Nonalignment of the magnetic hyperfine field of Ir in Fe†

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Previous discrepancies in the reported values of the nuclear magnetic dipole moment of the 171-keV level of ^{191}Ir measured by nuclear orientation and nuclear magnetic resonance techniques are resolved by an interpretation in terms of nonalignment of the static magnetic hyperfine field of Ir in Fe with the applied field. The angle between the applied and hyperfine field directions is deduced to be 15°. Other possible sources of the discrepancies in the reported moments are shown to be negligible. The best value of the moment is believed to be $\mu = (6.03 \pm 0.04)\mu_N$ measured by Eska *et al.* using NMR techniques.

[RADIOACTIVITY ^{191}Ir^m; measured $\gamma(\theta, T)$ in Fe; deduced nonalignment of hyperfine field.]

In a previous communication[1] dealing in part with the decay of ^{191}Ir^m oriented at low temperatures in Fe, we reported a deduced value for the magnetic dipole moment of the 171-keV ^{191}Ir level (see Fig. 1) of $\mu = (3.27 \pm 0.12)\mu_N$, based on the observed angular distribution of the 129-keV γ ray and on the assumption that the nuclear hyperfine field had the magnitude of the saturated value $H_{sat} = 1.405$ MOe.[2] We have subsequently become aware of a previous measurement of the magnetic moment of that level by Eska *et al.*,[3] who reported $\mu = (6.03 \pm 0.04)\mu_N$, derived from the observed resonant frequency for the destruction of the anisotropy of oriented ^{191}Ir^m in Fe. The result of the nuclear-orientation angular-distribution (NO/AD) measurement thus differs from that of the nuclear-orientation nuclear magnetic resonance (NO/NMR) measurement by nearly a factor of 2. In a subsequent communication,[4] we discussed the nonsaturation of the hyperfine fields of a number of impurities in Fe, among which was ^{191}Ir; it was concluded that applied fields H_{app} of magnitude less than 5 kOe are often not sufficient to saturate the hyperfine field and thus result in reducing the anisotropy of angular distributions measured by the NO/AD technique. It seems reasonable to conclude that our previous results using $H_{app} = 3$ kOe, from which the ^{191}Ir^m moment was deduced, suffered from this nonsaturation, that the discrepancy in the reported moments is more likely a discrepancy in the effective hyperfine fields, and that the result deduced by Eska *et al.*[3] represents a more nearly correct value for the ^{191}Ir^m moment. However, owing to the serious discrepancies in the literature in a number of moments deduced employing Fe(Ir) alloys (discussed in part in Ref. 4 and more completely by King, Grabowski, and Scharenberg[5]), it is of interest to attempt to use our previous result to obtain a greater understanding of the Fe(Ir) hyperfine field and its relationship to H_{app}.

Eska *et al.*[3] compare their result favorably with the value $\mu = (6.3 \pm 1.5)\mu_N$ deduced from a NO/AD experiment (with $H_{app} = 2$ kOe) by Cameron *et al.*[6] However, Cameron *et al.* used values for H_{sat} and for the angular-distribution parameter of the 129-keV γ ray which have been superseded by more precise values, in terms of which their result becomes $\mu \approx 4\mu_N$, in better agreement with

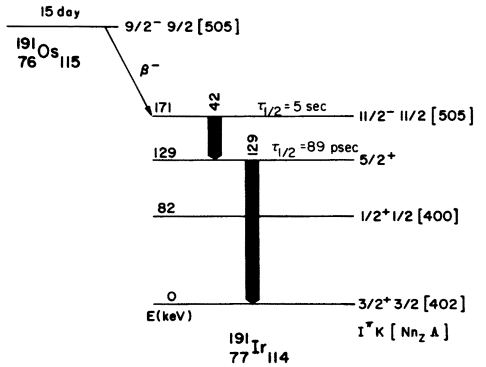

FIG. 1. The decay of ^{191}Os to levels of ^{191}Ir.

our previous NO/AD result[1] than with the NO/NMR result of Eska *et al.*[3] It thus appears that the difference in the deduced values of the moment possibly reflects a fundamental difference between the two techniques, and perhaps by extension a difference between resonant and nonresonant techniques in general.

