HYPERFINE MAGNETIC FIELDS ON DILUTE ACTINIDES IN IRON

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Résumé. — Nous avons mesuré le champ magnétique hyperfin créé par le thorium et l'uranium dilué dans le fer. Les mesures ont eu lieu à l'aide de la technique des perturbations de corrélation angulaire après une implantation ionique. Pour le thorium dans le fer à la température ambiante, le champ est de — 230 ± 80 kOe, et pour l'uranium dans le fer de — 580 ± 150 kOe. Le signe et l'ordre de grandeur des valeurs mesurées suggèrent que la contribution de la couche 5 f au champ total, est petite dans le cas de l'uranium, et donc différente de la contribution de la couche 4 f dans le cas des terres rares. Des résultats récents pour Np dans le nickel (H_{eff} = + 170 kOe) et pour Pu dans le fer (H_{eff} = + 620 kOe) montrent que, pour les actinides, les champs ne suivent pas la tendance qui apparaît pour la série des terres rares.

Abstract. — We have measured the hyperfine magnetic field on dilute thorium and uranium in iron metal using the ion implantation perturbed angular correlation technique. The field on thorium in iron at room temperature is — 230 ± 80 kOe and on uranium in iron — 580 ± 150 kOe. The sign and magnitude of the values suggests that the 5 f contribution to the field for uranium is small, unlike the 4f contribution for the rare earths. Recent results for Np in nickel (H_{eff} = + 170 kOe) and for Pu in iron (H_{eff} = + 620 kOe) indicate that fields on actinides do not follow the trend apparent for the rare earth series.

I. Introduction. — Since the discovery by Samoilov et al. [1] that the hyperfine magnetic field on gold nuclei in iron metal was − 1 280 kOe, there has been considerable interest in systematizing and understanding the hyperfine magnetic fields effective on dilute impurities in the ferromagnetic metals. Numerous measurements on more than 40 elements have been made using NMR, nuclear gamma resonance, and perturbed angular correlation techniques.

Several attempts have been made to theoretically understand the signs and magnitudes of the observed fields. The charge screening model of Daniel and Freidel [2] was especially successful in predicting the sign change near tin. Shirley et al. [3] have explained the fields in the 5 p region invoking a simple conduction electron polarization (CEP) mechanism, and Balabanov and Delyagin [4] have recently proposed a model emphasizing core polarization (CP) effects.

For the rare earths ($4f^n$) both core and CEP effects are smaller than the orbital contribution from the aligned unquenched angular momentum of the localized 4 f electrons. The latter leads to measured fields greater than one million Oe for most of the rare earths in iron [5].

With this in mind, it is interesting to extend experiment and possible models to heavier impurities and especially to the actinides ($5f^n$). For the latter, the implantation perturbed angular correlation method (IMPACT) seems best suited for experimentation, in spite of its inherent low precision, discussed below, because odd A stable isotopes for NMR work are unavailable, the few states available for Mössbauer studies exhibit complex hyperfine structure, and sample preparation problems are significant.

Using IMPACT, we have determined the field on thorium in iron to be − 230 kOe and on uranium in iron to be − 580 kOe, at room temperature, with uncertainties of about 30 %. Recent work by others (see table) measuring the hyperfine fields on Rn, Ra, U, Np, and Pu allow some general statements to be made about the relevant interactions.

II. Experimental details. — Many IMPACT experiments have been reported [6]. As schematically indicated in (Fig. 1), a beam of high energy heavy ions

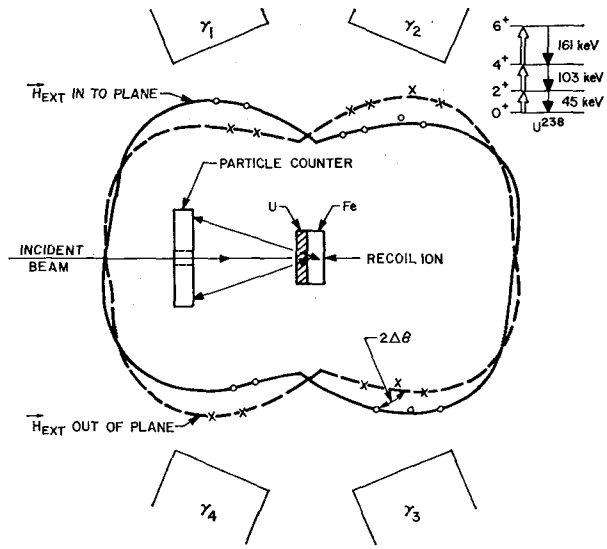

FIG. 1. — Schematic top view of apparatus. The polar drawings are of the $0 \to 4 \to 2$ ^{238}U perturbed angular correlation. The ^{238}U level scheme is indicated in the upper right.

impinges on a two layer target, and gamma rays from excited nuclei are detected in coincidence with back scattered ions. The coincidence condition insures a well defined recoil implantation energy as well as a known anisotropic gamma ray distribution. In the case illustrated, 80 MeV ^{32}S ions Coulomb excite the 2^+, 4^+ and 6^+ states of ^{238}U. The $4^+ \to 2^+$ gamma distribution is shown.

The unperturbed correlation appears rotated as a result of interaction with the aligned hyperfine magnetic field. Experimentally, one measures twice the total precession angle as the external polarizing field is reversed. It can be shown that for small angles [7]

$\Delta\theta = \omega_L \tau + \varphi$, where $\omega_L = -g\frac{\mu_N}{\hbar} H_{eff}$, is the Larmor precession frequency in the static hyperfine field, τ is the nuclear lifetime, and

$$\varphi = -g\frac{\mu_N}{\hbar}\int_0^\infty H_1(t)\,dt$$

is the precession due to the transient field [8].

