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Hyperfine Field and Diffusion of μ^+ in Fe Single Crystal*

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Abstract

Positive-muon spin rotation experiments have been performed in an Fe single crystal from 4.2°K to 300°K at the new meson facility TRIUMF. No signal was observed below 23°K. The μ^+ diffusion constants deduced from the relaxation times can be fitted to an Arrhenius law (activation energy 17 meV) above 70°K, but deviate from this at lower temperatures, indicating quantum diffusion. The μ^+ hyperfine field was found to have a temperature dependence slightly different from that of the saturation magnetization.

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In the last few years, the positive muon (μ^+) has been used extensively to probe phenomena related to polarized conduction electrons at interstitial sites in magnetic metals. The magnitude and width of the local magnetic field can be obtained from the precession and relaxation of the μ^+ spin via the time evolution of the asymmetric distribution of decay positrons (μSR method). Interstitial electron spin densities have thus far been studied in several ferromagnetic materials, in particular Ni[1,2] and Fe.[3,4]

A recent experiment showed an anomalous temperature dependence of the μ^+ hyperfine field in a Ni single crystal,[2] which enhanced our curiosity about the analogous phenomenon in Fe. However, a serious difficulty was anticipated: in bcc Fe various defects and/or vacancies were expected to suppress rapid diffusion of the μ^+, causing fast depolarization due to the inequivalent atomic dipolar fields at different interstitial sites or to inhomogeneous fields around the trapping center. In fact, previous experiments in both polycrystalline[3] and single crystal[4] Fe samples were unable to detect any signal below 150 K. We therefore carried out μ^+SR experiments on a carefully prepared single crystal of Fe, to study the temperature dependence of the hyperfine fields and the diffusion time.

This Letter represents the first μSR results from the new 'meson factory' TRIUMF; the experiments ran on the M20 muon channel using a 500-MeV proton beam of about 1 μA on a 10-cm Be production target. About 10^4 μ^+/sec with around 60% polarization were stopped in the Fe target, which consisted of five sheets of 50 mm × 48 mm × 1.1 mm single crystal plates cut from one large sheet grown by a strain-annealing

method.[5] The raw material was electrolytic Fe of high-purity grade; chemical analysis of an as-grown crystal showed that the largest impurity was 0.005 wt% of Ni. Linear defects were found to be less than 10^7 cm/cc. The target sheets were placed in the beam with the (011) plane perpendicular to the initial μ^+ spin direction, and decay positrons were detected at zero degrees in zero external field. The μ-e decay curve has the form

$$N(t) = N_0 \exp(-t/\tau_\mu)[1 - A\cdot G(t)\cos(\gamma_\mu B_\mu t)] + BG, \qquad (1)$$

where $\tau_\mu = 2.20$ μsec is the muon lifetime, B_μ is the local field felt by the μ^+, $\gamma_\mu/2\pi = 13.554$ kHz/G, A is the initial decay asymmetry, G(t) is the attenuation factor for the precessing μ^+ polarization, and BG is a constant background. The time spectra were measured by means of an EG&G Model TDC100 time-to-digital converter interfaced to a PDP-11/40 computer via a Bi-Ra Model MBD-11 microprogrammed CAMAC branch driver.[6]

The observed precession signals $A\cdot G(t)\cos(\gamma_\mu B_\mu t)$ are shown in Fig. 1. The relaxation time at room temperature is 5 times longer than in the previous measurement on a polycrystalline sample;[3] we were able to observe precession signals down to 23°K, a substantial improvement over previous attempts.[3,4] However, at 4.2°K no signal was visible, even after $\sim 2.5 \times 10^6$ events had been collected. The observed magnetic fields B_μ and relaxation rates λ, obtained by fitting the spectra to the form (1) with the assumption $G(t) = \exp(-\lambda t)$, are summarized in Table I.