In analyzing the nonsaturation of hyperfine fields of impurities in Fe, an effective reduction factor (dependent on H_{app}) of the parameters describing the orientation of the 171-keV level was deduced.[4] The use of this reduction factor [amounting to approximately 0.95 for Fe(Ir) at $H_{app}=3$ kOe] is not sufficient to resolve the discrepancy between the deduced moments. Moreover, the use of such an effective reduction factor ignores the mechanism which brings about the resultant nonsaturation, and in the following we attempt a somewhat more detailed consideration of the origin of the nonsaturation. As was done previously,[4] we postulate a nonalignment of the hyperfine field with the applied field; that is, due to local forces in the vicinity of the impurity atom, the impurity tends to orient along certain crystalline axes rather than along $\vec{H}_{app}$. This nonalignment has been considered in detail by Aharoni,[7] who has accounted for such an effect in terms of magnetostrictive, magnetocrystalline, exchange, and magnetostatic forces. According to this interpretation, the hyperfine field lies not along $\vec{H}_{app}$, but rather along the generating vector of a cone about $\vec{H}_{app}$. There is in fact a spectrum of possible cone angles representing orientations along various crystalline axes (we are dealing here with polycrystalline samples); however, we will represent this effect by an effective cone angle Θ which is a spatial rms average over the spectrum of possible cone angles. The angle Θ then represents an effective angle between the hyperfine field and $\vec{H}_{app}$, and is predicted to be of order 10 to 20°.[7] Our previous results[4] on a variety of different impurities in Fe showed evidence for nonsaturation of the hyperfine field for applied fields in the neighborhood of 5 kOe, with the degree of nonsaturation decreasing with increasing applied fields until virtually complete saturation was achieved at $H_{app}=15$ kOe. This effect was interpreted in terms of a cone angle Θ which decreased with increasing applied field as $\sin\Theta \propto (H_{app})^{-1}$; however no direct evidence was obtained for the presence of the cone angle. In the present communication we report a more direct evidence for the nonalignment of the hyperfine field and show that the NO/AD and NO/NMR results are in agreement for a well-defined value of Θ. Similar evidence of the existence of such nonalignment of the hyperfine fields acting on nuclei recoil implanted into Fe and Ni following Coulomb excitation has been given by Ben-Zvi *et al.*[8]

We represent the normalized angular distribution of γ rays from an oriented nucleus as

$$W(\theta) = 1 + Q_2 B_2 U_2 A_2 P_2(\cos\theta) + Q_4 B_4 U_4 A_4 P_4(\cos\theta). \tag{1}$$

The parameters of Eq. (1) are defined in Ref. 1. Using γ-ray detectors at 0 and 90° relative to the direction of the applied field, we observed, at a temperature $T = 19.2 \pm 0.2$ mK,

$$W_0 = 2.048 \pm 0.001$$
$$W_{90} = 0.493 \pm 0.001 \tag{2}$$

by normalizing the counting rates to the isotropic high-temperature limit. The subscripts 0 and 90 refer to the detectors at 0 and 90° relative to $\vec{H}_{app}$. Assuming the saturated hyperfine field to lie parallel to $\vec{H}_{app}$ [equivalent to assuming $\theta = 0°$ and $\theta = 90°$, respectively, for Eqs. (2)] results in the deduced values $B_2 = 1.511 \pm 0.015$, $B_4 = 0.778 \pm 0.055$, from which we previously deduced the magnetic moment $\mu = (3.27 \pm 0.12)\mu_N$.