Neither the nuclear g factor, nuclear lifetime, nor transient magnetic field are known exactly, but reliable theoretical estimates and experimental systematics, as discussed below, allow an unambiguous determination of the sign of the effective field and a good estimate of its magnitude.

The nuclei studied, ^{232}Th and ^{238}U, exhibit rotational level structure. The « cranking model » for rotational nuclei, which has been very successful in most cases, predicts g factors for the rotational excited states of 0.26 and 0.25 respectively [9]. The 2^+ state lifetimes for ^{232}Th and ^{238}U have been measured to 3 % precision [10]. The major uncertainty in the 4^+ and 6^+ state lifetimes, calculated using the rotational model [11], arises from the uncertainty in the internal conversion coefficients. We have used the theoretical values of Hager and Seltzer [12] to calculate lifetimes with an estimated uncertainty of about 10 %.

The transient field value is much less well known. Extrapolation of experimental data is impractical, because most of the existing data is for $Z \approx 50$. We have used the « uncorrected » form of the Lindhard-Winther theory [13] to obtain a transient field rotation of − 35 milliradians for the cases studied. Recent results by Heestand et al. [14] indicate that these values may be low by as much as a factor of 2.

In principle, a measurement of $\Delta\theta$ for both the 4^+ and 6^+ states determines the transient field correction directly [15]. In practice, the experimental uncertainties are such that only consistency with the transient field theory can be deduced from the data.

III. **Results and discussion.** — For ^{232}Th in iron :

$\Delta\theta(4 \to 2) = 31 \pm 7$ milliradians,
$\Delta\theta(6 \to 4) = -11 \pm 15$ milliradians.

For ^{238}U in iron :

$\Delta\theta(4 \to 2) = 78 \pm 8$ milliradians,
$\Delta\theta(6 \to 4) = 29 \pm 9$ milliradians.

Using a transient field contribution of − 35 milliradians ; $g_R = 0.26$ for Th and $g_R = 0.25$ for U ; $\tau^{4+} = 0.239$ ns, $\tau^{6+} = 0.050$ ns for Th, and $\tau^{4+} = 0.324$ ns, $\tau^{6+} = 0.085$ ns for U ; we extract values for the effective field on Th in iron of − 230 kOe and for U in iron − 580 kOe.

A similar experiment on the ^{238}U 4^+ state has been reported by Münchow et al. [16]. Their result of $\Delta\theta = 126 \pm 21$ milliradians is significantly greater than that quoted above. Possible reasons for the discrepancy are discussed in reference [15].

Table I lists the measured hyperfine magnetic fields, at room temperature, on impurities in iron from Rn through Pu. The value for neptunium, however, is for a nickel host. Systematics and most models would indicate that the field in iron is the same sign and possibly four times as large. The value quoted for uranium is a weighted average of the experiments described above. A very recent independent experiment at Aarhus [17] obtains a field of about − 660 kOe for uranium in iron.

The large positive field on radon is consistent with that on xenon in iron, and is presumably due to core polarization (CP) effects. The sign of the field on radium in iron is opposite to that found for lighter impurities with similar valence structure and opposite to the prediction of Balabanov and Delyagin [3], though the relatively small magnitude is consistent with other impurities having near closed shell electron structure. The modest negative field on thorium ($5f^0$, $6d^2$, $7s^2$) cannot be simply explained. Reference [3] would predict a positive field of approximately the observed magnitude. Systematics, however, of other impurities with d electrons in the conduction band would imply a large negative field, primarily due to conduction electron polarization (CEP).

In the first half of the rare earth series, the induced hyperfine fields are large and positive due to partially aligned inner 4 f shell angular momentum. The 5 f analog does not hold for uranium, where the measured field is negative and less than one million gauss. Friedel [18] has suggested a hybridized 5 f − 6 d band for uranium which overlaps the iron 3 d band. This picture has been successfully used to explain the susceptibility of U-Fe alloys [19] as well as the ferromagnetism of UFe_2 [20]. The negative field on uranium

TABLE I

Hyperfine fields on heavy impurities in iron. Uncertainties in the various measurements are discussed in the references.

Impurity	H_{eff}(Fe) kOe	References
$^{220}_{86}$Rn	+900	ANSALDO (E.), GRODZINS (L.) and KALISH (R.), *Physics Letters*, 1969, **30B**, 538.
$^{224}_{88}$Ra	−220	Ibid.
$^{223}_{88}$Ra	−105	LEVANONI (M.) and ZAWISLAK (F. C.), *Phys. Rev. C*, 1970, **2**, 672.
$^{232}_{90}$Th	−230	This work.
$^{238}_{92}$U	−660	Average of this work and reference [16].
$^{239}_{93}$Np	+170 (Ni)	ANSALDO (E.) and GRODZINS (L.), *Physics Letters*, 1970, **32B**, 6.
$^{238}_{94}$Pu	+620	RAVN (H.) et al., Proc. Intl. Conf. on Radioactivity in Nuclear Spectroscopy, Nashville, 1969.

in iron also suggests a d-band behavior similar to the 5-d transition elements. The positive fields on neptunium and plutonium however, suggest that in these cases, the 5 f angular momentum is not fully quenched and partially aligned, contributing a positive hyperfine field much larger than the negative CEP effects.

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