Measurement of the hyperfine anomaly between ^{187}Os and ^{189}Os

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NMR and spin echo measurements were performed on dilute samples of ^{187}Os and ^{189}Os in Fe. The magnetic hyperfine splitting frequencies $\nu_M = |g\mu_N B_{\mathrm{HF}}^{(\mathrm{eff})}/h|$ at 4.2 K were determined to be 111.17(1) and 371.00(5) MHz for ^{187}Os*Fe* and ^{189}Os*Fe*, respectively. Taking into account the known magnetic moments, the hyperfine anomaly is deduced to be $^{187}\Delta_{\mathrm{Fe}}^{189} = +0.0195(2)$, which is of opposite sign and much smaller than expected theoretically. [S0556-2813(96)01410-0]

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I. INTRODUCTION

If atoms are embedded as dilute impurities in a ferromagnetic host lattice such as Fe, Ni, and Co, the impurity nuclei are subject to a large magnetic hyperfine (HF) interaction,

$$\nu_M = |g\mu_N B_{\mathrm{HF}}^{(\mathrm{eff})}/h|, \tag{1}$$

where g is the nuclear g factor and $B_{\mathrm{HF}}^{(\mathrm{eff})}$ is the effective magnetic hyperfine field at the nuclear site. The different contributions to the magnetic hyperfine field may be separated into a contact field $B_{\mathrm{HF}}^{(\mathrm{c})}$ and a noncontact field $B_{\mathrm{HF}}^{(\mathrm{nc})}$ the latter normally being small. For high-Z nuclei, the contact field varies considerably over the nuclear volume. As a consequence, the ratio of hyperfine splittings of two nuclear states is in general different from the ratio of the respective g factors, which is denoted as hyperfine anomaly [1,2]. In a simplified picture, the spin and orbital parts of the nuclear magnetic moment experience different magnetic fields from the contact part of the magnetic hyperfine interaction. Thus, the hyperfine anomaly is large if the spin and the orbital contribution to the magnetic moment have opposite sign, i.e., if the nuclear magnetic moment is small. Actually, for the $I^\pi[Nn_z\Lambda] = 1/2^-[510]$ ground state of ^{183}W ($\mu = +0.1178\mu_N$ [3]), a large hyperfine anomaly with respect to 2^+ ^{184}W ($\mu = +0.578(14)\mu_N$ [4]) was found experimentally: $^{183}\Delta_{\mathrm{Fe}}^{184} = -0.130(23)$ [5]. [In the original work $^{184}\Delta_{\mathrm{Fe}}^{183} = +0.150(31)$ is given.] The ground state of ^{187}Os has the same Nilsson configuration as ^{183}W. The magnetic moment of ^{187}Os is even smaller, $\mu(^{187}\mathrm{Os}) = +0.064\,651\,89(6)\mu_N$ [6], i.e., that, with respect to $3/2^-$ ^{189}Os($\mu = +0.659\,933(4)\mu_N$ [7]), a huge hyperfine anomaly between ^{187}Os and ^{189}Os would be expected. Therefore NMR and spin echo (SE) measurements on ^{187}Os and ^{189}Os as dilute impurities in Fe were performed.

II. MAGNETIC HYPERFINE INTERACTION AND HYPERFINE ANOMALY

The magnetic hyperfine interaction of a nuclear state with magnetic moment distribution $\mu(r)$ in a magnetic hyperfine field with radial distribution $B_{\mathrm{HF}}(r)$ is given by

$$\mu B_{\mathrm{HF}}^{(\mathrm{eff})} = \int \mu(r) B_{\mathrm{HF}}(r) dr = \mu B_{\mathrm{HF}}(0)(1+\epsilon). \tag{2}$$

Here the so-called single-level anomaly ϵ describes the deviation from the point-dipole interaction. It depends on the radial dependence of the wave function of the nuclear state. This effect causes the ratio of the magnetic hyperfine splitting frequencies of two nuclear states in the same atomic environment to be different from the ratio of the respective g factors. The hyperfine anomaly (HA) is defined by the relation

$$\frac{\nu_1}{\nu_2} = \frac{g_1}{g_2}(1 + {}^1\Delta^2) \tag{3}$$

and can be expressed by

$${}^1\Delta^2 = \frac{\epsilon_1 - \epsilon_2}{1+\epsilon_2} \approx \epsilon_1 - \epsilon_2 . \tag{4}$$

III. EXPERIMENTAL DETAILS

The samples of ^{187,189}Os*Fe* were prepared as follows: ^{187}Os (isotopic enrichment 70.4%) and ^{189}Os (isotopic enrichment 95.6%) were melted with highly pure Fe (purity >99.999%) in an electron beam furnace, the Os concentration being 1 at. %. With further melting steps more diluted alloys were prepared, with concentrations of 0.01, 0.05, 0.10, 0.25, and 0.50 at. % Os. By coldrolling, foils with a thickness of ~1 μm were obtained. The final foils were annealed for 3 h at 650 °C in high vacuum.

