μSR NEWSLETTER

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DEFINITION OF μSR

μSR stands for Muon Spin Relaxation, Rotation, Resonance, Research, or what have you. The intention of the mnemonic acronym is to draw attention to the analogy with NMR and ESR, the range of whose applications is well known. Any study of the interactions of the muon spin by virtue of the asymmetric decay is considered μSR, but this definition is not intended to exclude any peripherally related phenomena, especially if relevant to the use of the muon's magnetic moment as a delicate probe of matter.

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GIANT HYPERFINE ANOMALY

BETWEEN BOUND NEGATIVE MUON AND Rh NUCLEUS IN Pd METAL

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ABSTRACT

The Kinght shift of negative muon (μ^-Pd) in Pd metal has been determined to be -9.0 ± 0.7 % at T = 11 K, revealing an unusually large hyperfine anomaly between μ^-Pd and its equivalent isotope Rh<u>Pd</u>; $H_{hf}(\mu^-\mathrm{Pd})/H_{hf}(\mathrm{Rh}\underline{\mathrm{Pd}}) = 0.60 \pm 0.05$, or $\varepsilon_\mu - \varepsilon_N = -40 \pm 5$ %. Its implication is discussed in terms of the spatial distribution of the electron spin density in transition metals.

The hyperfine anomaly, the change of the hyperfine field between different nuclear states of the same isotope, reflects spatial distributions of both nuclear magnetization and electron spin density. This problem was first studied theoretically by Bohr and Weisskopf [1], and then, rigorously by Stroke <u>et al</u>. [2]. They considered the radial decrease of s-electron density that is probed by nuclei of finite size. Another possible cause of hyperfine anomaly is the change of $|\psi_e(0)|^2$

due to the change of the charge distribution of the probe nucleus (so called Rosenthal-Breit-Crawford-Schawlow correction [3], as discussed in Ref.2). A large amount of experimental data has been explained by taking into account the nuclear magnetization distributions consistent with the nuclear wavefunctions, but neither the nuclear structure, nor the mechanism of the hyperfine field becomes clear from these studies simply because the nucleus is too small to produce a large effect sensitive enough to discriminate among models. It would be dramatic, if we could find a much more extended magnetic probe to detect the electron spin density.

In this context, we paid special attention to the hyperfine field probed by bound muons [4]. Polarized negative muons that have stopped in a material immediately reach the ground state ($1s_{1/2}$) of the muonic atom and stay for a certain length of time τ_μ (τ_μ varies from 2.2 μsec in the lightest atoms to 80 nsec in heavy atoms). The average polarization is decreased to about 1/6 due to the spin-orbit coupling, but is still large enough to observe spin precession (μ^-SR). The density of the bound μ^- is given by $|\psi^\mu_{1s}(r)|^2$. The muon wavefunction $\psi^\mu_{1s}(r)$ is, for a point nucleus, $\exp(-Zr/a_\mu)$ with $a_\mu = \hbar^2/m_\mu e^2 = 260$ fm, but, for heavy nuclei, where a_μ becomes close to the nuclear radius, it is modified due to the finite extension of nuclear charge. In either case, the bound muon is distributed largely outside the nucleus. However, when viewed from atomic electrons, the bound muon is still concentrated around the nucleus so that it should behave like an impurity nucleus of apparent charge (Z-1)e. Compared to its equivalent nucleus of true nuclear charge (Z-1)e, the bound muon has the following unique properties: i) a peculiar charge distribution - the genuine

nuclear charge Ze is surrounded by negatively charged cloud of distribution $|\psi^{\mu}_{1s}(r)|^2$, and ii) in the case of even-even nuclei (we will only consider the case I=0), the magnetization is carried only by the muon spin which is distributed with the density $|\psi^{\mu}_{1s}(r)|^2$ and thus can be calculated exactly, while the nuclear magnetization is confined within the nuclear sphere. Therefore, we expect a large hyperfine anomaly which may hopefully tell us the mechanism of the hyperfine field, but such a physical observable has never been known to date.

By this new method, we attempted to study the core polarization phenomena in magnetic hyperfine fields. The origin of negative hyperfine fields in magnetic ions has been explained by Freeman and Watson [5] in terms of the induced polarization of inner-shell s-electrons. Having such an extended magnetic probe as μ^-, we hoped to examine this mechanism on a firm experimental basis. For this purpose, the case of μ^-Pd in pure Pd metal was chosen because its nuclear counterpart, namely the Rh Knight shift in Pd metal, was known to have a very large negative value from a PAC measurement of ^{100}Rh in Pd metal by Rao et al. [6], indicating that the core polarization as well as enhanced moment localization plays important roles. The relaxation time T_1 of ^{103}Rh as well as its Knight shift was measured in an NMR experiment by Narath and Weaver [7]. If no hyperfine anomaly is present, we expect $T_1(\mu^-\text{Pd}) = 16$ μsec at 4 K which is much longer than $\tau_\mu = 80$ nsec.

In a very early experiment, Ignatenko [8] observed no μ^- precession signal in Pd at room temperature. Later, in our previous experiment [9], we cooled down a very pure Pd metal to 4 K, but no precession signal was observed. This result led us to a measurement of the

circular polarization of muonic K x-rays [10], which showed that the muon polarization survives until the muon reaches the ground state. In the present experiment we challenged this problem again, and finally obtained precession signals, yielding a large μ^- hyperfine anomaly for the first time.

The experiment was carried out at the stopped muon channel (SMC) of LAMPF. High-purity Pd wires with impurity concentration below 5 ppm were stacked together into approximate dimensions of 5 x 2 x 2 cm and were cooled down to 11 K in a helium-flow cryostat. A Varian 12-inch magnet with a pole gap of 5.2 cm was used to achieve a good field homogeneity; the field was monitored by an NMR gaussmeter to check the long-term field drift. The "backward" μ^- beam (80 MeV/c) was collimated to 1.9 cm in diameter and was stopped in the sample. The average stopping rate was 2×10^4 μ^-/sec. Two sets of electron telescopes, both placed at forward (down stream) directions, recorded μ - e decay time spectra. The time spectra were taken by two independent TAC-ADC systems interfaced to a PDP-11 computer via a MBD-11 microprogrammed CAMAC branch driver.

Measurements were performed at 3.6 kOe and at 5.0 kOe external fields. Each time spectrum was assumed to take the form

$$N(t) = N_1 e^{-t/\tau_1} [1 + A\cdot\cos(\omega t + \phi)] + N_2 e^{-t/\tau_2} + N_3 , \qquad (1)$$

where the second term represents a long-lived background from surrounding materials and the last term is a constant background. The following analysis was performed on the background-corrected data: we fixed the phase ϕ to be a "physical" value corresponding to the counter geometry and evaluated the asymmetry A and the χ^2 value for each

frequency point. To find the precession frequency, we imposed the criterion that the asymmetry of the (candidate of the) precession signal should come out with a correct sign. We also demanded consistencies between the two electron telescopes, and between the data taken at two different fields. In Fig.1, we present the Fourier analysis of 5.0 kOe measurement. The external field at the sample position was calibrated by observing the μ^- precession in a graphite sample.

