Magnetic Moments of the First 2^+ States in Os^{192} and Pt^{192} *

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ABSTRACT

PAC measurements of the magnetic moments of the first 2^+ states in Os^{192} and Pt^{192} give $0.797 \pm .036$ and $0.90 \pm .11 \mu_N$, respectively. Results agree with the $2(Z/A)$ values and differ appreciably from the Kumar and Baranger model predictions. Measurements using Fe(Ir) and Ni(Ir) alloys established the unsuitability of Fe(Ir) alloys for such experiments. NMR measurements of internal fields for Os^{187} in Fe and Ni give $H(296^\circ\text{K}) = -1093.43 \pm .65 \text{ kOe}$ and $H(295^\circ\text{K}) = -282.3 \pm 1.0 \text{ kOe}$, respectively.

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Previous measurements of the magnetic dipole moment of the first 2^+ state in Os^{192} have given inconsistent results. Recent perturbed angular correlation (PAC) measurements by Béraud et al.¹ and Levanoni et al.² using the internal field of Os in an iron lattice, gave $\omega_L \tau / H$ ratios of $0.545 \pm .078 \text{ rad/MG}$ and $0.527 \pm .065 \text{ rad/MG}$, respectively, whereas an earlier PAC measurement by Gilad et al.³, performed following Coulomb excitation and using an external magnetic field, gave an $\omega_L \tau / H$ ratio of $0.75 \pm .07 \text{ rad/MG}$. The value of the internal field used in the Béraud and Levanoni measurements is the 4.2°K NMR measurement of Kontani and Itoh⁴, corrected 2.6% for temperature, assuming that the temperature dependence of the internal field follows the temperature dependence of the saturation magnetization of iron. The possible incorrectness⁵ of this frequently made assumption, concerning

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the temperature dependence of the internal field, could account for the aforementioned differences in measured $\omega_L \tau / H$ ratios. In a number of cases the internal fields of solute atoms in ferromagnetic hosts have been observed to follow closely the temperature dependence of the saturation magnetization of the ferromagnetic host, but there have been some very notable exceptions⁶⁻⁸ in which the internal fields have been observed to fall off much more rapidly with increasing temperature than the corresponding saturation magnetization of the host.

To resolve this question, room temperature NMR measurements were made on Os¹⁸⁷ in Fe and Ni lattices. Samples were prepared by electron beam melting mixtures of 99.999% pure Fe and Ni with isotopically enriched Os¹⁸⁷ to achieve alloys containing 0.4 at. % Os. The homogeneity of the samples was established by electron micro-probe analysis. Samples were reduced to 20 μ particles and annealed, under vacuum, for 2 hours at 1000°C. The NMR measurements were made in zero external field using a frequency modulated and frequency swept marginal oscillator. Resonance signals were detected by means of a lock-in amplifier operating at the second harmonic of the modulation frequency.

The Os¹⁸⁷ in Fe resonance was observed at a frequency of $107.22 \pm .04$ MHz at a temperature of 296°K. The Os¹⁸⁷ in Ni resonance was observed at a frequency of $27.68 \pm .10$ MHz at a temperature of 295°K. Using the known⁹ magnetic dipole moment of the ground state of Os¹⁸⁷, the room temperature internal fields for Os in Fe and Ni hosts are calculated to be $-1093.43 \pm .65$ kOe and -282.3 ± 1.0 kOe, respectively. (The minus signs, which imply that the direction of the internal field is opposite to that of an external polarization field, do not follow from the NMR measurements, but rather from the PAC experiments which are discussed in the remainder of this paper.)

The room temperature (296°K) measurement for the internal field of Os in

Fe is in close agreement with what would be expected on the basis of the 4.2°K NMR measurement of Kontani and Itoh and the assumption that the temperature dependence of the internal field follows that of the saturation magnetization of Fe. This does not, therefore, account for the differences in the aforementioned $\omega_L \tau / H$ ratios.

To further investigate these differences, an extensive series of PAC measurements was made using the internal field of Os in both Fe and Ni lattices. The use of both Fe and Ni hosts provides for a convenient consistency check, in that the ratio of the $\omega_L \tau$ values in Fe and Ni base samples should be in agreement with the ratio of the measured internal fields for Os in Fe and Ni.

