Hyperfine field of einsteinium in iron and nuclear magnetic moment of ^{254}Es

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(Received 15 April 2009; published 29 June 2009)

The angular distributions of γ rays and α particles from oriented ^{250}Bk, ^{253,254}Es, and ^{255}Fm nuclei were investigated to extract hyperfine interaction information for these actinide impurities in an iron host lattice. The hyperfine field of einsteinium in iron was found to be $|B_{hf}(\mathrm{EsFe})| = 396(32)$ T. With this value the magnetic moment of ^{254}Es was then determined as $|\mu| = 4.35(41)\ \mu_N$.

DOI: 10.1103/PhysRevC.79.064322 PACS number(s): 21.10.Ky, 27.90.+b, 76.60.Jx

I. INTRODUCTION

Precise values of magnetic hyperfine fields [1] allow the determination of nuclear magnetic moments by experimental methods such as, e.g., integral perturbed angular correlation (IPAC) [2] and time differential perturbed angular distribution (TDPAD) [3], or also low temperature nuclear orientation (LTNO) [4] and nuclear magnetic resonance on oriented nuclei (NMR/ON) (see, e.g., [5]). Further, understanding and reproducing hyperfine fields is a strong test for *ab initio* condensed matter methods (e.g., [6–8]). Whereas the hyperfine fields for substitutional impurities in bcc Fe are at present well understood for almost all elements in the first five periods of the periodic table [8–12], sizable differences between theory and experiment remain for the heavier 5*d* impurities [13]. Recently, the hyperfine fields of lanthanide and actinide impurities in Fe were calculated with modern *ab initio* methods [14,15]. Here we present the first experimental determination of the magnetic hyperfine field of einsteinium in Fe, using the LTNO method. The same measurements also yielded the nuclear magnetic moment for the 7^+ ground state of ^{254}Es, as well as the magnetic hyperfine interaction strength (i.e., the product of the magnetic moment μ and the magnetic hyperfine field B_{hf}) for ^{250}Bk and ^{255}Fm in iron.

II. EXPERIMENT

The Es activity was produced by neutron irradiation of ^{252}Cf in a reactor in Dimitrovgrad (Russia), followed by radiochemical separation of the accumulated Es and irradiation of this separated Es activity to improve the isotopic content. Details of the radiochemical procedures used can be found in Ref. [16]. The resulting batch of activity contained the isotopes ^{253}Es ($t_{1/2} = 20.5$ d; 532 MBq), ^{254}Es ($t_{1/2} = 275.7$ d; 15.4 MBq), ^{255}Es ($t_{1/2} = 39.8$ d; 0.357 MBq), ^{255}Fm ($t_{1/2} = 20.1$ h; 0.328 MBq), ^{249}Cf ($t_{1/2} = 350.6$ y; 0.38 MBq) and ^{252}Cf ($t_{1/2} = 2.645$ y; <0.27 MBq). As the α transitions in the decay of the two californium isotopes have energies well below those for ^{253,254,255}Es and ^{255}Fm they did not pose a problem for the measurements. The activity was loaded into the oven of a positive surface ionization ion source of the isotope separator at Bonn (Germany), mass separated and implanted at room temperature at an acceleration voltage of 160 kV into a high purity (99.99%) annealed 100 μm Fe foil. The total number of ions implanted was about 10^8.

The sample thus obtained was soldered with Wood's eutectic onto a Cu sample holder and top-loaded into a ^{3}He-^{4}He dilution refrigerator [17] in Leuven (Belgium) for the nuclear orientation experiments. Two experimental campaigns of four weeks each were performed. The α decay of the sample was observed with up to six Si PIN-diodes mounted inside the 4.2 K shield of the refrigerator at angles of 15°, 78°, and 90° with respect to the nuclear orientation axis (i.e., the external orienting magnetic field). The energy resolution of these detectors at their operating temperature of about 10 K was 20 keV at 6 MeV. The part of the α spectrum containing the lines from ^{253}Es, ^{254}Es, and ^{255}Fm, registered by one of these detectors, is shown in Fig. 1.