The amplitude of μ^-Pd precession was 1.73 ± 0.45 %. As shown in Fig. 1, the precession signal is statistically significant. This amplitude is much smaller than that of μ^-C, 5.5 ± 0.8 %, even after a correction is made for the natural abundance (22.2 %) of ^{105}Pd (muons bound to odd-A nuclei ($I \neq 0$) are further depolarized due to the hyperfine coupling and do not contribute to the precession signal). The reason for this small amplitude is not clear, as the relaxation time of μ^-Pd is expected to be 6 μsec at 11 K from the T_1T value for Rh<u>Pd</u>. One possible reason may be the following: Due to the preceding K x-ray of about 3 MeV, the μ^-Pd 1s state is formed with a recoil energy around 50 eV, which is large enough to repel the muonic atom from its original site. This situation is somewhat similar to the final stopping stages of recoil atoms after neutron capture or nuclear reactions. As known from hyperfine-interaction studies in these cases, some significant fraction of the recoil atoms is settled down at another regular site, but some other fraction is splitted into various possible locations, thus giving rise to no precession signal (loss of orientation), since the internal field felt by a recoil nucleus depends on its location in magnetic materials, similar to the case of eqQ for $I \geq 1$ probes. Thus, the small amplitude of μ^-Pd precession may be due to this type of loss

of polarization, but the present μ^-Pd precession signal is most likely to correspond to the μ^-Pd sitting as a substitute of a Pd atom in a Pd metal, just like the Rh atom in a Rh<u>Pd</u> alloy.

After correcting each data for the g factor of the bound μ^-, which is -0.07 % for C and -2.0 % for Pd less than the free-muon g factor (since the experimental values are not accurate enough [11], we assumed the theoretical values by Ford <u>et al</u>. [12]). We finally deduced the μ^-Pd Knight shift in Pd to be $K_\mu = -9.0 \pm 0.7$ % at T = 11 K. Compared with the Knight shift of Rh in Pd, K(Rh<u>Pd</u>) = -15.0 % at 11 K, this leads to a surprisingly large hyperfine anomaly,

$$\Delta = \varepsilon_\mu - \varepsilon_n = \frac{K(\mu^- Pd) - K(Rh\underline{Pd})}{K(Rh\underline{Pd})} = -40 \pm 5\ \% . \qquad (2)$$

In the following we will give a qualitative account of the present experiment.

The hyperfine anomaly defined with respect to the hypothetical point-nucleus value can be expressed by

$$\varepsilon = -\int_{probe} m(R) \frac{\Delta\rho(R)}{\Delta\rho(0)} d\tau_R , \qquad (3)$$

where m(R) is the normalized magnetization distribution of a probe (nucleus or muonic atom) and $\Delta\rho(R)$ is the electron spin density. If the spin density is produced by a $s_{1/2}$ electron as in a free atom, $\Delta\rho(R)$ is $|\psi_s(r)|^2$ in the non-relativistic expression, which takes the following form in the vicinity of the nucleus (neglecting the RBCS effect),

$$|\psi_s(r)|^2 = |\psi_s(0)|^2 \times \begin{cases} 1 - b_{in}\dfrac{r^2}{R_0^2}, \; b_{in} = \dfrac{ZR_0}{a_0} & \text{for } R < R_0, \quad (4a) \\[2ex] 1 + b_{in} - b_{out}\dfrac{r}{R_0}, \; b_{out} = \dfrac{2ZR_0}{a_0} & \text{for } R > R_0, \quad (4b) \end{cases}$$

where R_0 is the nuclear radius, and expressions for b_{in} and b_{out} are for the case of uniform nuclear charge distribution. In the relativistic expression, $\Delta\rho(R)$ is given by $\int_R^\infty FGdr$, and the coefficient ,

$$b_{in} = \int_0^{R_0} FG\, dr \Big/ \int_0^\infty FG\, dr \qquad (5)$$

can be calculated numerically, as given in Ref.2.

In the case of nuclear hyperfine anomalies the integration in Eq.(3) is confined within the nuclear sphere, while, in the present case, $m(R)$ distributes outside the nuclear sphere so that

$$\varepsilon = -b_{in}\int_0^{R_0} m(R)\,\frac{R^2}{R_0^2}\,d\tau_R - b_{out}\int_{R_0}^\infty m(R)\,\frac{R}{R_0}\,d\tau_R\,. \qquad (6)$$

In the case of μ^-Pd, $b_{in} = 0.72$ % and $b_{out} = 0.97$ %. We evaluated the integrals numerically using the $1s_{1/2}$ muonic wavefunction, and obtained $\varepsilon_\mu \simeq -4$ %. The nuclear hyperfine anomaly is around $\varepsilon_n \simeq -1$ %. The difference, $\Delta = \varepsilon_\mu - \varepsilon_n \simeq -3$ %, is much smaller than the observed value $\Delta = -40$ %. Then, we ask why such a large discrepancy could occur. A more realistic evaluation of the electron and muon wavefunctions taking into account the nuclear charge density cannot help this situation. Note that in Eq.(4) the radial gradient is $2Z/a_0$, independent of the principal quantum number. In this sense, there is no

way of removing this discrepancy, unless we assume a steeper gradient of $\Delta\rho(r)$.

In magnetic ions, where the s-d interaction induces polarization of inner s-shell electrons by the d-shell electrons of the atom according to Freeman and Watson [5], the electron spin density is due to a small difference of large $|\psi_s^{\uparrow}(r)|^2$ and $|\psi_s^{\downarrow}(r)|^2$, which behaves differently from $|\psi_s(r)|^2$ itself, as illustrated in Ref.5. The asymptotic form of $\Delta\rho(r)$ is not known, but let us assume that the gradient of $\Delta\rho(r)$ outside is $2Z/a_0^{CP}$ with a_0^{CP} being the effective Bohr radius. Then, ε is increased by a factor of a_0/a_0^{CP}. The present experiment seems to infer that $a_0^{CP} \simeq (1/10)a_0$. This is an entirely new type of information which can be obtained only by the negative muon as a probe outside the nucleus. Whether or not this interpretation is consistent with the core polarization theory is an open question. The present fact may also require some other mechanism for the hyperfine field.

The present problem raises also an interesting question: How well does a muonic atom $\mu^- Z$ behave like an impurity nucleus (Z-1)? In a strict sense, the muonic atom is a special "three body" system consisting of the nucleus, μ^- and electrons. Recently, Mallow et al. [13] have performed Dirac-Fock calculations for a variety of muonic atoms, by explicitly taking account of the muon in the self-consistent field theory. Their results show that higher states of muonic atoms are appreciably affected by the presence of atomic electrons and visa versa. It is an important, but still open question, to what extent the ground state muonic atom changes the electronic configuration and the local band structure.

In the case of light nuclei, where the nuclear finite-size effect as well as the relativistic effect is unimportant for both electron and muon, we can derive a simple expression

$$\varepsilon_\mu = -3 \frac{a_0}{a_0^{CP}} \frac{m_e}{m_\mu} = -1.5 \frac{a_0}{a_0^{CP}} \ \% , \qquad (7)$$

irrespective of Z. The nuclear hyperfine anomaly is, of course, negligibly small. Therefore, an experimental value of ε_μ will lead to a_0^{CP} in a straightforward way. We have already observed a large negative Knight shift for μ^-Si in a weak itinerant magnet MnSi. An experiment to obtain the Knight shift of its partner, Al as a substitute for Si in MnSi, is in progress.

