experimental conditions. The reader is referred to other papers on the temperature dependence of the diffusion coefficient to fully appreciate the various diffusion mechanisms that have been proposed.

Equilibrium defects such as thermally induced vacancies are known to exist where sample temperatures approach the melting point; however it has not been shown that the muon is localized at a unique substitutional site.⁴ The lack of a measurable depolarization rate at high temperatures eliminates a large class of experiments that could be performed by this technique. If methods do become available to measure depolarization times of many μ secs, the muon should become useful in the study of these equilibrium defects.

Non-equilibrium defects on the ppm level have been shown to be effective traps for the muon. For example, monovacancies have been shown to localize the muon at low temperatures in high-purity Al.¹⁷⁻¹⁹ Other shallow traps such as dislocations,¹⁹ impurity clusters¹⁹ and loops will also be important in these low temperature investigations; therefore defect characterization will be of the utmost importance.

TRAPPING MODELS

In applying the trapping model to either probe one must at least have a primitive knowledge of the diffusion mechanism and its temperature dependence, in order to estimate defect concentrations. The mechanics of muon and positron diffusion will be specifically discussed in their relation to the trapping model. A number of diffusion models have been proposed for the muon which are not only applicable to different temperature regimes but their initiation can be dependent on the defect concentration and type existing in the metal.²⁰ The extraction of information concerning defect concentrations in a diffusion limited case (i.e. where the trapping rate into a defect is determined by the diffusion time) is difficult to accomplish. The trapping rate per unit trap concentration (κ) is usually written as^{21,22} $\kappa = 4\pi D r_0/V_a$ where D is the diffusion constant, r_0 the effective capture radius of the traps and V_a the atomic volume. One can easily surmise that if the temperature dependence of D ²⁰ is not known an unambiguous derivation of κ is close to impossible, without the defect concentration and distribution being known.

The product κC , where C is the atomic concentration of trapping sites, determines the fraction of trapped muons or positrons at defects. Therefore, if $\kappa C\tau \gg 1$, "saturation trapping" occurs. If $\kappa C\tau \ll 1$ then most of the particles will decay while diffusing in the perfect lattice. One can easily see that the difference in the mean lifetime (τ) of the positron as compared to the muon in metals should be easily offset by the difference in the respective diffusion constants.^{23,24}

Figure 1 shows the theoretical prediction of the positron diffusion constant as a function of sample temperature for pure Al²³ and Al -0.01 at.% Si. The diffusion constant at high temperatures is mainly limited by the positron-phonon interaction which has been treated by the deformation potential model.²³ From Figure 1 it can be seen that at low temperatures dilute impurities assume an important role in determining the positron diffusion constant. In this case of Si in Al, a difference in the potential of 0.01 Ry was assumed between the Al host and the Si impurity.

One can make a rough estimate of the average diffusion length (L) for both the positron and the muon by the following relation, $L = \sqrt{2D\tau}$. Using values of $D_{e+} \sim 0.2 \text{ cm}^2/\text{sec}$ ²⁵ and $D_{\mu+} \sim 3.0 \times 10^{-5} \text{ cm}^2/\text{sec}$ for 298K one can estimate a diffusion length of $\sim 1000 \text{ \AA}$ for both the positron and the muon. Since the muon diffusion constant is not known for Al one can only guess that this would be a

Figure 1. The log of the positron diffusion constant versus temperature for pure Al and Al-0.01 at.% Si derived from the theoretical equation of Ref. 23.

reasonable value of L . Since one experimentally measures the product κC , and κ is unknown, except for Fe,²⁶ one cannot determine C . Even in Fe the fundamental diffusion mechanism may depend on the number and type of defects. Another consideration is that the presence of impurity concentrations of a few ppm or more induces temperature dependent depolarization rates. High purity samples are therefore necessary to ensure that muon-impurity trapping is not occurring in the low temperature regions where data could be misinterpreted as evidence for an intrinsic defect property.

