

# Investigation of the temperature dependence of the effective magnetic fields on Pu nuclei in nickel and iron matrices

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To determine more accurately the mechanism of production of hyperfine magnetic fields on actinide nuclei implanted in ferromagnetic matrices, an investigation was made of the temperature dependence of the effective magnetic fields on Pu nuclei in Ni and Fe. The measurements were made by the method of perturbed  $\alpha$ - $e$  correlations. It was shown that at  $T = 300$  K the fields are nearly equal:  $H(\text{PuNi}) = +(1.5 \pm 0.2)$  MOe,  $H(\text{PuFe}) \approx +1.3$  MOe. A significant temperature dependence of  $H(\text{PuNi})$  was observed, whereas  $H(\text{PuFe})$  is almost constant over the interval 40-490 K. The positive sign of the effective magnetic field and the presence of temperature dependence of the field indicate strong localization of the  $5f$  electrons in the PuNi system. Processing of the experimental data in the localized-moment, exchange-field model gives  $H_{\text{ex}}(\text{PuNi}) = -(2.2 \pm 0.2)$  MOe; the contribution to the hyperfine magnetic field resulting from polarization of the core and of the conduction electrons gives  $H_{\text{cp}} + H_{\text{cep}} = -(1.2 \pm 0.2)$  MOe. The absence of temperature dependence for  $H(\text{PuFe})$  is discussed on the basis of a larger exchange field than in PuNi ( $|H_{\text{ex}}| \geq 7$  MOe) and of the presence of quadrupole interactions.

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## 1. INTRODUCTION

Investigations of the hyperfine magnetic fields of actinide atoms in metals, alloys, and compounds give important information about the electronic structure of the actinides.

The uniqueness of the properties of the actinides is to a large degree determined by the closeness of the binding energies of the  $6d$ ,  $7s$  and  $5f$  electrons. The  $5f$  electrons play an essential role in shaping the magnetic properties of the actinides; the conditions for their localization depend both on the atomic number of the actinide itself and on the properties of the neighboring atoms. Competition between Coulomb correlations and the centrifugal potential produces a tendency toward localization of the  $5f$  electrons with increase of  $Z$ .<sup>[1]</sup> Ferromagnetic matrices produce more favorable conditions for localization of  $5f$  electrons of actinides, dissolved in such matrices, than do elemental actinide metals. This conclusion can be drawn from the similarity of the magnitudes and signs of the effective fields for Np, Pu and Cm to the corresponding data for the rare-earth elements.<sup>[2]</sup> The principal mechanism for production of the effective magnetic fields for lanthanides in ferromagnetic matrices is the orbital magnetism of the strongly localized  $4f$  shell.

It has been shown recently<sup>[3,4]</sup> that in rare-earth ions Nd, Sm, and Tm implanted in Ni and Fe, there is a strong temperature dependence of the effective magnetic fields; this is due to the presence of exchange interaction between the strongly localized magnetic moment and the matrix. One might expect a similar dependence for the actinides also; its presence would be a good indicator of localization of the  $5f$  electrons. From the temperature dependence of the fields one can also obtain information about the quantities that describe the interaction of impurity atoms with the matrix.

The aim of the present work is the investigation of the temperature dependence of the effective magnetic fields in Pu atoms implanted in iron and nickel matrices. The method used is that of integrated perturbed angular correlations. For investigation over a wide range of temperature, the method of perturbed angular correlations has a great advantage over other methods using nuclear radiation (nuclear gamma resonance, oriented nuclei).

## 2. RESULTS OF THE MEASUREMENTS

In this work, use was made of the cascade  $0^+ \alpha 2^+ e 0^+$  ( $e_M$  are internal-conversion electrons of the  $M$  shell), excited in the decay  $^{244}\text{Cm} \alpha ^{240}\text{Pu}$ . The source of  $^{244}\text{Cm}$  was deposited on the surface of an iron or nickel foil; the implantation of the Pu atoms into the corresponding matrix was accomplished by virtue of the recoil energy during the  $\alpha$  decay. The level  $2^+$  (42.8 keV) has lifetime  $\tau_n = 0.24$  nsec and is populated, in the  $\alpha$  decay, with probability 24%. In work with cascades that include  $\alpha$  particles and internal-conversion electrons ( $\alpha$ - $e$  cascades), it is necessary to apply a special methodology, which has been described in detail.<sup>[5]</sup> The research was performed with completely magnetized foils of iron and nickel, whose temperature could be varied from 40 to 675 K.