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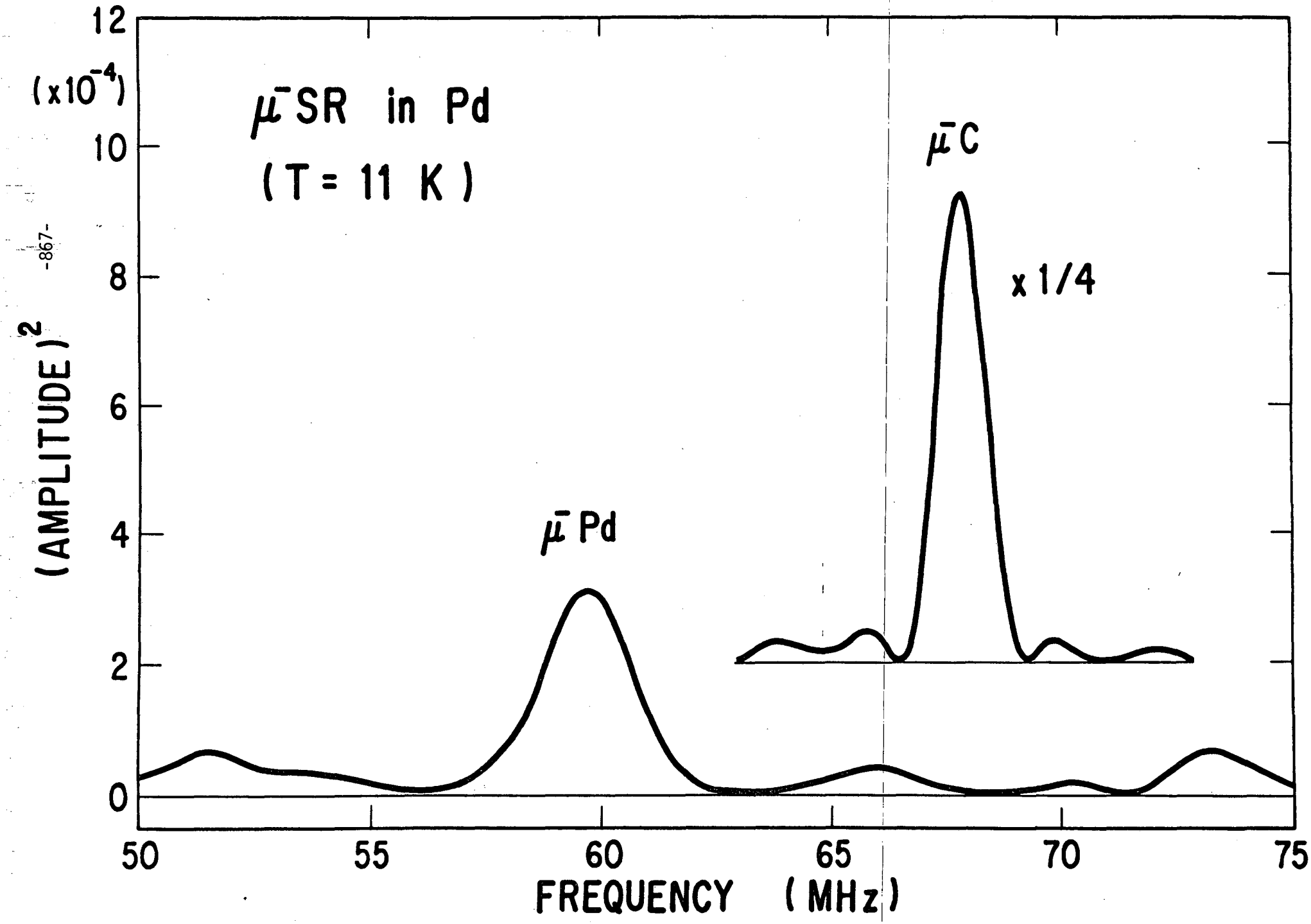
Figure caption

Fig. 1 Fourier power spectra, $|A|^2$ vs f of the μ^-Pd (at 11 K) and μ^-C precessions at H_0 = 5.0 kOe. The statistical fluctuations of $|A|^2$ is 0.2×10^{-4} for μ^-Pd and 0.6×10^{-4} for μ^-C.


μ⁻SR in Pd
(T = 11 K)
μ⁻C
x 1/4
μ⁻Pd
(AMPLITUDE)²
(x10⁻⁴)
12
10
8
6
4
2
0
50
55
60
65
70
75
FREQUENCY (MHz)

Test of Macroscopic Muon Diffusion in Aluminum.

S.S. ROSENBLUM, C.W. CLAWSON, K.M. CROWE, Lawrence Berkeley Lab*, J.H. BREWER, TRIUMF, and B.D. PATTERSON, Simon Fraser Univ.--There has been much recent interest in the diffusion and trapping behavior of positive muons in pure metals. In several cases the muon depolarization rate is found to exhibit sharp minima as a function of temperature. Aluminum is anomalous in that there is practically no depolarization down to 2K. In order to determine if this was due to macroscopic muon diffusion, a high purity Al foil, 0.25 mm thick, was cathodically etched in vacuum to remove oxides and then sputtered with 0.3 μm of copper[1]. A stack of 30 of these foils was used as a stopping target for a thermalized muon beam. At 4.2K the on-line data analysis showed no difference in depolarization rate between this target and a high purity polycrystalline Al target. Since the muon shows a large depolarization in copper at this temperature, this shows that the muon is not diffusing over distances as large as ~25 μm. Results of a more thorough analysis as well as results on high purity aluminum powder will also be presented.

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MUON HYPERFINE FIELDS IN Fe(Al) ALLOYS

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Abstract

The hyperfine field on the muon, B_{hf}, at interstitial sites in dilute Fe(Al) alloys has been measured for four different concentrations of Al and as a function of temperature by the muon spin rotation method. The magnitude of B_{hf}, which is negative, decreases at rates ranging from 0.09 ± 0.04% per at.% Al at 200 K to an asymptotic limit of 0.35 ± 0.03 above 440 K. This behavior shows that sites near the Al impurity are weakly repulsive to the muon, with an interaction potential of 13 ± 3 meV. In order to fit the temperature dependence of the hyperfine field, it is necessary to hypothesize the existence of a small concentration of unidentified defects, possibly dislocations, which are attractive to the muon. Although the Al impurity acts as a non-magnetic hole in the Fe lattice, the observed decrease in B_{hf} is only 35% of the decrease in the bulk magnetization. We conclude that B_{hf} is determined mainly by the enhanced screening of conduction electrons in Fe and Fe(Al). Since the influence of the Al impurity on the neighboring Fe moments is very small, most of the change in B_{hf} is therefore attributed to the increase in conduction electron polarization at the Al impurity. [PACS 75.50.Bb].

1. Introduction

Knowledge of the local electronic structure around the isotopes of hydrogen, when present as impurities in metals and alloys, is fundamental to a basic understanding of the diffusion of these particles and their poorly-understood interaction with defects and impurities, particularly in Fe. Measurements of the Fermi contact or hyperfine field on positive muons can help provide this information [1-3]. We have studied Fe(Al) alloys with muon spin rotation methods (μSR) to help formulate a physical picture of the local magnetic field and how the host electrons participate in screening the muon's positive charge. Information is also obtained on the mutual interaction between the positive muon and the Al impurities in the Fe host.