Considering the possibility of a nonvanishing cone angle Θ, the results can be analyzed as follows: The detector in the 0° direction (along the applied field) will always be oriented at an angle Θ relative to the nuclear polarization direction (which is in the direction of the cone generatrix). We thus take the angular distribution previously ascribed to 0° to give now $W(\theta = \Theta) = 2.048 \pm 0.001$, and assuming values for B_2 and B_4 computed from the NMR value of μ and the fully saturated hyperfine field ($B_2 = 1.685 \pm 0.002$, $B_4 = 1.152 \pm 0.005$), we compute

$$\cos^2\Theta = 0.931 \pm 0.004$$

or

$$\Theta = 15.2° \pm 0.4°. \tag{3}$$

The orientation of the nuclei relative to the 90° detector is not as straightforward to determine. We assume the nuclei to be distributed randomly along the cone generatrix. The true angle θ between the direction of nuclear polarization and the 90° detector axis is given by

$$\cos\theta = \sin\Theta \cos\phi, \tag{4}$$

where ϕ is the azimuthal angle of the generatrix. The random distribution along the cone is equivalent to assuming the equal probability of all values of ϕ, and average values of the Legendre polynomials P_2 and P_4 may be computed as

$$\langle P_k \rangle = \frac{1}{2\pi} \int_0^{2\pi} P_k(\cos\theta)\, d\phi \tag{5}$$

from which we obtain

$$\langle P_2 \rangle = \tfrac{3}{4}\sin^2\Theta - \tfrac{1}{2}, \quad (6)$$

$$\langle P_4 \rangle = \tfrac{105}{64}\sin^4\Theta - \tfrac{15}{8}\sin^2\Theta + \tfrac{3}{8}. \quad (7)$$

Using the measured W_{90} for $\langle W(\theta)\rangle$, along with the values of B_2 and B_4 used above, we obtain

$$\sin^2\Theta = 0.066 \pm 0.006$$

or

$$\Theta = 14.9^\circ \pm 0.7^\circ \quad (8)$$

in excellent agreement with the result deduced for the 0° detector.

It is also possible to observe a reduced value of the hyperfine field resulting from macroscopic or microscopic alloy inhomogeneities. In the former case, one would observe a reduced average hyperfine field, in the present situation amounting to $0.54H_{sat}$; we believe it to be unlikely that our $\frac{1}{4}$ at.% alloy could produce such a drastic reduction in the average internal field. Microscopic inhomogeneities would result in a fraction f of the nuclei experiencing a small (possibly vanishing) field while the remainder experience the full H_{sat}. Our reduced value of B_2 is consistent with a fraction $f = 0.10 \pm 0.01$ of the nuclei at nonmagnetic sites, but this value of f is not consistent with the reduction observed in the value of B_4, which would require $f = 0.30 \pm 0.03$. Thus we conclude that only an interpretation in terms of nonalignment presents a reasonable and consistent explanation of the apparent discrepancies in the effective hyperfine field of Fe(Ir) alloys. It should be noted that the nonalignment does not alter the results of the NO/NMR experiment, in which the resonant frequency depends only on the magnitude of the field acting at the nucleus and not on its direction.

A number of alternative explanations (which alter the angular distribution itself rather than the internal field) for the low value of μ deduced from the NO/AD experiments may be postulated; these are discussed below. All of these effects are such as to attenuate the anisotropy.

(1) Deorientation: We consider attenuation arising from deorientation of the magnetic substates caused by the unobserved 42-keV γ ray, which is intermediate between the 171-keV initial oriented level and the 129-keV level from which the 129-keV γ ray is emitted. The preceding discussion has assumed the 42-keV $\frac{11}{2}^- - \frac{5}{2}^+$ transition to be pure $E3$. A possible $M4$ component would cause a reduction of the deorientation coefficients U_2 and U_4 relative to their values for a pure $E3$ transition. If we assume the hyperfine field to lie parallel to the applied field, and use the full saturation values of B_2 and B_4 used above, it is possible to deduce the magnitude of the $M4$ component which might produce such a reduction of the anisotropy. From U_2 we deduce $M4 = (14 \pm 1)\%$ and from U_4, $M4 = (13 \pm 2)\%$; although these values are internally consistent, they differ considerably from the value $M4 < 10^{-3}\%$ deduced by Deutsch and Hornshoj[9] from L-subshell conversion-electron measurements. It thus seems unlikely for the reduced anisotropy to arise from this source.