A stack of foils of a given composition were insulated from each other and placed in an untuned slow wave structure [8] forming part of an automatic spin echo spectrometer. The rf power was sufficiently low for only domain wall enhanced NMR to be observed. At each frequency the echo was integrated at (unknown) phase ϕ with respect to the reference and then at ($\phi + \pi/2$). The intensity was then found from $S = (S_\phi^2 + S_{\phi+\pi/2}^2)^{1/2}$. All measurements were performed at 4.2 K.

TABLE I. Resonance frequencies of ^{187}Os and ^{189}Os in Fe at 4.2 K, hyperfine field, and full width at half maximum of the distribution, measured by swept frequency NMR.

	^{187}Os			^{189}Os		
Os conc. at. %	ν (MHz)	$B_{HF}^{(eff)}$ T	FWHM T	ν (MHz)	$B_{HF}^{(eff)}$ T	FWHM T
0.01	111.175	-112.80	0.27	370.95	-110.61	0.38
0.05	111.17	-112.79	0.29	371.08	-110.65	0.37
0.10	111.16	-112.78	0.37	370.97	-110.62	0.42
0.25	111.16	-112.78	0.43	371.05	-110.64	0.47
0.50	111.16	-112.78	0.54	370.67	-110.53	0.78
1.00	111.19	-112.82	0.99	371.16	-110.68	0.78

IV. RESULTS AND DISCUSSION

The results are compiled in Table I. It is evident that at a concentration of 1 at. % Os, a level often considered dilute in the past, there is appreciable line broadening and also a (weak) shift in the peak position from that found at lower concentrations. Below an impurity concentration of 0.5 at. % there is little change in the line position although the line width continues to decrease to near the minimum concentration.

The accuracy with which the peak frequency of a distribution of hyperfine fields can be measured in swept frequency NMR depends upon the line width and the signal to noise ratio and to some extent upon the asymmetry of the distribution. Fortunately the accuracy obtainable for the frequencies of the ^{187}Os and ^{189}Os NMR in the present experiment is such that it is not difficult to take the dilute limit in order to calculate the hyperfine anomaly. From Table I we find that, at $T = 4.2$ K,

$$\nu_M(^{187}\mathrm{Os}Fe)=111.17(1)\ \ \mathrm{MHz},$$

$$\nu_M(^{189}\mathrm{Os}Fe)=371.00(5)\ \ \mathrm{MHz}.$$

Our results are consistent with earlier spin echo measurements of lower accuracy obtained on ^{189}Os in samples containing $\sim$1 at. % Os [9]. Assuming that the Os NMR has the same temperature dependence as that of ^{57}Fe in pure Fe the extrapolation to $T = 0$ K yields

$$\nu_M(^{187}\mathrm{Os}Fe)=111.17(1)\ \ \mathrm{MHz},$$

$$\nu_M(^{189}\mathrm{Os}Fe)=371.01(5)\ \ \mathrm{MHz}.$$

For the ratio of resonance frequencies our final result is

$$\frac{\nu_M(^{187}\mathrm{Os}Fe)}{\nu_M(^{189}\mathrm{Os}Fe)}=0.299\,642(50).$$

Taking the ratio of magnetic moments given in Ref. [7], $\mu(^{189}\mathrm{Os})/\mu(^{187}\mathrm{Os})=10.2075(1)$, the ratio of g factors is

$$\frac{g(^{187}\mathrm{Os}Fe)}{g(^{189}\mathrm{Os}Fe)}=0.293\,902(3).$$

With these values, we get for the hyperfine anomaly

$$^{187}\Delta_{\mathrm{Fe}}^{189}=+0.0195(2).$$

This is, contrary to the expectation, (i) in absolute value much smaller than the experimental hyperfine anomaly for $1/2^-$ ^{183}W and 2^+ ^{184}W, $^{183}\Delta_{\mathrm{Fe}}^{184} = -0.130(23)$ [5] and (ii) has opposite sign.