The muon probes the magnetic fields at interstitial sites, so that it is specifically sensitive to the extent to which electron states in metals are delocalized, or band-like. As an example of how this is accomplished, we refer to the recent work of Hayano et al. who have compared μSR and host NMR measurements in a study of the itinerant magnetism of the helimagnet MnSi [4]. General applicability of this aspect of such measurements is of course contingent on being able to correctly account for the perturbation which the muon creates in the metal. The problem appears to be tractable and several theoretical approaches have been advanced [1-3,5]. The pure elemental ferromagnets Ni [6-10], Fe [6,7,9-12], Co [7,9,13,14], Gd [15,16], and Dy [17] have been among the metals studied with muons.

The basic properties of the muon in metals may be derived from the assumption that the s-like conduction band electrons are mainly responsible for screening the muon's charge. This interpretation has been proposed for the case of Ni [1,2] assuming d-like states contribute very weakly to the screening at the muon site, even though there is a high density of d states at the Fermi level. However, the s-like bands of Ni have little or no polarization, and the muon therefore experiences a negative hyperfine field which is dominated by the tails of the minority-spin d-like wavefunctions in the

interstitial region. The theoretical basis for this model was discussed by Petzinger and Munjal [1] and by Petzinger [2]. This model takes the opposite view to what had been previously supposed for hydrogen in transition metals [18]. Patterson and Keller [5] recently carried out a finite cluster calculation which lends additional support to this model, in that the d-electrons remain on the neighboring Ni atoms in the cluster.

The above model is thought to be applicable to Fe in spite of the more complicated band structure, where more significant s-d hybridization is found [19]. Jena has discussed the non-linear screening of the muon in Fe in terms of a free-electron gas [3], where the ambient spin density is increased by a factor of 9.8. The measured hyperfine field on the muon is -11 kG, so that on applying this model one finds that the ambient polarization density is effectively $-0.014\,\mu_B\text{Å}^{-3}$. By comparison, neutron diffraction measurements [20] give a value of $-0.014 \pm 0.004\,\mu_B\text{Å}^{-3}$, averaged over a 0.5 Å cube centered at the tetrahedral interstitial site where the muon is presumed to reside [21]. The neutron data also show that there is a delocalized background polarization of $-0.21\,\mu_B$ per atom, which is equivalent to a homogeneous polarization density of $-0.018\,\mu_B\text{Å}^{-3}$ [18]. This indicates that the muon hyperfine field is obtained following two quite different assumptions, that the muon experiences either the average conduction electron polarization or the local polarization at a particular site. Band structure calculations have found that the 4s electron contribution to the polarization is between -0.04 and $-0.07\,\mu_B$ per atom [19]. This does not include the contribution from itinerant d states, which is apparently larger. Thus, our view is that previous work indicates that itinerant states in Fe might be treated as a free electron gas in screening the muon.

Alloys of various non-transition elements in Fe have been studied by bulk magnetization measurements [22,23], neutron diffraction [24,25], NMR [26-29] and the Mössbauer effect [29-31]. These measurements found that the Al impurity produces simple magnetic dilution. Neutron diffraction studies have confirmed that the Al is

non-magnetic and that the surrounding Fe neighbors exhibit very little perturbation on their moments. The NMR and Mössbauer satellite lines have been associated with Al in various neighbor shells around the Fe. Grüner et al. [29] have shown that these results can be explained by the spin-polarized conduction electron cloud around the Al. More recently, Terakura has proposed a theoretical explanation based on an ab initio calculation of the electronic densities and polarizations at the Al site [32]. He found a net increase in the s-p polarization at the Al site. His predictions for Si impurities are quite similar to those for Al.

This paper is an elaboration and an extension of our previous studies of Fe(Al) with muons [33]. Additional studies of ferromagnetic alloys were recently reported by Kossler et al. [34] for Ni(Co) and Ni(Cu) and by Nishida et al. [35] for Ni(Cr), Fe(Si) and Fe(Ti).

2. Experiment

μSR is a perturbed angular distribution technique and it has been discussed extensively in the literature [6,36,37]. Spin-polarized positive muons are implanted into the sample under investigation and the time intervals for individual positron decays are recorded. The anisotropy of the positron decay and the precession of the muon spin in the local magnetic field give rise to an oscillatory component in the time dependence of the measured positron emission rate in a given direction. The present studies were done at zero external magnetic field on the samples and used positron detectors at 0° and 180° with respect to the μ^+ beam. The time distribution of the positrons is given by the following formula:

$$N_{e^+}(t) = N_o \exp(-t/\tau_\mu)\,[1 + a + b\exp(-\lambda t)\cos(\omega t + \phi)] + N_{back}. \qquad (1)$$

N_o is an overall normalization, τ_μ the muon lifetime of 2.2 μsec, λ the depolarization

rate, ω the spin precession angular frequency and ϕ a geometrical phase, essentially the angular coordinate of the positron detector. The values of a and b depend upon the distribution of the orientations of the local fields in the sample and upon the angle subtended by the detector [37]. Taking the case of an unmagnetized ferromagnetic sample, where the local fields are isotropically distributed, and longitudinal placement of the detector, we have $a = P/3$ and $b = 2P/3$, where P is a function of the polarization of the muon beam and the energy dependence of the positron detection efficiency [37]. For our experiment $P \approx 0.1$. The term N_{back} is included to account for accidental background counts in the data.

The experiments were performed at temperatures ranging from 80 to 433 K. The data were fitted with Eq.(1) using a multi-parameter least-squares fitting routine. We did not attempt to find the term proportional to a, but rather include it into the definition of N_o and b in fitting the data. The field on the muon is given by $B_\mu = \omega \gamma_\mu^{-1}$, where $\gamma_\mu = 8.51 \times 10^4 \mathrm{sec}^{-1} \mathrm{G}^{-1}$.

Four spherical samples were fabricated, one of 99.99%-pure Fe, the others containing 1.85, 4.29 and 5.53 at.% Al. The sample materials were melted in a MgO crucible in a He atmosphere by rf induction. The Al was added by including Fe(Al) alloy in the melt. The melt was poured into an Al_2O_3-coated Fe mold and allowed to cool in the furnace. The castings were 6 cm in diameter and 20 cm in length. Each casting was reheated to 1273 K and hot pressed, reducing the length by a factor of 2. This is a standard procedure for removing most of the casting structure in Fe and its alloys. The castings were then machined to 5.715 ± 0.001-cm-diameter spheres and annealed in H_2 at 1088 K for one hour. At the concentrations used here, the Al is in a random solid solution [38]. The results of a chemical analysis on the samples is given in Table I. After the μSR runs, the spheres were sectioned for analysis. The sizes of the macroscopic crystallites vary between 0.1 and several mm. The 4.29%-Al sample contains, in addition, dispersed Al_2O_3 precipitates on the order of 1 μm across, which

accounts for the high oxygen concentration in the analysis (There is a thermite reaction between aluminum and oxygen). From transmission electron microscopy, the dislocation density is on the order of $10^8 cm^{-2}$ and the subgrain cells are about 1 μm accross.

3. Data Analysis

3.1 Extraction of the Hyperfine Field

The results for the magnetic fields on the muon and the depolarization rates are listed in Table II. Values of B_μ given in brackets were obtained by linearly interpolating the temperature dependence of the results on pure Fe given by Nishida et al. [11]. The hyperfine fields were extracted in the following manner.

