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Anomalous Temperature Behaviour of the Local Magnetic Field on ^{207}Pb in Fe*

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The hyperfine magnetic field acting on dilute ^{207}Pb impurity in Fe host has been investigated as a function of temperature between 87°K and 983°K. The measurements have been performed by the Perturbed Angular Correlation technique through the 1064 - 570 keV gamma-gamma cascade in ^{207}Pb. A strongly anomalous temperature behaviour of the field is observed.

O campo magnético hiperfíno atuando sobre impureza diluída de ^{207}Pb em matriz de Fe foi investigado como função da temperatura entre 87°K e 983°K. As medidas foram feitas pela técnica da correlação angular perturbada através da cascata gama-gama de 1064 – 570 keV no ^{207}Pb. Observa-se um comportamento fortemente anômalo do campo com a temperatura.

1. Introduction

Various measurements of the hyperfine magnetic field acting on Pb isotopes diffused or implanted into ferromagnetic hosts have been reported recently. Pramila *et al.*[1] measured a field of +260 kOe on ^{207}Pb in Fe (^{207}Pb$\underline{\text{Fe}}$) making use of a source of ^{207}Bi electroplated on an iron wire and diffused. Bowman and Zawislak[2] obtained a field of + 660 kOe for ^{207}Pb$\underline{\text{Fe}}$ with a source prepared by a different metallurgical procedure; mixing ^{207}Bi activity with iron powder, reducing in H,, coining into a small cylinder and annealing. The latter experiment was repeated by Kaufmann[3], who also obtained a field of the order of + 600 kOe. The field on ^{208}Pb$\underline{\text{Fe}}$ was reported[2] to be + 280 kOe. In this last experiment, ^{208}Tl the parent of ^{208}Pb was implanted in Fe with a recoil energy of ~ 100 keV due to the preceding 6 MeV alpha emission from ^{212}Bi. More recently, Bacon *et al.*[4] determined a field of + 260 kOe for ^{204}Pb$\underline{\text{Fe}}$ using a sample obtained by melting and quenching a

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mixture of ^{204}Bi with Fe. All the above measurements were performed at room temperature.

A study of lattice location of Bi and Pb implanted in an Fe single crystal has been made by means of the channeling technique[S]. These experiments suggest that the smaller field (~ 300 kOe) corresponds to Pb atoms located substituionally at regular sites and that the higher field (-650 kOe) is associated with Pb atoms located off regular lattice sites in the Fe matrix.

In addition to the problem of the location of the impurity atoms in the host matrix, a further important question in understanding the origin of the hyperfine fields concerns the nature of the coupling between the impurity and the ferromagnetic host. In this paper, we describe a measurement of the hyperfine magnetic field on ^{207}PbFe as a function of temperature in the range from 87°K to 983°K in an effort to clarify the nature of this coupling.

2. Source Preparation and Experimental Procedure

The source was prepared by depositing carrier free ^{207}Bi activity in dilute HCl solution on 300 mg of pure iron (99.99) powder. The mixture was allowed to stand for one minute, was washed with distilled water, dried and then reduced in a H, atmosphere at 600°C during two hours. The reduced powder was allowed to cool and immediately formed into a right circular cylinder under a pressure of -2×10^4 atm. The source was 3 mm in diameter and 5 mm high, containing less than 0.01 atomic per cent of ^{207}Bi impurity.

The measurements were performed by the integral perturbed angular correlation method through the 1064 – 570 keV gamma-gamma cascade in ^{207}Pb, using a standard fast-slow electronic system with two NAI(Tl) scintillation spectrometers. The results have been analyzed in terms of the ratio

$$R(\theta) = 2\frac{W(\theta, +H) - W(\theta, -H)}{W(\theta, +H) + W(\theta, -H)},$$

where

$$W(\theta, \pm H) = \sum_{\substack{even \\ k}} \frac{b_k}{[1 + (k\omega\tau)^2]^{1/2}} \cos k(\theta \mp \omega\tau)$$

is the expression for the angular correlation function in presence of a perpendicular magnetic field.

Since, for the present cascade in ^{207}Pb, $b_4 \ll b_2$, we have

$$R(\theta = 135°) \simeq \frac{4b_2\,\omega\tau}{1 + (2\omega\tau)^2},$$

where $\omega\tau$ is the average value of the spin precession angle in the magnetic field.

Preliminary measurements of the ratio $R(\theta = 135")$ as a function of annealing time at 800°C in a H, atmosphere have shown that, after 6 hours of annealing, a hyperfine field of $\sim$ 700 kOe was obtained in agreement with the value reported in Ref. 2. In a next step, the source was placed in a small furnace designed to fit between the polecaps of the polarizing electromagnet. The temperature of the heating device was controlled to better than 0.3%. A copper cold finger was used for the measurement at 87°K.

The angular correlation function was measured with the sample unoriented in the magnet and heating device geometry giving $b_2 = +0.110 \pm 0.003$ and $b_4 = -0.007 \pm 0.004$. These values are corrected for random coincidences but not for geometry and polycrystalline magnetic attenuation. The measurements of the ratio $R(O = 135°)$ were performed with an external polarizing field of 7 kOe which was enough to saturate the sample magnetically.