### Pu in nickel (Pu/Ni)<sup>1)</sup>

The effective magnetic field for PuNi has not been known previously. Figure 1 shows the variation of

$$R = 2 \frac{N_i(45^\circ) - N_i(45^\circ)}{N_i(45^\circ) + N_i(45^\circ)} \quad (1)$$

with the value of the field  $B_e$  that magnetizes the foil with the source, for various temperatures;  $N_i(45^\circ)$  and

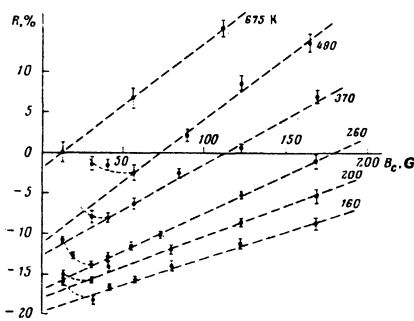


FIG. 1. Variation of  $R$  with  $B_c$  at various temperatures, for  $\text{PuNi}$ .

$N_{\pm}(45^\circ)$  are the numbers of  $\alpha$ - $e$  coincidences for two opposite directions of the field, when the angle between the  $\alpha$  and  $e$  detectors is  $\theta = 45^\circ$ .

It was shown in Refs. 2 and 5 that from this variation one can derive the perturbation parameters of the angular correlation function, namely: the angle  $\Delta\theta_L$  resulting from Larmor precession of the nuclear spin in the internal field, and the suppression factor  $G_{22}$ .

The strong temperature dependence of  $R(B_c)$  (Fig. 1) means that an important role in the perturbation of angular correlation (PAC) is played by dynamic interactions; that is, interactions of time-fluctuating fields with the electromagnetic moments of the nuclei. In this case, to derive the parameters of the perturbation of angular correlation it is reasonable to use the theory of Abragam and Pound.<sup>[6, 7]</sup> The parameters thus found, for various temperatures, are shown in Table I.

According to theory<sup>[6, 7]</sup>

$$\text{tg } 2\Delta\theta_L = 2G_{22}\omega_L\tau_n, \quad (2)$$

$$G_{22} = (1 + \lambda_2\tau_n)^{-1}, \quad \lambda_2 = 2\omega_m^2\tau_c, \quad (3)$$

where

$$\omega_L = -(g\mu_n/\hbar)\langle H_{\text{int}}^z \rangle, \quad \omega_m = -(g\mu_n/\hbar)\langle H_{\text{int}}^2 \rangle^{1/2};$$

$\langle H_{\text{int}}^z \rangle$  and  $\langle H_{\text{int}}^2 \rangle^{1/2}$  are averaged values of the fields along the external magnetic field and along the ionic field, respectively. The theory is applicable when  $\tau_c \ll \tau_n$  and  $\omega_m\tau_c \ll 1$ , where  $\tau_c$  is the correlation (relaxation) time.

The applicability of the Abragam-Pound theory in the case of  $\text{PuNi}$  can be tested by making an estimate of the relaxation time of the Pu and Ni atoms. Here we may start from the fact that  $\tau_c \sim T^{-1}$ , and consequently, also,  $\lambda_2\tau_n \sim T^{-1}$ . Figure 2 shows the variation of  $\lambda_2\tau_n$  with  $T^{-1}$ , calculated by formula (3) from the  $G_{22}$  data. It is evi-

TABLE I. Data on perturbed angular correlations for  $^{240}\text{Pu}$  in Ni.

T, K	$G_{22}$	$\omega_L\tau_n$ , rad	H, MOe	T, K	$G_{22}$	$\omega_L\tau_n$ , rad	H, MOe
40	0.23±0.05	1.4±0.4	4.0±1.1	310	0.37±0.04	0.53±0.05	1.52±0.14
80	0.26±0.02	1.09±0.13	3.1±0.4	370	0.41±0.03	0.35±0.02	1.00±0.06
160	0.27±0.02	1.16±0.08	3.3±0.2	495	0.51±0.06	0.16±0.02	0.46±0.06
200	0.32±0.02	0.93±0.07	2.7±0.2	570	0.66±0.12	0.100±0.025	0.29±0.07
260	0.36±0.01	0.80±0.03	2.3±0.1	675	0.55±0.06	0.03±0.2	0.10±0.05