The temperature dependence of the relaxation rate upon temperature is shown in Fig. 2(a). The data for $T \geq 73$°K are well represented by a

single exponential law which can be explained in terms of dipolar broadening narrowed by μ^+ diffusion. There are two magnetically inequivalent interstitial sites in a bcc Fe single crystal; these sites occur with relative probability 1:2 and have dipolar fields $2H_1$ and $-H_1$, respectively. Thus, if the μ^+ diffusion is fast enough, such dipolar fields will cancel. A one-dimensional random-walk model predicts

$$\lambda = (\gamma_\mu H_1)^2/\nu, \qquad (2)$$

where ν is the jumping frequency of the μ^+ among the interstitial sites, which can be expressed by the Arrhenius formula

$$\nu = (\alpha/a^2)D = (\alpha/a^2)D_0 \exp(-E_a/k_B T), \qquad (3)$$

where D is the diffusion constant, a is the lattice parameter (2.86 Å), α is 64 for tetrahedral-tetrahedral jumps and 24 for octahedral-octahedral jumps, and E_a is the activation energy. The observed temperature dependence of λ is represented by Eqs. (2) and (3) from 300°K down to 73°K, yielding $E_a = 17 \pm 0.3$ meV ($E_a/k_B = 185 \pm 3$°K). Without knowing the location of the μ^+ and the magnitude of H_1, it is impossible to learn D_0 from λ. Making a classical estimate gives $H_1 = -2.6$ kG (9.4 kG) for tetrahedral (octahedral) interstices and $D_0 = 1.2 \times 10^{-5}$ (7×10^{-5}) cm^2/sec.

Diffusion of H^+ in Fe has been studied extensively,[7] but the observed diffusion constants scatter considerably from sample to sample. If we take the smallest value (47 meV) for the H^+ activation energy, the present value for the μ^+ is 3 times smaller. Taking these values to represent intrinsic activation energies in the absence of traps, we may conclude that the difference is due to the lighter mass of the μ^+

($M_\mu/M_H \sim 1/9$). The obvious suggestion is that E_a depends on the mass as $\sqrt{M}$, in conflict with the quantum theory of Flynn and Stoneham,[8] where a self-trapping mechanism predicts a dependence of E_a on M^{-1}.

Below 44°K, the relaxation data deviate from the exponential law extrapolated from the higher temperatures. This suggests that fast quantum diffusion dominates over thermally activated diffusion at low temperatures, as earlier suggested for μ^+ in Nb.[9] The deviation in Fig. 2(a) is too strong to be explained by a T^7 law predicted by Flynn and Stoneham.[8] Moreover, experimental results show the absence of any visible precession signal at 4.2°K, where the μ^+ still seems to be diffusing, instead of being 'frozen'.

In zero external field, the local field B_μ felt by the μ^+ can be decomposed as

$$B_\mu = (4\pi/3)\, M + \langle H_d \rangle + H_{int}, \qquad (4)$$

where the first term is the Lorentz field, the second the average dipolar field within the Lorentz cavity, and H_{int} is the contact hyperfine field due to polarized conduction electrons. The dipolar field $\langle H_d \rangle$ is assumed to average to zero in a random walk with $\nu >> \gamma_\mu H_l$. The sign of B_μ in Fe is known to be negative.[3] Thus, using known values of the saturation magnetization M,[10] we obtain H_{int} at various temperatures, as shown in Table I.

At the low temperature limit H_{int} becomes -11.1 kG. It is interesting to relate this hyperfine field to the conduction electron polarization observed with polarized neutrons.[11] For the μ^+ located at a tetrahedral site, which is known to have the deepest negative magnetization of $4\pi M_{int} = -1.6 \pm 0.5$ kG, the hyperfine field would be

$(8\pi/3)M_{int} = -1.1 \pm 0.3$ kG in the absence of the screening effect of polarized electrons. The observed H_{int} is an order of magnitude larger than this value. This surprisingly large discrepancy can be attributed to pronounced screening of μ^+ by polarized conduction electrons in Fe, while in Ni such a screening enhancement appears minimal;[12] further theoretical study is clearly required. The calculation of Jena[13] based on the Daniel-Friedel model predicts that H_{int}(Fe) = -5.6 kG. In this model the conduction electron spin density is assumed to be uniform but, if the μ^+ prefers a tetrahedral site with subsequent conduction electron screening, the predicted value would be close to the observed one.