To get the hyperfine anomaly in a pure-contact field $^{187}\Delta_{\mathrm{c}}^{189}$, the fraction of contact field to the hyperfine field $B_{\mathrm{HF}}^{(\mathrm{c})}/B_{\mathrm{HF}}$ has to be taken into account. (For Ir and Au the ratio of the noncontact field to the hyperfine field is $B_{\mathrm{HF}}^{(\mathrm{nc})}/B_{\mathrm{HF}}\sim-0.15$, i.e., $B_{\mathrm{HF}}^{(\mathrm{c})}/B_{\mathrm{HF}}\sim1.15$ [10].) It has been speculated that the noncontact field of Os in Fe may be negative [11]. Assuming that the absolute magnitude of the noncontact field is similar to those for Ir and Au in Fe, we take $B_{\mathrm{HF}}^{(\mathrm{c})}/B_{\mathrm{HF}}\sim1.00(15)$ as a very conservative estimate. Then the hyperfine anomaly between ^{187}Os and ^{189}Os in a pure-contact field is

$$^{187}\Delta_{\mathrm{c}}^{189}=+0.020(3).$$

For Hg isotopes, Moskowitz and Lombardi found an empirical relation for the hyperfine anomaly [12], which was later put on a theoretical basis by Fujita and Arima [13]. According to this relation the single-level hyperfine anomaly is

$$\epsilon=\frac{\alpha_s}{\mu}. \tag{5}$$

Here μ is the nuclear magnetic moment and $\alpha_s=\pm1.13\times10^{-2}\mu_N$ for $I=l\pm1/2$, where l is the orbital angular momentum of the odd neutron. Meanwhile it became evident that the Moskowitz-Lomabardi rule is not restricted to Hg, but seems to have a more general applicability, if the constant α_s is properly scaled by Z^2, where Z is the atomic number. Thus, according to Eq. (5), the hyperfine anomaly is large for nuclear states with a small magnetic moment. Applying the Moskowitz-Lombardi rule of Eq. (5) to ^{187}Os and ^{189}Os, we get $\epsilon^{(187)}=-0.153$, the arguments for the minus sign are given in Ref. [5], $\epsilon^{(189)}=-0.015$, and hence $^{187}\Delta_{\mathrm{c}}^{189}=-0.14$ for a pure contact field. It should be mentioned that the experimental hyperfine anomaly between $1/2^-$ ^{183}W and 2^+ ^{184}W could be explained moderately well with the Moskowitz-Lomabardi rule [5]. Thus the small positive hyperfine anomaly between $1/2^-$ ^{187}Os and $3/2^-$ ^{189}Os is unexpected and we cannot offer any explanation for it. A very simplified explanation of the hyperfine anomaly of single-particle states is as follows: The nuclear magnetic moment of a single-particle state is a composition of an orbital magnetic moment and the spin magnetic moment. The orbital magnetic moment can be described as if resulting from an electric current; thus it senses the average magnetic field, $B_o=\int_0^R B(r)dr$, whereas the spin magnetic moment senses the field according to the wave function of the single-particle state $\psi(r)$, $B_s=\int B(r)|\psi(r)|^2dr$. Thus, in general $|B_s|<|B_o|$. In this way, the large positive hyperfine anomalies of the proton $3/2^+$ states ^{191}Ir, ^{193}Ir, and ^{197}Au can be understood easily. For neutron single-particle states the interpretation is not straightforward because of the missing orbital contribution to the magnetic moment. Small magnetic moments of single-particle neutron states must be mainly due to (i) quenching of the spin magnetism by core polarization and (ii) a magnetic contribution by collective admixtures in the wave function. If the radial wave function connected with these magnetic contributions is not too different from the single-particle wave function, the hyperfine anomaly may indeed be small, despite the fact that the magnetic moment is small. Then, the Moskowitz-Lombardi rule is no longer applicable. Thus, the experimental hyperfine anomaly ${}^{187}\Delta^{189}_{\mathrm{Fe}}$ indicates that the radial distribution of the magnetic moment density is very similar for $1/2^-$ ^{187}Os and $3/2^-$ ^{189}Os. It indicates further that the radial distribution of the magnetic moment density changes drastically between ^{183}W and ^{187}Os, despite the fact that the unpaired neutron is in a $I^{\pi}[Nn_z\Lambda]=1/2^-[510]$ Nilsson state. To clarify the situation, nuclear structure calculations would be valuable.

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