The field experienced by the muon may be decomposed [39] into

$$B_\mu = B_{ext} - B_{DM} - B_L + B_{dip} + B_{hf}, \tag{2}$$

where B_{ext} is the applied external field, B_{DM} is the demagnetizing field which depends upon sample geometry, B_L is the Lorentz cavity field $4\pi M/3$ where M is the domain magnetization, B_{dip} is the sum of the dipole fields inside the Lorentz sphere, and B_{hf} is the Fermi contact or hyperfine field. Because we performed the experiment with zero external field, and since the μ^+ particles hop rapidly from site to site, averaging B_{dip} to zero, the expression for B_{hf} reduces to

$$B_{hf} = B_\mu + B_L. \tag{3}$$

The temperature dependence of the Lorentz cavity field was computed following the parameterization of magnetization data for pure Fe given by Redi [40]:

$$M = M_o f(T) = M_o[1 - A(T/T_\partial)^{3/2} - B(T/T_\partial)^{7/2}], \tag{4}$$

where $A = 0.102 \pm 0.005$, $B = 0.33 \pm 0.07$, and $M_o = 1749\,\mathrm{G}$. We have assumed that the temperature dependence scales with the Curie temperature of the alloy, given as [41]

$$T_c = T_{co}(1 - 0.1\,c), \tag{5}$$

where $T_{co} = 1044\,\mathrm{K}$ is the Curie temperature of pure Fe and c is the Al concentration. The magnetization is also corrected for the presence of Al in the sample by summing the effects of simple dilution, i.e., replacing Fe atoms with Al atoms and taking into account the known change in lattice spacing [42]. The change in lattice spacing Δa_o gives a density factor $3\Delta a_o/a_o$, which becomes $0.157\,c$. Combining these factors yields

$$B_L = \frac{4\pi M}{3}(1 - 1.157\,c), \tag{6}$$

with M given by Eq.(4).

The resulting values of B_{hf} are listed in Table II. In calculating B_L for sphere No.4 (5.53%), we took into account the Si impurity in the sample. The data and fit for B_{hf} at 301 K as a function of concentration are plotted in Fig. 1. The change in B_{hf} is $-0.23 \pm 0.01\%$ per at.% impurity at 301 K.

The reduced quantity $B_{hf}/c\,B_{hf}$ for $c = 0.0429$ is plotted as a function of temperature in Fig.2. Muon precession oscillations were observed only at 200 K and above. It is seen that the fractional change in hyperfine field is temperature dependent.

3.2 Temperature Dependence of B_{hf}

The temperature dependence of the change in B_{hf} with Al concentration (Fig.2) can be explained if the diffusing muons do not randomly sample the interstitial sites in the alloy. In general, the average hyperfine field is obtained by summing local contributions from all available sites, weighted according to Boltzmann population factors;

$$B_{hf} = \sum_i B_{hf}{}^i \exp(-\beta E^i) \,/\, Z, \tag{7}$$

where $\beta = (kT)^{-1}$, E^i the state energies of the muon at sites i, B_{hf} the local hyperfine fields at sites i, and Z the thermodynamic partition function.

In order to give this effect a quantitative treatment, we consider the following model. Firstly, we assume that the E^i vary significantly only at sites immediately adjacent to impurities and defects. Secondly, we assume that the muon energy is changed by an amount E' for a fraction c' of sites which are near the Al impurity. Defects are taken into account by including a fraction c'' of sites with energy E'' relative to the unperturbed sites. Since there are 24 tetrahedral interstitial sites around each Al atom, as opposed to the 6 tetrahedral sites per Fe atom, we take $c' = 4c$, for consistency. If we write the field near the Al sites as B_{hf}', the field at the defect sites B_{hf}'', and the field elsewhere as $B_{hf}{}^o$, then the average hyperfine field is given by

$$\Delta B_{hf}/B_{hf}{}^o = [f' c' \exp(-\beta E') + f'' c'' \exp(-\beta E'')]$$

$$/[1 - c' - c'' + c' \exp(-\beta E') + c'' \exp(-\beta E'')], \tag{8}$$

where we have used the reduced parameters

$$f' = 1 - B_{hf}'/ B_{hf}{}^o$$

and

$$f'' = 1 - B_{hf}''/ B_{hf}{}^o.$$

The data of Fig.2 were fitted with Eq.(8) by treating f', f'', c'', E', and E'' as adjustable parameters. The results are listed in Table III.

We find that the rapid dependence of $\Delta B_{hf}/c\, B_{hf}$ near 250 K, followed by a comparatively gentle dependence at higher temperatures, can therefore easily be explained in terms of a small concentration of strongly attractive defect sites and weakly

repulsive Al sites. The reduced χ^2, for 3 degrees of freedom, is 0.54 for the fit given in Table III. For comparison, we also tested an alternative fit to the data by omitting the defect term, *i.e.*, with $c'' = 0$. The asymptotic value of $\Delta B_{hf}/c\, B_{hf}$ turns out to be 40 % larger and E' a factor of 2 larger. However, the reduced χ^2, for 6 degrees of freedom, increases to 10. The statistical probability that the defect term gives a better fit is therefore 0.99.

The asymptotic value of $\Delta B_{hf}/c\, B_{hf}$ is -0.35 ± 0.03 at high temperatures, and we take this to be the average hyperfine field shift for random sampling by the muon. The hyperfine field on the muon at sites near Al is weaker by $8 \pm 1\%$ with respect to pure Fe. The three parameters characterizing the defect sites might be determined better if we had more data in the low-temperature region. We therefore regard these values as approximate. From the small value of the defect concentration found in the fit, $c'' \approx 10^{-9}$, it seems that dislocations and subgrain boundaries would be the likely explanation. Our result does, however, indicate that the shift in the hyperfine field with impurity doping can be developed as a technique for studying the muon-impurity interaction. It could be complementary to depolarization measurements which study the motional narrowing effect of muon motion [9-12]. These points are discussed further in the following sections.

3.3 Depolarization Rates

The μSR signal is observable in Fe and Fe(Al) when the rapid motion of the muon nearly averages out the dipolar field. From the depolarization rate the correlation time of the local dipolar field on the muon can be calculated, and this is nearly the same as the mean time of stay at an interstitial site. The second moment of the dipolar field distribution is about $\overline{(\Delta B_{dip})^2} = (2.6\,\mathrm{kG})^2$, owing to the existence of two magnetically inequivalent tetrahedral sites in pure Fe [10]. The value is nearly the same for the Fe(Al) alloy, although the spatial distribution differs. The depolarization

rate measured at 301 K for our samples is about a factor of 10 larger than those measured in high-purity Fe [12] and does not appear to be sensitive to the Al concentration. Since samples of Fe of nominally lesser purity typically show larger depolarization rates, this effect has been attributed to longer mean times of stay for muons at sites near or at defects. We could assume, therefore, that the diffusing muons spend part of their lifetime sampling some kind of defect sites throughout the temperature range covered. Clues as to the nature of these defects are obtained by comparing the hyperfine field shift and depolarization data.