To measure γ spectra, two large volume HPGe detectors were installed at 0° and at 90° outside the refrigerator. The sample temperature was monitored with a ^{54}MnNi nuclear orientation thermometer [18] that was soldered onto the back side of the sample holder and that was previously calibrated against a ^{60}CoCo single crystal nuclear orientation thermometer [19]. Count rates $N(\theta)$ were recorded for both oriented [$N(\theta)_{\mathrm{cold}}$ at $T < 300$ mK] and nonoriented [$N(\theta)_{\mathrm{warm}}$ at $T \approx 4$ K] nuclei. From these the angular distribution

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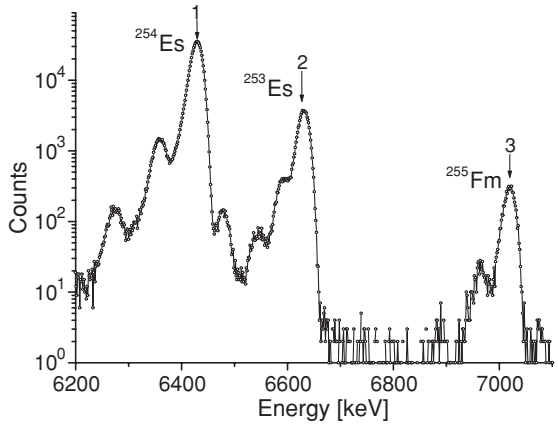

FIG. 1. α spectrum observed with one of the Si PIN-diode particle detectors. The energies of the three α lines for which anisotropies were determined are 6429 keV (line 1), 6633 keV (line 2), and 7022 keV (line 3).

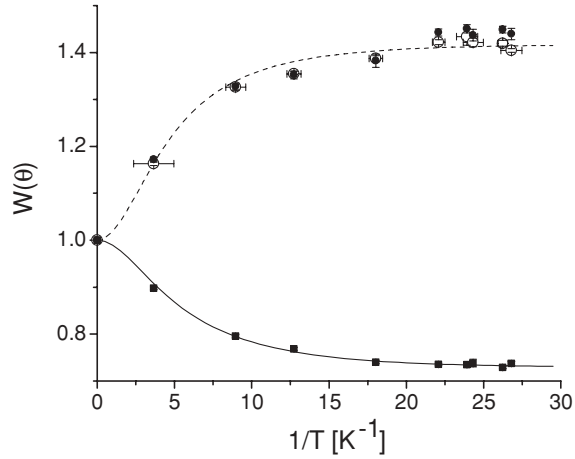

FIG. 2. Simultaneous fit of the anisotropies $W(\theta)$ for the favored $7/2^+ \rightarrow 7/2^+6633$ keV α transition of ^{253}Es observed with two detectors at 15° (dashed line) and one at 90° (solid line). Where error bars are not shown they are smaller than the size of the data point.

$W(\theta) = N(\theta)_{\text{cold}}/N(\theta)_{\text{warm}}$ is obtained which, for α particles and γ rays, can be fitted to the theoretical function [20]

$$W(\theta) = 1 + f \sum_{k=\text{even}} A_k U_k B_k Q_k P_k(\cos\theta). \qquad (1)$$

Here the implantation parameter f represents the fraction of nuclei experiencing the full orienting hyperfine interaction, whereas the rest (1-f) is assumed to feel no hyperfine interaction at all. The reliability of this model was shown before [21,22] (see also Sec. IV). The B_k describe the nuclear orientation and depend on the ratio $\Delta_M/T = \mu B/Ik_BT$, with μ the nuclear magnetic moment, B the total magnetic field at the site of the nucleus ($B = B_{\text{ext}} + B_{hf}$ with B_{ext} the external magnetic field and B_{hf} the hyperfine field), I the spin of the oriented state, k_B the Boltzmann constant and T the temperature of the sample. Further, A_k are the angular distribution coefficients which depend on the properties of the transition observed, P_k are the Legendre polynomials and Q_k the solid angle correction factors which account for the finite size of the source and detectors. The coefficients U_k are unity for the detection of α radiation originating directly from the oriented nuclear state. For the detection of following γ radiation they account for the reorientation by intermediate radiation transitions.

Experimental anisotropies could be determined for α particles from the decay of ^{253,254}Es and ^{255}Fm (daughter isotope of ^{255}Es), and for the 1031 keV γ transition in the decay of ^{250}Bk (daughter isotope of ^{254}Es). Data are shown in Figs. 2–5. The α anisotropies for the Es and Fm nuclei previously already provided valuable information on the α particle emission process in the case of oriented nuclei through the A_k angular distribution coefficients [16]. However, since for the Es isotopes ^{253,254}Es full saturation of orientation and for ^{255}Fm nearly full saturation was reached, the temperature dependent parameters, B_k in Eq. (1), could be obtained independent of all other (i.e., temperature independent) parameters as, e.g., the A_k coefficients. As the B_k parameters depend on the hyperfine interaction strength μB this then allowed extracting information on the magnetic moment μ and the magnetic hyperfine field B_{hf} of the einsteinium isotopes studied, independent of the other parameters that determine $W(\theta)$. It was verified that the term depending on A_6 could be neglected [16] such that one could restrict Eq. (1) to the $k = 2, 4$ terms. Also, it was not necessary to assume a combined magnetic and electric interaction as calculations [15] and experimental results [16] have shown that there is no appreciable electric field gradient for Es impurities in Fe. This is due to an almost exact cancellation of the $5f$ charge anisotropy by the $6p$ core anisotropy with opposite sign that is induced by the $5f$ anisotropy. Calculations using both the local density approximation (LDA) and B3PW91-like hybrid functionals predict an electric field gradient for Es impurities in Fe well below 5×10^{21} V/m^2 [15].