The apparent success of the two-state trapping³ model in positron annihilation studies suggests that a similar model may also be applicable to muon studies. The final trapping rate equation for both the positron and muon will be shown only for the simple two-state situation (free and trapped). For cases involving multiple traps the reader is referred to the original works. In both cases it will be assumed that no epiternal trapping occurs, i.e. pre-thermalized trapping. In these equations n_f and n_t are the number of free and trapped positrons, respectively, and κC is the trapping rate. The associated decay of the positron in the lattice is represented by λ_f , and λ_t is the decay in the trap, whereas the muon has a fixed decay rate (λ_u). The positron rate equations are as follows:

$$\begin{aligned} \frac{dn_f(t)}{dt} &= -\lambda_f n_f - \kappa C n_f \\ \frac{dn_t(t)}{dt} &= -\lambda_t n_t + \kappa C n_f \end{aligned} \quad (1)$$

The first rate equation represents the change in the fraction of positrons in the lattice per unit time which is determined by the fraction lost through decay and the fraction lost through trapping. The second equation represents the change in the fraction of positrons trapped in a defect per unit time which is given by the fraction lost through decay plus the fraction gained by trapping. Solving for the number of positrons remaining at some time $[N(t)]$:

$$N(t) = \frac{d}{dt}[n_f(t) + n_t(t)],$$

one finds

$$N(t) = \frac{\kappa C \exp(-\lambda_t t)}{\lambda_f - \lambda_t + \kappa C} + \left[1 - \frac{\kappa C \exp(-(\lambda_f + \kappa C)t)}{\lambda_f - \lambda_t + \kappa C} \right]. \quad (2)$$

This expression is the sum of two exponential components whose decay rates and intensities depend on λ_f , λ_t and κC and is directly measurable with positron lifetime measurements. A similar equation can be derived for results obtained from angular-correlation and Doppler-broadening experiments, as both the perfect lattice and the defect have unique momentum profiles. One characterizes this profile by some lineshape parameter which indicates that either all the positrons annihilate in the free lattice or in defects. With these two limits one can therefore apply the same two-state model. Although it should be noted that when comparing positron lifetime to either Doppler-broadening or angular-correlation measurements two different physical measurements are involved. The lifetime is determined by the electron density at the site of the positron, whereas the latter two measurements, when analyzed in terms of some lineshape parameter or peak counting rate, are biased toward annihilations with electrons having narrowly restricted momenta; this can produce differences between the measurements.

A brief discussion of the Petzinger et al.²⁷ trapping model for muons will be made and compared to the available Al data. This model assumes that the muon hopping rate in the pure host is monotonically increasing as a function of temperature, which decreases the depolarization rate as T increases. This assumption is incorrect if the muon motion is described by a band like propagation.²⁰ If we represent the quantity of interest, the time-dependence of the muon spin polarization $[P(t)]$ as equalling $\exp[-\gamma(t)]$ then the muon spin decay rate is defined as $d\gamma(t)/dt$ and is weighted according to the radioactive decay law. Using a dimensionless depolarization parameter, α , as suggested by Fiory,²⁸

$$\alpha = \int_0^\infty \exp(-\lambda_\mu t) \left(\frac{d\gamma(t)}{dt} \right) dt \quad (3)$$

where α is evaluated by fitting the experimental time histogram with a Gaussian depolarization function,

$$\gamma(t) = \frac{\alpha \lambda_m^2 t^2}{2}.$$

A motional-narrowing theory for $\gamma(t)$ ^{27,29} is employed to describe muon diffusion and trapping by the use of the correlation function

$$\gamma(t) = \sum_i \sigma_i^2 \int_0^t dt' \int_0^{t'} dt'' F_{ij}(t'') G_i(t' - t''). \quad (4)$$