Relative values of $H_{int}(T)$ are shown in Fig. 2(b), along with the temperature dependences of the saturation magnetization (M) and the nuclear hyperfine field (H_n).[10] Clearly H_{int} deviates from M in the same sense as H_n, but more strongly. However, just the opposite but similarly pronounced effect is seen in Ni: H_{int} remains nearly constant up to 300°K, while the magnetization decreased by 7%.[2]

The presence of the interstitial μ^+ perturbs the host metal in two ways: i) screening of the positive charge by polarized conduction electrons; and ii) static distortion of the neighbouring lattice. Our results imply that the μ^+ in Ni perturbs the neighbouring atoms in such a way that they have a stronger ferromagnetic coupling than the bulk atoms, but that the μ^+ in Fe does not. This conclusion seems relevant to several known differences between Ni and Fe: while the host magnetic moment of Ni is strongly affected by the presence of magnetic impurities, that of Fe shows only simple dilution;[14] while the pressure

dependence of the Curie temperature is positive for Ni, there is none for Fe; and so on. Another important difference is the spatial distribution of the unperturbed interstitial spin density. In fcc Ni it is almost constant,[15] but in bcc Fe it varies dramatically within one interstitial site.[11] Thus, if the μ^+ position is broadened with increasing temperature, the magnitude of H_{int} could be attenuated.

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Table I. Summary of μ^+SR experiment in Fe at H_{ext} = 0G.

T (K)	B_μ (G)	H_{int} (G)	λ (μsec^{-1})
22.5 (25)	-3755 (96)	-11 092 (96)	19.2 (37)[a]
44.0 (20)	-3711 (37)	-11 044 (37)	13.3 (8)
73.4 (39)	-3773 (10)	-11 099 (10)	5.1 (2)
96.9 (8)	-3763.6(32)	-11 081.6(32)	2.8 (2)
121.7 (15)	-3740.9 (7)	-11 050.9 (7)	2.4 (1)
154.2 (11)	-3710.6 (2)	-11 003.6 (2)	1.48 (1)
291.0 (10)	-3595.0 (1)	-10 783.0 (1)	0.80 (0)

[a]Due to low statistics and fast relaxation, the initial amplitude was held fixed at the values (8.5%) fitted for higher temperatures.

Figure Captions

Fig. 1 μ^+ precession signals in Fe at representative temperatures. Solid lines represent best fits to the data.

Fig. 2 (a) μ^+ relaxation rate in Fe as a function of inverse temperature. (b) Fractional change of saturation magnetization (M), nuclear hyperfine field (H_n) and μ^+ hyperfine field (H_{int}) in Fe as a function of temperature.