The depolarization measurements for the 4.29 % alloy given in Table II show a monotonic decrease with temperature, with a tendency towards saturation at high temperatures. Thus it appears that the mean time of stay decreases steadily with temperature and that there is a background contribution to the depolarization rate. We consider expressing these data as follows:

$$\lambda = \gamma_\mu^2 \overline{(\Delta B_{dip})^2} f^2(T)\, \tau_c + \gamma_\mu\, \Delta B\, f(T). \tag{9}$$

The first term represents the motionally-narrowed local dipolar inhomogeneity and the second term the macroscopic magnetization inhomogeneities. τ_c is a correlation time, which we assume has an Arrhenius temperature dependence;

$$\tau_c = \tau_o \exp(U/kT). \tag{10}$$

The function $f(T)$ is the same as that given in Eq.(4) and it is included in Eq.(9) in order to correct for the temperature dependence of the dipolar fields. The data do not show any evidence for diffusion limited capture by deep traps (no de-trapping), which would have a τ_c^{-1} dependence [43,12]. A trapping term had been considered by Kossler et al.[43] in their interpretation of the non-Arrhenius temperature dependence of the depolarization rate in Cr and by Nishida et al.[12] in a study of Fe.

The results of a non-linear least-squares fit of Eqs.(9) and (10) to the depolarization data yields the Arrhenius plot of the correlation time given in Fig.3. The parameters of the fit are $\Delta B = 8.8 \pm 0.2\,\mathrm{G}$, $\tau_o = 0.3\,\mathrm{ps}$, and $U = 0.11 \pm 0.02\,\mathrm{eV}$. The statistical uncertainty in τ_o is about a factor of 10.

We note that the activation energy U for the muon jump processes is about a factor of 4 smaller than the magnitude of the defect interaction energy E'' (see Table III) found in the fit of the hyperfine field shift. This difference needs explanation. One consistency check concerns whether the muon jump rate is high enough for the muons to reach the defects. A fair test can be made at $T = 250\,\mathrm{K}$, where the change in the hyperfine field with temperature is most rapid. At this temperature $c''\exp(-\beta E'') \approx 1$, with muons spending about one-half their time at the defect sites, according to our previous analysis. It can be readily estimated that over the mean duration of the measurement, which is $\lambda^{-1} \approx 0.3\,\mu\mathrm{sec}$ at 250 K, the muon executes $N \approx \lambda^{-1}\tau^{-1}$ jumps, where τ is the jump time. A possible value for τ is 5 ps, obtained by Nishida et al. [12] for high-purity Fe at 250 K, with the result $N \approx 6\times10^4$. Equilibrium sampling of defects would be approached when $N c'' \approx 1$. On the contrary, we find $N c'' < 10^{-4}$. To resolve the apparent discrepancy we propose the following: (a) The defects are dislocations and subgrain boundaries, whose effective site concentration is very small; (b) E'' is some average muon-dislocation binding energy; (c) the muons are mobile along the dislocations, with an activation energy of migration U; and (d) the muon mean time of stay at unperturbed interstitial sites in Fe is probably much smaller than 5 ps at 250 K.

4. Discussion

A simple view of the effect of dilute Al impurities is first that the Al is substitutional in Fe and the contribution from the d-states of the removed atom can be simply subtracted. Invoking the observation that the moments of the Fe atoms are nearly unperturbed, we then assume that the d-electron contribution to the average hyperfine field on the muon is simply decreased by 1% per atomic per cent Al. Further support for this assumption comes from the fact that the magnetic disturbance has only a weak dependence on the valence of the impurity atom [24,25]. There is a similarly small perturbation on the neighboring Fe moments for Si impurities, which have a valence difference of 3 with respect to Fe [25]. The valence difference is 2 for Al. Terakura [32] has discussed the d-band filling and emptying effects for non-transition element impurities in Fe and concludes that these nearly cancel each other. Simple dilution of the d-electron contribution to the hyperfine field on the muon, if that were the main contribution, would lead to $\Delta B_{hf}/c\, B_{hf}$ of -1.0 which is three times larger than the data indicate. The apparent disagreement can be resolved with an explanation in terms of the contribution from conduction electrons.

NMR and Mössbauer measurements have found that the hyperfine field on Fe nuclei at sites nearest neighbor to Al impurities is less negative by 7% and this has been attributed to a small reduction in the local, negative conduction electron polarization [29]. At the Al site, the conduction electron polarization is negative due to the negative hyperfine field on the Al [40]. From a different point of view, Stearns has treated the pick-up of conduction electron polarization at impurities from the neighboring Fe atoms in terms of the volume misfit of the impurity atom [44,45]. This effect is small for the Al impurity and can be neglected. Referring to the theoretical results calculated by Terakura [32], the fractional change in the s-electron polarization within the Wigner-Seitz sphere at the Al is $+0.08$ and the p-polarization change is -0.03, for a total of $+0.05$. These findings can be used to make a prediction for the

change in the average B_{hf} for muons per unit concentration of Al impurity, taking into account the free electron spin density enhancement factor of 9.8:

$$\Delta B_{hf}/c = 9.8 \frac{8\pi}{3} [0.05 \times 2\mu_B/a_o^3] = 3.3\,\mathrm{kG}. \tag{11}$$

This is equivalent to $\Delta B_{hf}/c\, B_{hf} = -0.30$ and is close to the measured asymptotic value of -0.35 ± 0.03.

The average change in the hyperfine field at the Fe sites in Fe(Al) has been given by several authors [46,47]. The fractional change is in the range -0.4 to -0.5.

Nishida et al. [35] have recently measured B_{hf} for Fe doped with 5% Si at 300K. They find $B_{hf}/c\, B_{hf} = -0.24$. This is the same as the value we find for our 301 K measurements on Fe(Al). Although the asymptotic limit was not checked in the Fe(Si) measurement, this result suggests an insensitivity to the valence difference between Al and Si [30]. The magnetic properties of Fe(Al) and Fe(Si) are also similar [27].

Our explanation for the change in the hyperfine field on muons assumes that distortions in the localized d-wavefunction amplitudes at interstitial sites near the Al impurity can be neglected. This is based on the assumption that local changes in electron spin density associated with these states are not enhanced when the muon is present, as is the case for itinerant states. Thus we are assuming that the μ^+ impurity acts in a manner similar to Al and Si impurities by creating minimal magnetic disturbance in their vicinity. Nevertheless, we cannot completely rule out the possibility that the weak dependence of B_{hf} on Al concentration may arise in part from a large increase in the minority-spin d-like wavefunctions at interstitial sites near the Al. It would be interesting, therefore, to investigate the systematic behavior of B_{hf} for other non-magnetic impurities in Fe as well.

Owing to the apparently extreme sensitivity of B_{hf} to attractive defect potentials, it would be interesting to investigate the systematic effects of dislocations and

impurities such as C, O, and N in Fe alloy samples. Our interpretation predicts a logarithmic dependence of the temperature of inflection in Fig. 2 on the defect concentration. This could be tested with deformed alloys.

Our basic conclusion is that one obtains consistency by treating both the local magnetic perturbation around Al impurities [29] and the hyperfine field on the muon [3] as problems of conduction electron screening and spin density perturbation. It is hoped that these measurements will motivate more fundamental calculations for determining the origins of the muon hyperfine field and the local muon potential, such as finite cluster calculations treating the ternary system μ^+-Al in Fe.

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TABLE I. Properties of the Fe(Al) spheres used in this study. The impurity concentrations measured by atomic absorption analysis for Al, and mass spectrographic analysis for the other elements, are given in atomic per cent under the element headings. Vacuum fusion analysis of sample No.3 gave C - 0.013%, O - 0.3%, and N - 0.01%. He and H were below the limit of detectability, 0.0001% and 0.0005% respectively.