Here F_{ij} is the probability that a transition from site i to site j has occurred in time t'' , G_i is the auto-correlation function for site i and σ_i^2 is the second moment of the frequency distribution attributable to site i in the lattice where the sum extends over all the interstitial sites. The above equation can be rewritten as a rate equation to determine F_{ij} which represent equivalent unperturbed sites in the host and a trapping site. The rate equations used in muon studies are similar to those used in positron studies but must be modified slightly. The quantity f_0 is defined as the probability that any host site is occupied, f_t is the probability that a trap of concentration c_t is occupied, and the autocorrelation function of the trap is represented by $g_t = \exp(-p_t t)$.²⁹ The rate equations can be expressed as¹⁹

$$\frac{df_0}{dt} = -p_0 c f_0 + p_t f_t \quad [f_0(0) = 1 - c] \quad (5a)$$

$$\frac{df_t}{dt} = p_0 c f_0 - p_t f_t \quad [f_t(0) = c_t] \quad (5b)$$

where the muon decay is taken into account by α and in this single trap case $c = c_t$. These equations assume one trap although they are simply generalized by letting t sum through the number of different traps. These equations are easily solved by Laplace transforms and the expression for α is the Laplace transform of the convolution in Eq. (4) and is the simple product of the transforms of $F_{ij}(t)$ and G_i . In this case p_t is a detrapping rate given as $p_t = p_0 \exp(-H_t/kT)$ with H_t representing the binding enthalpy to the trapping site, t . p_0 is defined as the renormalized jump rate between host sites. Now the expression for α can be represented by a single trap as

$$\alpha = \frac{\sigma_0^2 \bar{f}_0(\lambda_\mu)}{\lambda_\mu + p_0} + \frac{\sigma_t^2 \bar{f}_1(\lambda_\mu)}{\lambda_\mu + p_t}$$

where $\bar{f}_1(\lambda_\mu)$ is the transform of $f_t(t)$ and described as

$$\bar{f}_1 = \frac{c(p_0 + \lambda_\mu)}{[\lambda_\mu(\lambda_\mu + p_0 c + p_t)]}$$

Different functional forms can represent p_0 ^{7,19,27,30} although they are not based on a sound theoretical approach.

In order to extract trap information, both the positron measurements and the muon depolarization rate data have to be compared to rather complicated nonlinear expressions. The fitting of these models makes it difficult to obtain unambiguous results, especially if two traps or detrapping phenomena are present. These necessary fitting procedures limit both techniques in resolving the complicated defect structures which exist in many metals. The complex structure observed in the depolarization rate data has evoked many different interpretations,^{19,30,31} e.g. different diffusion mechanisms, Anderson localization, as well as impurity and defect trapping. To help resolve this situation Petzinger³² has extended the theory of Kubo and Toyabe.³³ He suggests that by measuring the longitudinal depolarization in zero magnetic field one can determine whether the structure observed in the depolarization rates, as deduced from transverse field experiments, is associated with defect trapping or with the initiation of a new diffusion mechanism. In other words zero field measurements can fundamentally distinguish between diffusion away from a trap (detrapping) or diffusion toward one. Zero field measurements will be critical in resolving much of the structure found by transverse field measurements. Owing to limited space this model will not be discussed further. Both transverse and longitudinal field measurements should be made on all samples where confusion over the interpretation of the data exists.

If the muon is localized in the lattice or at a defect the spin of the muon depolarizes with a characteristic time (σ_0) which is related to the second moment of the dipolar local-field distribution. Assuming one is in the high field limit³⁴ and the muon is localized, one can determine the position of the implanted muon in the lattice by the resulting depolarization rate. The values shown in Table 1 are the calculated dipolar linewidths for an undistorted Al lattice in the high field limit assuming no spread on the muon wavefunction.^{18,35} As can be seen in Table 1 the values of σ are different enough to determine if the muon is, for example, positioned at an interstitial site or at a vacancy by the transverse measurement. This method is not as useful for determining larger vacancy clusters as the sensitivity is reduced in contrast to positron annihilation experiments. For example, the value of σ is about 4% less for a divacancy compared to a monovacancy. This insensitivity to larger clusters is attributable to the static dipolar field which depends inversely on the third power of the distance between the nuclei and the muon.

Table 1

Gaussian depolarization rate parameters, σ , calculated for muon localized at sites in an undistorted lattice.