Our results for the average value of the spin precession angle $\omega\tau$, calculated from the ratio $R(\theta = 135°)$ and measured at nine different temperatures are summarized in Table 1. The last column of the table shows the corresponding hyperfine magnetic fields obtained by comparing these results with the weighted average of two determinations[6,7] of $\omega\tau$ for the same state in ^{207}Pb in an external magnetic field. The field at 87°K is about 15% larger than at room temperature. Fig. 1 shows the values of $\omega\tau$ as a function of temperature normalized to the spontaneous magnetization curve of pure Fe at 87°K. These data display the strong anomalous temperature dependence of the hyperfine magnetic field acting on Pb dilute impurity in the Fe matrix. The measurement of the hyperfine field as a function of temperature was performed twice in ihe same sample giving the same result. This is an indication that the impurity atoms occupy well defined positions in the host lattice.

Temperature (°K)	$\omega\tau$ (radians)	H_{hf} (kOe)
87	0.1940 ± 0.0086	+ 798 ± 58
303	0.1689 ± 0.0085	+ 695 ± 53
383	0.1675 ± 0.0084	+ 689 ± 53
483	0.1211 ± 0.0070	+ 498 ± 41
583	0.0953 ± 0.0070	+ 392 ± 37
683	0.0752 ± 0.0055	+ 309 ± 29
783	0.0501 ± 0.0058	+ 206 ± 27
883	0.0492 ± 0.0052	+ 202 ± 24
983	0.0218 ± 0.0080	+ 90 ± 33

Table 1 - Mean precession angles $\omega\tau$ and extracted hyperfine magnetic fields on ^{207}Pb<u>Fe</u> at different temperatures. **o** = $= g\mu_N H/\hbar$ is the Larmor frequency and τ is the lifetime of the nuclear level.

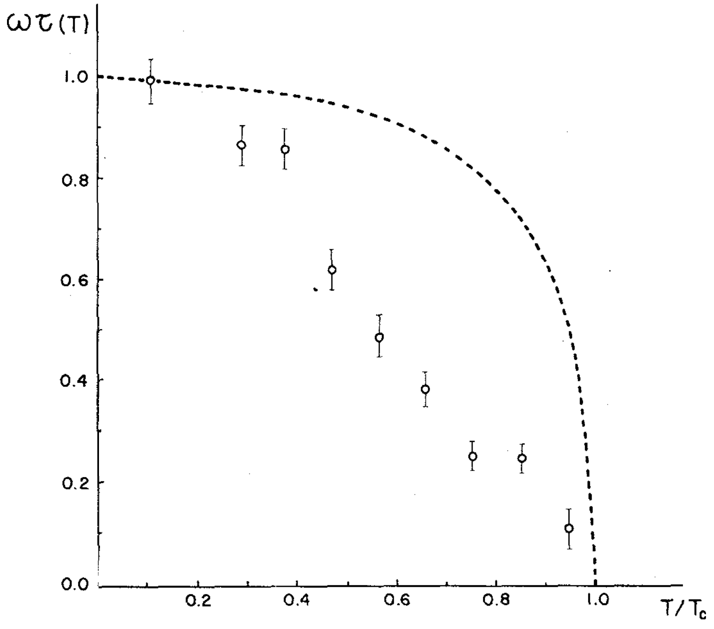

Figure 1 - Measured values of $\omega\tau$ for ^{207}Pb<u>Fe</u> as a function of temperature, normalized to the magnetization curve of Fe. The broken curve shows the spontaneous magnetization of Iron.

3. Discussion

A number of approaches have been proposed in the literature to explain the deviations between the temperature dependence of local magnetic fields at impurity nuclei and the host magnetization. The molecular field treatments[8,9] are based on the existence of a localized moment at the impurity. When a local moment exists at the impurity, the hyperfine magnetic field may display a temperature dependence differing from that of the host bulk magnetization. A fitting of the experimental points in Fig. 1, using the molecular field mode1[8], gives equally good fittings for the impurity spin S in the range $1/2 \leq S \leq 7/2$. This means that S cannot be determined only from the temperature dependence measurement of the hyperfine magnetic field. On the other hand, the transition model[10] based on Friedel's description of transition impurities in ferromagnetic metals suggests that in a case like ^{207}Pb in Fe the temperature behaviour of the hyperfine field at the impurity should not be anomalous.

It is interesting to note that similar anomalies have been observed for the hyperfine magnetic field on ^{119}Sn in Fe, Co and Ni hosts[11,12,13]. The case of ^{119}Sn<u>Co</u> displays an especially strong anomalous temperatiire dependence. Sn is located in the same column of the periodic table of elements as Pb but belongs to period V. Cranshaw suceeded in fitting the anomalous temperature behaviour of the field on ^{119}Sn<u>Co</u> assuming that the effective field on Sn has a component strongly dependent on the overlap between the Sn 5s-wavefunctions and the 3d-wavefunctions of the matrix and that this overlap depends on the temperature due to the thermal oscillations of the atoms.

The channeling experiments on Pb implanted in an iron single crysta1[5] show that by increasing the temperature of the sample the Pb atoms move from substitutional into non-substitutional sites. Our source was prepared with polycrystalline iron and by annealing at high temperature. It is then possible that in the present case ^{207}Pb is located in non-substitutional sites, which could be interstitials (although, because of their larger size, it is difficult to believe that Pb atoms are located interstitially in Fe). If the Pb atoms are located interstitially, an approach similar to the one used by Cranshaw could also be applied for this case. Because Pb atoms are 50% larger than Fe atoms, a considerable overlap with the 3d-wave functions could be expected in the interstitial site.

It is difficult to understand this anomaly and very probably the interpretation of the experiment will depend on additional experimental studies, especially concerning the location of the impurity in the lattice, before a more definitive conclusion can be reached.

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