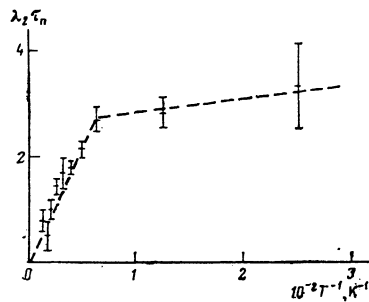


FIG. 2. Variation of  $\lambda_2\tau_n$  with  $T^{-1}$  for  $\text{PuNi}$ .

dent that over a broad range of temperature, from 160 to 675 K, linearity is maintained:  $\lambda_2\tau_n = (440 \pm 40)T^{-1}$ ; for  $T \approx 160$  K, a clear breakdown of the linearity occurs. This breakdown can apparently be attributed to inapplicability of the Abragam-Pound theory when  $T < 160$  K. If at the critical point  $T = 160$  K we make an estimate of the relaxation time by formula (3), then from the experimental value of  $\lambda_2\tau_n$  we get  $\tau_c \approx 60$  nsec. If we take into account that  $\tau_n = 240$  nsec, then it is evident that when  $T < 160$  K, the condition for applicability of the Abragam-Pound theory,  $\tau_c \ll \tau_n$ , ceases to be satisfied. Therefore in the further calculations, the experimental values obtained at  $T = 40$  and  $80$  K were not used. The field  $H$  for 675 K was also disregarded, because it was obtained at a temperature above the Curie point for nickel (631 K).

In the case  $\tau_c \approx \tau_n$ , the theory becomes considerably more complicated.<sup>[8]</sup> Suppression factors of the angular correlation have been calculated<sup>[3, 9]</sup> as functions of  $x = \tau_c/\tau_n$  with allowance for the magnetic interaction alone. No general analytic expression of the type (3) was given, and the correlation time  $\tau_c$  at room temperature (300 K) appears as a parameter. In an experiment with  $\text{TmFe}$  it was shown that the Abragam-Pound theory is inapplicable for  $T < 200$  K; but the variation of the parameter  $\tau_c(300 \text{ K})$  does not lead to a significant change of the results of the fitting, and  $\tau_c(300 \text{ K})$  is close to the value obtained according to the theory of Refs. 6 and 7.

### Pu in iron ( $\text{PuFe}$ )

In this case, the parameters of the perturbation of angular correlation shown no appreciable variation with temperature. For  $\text{PuFe}$ , in addition to measurement of the values of  $R$ , the value of the anisotropy  $A = [N(0) - N(90^\circ)]/N(90^\circ)$  was measured, since this quantity is more sensitive to the suppression factor.

Figure 3 gives the data for values of  $\Delta\theta_L$  and of the anisotropy  $A$  as functions of  $T$ . If one processes the data according to the theory of Refs. 6 and 7, and if one represents  $G_{22}$  and  $H$  as linear functions of the temperature, then ( $H$  is in kilosterds)

$$G_{22} = (0.36 \pm 0.01) + (0.65 \pm 0.18) \cdot 10^{-4} T, \quad H = (1370 \pm 60) - (0.14 \pm 0.17) T.$$

The absence of appreciable temperature dependence for  $\text{PuFe}$  suggests that in this case the principal role in the suppression of angular correlation is played by

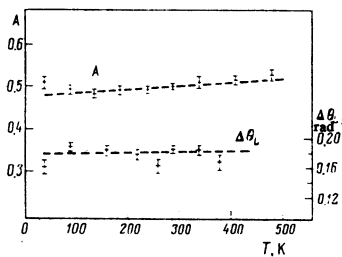


FIG. 3. Anisotropy  $A$  and angle of shift of the angular correlation function,  $\Delta\theta_L$ , at various temperatures, for  $\text{PuFe}$ .

static fields. It has been shown<sup>[10]</sup> that in the case of Rn and Ra in an iron matrix, there is a strong static quadrupole interaction, due to the presence of a quadrupole moment in the excited state of the nucleus and to randomly directed static gradients of the electric field. Such gradients, in the cubic lattice of iron, can occur as a result of radiational defects<sup>[11]</sup> produced during implantation of the atoms in the matrix. In the case of Pu, the static quadrupole interaction may be still larger in view of the fact that the quadrupole moment of the state  $2^+ 42.8$  keV of  $^{240}\text{Pu}$  is about three times larger than that of the state  $2^+ 241$  keV of  $^{220}\text{Rn}$ . Apparently the strong suppression of the angular correlation in the case of Pu in Fe must be attributed principally to the quadrupole interaction, and the processing should be carried out according to a theory that allows for the simultaneous action of a transverse magnetic field and of static quadrupole interaction.<sup>[12]</sup> Such an analysis is difficult to carry out in our case because of the proximity of the observed suppression to the "rigid core" value  $G_{22} = 0.37$ .