Field Direction	Octahedral	Tetrahedral	Vacancy
Al(100)	0.322	0.081	0.096
Al(110)	0.172	0.292	0.115
Al(111)	0.070	0.334	0.120

COMPARISON OF POSITRON AND MUON DATA ON Al

By way of an example, muon and positron data for Al will be compared emphasizing the differences that exist between the sensitivity of the probes to defects. As has been previously mentioned both the positron and the muon show signs of trapping at monovacancies at low sample temperatures. In Fig. 2 one can observe the differences in the two sets of measurements over a wide temperature region. The positron-annihilation Doppler-broadening data on quenched Al of ($T_q = 853K$) 5N purity is that of Alam et al.³⁶

Figure 2. The temperature dependence is shown both for the muon depolarization rate (σ) and the positron Doppler-broadening lineshape parameter (S) for high purity Al. The circles represent depolarization rate data from Brown et al.¹⁷ on quenched Al and the dots are depolarization data on well-annealed Al from Ref. 4. The solid line represents a fit to the trapping model.²⁷ The x's are the lineshape parameters extracted from Doppler-broadening data on quenched 5N Al of Alam et al.³⁶ and the squares represent the lineshape data of Lynn et al.³⁸ in a temperature equilibrium measurement.

The changes, which occur in the positron lineshape data vs. temperature (Fig. 2), are characterized by a lineshape parameter, S . This S parameter is defined as the ratio of the total counts in the central region to the total counts in the full energy peak. Since the positrons localize at low ion density regions, the probability of annihilation with core electrons (high momentum electrons) decreases. This produces a narrowing of the annihilation photopeak and therefore an increasing value of S . For example, a defect such as a cluster of vacancies will produce an even narrower photopeak than that of a monovacancy. The increase in the S parameter at 220K is associated with Stage III (vacancy migration)³⁶ and is described in terms of vacancy agglomeration. After the vacancy cluster reaches between 5 to 10Å in radius the peak is no longer expected to narrow with increasing cluster size.³⁷ The decrease in S above 250K is associated with a decrease in the concentration of traps and the formation of dislocation loops from the previously formed vacancy clusters. Above 440K the dislocation loops and remaining vacancy clusters become unstable and disappear corresponding to S values obtained for the well-annealed samples. Larger voids (>250Å radius)

produced by neutron irradiation have been found to be stable to temperatures greater than 500K before they anneal out of the crystal.³⁶ Figure 2 shows the onset of positron trapping at thermally generated monovacancies which occurs when the sample temperature above 500K.³⁸ The two-state trapping model can be fit to the higher temperature data which allows extraction of the vacancy formation enthalpy.³ Analysis of the equilibrium data from both positron lifetime and momentum measurements generally produces a monovacancy formation enthalpy (E_{1v}) of approximately 0.64 ± 0.02 eV for Al.³

In contrast to the above picture for positrons, muons at high sample temperatures show no signs of trapping at thermally induced monovacancies.⁴ The absence of this depolarization associated with muon trapping of vacancies has three possible explanations: (1) a small binding energy of the muon to the vacancy (<1.0 eV) resulting in significant thermal detrapping; (2) the muon-vacancy complex migrates appreciably and does not show signs of motional narrowing,^{39,40} (3) even at high sample temperatures the muons diffuse too slowly to trap at vacancies during a muon lifetime.⁴ The latter possibility seems unlikely as it would require the muon to have a diffusion constant much smaller than that of hydrogen. As previously mentioned the muon has already been shown in Al to be sensitive to less than 100 ppm of Mn impurities at low temperatures.⁵ This impurity concentration is significantly less than the vacancy concentration expected near the melting temperature. For the monovacancy complex to migrate the muon must first be rejected during the vacancy migration therefore raising the migration energy barrier by almost the full binding energy of the muon to the vacancy. Doyama et al.⁴⁰ have suggested that this complex could still make 600 jumps within the muon lifetime. This mechanism seems more unlikely than simple thermal detrapping of the muon from the vacancy in a time short compared to the muon lifetime.

