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INTERNAL MAGNETIC FIELDS IN HCP-IRON^a

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ABSTRACT

The magnetic behavior of hcp-Fe (epsilon iron) has been investigated by the Mössbauer Effect over a wide range of temperature T , pressure P , and applied magnetic field H_0 . The internal field H_i of the induced moment is given by $H_i = 0.20 \pm .01 H_0$ and is independent of T and P . Enhanced paramagnetism is suggested as the most likely origin for a contribution of this magnitude to H_i .

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INTRODUCTION

Two phases of iron are stable at room temperature - ordinary bcc- or α -Fe and hcp- or ϵ -Fe. The hcp phase exists over a wide range of temperature but is formed only at pressures in excess of about 11.5 GPa (115 kbar). Much of the experimental and theoretical work reported on ϵ -Fe has addressed the question of magnetism. Band theory calculations [1,2,3] generally support the premise that ϵ -Fe will not order either ferromagnetically or antiferromagnetically. On the other hand certain hcp-alloy data [4,5] extrapolated to "pure ϵ -Fe" suggest antiferromagnetism. Most of the experiments searching for spontaneous magnetic order in ϵ -Fe have employed the Mössbauer Effect (ME) of ^{57}Fe [6,7], carried out during the past decade to successively lower temperatures. The presence of magnetic ordering should produce a hyperfine splitting or at least a broadening in the ME spectra. Other effects can lead to a temperature-dependent broadening, and indeed slight broadening was observed in some of the earliest measurements. The latest ME data [7] clearly show ϵ -Fe is not magnetically ordered at T down to 0.030 K and at 19.1 GPa. We report here an extension of this work using an external magnetic field and a new iron sample in our diamond-anvil pressure cell.

The ME of ^{57}Fe has been used to study the magnetic response of a number of dilute or weakly magnetic systems [8,9]. The ^{57}Fe acts as a probe of the local magnetization. Measurements of the average magnetic field at the ^{57}Fe ME nucleus in ϵ -Fe as a function of temperature, pressure, and applied field should help establish the size of any induced magnetic moment present in ϵ -Fe.

EXPERIMENTAL

The experimental procedures employed were the same as those given before [7], except a different iron source was used, the temperature of the source could be varied continuously from 4.2 to 300 K, and an external magnetic field of up to 5 T could be applied. The z-axis of the solenoidal field was

If we assume that H_i indeed is a measure of the local magnetization [8], the corresponding magnetic susceptibility of ϵ -Fe is constant, independent of temperature (and pressure). A temperature-independent susceptibility suggests that ϵ -Fe shows pure Pauli paramagnetism. While this may be the simple explanation of the results, we are struck by the magnitude of the effect on H_{eff} . Since the proportionality constant relating the magnetization and the internal field is not rigorously known, we consider alternative viewpoints.

Polarization of the conduction-band electrons in a metal by H_0 produces a local field at the nucleus proportional to H_0 and essentially independent of temperature. The Knight shift is a measure of this effect. For most metals there is field enhancement, but negative shifts have been found in the transition metals. For ME measurements of localized moments of Fe in various host metals, this contribution to H_i is small and usually has been ignored; however, for Fe in V, Nb, and Ta the field enhancement has been seen [8]. (Fe has no moment in these superconductors.) In these three cases the enhancement at the ^{57}Fe nucleus was just that given by the Knight shift of the host, that is, $H_i = \Lambda H_0$, where Λ varies from +0.007 for V to +0.015 for Ta. The numerical agreement with the Knight shift is probably fortuitous, but the sign and the trend with host mass are no doubt real. It seems unlikely that such a large negative value of $\Lambda \approx -0.20$ for ϵ -Fe arises solely from a Knight-shift type of argument. Generally, large negative Knight shifts are observed only when a significant moment localized on the d-shell produces a negative contribution to H_i through core polarization [11].