Thus it is not possible to derive a value of  $H(\text{PuFe})$  in correct form from our experimental data; the true value of  $H(\text{PuFe})$  may prove to be significantly larger than the value obtained above according to the Abragam-Pound theory, and therefore this value must be regarded as only an estimate: for  $T = 300$  K,

$$H(\text{PuFe}) \approx +1.3 \text{ MOe.}$$

### 3. ANALYSIS OF THE TEMPERATURE DEPENDENCE

Thus the effective magnetic fields for the systems  $\text{PuFe}$  and  $\text{PuNi}$  have positive signs and are comparable in value at  $T = 300$  K, but they show significantly different temperature variations:  $H(\text{PuNi})$  changes by an order of magnitude over the temperature interval investigated, whereas for  $H(\text{PuFe})$  no variation was detected.

The character of the temperature variation of  $G_{22}$  and  $H$  for Pu in nickel is reminiscent of a similar variation for Nd, Sm, and Tm in Ni and Fe<sup>[3, 4]</sup> and may indicate localization of the  $5f$  electrons in Pu and Ni. As a first step in the processing of the data, we use the simplest model:

1) We suppose that the experimentally found field at the nucleus is entirely determined by the completely localized  $5f$  shell of Pu:  $H = H_{5f}$ ;

2) The interaction of the Pu ion with the matrix occurs solely by virtue of exchange interaction—we ne-

glect crystalline fields;

3) We assume that Pu in a nickel matrix is in the state  $\text{Pu}^{4+}$  with electronic configuration  $5f^4$ , described by the term  $^5I_4$ .

In the presence of a localized moment, the temperature dependence of the hyperfine magnetic field is expressed in terms of the Brillouin function  $B(X)$ :

$$B(X) = H_{5f}(T)/H_{5f}(0) = \langle J_z \rangle / J, \quad (4)$$

$$X = 2\mu_B(g_J - 1)JH_{ex}/kT, \quad (5)$$

where  $\mu_B$  is the Bohr magneton,  $g_J$  is the Landé factor,  $J$  is the spin of the atom,  $H_{ex}$  is the exchange field, and  $H_{5f}(0)$  is the value of the hyperfine magnetic field of the free ion at  $T = 0$  K; for  $\text{Pu}^{4+}$  we have  $H_{5f}(0) = 5240$  kOe.<sup>[13]</sup> For the exchange field  $H_{ex}$ , the following temperature dependence is assumed:  $H_{ex} = H_{ex}(0)\sigma(T)/\sigma(0)$ , where  $\sigma(T)$  is the magnetization of the matrix.

If we start from this simple model and process the experimental data by the method of least squares, then with  $\chi^2 = 15$

$$H_{ex}(0) = -(1.13 \pm 0.13) \text{ MOe.}$$

It is clear that the model adopted is very crude. First of all, it must be taken into account that in actinide atoms the fields  $H_{cp}$  due to polarization of the atomic core by the  $5f$  shell and  $H_{cep}$  due to polarization of the conduction electrons of the matrix are very large. Estimates<sup>[14]</sup> show that the values of these fields for actinides in Ni and Fe are comparable in value with the fields  $H_{5f}$ ;  $H_{cp}$  and  $H_{cep}$  are opposite in sign, and  $H_p = H_{cp} + H_{cep} \approx -1$  MOe. Since there are at present only crude estimates for the field  $H_p$ , we shall try to take account of it by introducing into the model still another parameter:  $H_p(0)$ . Thus the observed experimental field is  $H(T) = H_{5f} + H_p$ , where  $H_p(t) = H_p(0)\sigma(T)/\sigma(0)$ .

Processing of the experimental data with allowance for two parameters,  $H_{ex}(0)$  and  $H_p(0)$ , gives  $H_{ex}(0) = -(2.2 \pm 0.2)$  MOe and  $H_p(0) = -(1.2 \pm 0.2)$  MOe, with  $\chi^2 = 4.7$ .