The quenched Al data ($T_q = 873$ K) of Brown et al.¹⁷ shown in Fig. 2 as the open circles were interpreted in terms of the Petzinger et al.²⁷ trapping model and are quantitatively consistent with muon trapping. The solid line through the data represents the fit of the 3 trap model to the data. The traps are associated with the maxima occurring at ~ 20 K and ~ 100 K and the plateau beginning around 140K. Assuming a random hopping rate increasing monotonically with temperature the authors associated the ~ 100 K peak and the ~ 140 K plateau with muon trapping at monovacancies and divacancies, respectively. Above 200K the vacancies are mobile and form vacancy clusters thereby decreasing the total concentration of traps. The muon most likely is in a diffusion limited regime or is not as sensitive to these large vacancy clusters. One should mention that the large number of adjustable parameters and the lack of uniqueness of the fit are inherent problems with this approach. It is recommended that in future studies more data be taken so that a quantitative comparison can be made to the trapping model. It would also be useful where possible to reduce the number of traps present in the sample.

Recent quenching results⁴² ($T_q = 873$ K) on 6N oriented Al single crystals are consistent with those results on polycrystals.¹⁷ Small differences do exist in that the lower peak which was observed at ~ 20 K moved up on the single crystal results to around 40K, and the minimum (~ 130 K) found in the polycrystalline data was reduced in magnitude. Brown et al.¹⁷ associated this lower peak (~ 20 K) with extended traps arising from strains developed in the polycrystal during quenching. The fact that this peak was also observed in the single crystal demonstrated that it is not associated with the polycrystalline nature of the sample. It is still associated with a quenched-in defect and most likely an extended type defect. The measured depolarization rate at 100K in both the single and polycrystalline samples is consistent with the muon sitting in the center of the vacancy, in contrast to channeling results on protons in Al.⁴³ Channeling results are interpreted such that the protons occupy tetrahedral sites next to monovacancies.

Using the deduced parameters found by Brown et al.¹⁷ in fitting the trapping model to their Al data and employing typical values of the monovacancy and divacancy formation enthalpies ($E_{1V} = 0.64$ eV and $E_{2V} = 1.09$ eV), one can observe the values predicted by the trapping model²⁷ in Fig. 3 for two quench temperatures ($T_Q = 776$ and $T_Q = 873$ K). This figure illustrates the predicted sensitivity of the muon to monovacancies and divacancies with the above interpretation. Zero field measurements could also be performed in these cases to determine if detrapping is occurring for the region between 100-140 K. Both zero field measurements and data obtained for a different quench temperature would be useful in confirming the interpretation of Brown et al.¹⁷ and for a quantitative understanding of the muon-vacancy interaction. Low temperature electron irradiation experiments would be useful, as they remove the effect of divacancies produced in quenching experiments.

Figure 3. A plot of the theoretical prediction for different quenching temperatures in Al from the trapping model of Petzinger et al.²⁷ using the parameters derived from the fit of the quenching data of Brown et al.¹⁷ with a monovacancy and divacancy formation enthalpy of 0.64 and 1.09 eV, respectively.

Studies performed on neutron irradiated Al^{18,44} have the added difficulty that more complicated defects are generated as well as transmutation products. For example a thermal neutron fluence of 0.9×10^{22} n/cm² corresponding to a fast fluence of 4×10^{21} n/cm² ($E > 0.1$ MeV) produces approximately 0.23 at.% Si.³⁶ Figure 4 shows the extracted depolarization rate found in a well-annealed polycrystalline Al sample containing only 0.09 at.% Si.⁴⁵ Different cooling

Figure 4. Temperature dependence of the muon spin depolarization rate for Al-0.09 at.% Si sample. The data were initially fit to a Gaussian depolarization, and therefore $\alpha = 2\sigma^2/\gamma_\mu^2$.

rates on this Al-0.09 at.% Si sample showed that the same peak structure occurred as that shown in Fig. 4, although the sharpness of the peaks did vary depending on the cooling rate. One can see that this already shows a complicated structure, independent of defects produced by neutron irradiation, presumably associated with muon trapping at Si impurities. These Si impurities produced from transmutation during neutron irradiation could explain why Herlach et al.¹⁸ found a lack of recovery in Al after isochronal anneals above 300K. Impurities produced by nuclear reactions will complicate any interpretation on what were initially considered high purity metallic samples, but might be a useful way to dope a sample with a particular impurity.