No.	Mass (g)	Al	Si	Ni	Zr	Ti	Mg	Cu
1	766.91	0.001	0.004	0.005	0.003	0.0005	0.0005	0.0002
2	760.84	1.85	0.04	0.005	0.003	0.0005	0.0005	0.0002
3	743.22	4.29	0.008	0.005	0.003	0.0005	0.0005	0.0002
4	739.88	5.53	0.4	0.005	0.003	0.0005	0.0005	0.0002

TABLE II. Observed magnetic fields on the muon B_μ, muon-spin depolarization rates λ, calculated Lorentz cavity fields B_L, and resultant Fermi-contact hyperfine fields B_{hf}. Errors of measurement are enclosed in parenthesis. Values of B_μ enclosed in brackets were interpolated from the data of Nishida et al. [11]. c is the Al impurity concentration in atomic per cent.

c	T (K)	B_μ(G)	λ (μ sec^{-1})	B_L(G)	B_{hf}(G)
0	200	[−3672(2)]		7256	−10,928
	240	[−3638(2)]		7230	−10,868
	260	[−3621(2)]		7215	−10,836
	280	[−3604(2)]		7198	−10,802
	301	[−3590(1)]		7179	−10,769
	301	−3592(3)	3.0(3)	7179	−10,771
	343	[−3530(4)]		7136	−10,666
	373	−3477(2)	2.2(2)	7101	−10,578
	433	−3379(3)	1.5(1)	7016	−10,394
1.85	301	−3687(2)	1.4(2)	7024	−10,711
4.29	200	−3990(14)	9(2)	6896	−10,886
	240	−3954(5)	3.8(4)	6870	−10,824
	260	−3902(2)	2.9(2)	6856	−10,758
	280	−3866(2)	2.0(1)	6840	−10,706
	301	−3842(2)	1.7(3)	6822	−10,664
	343	−3772(1)	1.5(1)	6781	−10,553
	373	−3719(1)	1.0(1)	6746	−10,466
	433	−3612(1)	1.0(1)	6665	−10,277
5.53	301	−3930(2)	1.3(2)	6685	−10,615

TABLE III. Parameters fitting Eq.(8) to the temperature dependence of the hyperfine field in the 4.29% Fe(Al) alloy. f' and f'' are the fractional changes in the hyperfine field, c' and c'' the site concentrations, E' and E'' the site energies, for Al and defect sites, respectively. The parameter $c' = 4c$ was held constant.

f'	$-0.087(7)$
f''	$-0.0032(8)$
c'	0.172
c''	$10^{-11.7\pm3.6}$
E'	0.013(3) eV
E''	$-0.6(2)$ eV

Figure Captions

Figure 1. The Fermi-contact hyperfine field on the positive muon in Fe(Al) as a function of impurity concentration at a temperature of 301 K. The line is a least-squares fit.

Figure 1. Temperature dependence of the fractional change in the hyperfine field with aluminum concentration. Data were taken for $c = 0.0429$. The curve is the fitted function Eq.(8) divided by c, using the parameters of Table III.

Figure 3. Dependence of the local field correlation time upon inverse temperature, obtained from depolarization rates according to Eqs.(9) and (10). The curve is the fitted function.