(2) Perturbations: One other effect which might give rise to an attenuation of the angular distribution would be external perturbations acting on the 129-keV level ($\tau_{1/2} = 89$ psec). It is not possible for the hyperfine field itself to produce such a perturbation, a conclusion which follows from the results of angular-correlation theory that an axially symmetric static perturbing field parallel to the orientation axis of an axially symmetric oriented system produces no effect on the angular correlation.[10] Other possible perturbing mechanisms would be randomly oriented time-dependent magnetic interactions or static electric quadrupole interactions. The latter possibility may be disregarded based on the results of Salomon and Shirley,[11] who determined the static quadrupole coupling of the ^{193}Ir ground state in Fe. Extrapolating those results to the present case, the quadrupole interaction frequency ω_0 is estimated to be 0.7 MHz, and thus $\omega_0\tau \approx 10^{-4}$, indicating such quadrupole effects are negligible. A possible randomly oriented time-dependent magnetic interaction results from the continual random variation in the direction of the electronic spin. An applied field of sufficient magnitude can "decouple" the electronic and nuclear spins, thus aligning the electron spins; no effect on the angular distribution would result in such a case, for the reason discussed above. However, it is unlikely that our 3-kOe applied field is of sufficient strength to result in such decoupling. Rather, the rapidly fluctuating field $\vec{H}(t)$ may be decomposed into a component parallel to the applied field (which cannot perturb the angular distribution) and components normal to $\vec{H}_{app}$, the effect of which may be an additional contribution to the cone angle Θ. That such effects are likely negligibly small follows from the short lifetime of the 129-keV level and from the short relaxation times (of order 10^{-12} sec) associated with the electronic shells.[10]

(3) Relaxation: The measured orientation parameters have been assumed to be characteristic of the 171-keV ^{191}Ir^m ($\tau_{1/2} = 5$ sec) rather than of the ^{191}Os parent state which decays by β emission to the ^{191}Ir level. This assumption is based on the observed nuclear spin-lattice relaxation times

T_1 associated with the 129-keV anisotropy. Eska *et al.*[3] observed $T_1 = 0.25 \pm 0.10$ sec for Ni(Ir); since $T_1 \propto H^{-2}$, we estimate $T_1 = 0.03 \pm 0.01$ sec for Fe(Ir), a value considerably less than the lifetime of the $^{191}Ir^m$. This NMR-deduced value agrees with our NO/AD value of $T_1 \lesssim 0.1$ sec. Assuming the time dependence of the anisotropy to be approximately an exponential characterized by this relaxation time, one expects that a fraction $T_1/(T_1 + \tau)$ of the 129-keV γ rays will show anisotropy characteristic of the ^{191}Os; for the present case this fraction amounts to $(0.39 \pm 0.15)\%$. The anisotropy of the 129-keV γ rays characteristic of the ^{191}Os parent is estimated to be $W_{O_5}(\theta) = 1 + 0.41P_2(\cos\theta)$ based on a calculated value for the ^{191}Os ground-state moment of $\mu = 1.2\mu_N$ (assuming this state to be described by a pure $\frac{9}{2}^-[505]$ Nilsson configuration); correcting the appropriate observed anisotropies [Eq. (2)] results in variations in the two deduced values of Θ by only one third of the quoted uncertainties. Consequently, we assume this to be a negligible effect.

Finally, we note that while the preceding considerations have a substantial effect on the value of the nuclear magnetic moment of $^{191}Ir^m$ (and possibly also of ^{193}Os) deduced in Ref. 1, the multipole mixing ratios deduced for the various β- and γ-radiation fields are not affected since the mixing ratios were deduced from the relative angular distributions of two or more γ rays.

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