Recent positron annihilation results⁴⁶ on quenched Al-0.23 at.% Si exhibit behavior similar to that shown in Fig. 2 for the high purity quenched Al sample results. Small but significant differences were found, i.e., the rise occurring at ~220K continued to values larger than 0.53 and this peak broadened extending to almost 300K. This indicates that the Si impurities appear to stabilize these vacancy clusters. It is clear from these results that positrons are not as sensitive as muons to these individual impurity clusters.

The relative importance of the particle interactions with impurities, defects or the defect-impurity complexes will certainly depend on both the type and concentration of the impurities and defects and the relative particle affinity to each type. Very little research has been performed in this area of positrons or muons. Careful and systematic studies are needed using the two probes in complementing studies.

The lack of depolarization of muon trapping at lattice defects produced by irradiation of Al foils at 77K with 4 MeV protons is not understood.⁴⁷ One possible explanation is that the recovery of the resistivity observed in similar samples occurred at higher sample temperatures than expected from previous studies.⁴⁸ This could indicate that possibly dislocation loops, and not simple point defects, were formed during irradiation; therefore the concentration of defects was overestimated. This factor could decrease the probability of trapping to a value where the fraction trapped would be below detection. A second possibility is that the muon after trapping could continue to diffuse in the dislocation loop. Assuming no difference between interstitial and vacancy defects and assuming no agglomeration of the vacancies which could have been produced by thermal spikes, the authors concluded that a muon jump rate of less than 10^{10} sec^{-1} at 250K would be consistent with their results.⁴⁷ Assuming diffusion occurs over octahedral interstitial sites one finds a maximum diffusion constant of $3 \times 10^{-6} \text{ cm}^2/\text{sec}$.

Kossler et al.¹⁹ have also reported muon trapping at both vacancies and dislocations produced in a 5N Al single crystal sample deformed at 77K. The deduced value of the depolarization rate is consistent with muon localization at a substitutional site. One might expect that a muon would trap at an edge dislocation, although diffusion along the dislocation would continue until a jog (i.e. point-like defect) was encountered providing a deeper trap. An unambiguous identification of individual traps is very unlikely in plastically deformed samples where many different defects coexist as possible trapping sites. The determination of muon trapping at dislocations could, however, be done by careful bending of oriented single crystals.

Muon trapping has also been observed at 293K by the longitudinal field method in non-stoichiometric aluminum alloys⁴⁰ containing excess vacancies. The main trap is associated with monovacancies although the authors suggested a possible second trap, i.e. stacking faults, to explain a small minimum occurring in the data at 3 μsec . Further studies are necessary to confirm this latter possibility.

SURFACE INTERACTIONS

Recently monoenergetic beams of slow positrons⁴⁹ have been developed which can be used as unique probes in surface physics studies. One can imagine that with the high intensity surface muon beams presently available, individual surfaces or those surfaces associated with a number of thin films could be studied. There is no doubt that a chemisorbed muon state exists on the surface of metals, as has been found with both positrons⁵⁰ and hydrogen.⁵¹ One could then measure such properties as surface magnetism, thermal desorption of the muon into muonium and possibly surface defects and impurities.

With reasonable estimates of the muon diffusion coefficient, reflection coefficient and foil thickness one can predict by employing the model of Leon and Heffner,⁵² that a significant fraction of muons will reach the surface of a trap-free film before decaying. This model assumes that the muons stop homogeneously throughout the foil and that no non-thermal muons escape by forming muonium. As suggested by the above authors these thin foils could be used to measure macroscopic diffusion constants (10^{-7} cm²/sec) and their temperature dependence. Figure 5 shows the solution to the equations presented in Ref. 52. The log of the thickness is plotted versus the efficiency of the muon in reaching the surface of the foil for diffusion constants ranging from 10^{-8} to 10^{-2} cm²/sec. A value of 10^6 for β was used for the radiation boundary condition.²¹ These foils would have to be annealed in situ to remove most of the defects produced in fabrication and handling. With the high intensities of surface muons expected at SIN one could use a single bulk sample and simply retard the muon energy so that it would only interact with the first few monolayers of the sample. Strong consideration should be given to this area in the future.