The Brillouin function with these parameters is shown in Fig. 4 together with the experimental points.

Further improvement of the model, with the available experimental material, is scarcely feasible. Allowance

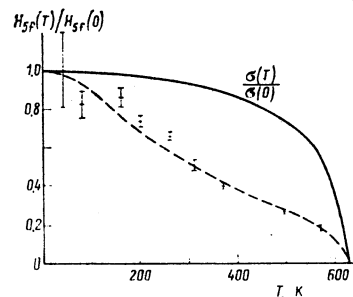


FIG. 4. Reduced values of  $H$  for  $\text{PuNi}$  as a function of temperature. The dotted curve was obtained by fitting by the method of least squares, for the model of pure exchange interaction of localized  $5f$  electrons with the parameters  $H_{ex}(0) = -(2.2 \pm 0.2)$  MOe and  $H_p = -(1.2 \pm 0.2)$  MOe.

for crystalline fields leads to a departure from the Brillouin curve only at small  $T$ ,<sup>[4]</sup> where our experimental errors are large. Moreover, a correct calculation of these fields is difficult, because the true environment of the impurity atoms is unknown. In Ref. 4, allowance for the crystalline field was made on the assumption that the impurity atoms are located on sites of the undisturbed cubic Ni or Fe lattice.

The actual situation, because of radiational defects of the lattice, is much more complicated<sup>[15]</sup>; furthermore, the environment of an impurity atom may be different at different temperatures, and this leads to a distortion of the temperature dependence  $H(T)$ .

We shall now consider the problem of the absence of noticeable temperature variation of the hyperfine field  $H$  in Fe. The fact that the field in Fe is positive certainly suggests that the  $5f$  electrons play a decisive role in its production.<sup>[2, 13]</sup> Therefore one must attempt to explain the temperature independence of the field within the framework of a localized  $5f$ -shell model. This proves possible if one assumes that the exchange forces are several times larger for  $\text{PuFe}$  than for  $\text{PuNi}$ . With increase of  $H_{ex}$ , the splitting of the atomic levels increases; therefore  $\langle J_z \rangle \rightarrow J$ , and the Brillouin function approaches the  $\sigma(T)/\sigma(0)$  curve corresponding to the matrix, and this leads to a lessening of the temperature dependence over the temperature interval investigated, 40–490 K. It should be remembered that the Curie temperature for iron is 1043 K. By processing the experimental data on the temperature variation of  $H(\text{PuFe})$ , we get  $|H_{ex}(\text{PuFe})| \approx 7 \text{ MOe}$ .

With this approach, one would expect that at  $T = 300 \text{ K}$  the value of  $H(\text{PuFe})$  would be significantly larger than  $H(\text{PuNi})$ . It must be kept in mind, however, that, as was discussed above, the strong static quadrupole interaction in  $\text{PuFe}$  makes correct determination of  $H(\text{PuFe})$  difficult; and the field obtained,  $H \approx +1.3 \text{ MOe}$ , must be considered only a lower bound. Furthermore, the effects of polarization play a greater role in Fe than in Ni, and this may lead to an appreciably smaller field in Fe as compared with Ni at  $T = 0 \text{ K}$ .

The large exchange fields in the case of  $\text{PuFe}$  lead to a practical disappearance of time-dependent fields, and the method of integrated perturbations of angular correlations, which measures resultant effects, becomes sensitive to the presence of static quadrupole interactions. One must also keep in mind that the quadrupole interaction is stronger in Fe than in Ni; for the lanthanides, it is two to five times larger in Fe than in Ni.<sup>[15]</sup> In the case of  $\text{PuNi}$ , because of the smaller exchange interactions, a larger role is played by time-dependent

fields; against their background, in experiments with integrated perturbations of angular correlations, the effect of static fields becomes less noticeable.

Thus the experimental data on Pu in nickel and iron can be explained from a single point of view, starting from the assumption of localization of the  $5f$  electrons and of the presence of exchange interactions several times larger in the case of  $\text{PuFe}$  as compared with  $\text{PuNi}$ . The data on lanthanides in Fe and Ni also indicate that the exchange fields of lanthanides in iron are six to eight times larger than in Ni<sup>[4, 15]</sup>; at the same time, these fields are several times smaller than the exchange fields for Pu in Ni and Fe.

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<sup>1)</sup>The chemical symbols of the elements that constitute the matrix are represented throughout the article by italic letters.

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