Another approach to interpreting the ^{57}Fe ME data is to scale the moment using the 3d-free-ion value of about $-1.7 \mu_B/\mu_B$ [11]. If we assume such scaling for Fe in ϵ -Fe, we can calculate an induced moment and a susceptibility. At 5 T the induced moment is $\sim 0.03 \mu_B$, and the susceptibility is 9.4×10^{-4} emu/mole. Somewhat similar results have been reported [9] for Fe in $\text{Dy}_2\text{Fe}_{14}\text{Si}_{12}$. Below 4 K the Dy aligns antiferromagnetically, but Fe apparently doesn't order. Only on an applied field is a moment induced on the Fe atoms.

RESULTS

for the magnetic response at an ^{57}Fe nucleus in $\epsilon\text{-Fe}$ at 21.5 and 15.0 GPa as a function of H_0 are presented in Fig. 1. About half the data were taken at each pressure; no distinction is made in Fig. 1 as there were no trends with pressure. Essentially the same was true of temperature, although the points at 280 K possibly lie somewhat above the line $H_{\text{eff}}/H_0 = 0.80 \pm 0.01$. These results were fully confirmed by preliminary field data using a different, weaker $\epsilon\text{-Fe}$ source at a pressure of 21.5 GPa in the temperature range 1.2 to 35 K. The data at the lowest fields were less reliable because the line width and the effective field splitting are not fully independent parameters for partially resolved ME spectra.

Since we have defined $H_i = H_{\text{eff}} - H_0$, it follows that $H_i = -0.20 H_0$. In summary, we find that the field at the ^{57}Fe ME nucleus in $\epsilon\text{-Fe}$ is proportional to the applied field (1 - 5 T) and is independent of temperature (1.2 - 280 K) and pressure (15.0 - 21.5 GPa).

DISCUSSION

It is difficult to compare quantitatively bulk susceptibility measurements with theory because there are so many different contributions to the susceptibility. Furthermore, it is difficult to compare ME hyperfine fields directly with bulk magnetization measurements as they are only approximately measurements of the same thing. On the other hand, microscopic measurements, such as NMR and ME, should complement bulk measurements, not just complicate them. The ME is uniquely suited for magnetic measurements in a diamond cell, while techniques for conventional magnetization or susceptibility measurements are not available.

The high susceptibility of ϵ -Fe implied from these measurements strongly suggests enhanced paramagnetism. Recently, Shimizu [12] has reviewed the theory and has included many applications. The susceptibilities of the paramagnetic transition metals are typically high (7.2×10^{-4} emu/mole for Pd at 4 K) and temperature dependent. The value derived here for ϵ -Fe is an order of magnitude larger; however, enhanced susceptibilities still larger have recently been reported by Felner and Nowik [13]; from magnetometer measurements, they find the bulk susceptibility of YRh_6B_4 and LuRh_6B_4 at 4.1 K to be 240 and 220×10^{-4} emu/mole, respectively. Upon addition of 0.5 at% Fe, these compounds magnetically ordered at 150 and 220 K, respectively. Itinerant electron magnetism surely plays a role in the magnetic behavior of ϵ -Fe.

Superconductivity in ϵ -Fe at $T \leq 0.25$ K has been proposed [3] based on an analysis of the band theory calculations. The absence of superconductivity has also been predicted [14]. Unfortunately, the ME is not sensitive to a transition to superconductivity, so the zero-field low-temperature results [7] neither support nor deny superconductivity. In some superconductors, e.g., W and Mo, Fe impurities develop a localized moment; in others, e.g., Nb and Ta, there is no local moment. For those superconductors in which a localized moment is formed, the addition of Fe has dramatic effect on T_c , the superconducting transition temperature. For those in which no localized moment is found, the addition of Fe has a small effect on T_c . Although Fe is hardly an impurity in ϵ -Fe, the significant enhanced paramagnetism observed may suppress superconductivity.

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Fig. 1. The relative effective field H_{eff}/H_0 at the ^{57}Fe nucleus as a function of the applied field H_0 and pressure (see text).

FIG. 1