Figure 5. The predicted probability that positive muons will reach the surface of foils of varying thicknesses and diffusion constants before decaying. These solutions are based on the equations presented in Ref. 52. The radiation boundary condition which determines β was chosen to be 10^6 for these plots, where values of β of zero and infinity mean a perfect reflector or absorber, respectively.

CONCLUSIONS

A brief but hopefully general comparison has been made between muons and positrons as probes for the study of defects in metals. Since muon experiments are not only more demanding in manpower, cost and availability than positron experiments, they should be carefully designed in light of the knowledge that the muon is extremely sensitive to both intrinsic and extrinsic defects. Initial experiments should provide estimates of the muon diffusion coefficients as a function of sample temperature. High temperature hydrogen diffusion measurements do provide guidelines, although so far most of the observations of muon trapping have been made at low sample temperatures where hydrogen diffusion data do not exist. Given that the diffusion constant is known as a function of temperature, high-purity Fe²⁶ is therefore a good candidate to study with muons after low temperature electron irradiation. Since the defect type and concentration can be controlled in electron irradiated samples, such investigations could confirm the stated values of the diffusion constants in Fe thereby providing a new method for evaluating diffusion coefficients below Stage III in different metals.

All the samples should be initially characterized by other less costly techniques to obtain, where possible, the concentration and type of extrinsic and intrinsic defects. Both transverse and longitudinal measurements should be made to unravel the question of different diffusion mechanisms versus defect-, impurity- or self-trapping.

Provided that the full capabilities of the muon are utilized and coupled with complementary techniques, i.e., positrons, the muon will constitute a useful new probe in deepening our understanding of defects in metals.

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REFERENCES

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- [1] R. L. Garwin et al., Phys. Rev. 105 (1957) 1415.
- [2] For general reviews on the muon techniques see: A. Schenck in Nuclear and Particle Physics at Intermediate Energies, J. B. Warren (ed.) (Plenum Press, New York, 1975), p. 159; and J. H. Brewer and K. M. Crowe, "Advances in Muon Spin Rotation," Ann. Rev. Nucl. Part. Sci. 28 (1979) 239.
- [3] A recent series of reviews of positrons, Positrons in Solids, P. Hautojärvi (ed.), Topics in Current Physics (Springer-Verlag, 1979). In particular see R. N. West, Positron Studies of Lattice Defects in Metals, p. 89.
- [4] W. B. Gauster et al., Solid State Commun. 24 (1977) 619.
- [5] O. Hartmann et al., Phys. Rev. Letts. 44 (1980) 337.
- [6] R. M. J. Cotterill et al., Nature 239 (1972) 101.
- [7] C. H. Hodges, Phys. Rev. Letts. 25 (1970) 284.
- [8] C. H. Hodges, J. of Phys. F 4 (1974) L230.
- [9] R. M. Nieminen et al., Phys. Rev. B19 (1979) 1397 and references therein.
- [10] J. P. Gupta and R. W. Siegel, Phys. Rev. Lett. 39 (1977) 1212.
- [11] V. L. Sedov, V. A. Teimurazova, K. Berndt, Phys. Lett. A33 (1970) 319.
- [12] P. Hautojärvi et al., Phys. Rev. Lett. 44 (1980) 1326.
- [13] P. J. Schultz et al., Phys. Rev. Lett. 44 (1980) 1629.
- [14] K. G. Lynn and E. Lutz, Rev. Sci. Instru. 7 (1980) 977 and references therein.
- [15] Muon spin rotation, Proc. of the First International Topical Meeting on Muon Spin Rotation, F. N. Gyax, W. Kundig and P. F. Meir (eds.) (North-Holland, 1979).