The integral PAC measurements were performed with an automatic $\gamma - \gamma$ spectrometer incorporating a 25 cm³ coaxial Ge(Li) detector, an integrally mounted 3 x 3 inch NaI(Tl) detector with high voltage stabilization, and a conventional coincidence circuit. The 485 - 206 keV $\gamma - \gamma$ cascade in Os¹⁹² was used, with the 206 keV γ being selected in the Ge(Li) detector and the 485 keV γ in the NaI(Tl) detector. The samples were polarized for the "field up" and "field down" measurements by means of an electromagnet. Coincidence counts were recorded at seven different angles and were corrected for accidentals, background, and normalization, to give the true coincidence data. The data were computer fitted to the function:¹⁰

$$W(\theta, \infty, H) = \sum_{k=0,2,4} \frac{b_k}{[1 + (k\omega_L \tau)^2]^{\frac{1}{2}}} \cos(k\theta - \arctan k\omega_L \tau),$$

from which the $\omega_L \tau$ (product of Larmor frequency and mean life) values were determined.

The samples used in the PAC measurements were in the form of small, 1 mm

diameter, spheres which were individually prepared by electron beam melting mixtures of 99.999% pure Fe and Ni with unenriched Ir, to give pellets containing about 0.6 - 0.8 at. % Ir. The Ir^{192} parent was subsequently produced by neutron irradiation of the samples.

The results of the PAC measurements are summarized in Table I (Ni) and Table II (Fe). The A_{22} values are the measured coefficients for the standard Legendre polynomial expansion of $W(\theta)$.¹⁰

The ratio of the NMR - measured internal fields is 3.87 and, therefore, on the basis of the average, $\langle \omega_L \tau \rangle = 0.223$ recorded for the Ni base alloys, one expects to observe $\omega_L \tau$ values on the order of 0.863 radians for the Fe base alloys. As Table II indicates, this is considerably larger than any measured value. Thus, there is an appreciable disparity between the Fe and Ni results.

Considerable evidence favors the correctness of the Ni data over the Fe data. Table II shows a significant variation among the results from the different iron samples (compare with Table I). It is observed that annealing produced a marked decrease in the $\omega_L \tau$ value of one of the "better" iron samples, and yet the identical annealing of nickel samples produced no measurable change in the $\omega_L \tau$ values. Evidently the state of the Fe(Ir) alloy is strongly dependent on the heat treatment of the alloy, and quite possibly on the manner in which the alloys are cooled from the molten state. The A_{22} coefficients for the Ni data (Table I) are in agreement with the liquid source value, $A_{22} = -0.274 \pm .011$, reported by Grabowski.¹¹ By comparison, the A_{22} coefficients from the Fe data (Table II) are clearly attenuated with respect to the liquid source value, suggesting the presence of additional perturbations on the nuclei, which could result, also, in an apparent attenuation of the $\omega_L \tau$ values. It is also noted that the $\omega_L \tau / H$ ratio for the

Ni data, 0.79 rad/MG, is in good agreement with the value of $0.75 \pm .07$ rad/MG observed by Gilad *et al.*³ in the PAC measurement following Coulomb excitation, in which an external magnetic field was used. Iron samples 2 and 4 and Ni samples 1 and 2 (see Tables I and II) were polished, etched, and subjected to a metallurgical examination. There were regions of microporosity evident in the microstructure of both of the iron samples. These regions were considerably more prominent in Fe(Ir) sample 2, which, notably, gave a significantly lower $\omega_L \tau$ value than sample 4 (Table II). This suggests that the annealing effects and $\omega_L \tau$ and A_{22} attenuations observed with the Fe samples might possibly be caused by vacancy - impurity (Ir) pair formation and the segregation that can result in the vicinity of pores as a result of the vacancy - impurity pair migrations. Porosity segregation has been observed, e.g., with a Mn solute in copper.¹² The nickel samples did not show either microporosity or a microstructure suggestive of a second phase.

The above evidence points convincingly to the conclusion that there are serious metallurgical problems which inhibit the preparation of a microscopically homogeneous Fe(Ir) alloy, and the disparity between the Fe and Ni results must be resolved in favor of the Ni data.