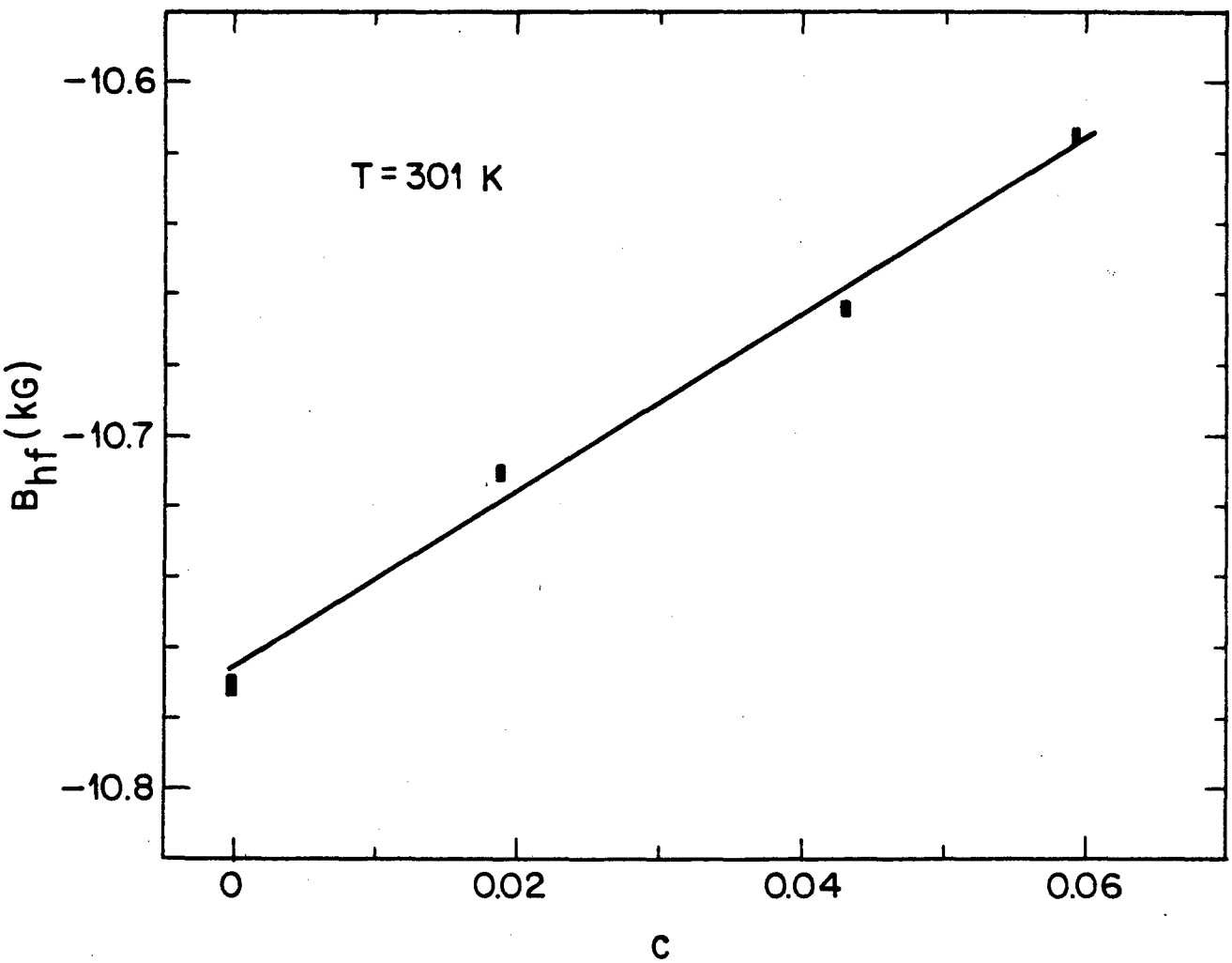

FIGURE 1

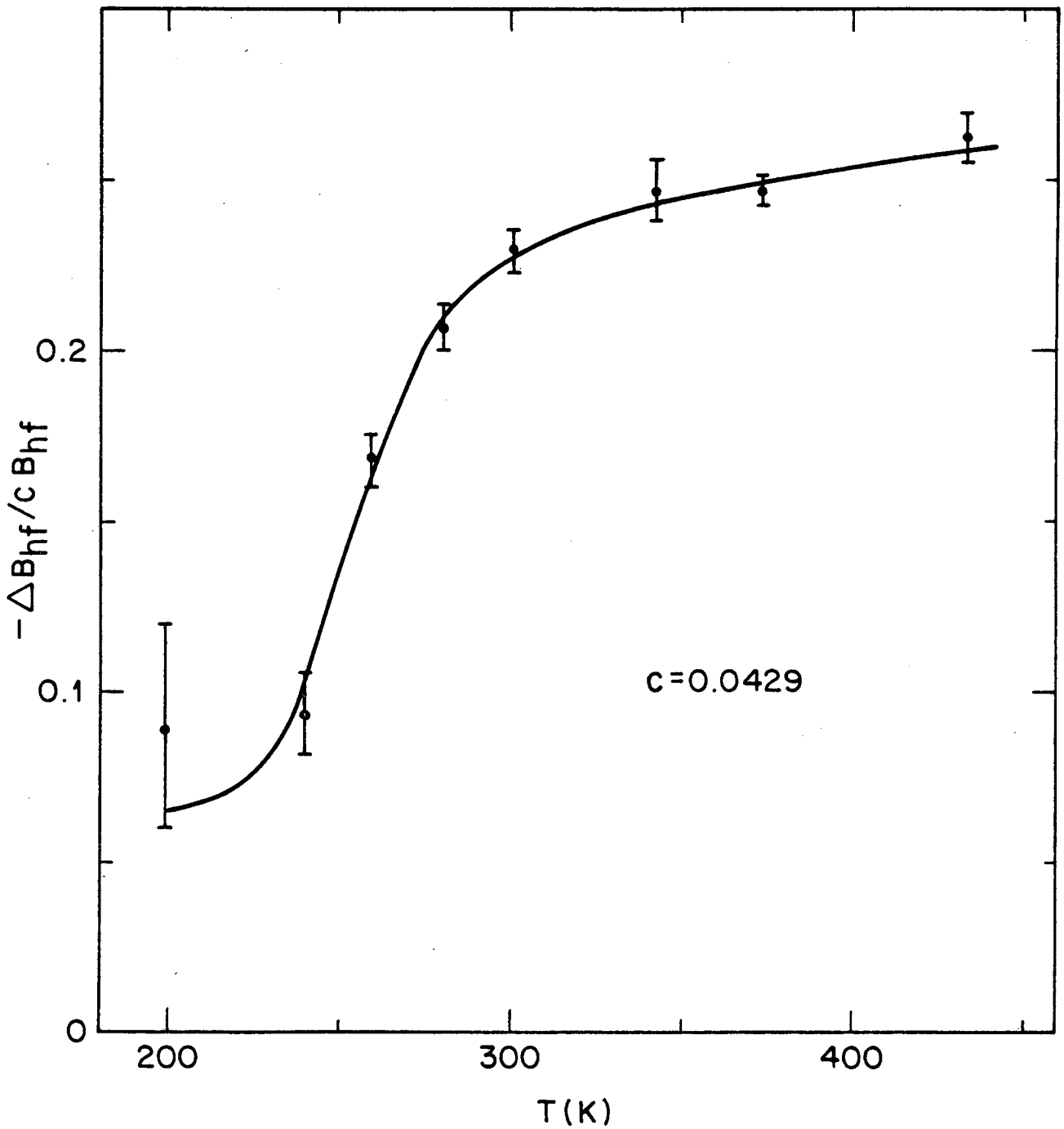

FIGURE 2

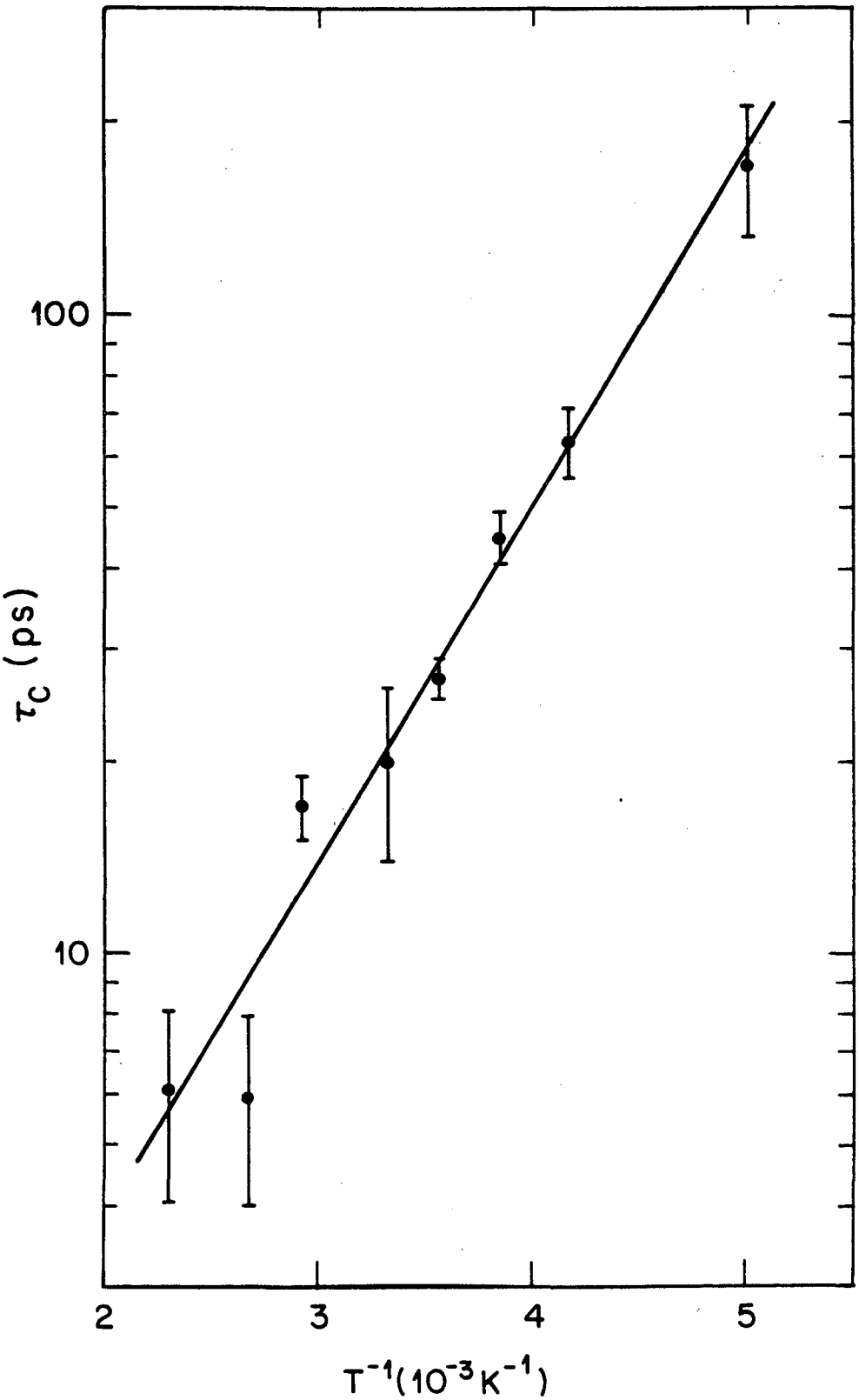

FIGURE 3

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