- [16] Y. Tamazaki, *Hyp. Int.* 6 (1979) 115.
- [17] J. A. Brown et al., *Phys. Rev. Lett.* 43 (1979) 1513, 1899(E).
- [18] D. Herlach et al., *Hyp. Int.* 6 (1979) 323.
- [19] W. J. Kossler et al., *Phys. Rev. Lett.* 41 (1978) 1558.
- [20] This is a rapidly developing field. Therefore the reader is referred to other contributed papers from this conference. A recent published review is O. Hartmann, *Hyp. Int.* 6 (1979) 203.
- [21] T. R. Waite, *Phys. Rev.* 107 (1957) 463.
- [22] A. Seeger, *Appl. Phys.* 4 (1974) and references therein.
- [23] B. Bergerson et al., *Sol. State Commun.* 15 (1974) 1377.
- [24] A. Seeger in: *Hydrogen in Metals*, Vol. I, G. Alefeld and J. Volkl (eds.) Topics in Current Physics (Springer-Verlag, 1979), p. 348.
- [25] K. G. Lynn and H. Lutz, to be published *Phys. Rev. B*.
- [26] H. Graf et al., *Phys. Rev. Letts.* 44 (1980) 1333.
- [27] J. G. Petzinger et al., *Hyp. Int.* 6 (1979) 223.
- [28] A. T. Fiory, *Hyp. Int.* 6 (1979) 261.
- [29] T. McMullen and E. Zaremba, *Phys. Rev. B* 8 (1978) 3026.
- [30] O. Hartmann, *Phys. Rev. Lett.* 41 (1978) 1055.
- [31] V. G. Grebinnik et al., *Pisma Zh. Eksp. Teor. Fiz.* 25 (1977) 322.
- [32] K. G. Petzinger, *Phys. Letts.* 75A (1980) 225.
- [33] R. Kubo and T. Toyabe in *Magnetic Resonance and Relaxation*, R. Blink (ed.) (North-Holland, 1967) p. 810.
- [34] O. Hartmann, *Phys. Rev. Letts.* 39 (1977) 832 and M. Camani et al., *Phys. Rev. Letts.* 39 (1977) 836.
- [35] G. Straub, D. Parkin and K. G. Lynn, *J. Nucl. Mater.* 69 & 70 (1978) 613.
- [36] A. Alam et al., *Rad. Eff. Lett.* 50 (1979) 27 and references therein.
- [37] P. Hautojärvi et al., *Phil. Mag.* 35 (1977) 974.
- [38] K. G. Lynn et al., *Phys. Rev. B* 20 (1979) 3566.
- [39] A. Seeger, *Proc. 5th Int. Conf. on Positron Annihilation*, Japan, 771 (1979).
- [40] M. Doyama et al., *Proc. 5th Int. Conf. on Positron Annihilation*, Japan, 763 (1979).
- [41] W. B. Gauster et al., *J. Nucl. Mater.* 69 & 70 (1978) 147.
- [42] R. H. Heffner, private communication.
- [43] J. P. Bugeat et al., *Phys. Lett.* 58A (1976) 127.
- [44] K. Dorenburg et al., *Z. Phys.* B31 (1978) 165; D. Herlach et al., *Proc. 5th Int. Conf. on Positron Annihilation*, Japan, 767 (1979).
- [45] A. T. Fiory et al., unpublished data and W. J. Kossler, *Hyp. Int.* 6 (1979) 295.
- [46] R. N. West, private communication.
- [47] N. Echt et al., *Phys. Lett.* 67A (1978) 427.
- [48] L. Trieb and K. Siebinger, *Z. Metall.* 62 (1971) 682.
- [49] A. P. Mills, Jr., *Phys. Rev. Letts.* 41 (1978) 1828; K. G. Lynn, *J. Phys. C* 12 (1979) L435.
- [50] K. G. Lynn, *Phys. Rev. Lett.* 43 (1979) 391.
- [51] A recent review, *Interactions on Metal Surfaces*, R. Gomer (ed.) Topics in Applied Physics (Springer-Verlag, 1975) p. 64.
- [52] M. Leon and R. H. Heffner, *Hyp. Int.* 6 (1979) 301; M. Leon and R. H. Heffner, *Phys. Letts.* 69A (1978) 235.