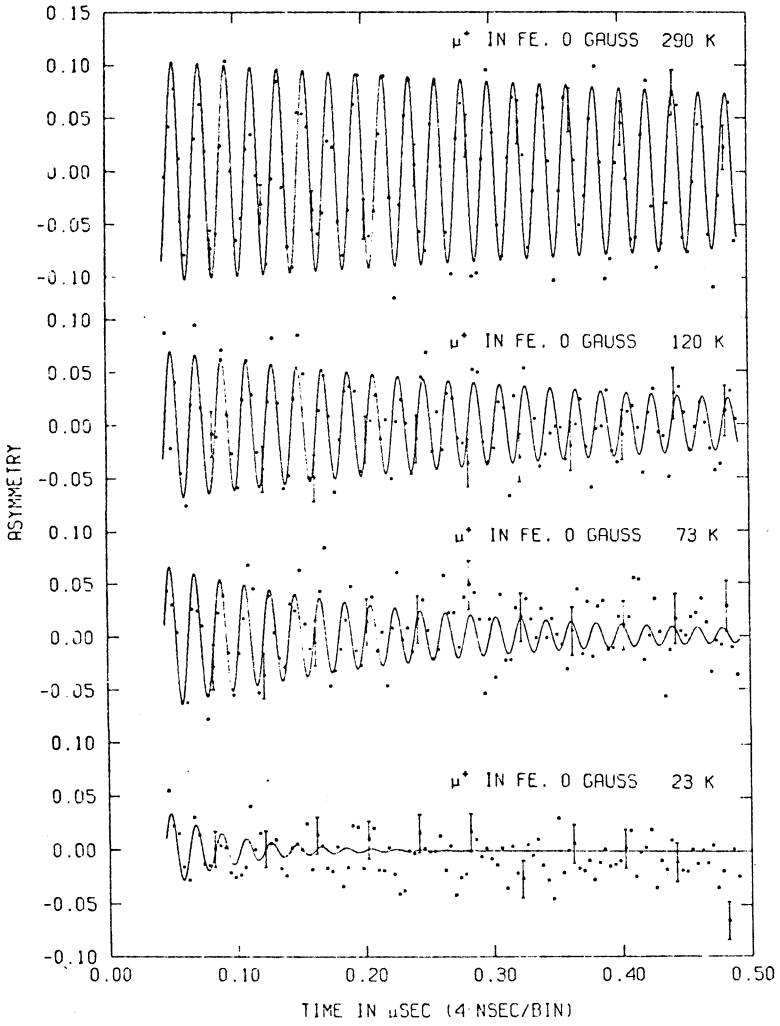

Fig. 1

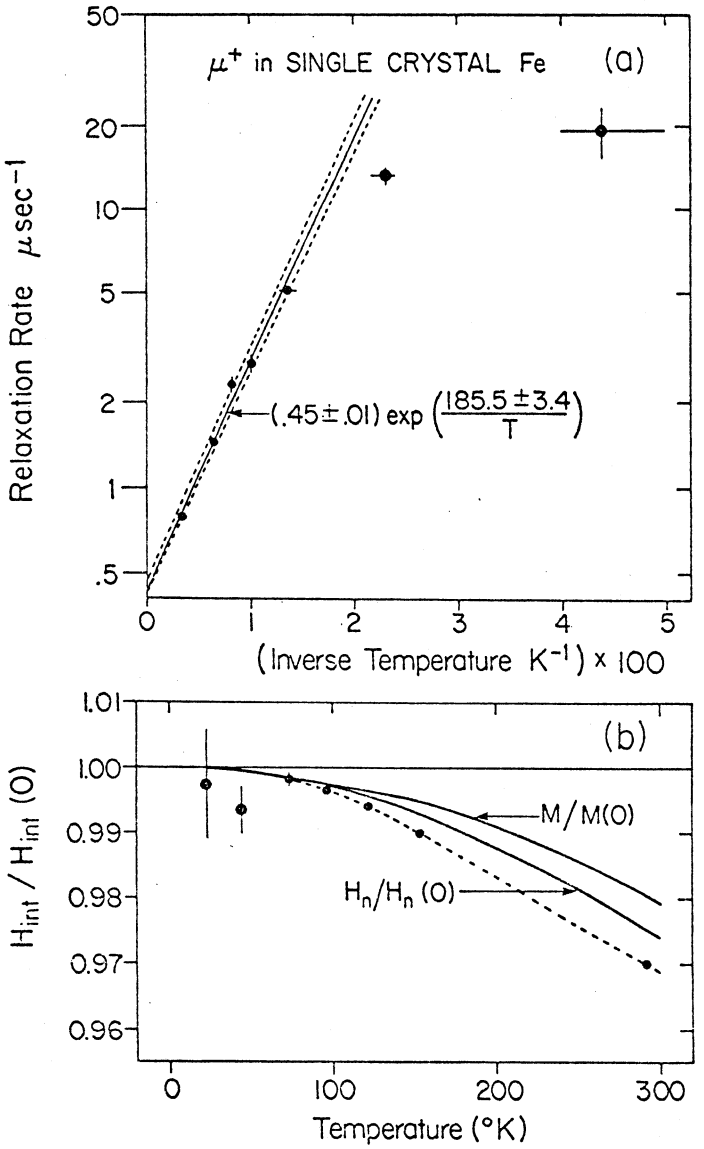

Fig. 2