Since the $\omega_L \tau$ values from Fe samples 1 and 4 are in agreement with the highest previously observed^{1,2} $\omega_L \tau$ values in Fe(Ir) alloys, it is likely that all PAC experiments which have attempted to use Fe(Ir) alloys have given erroneous results. A case in point would be the PAC measurements of the magnetic dipole moments of the excited states in Pt¹⁹². Because of the short lifetimes involved, all such measurements have made use of the large internal field seen by Pt¹⁹² in an Fe lattice and, in all cases, this has been accomplished by starting with Fe(Ir) alloys. To confirm the suspicions regarding previous measurements of Pt¹⁹², integral PAC measurements were made on the

first 2^+ state of Pt^{192} using the 468 - 316 keV γ - γ cascade and a Ni(Ir) alloy. The experimental arrangement was identical to that used in the Os^{192} measurements.

The results of both the Os^{192} and the Pt^{192} measurements are summarized in Table III. Since the expected resonance frequency of Pt^{195} is outside the range of the present spectrometer, no attempt was made at a room temperature NMR measurement of the internal field for Pt in Ni. Instead, the 4.2°K measurement of Kontani and Itoh has been used in the calculation of the Pt moment.

Previous measurements^{1,17} of the magnetic dipole moment of the first 2^+ state in Pt^{192} have given $\mu \lesssim 0.6\mu_N$. As expected, the present result, using a Ni base alloy, is appreciably higher.

Significantly, the magnetic dipole moments of the first 2^+ states in Os^{192} and Pt^{192} are observed to be in agreement with the hydrodynamical $2(Z/A)$ values, and appreciably higher than the predictions of the Kumar - Baranger pairing-plus-quadrupole model¹⁸ (see Table III).

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TABLE I. Integral PAC results for the 485 - 206 keV
 γ - γ correlation in Os¹⁹² using Ni base alloys.

Sample	$\omega_L \tau$ (radians)	A_{22}^a
1	$0.235 \pm .014$	(not measured)
2	$0.225 \pm .014$	$-0.286 \pm .005$
2 ^b	$0.216 \pm .017$	$-0.292 \pm .006$
3	$0.217 \pm .008$	$-0.295 \pm .005$
4	$0.227 \pm .012$	$-0.304 \pm .005$
4 ^b	$0.225 \pm .028$	$-0.281 \pm .016$
Wt.Av.: $\langle \omega_L \tau \rangle = 0.223 \pm .005$ $\langle A_{22} \rangle = -0.294 \pm .003$		

^a Corrected for geometry.

^b Samples were annealed for 5 hours at 1000°C and
 allowed to cool slowly to room temperature.

TABLE II. Integral PAC results for the 485 - 206 keV
 γ - γ correlation in Os¹⁹² using Fe base alloys.

Sample	$\omega_L \tau$ (radians)	A_{22}^a
1	$0.591 \pm .050$	$-0.235 \pm .015$
1 ^b	$0.417 \pm .035$	$-0.236 \pm .010$
1 ^c	$0.268 \pm .038$	$-0.201 \pm .009$
2	$0.437 \pm .055$	$-0.220 \pm .018$
3	$0.497 \pm .052$	$-0.220 \pm .015$
4	$0.559 \pm .027$	$-0.218 \pm .010$
5	$0.486 \pm .037$	$-0.242 \pm .011$

^a Corrected for geometry.

^b Sample was annealed under vacuum for 5 hours at
 1000°C and allowed to cool slowly to room
 temperature.

^c Sample was reheated and quenched.

TABLE III. Magnetic dipole moments of the first 2^+ states in Os^{192} and Pt^{192} as determined from integral PAC measurements using Ni base alloys.

Nucleus	(state)	$\omega_L \tau$ (radians)	τ (psec)	H (kG)	μ (μ_N)	Kumar Baranger ^a (μ_N)	2(Z/A)
Os^{192}	(2^+)	$0.223 \pm .005$	414 ± 16^b	282.3 ± 1.0^e	$0.797 \pm .036$	0.590	0.792
Pt^{192}	(2^+)	$0.036 \pm .004$	51.8 ± 2.0^c	323 ± 10^d	$0.90 \pm .11$	0.425	0.813

^a Reference 18

^b Calculated from the B(E2) values of references 13, 14, and 15.

^c Weighted average of the measurements of references 1 and 16, and the value calculated from the B(E2) measurements of references 13 and 15.

^d Reference 4, including a $4.9 \pm 1\%$ temperature correction.

^e This work.