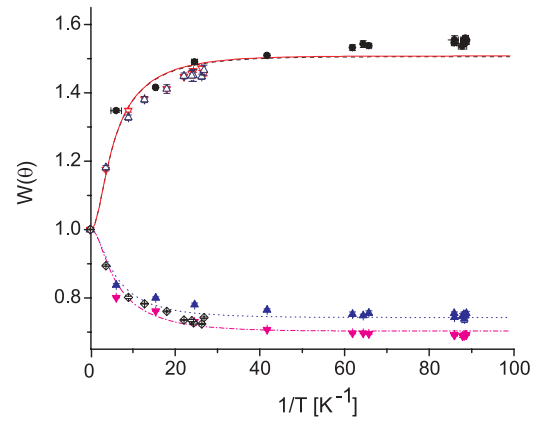

FIG. 3. (Color online) Simultaneous fit of the anisotropies $W(\theta)$ for the favored $(7^+) \rightarrow (7^+)$ 6429 keV α transition of ^{254}Es observed with three detectors at 15° (upper line), one at 78° (dotted line at bottom), and two at 90° (dot-dashed line at bottom).

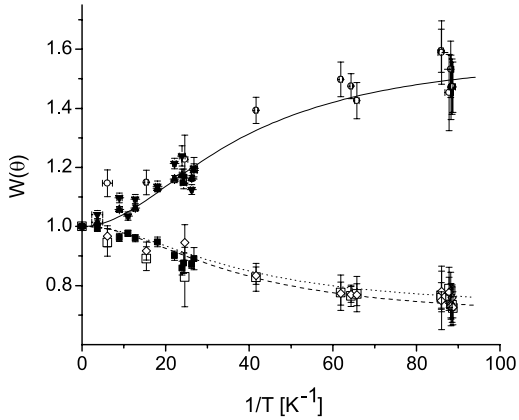

FIG. 4. Simultaneous fit of the anisotropies $W(\theta)$ for the favored $7/2^+ \rightarrow 7/2^+$ 7022 keV α transition of ^{255}Fm observed with three detectors at 15° (solid line), one at 78° (dotted line), and two at 90° (dashed line).

III. RESULTS

The anisotropy curves obtained for ^{253}Es (see Fig. 2) were fitted simultaneously with three free parameters, i.e., fA_2 and fA_4, as well as the Zeeman splitting Δ_M of the magnetic substates of the $I^\pi = 7/2^+$ ground state of ^{253}Es, i.e.,

$$\Delta_M[\mathrm{mK}] = \frac{0.366\ \mu[\mu_N]\ B[\mathrm{T}]}{I[\hbar]}. \qquad (2)$$

Assuming different values for the electric field gradient, ranging from $V_{zz} = -20 \times 10^{21}$ V/m^2 to $V_{zz} = +20 \times 10^{21}$ V/m^2, the results listed in Table I were obtained. As can be seen, varying V_{zz} changes the fitted values for fA_2 and fA_4 only within one or two standard deviations, while affecting significantly the value for Δ_M (and thus of B_{hf}). Indeed, due to the saturation of the α anisotropy (see Fig. 2) the correlation between the temperature dependent Zeeman splitting Δ_M and the electric field gradient V_{zz} on the one hand, and the temperature independent parameters fA_2 and fA_4 on the other hand, is small. Conversely, this also implies that the value of Δ_M (and thus the hyperfine field value), as derived, is not influenced by the fraction at good sites f.

TABLE I. Values obtained for fA_2, fA_4, and Δ_M, assuming different values for the electric field gradient V_{zz} and simultaneously fitting the three anisotropy curves for the 6633 keV favored α transition in the decay of ^{253}Es (Fig. 2). For the quadrupole moment the value $Q = 6.7(8)$ eb [23] was used. As can be seen the values obtained for Δ_M depend strongly on V_{zz}, whereas fA_2 and fA_4 are almost insensitive to V_{zz}. The error bars are purely statistical. The values for B_{hf} are obtained from Δ_M with Eq. (2) using $\mu(^{253}\mathrm{Es}) = +4.10(7)\ \mu_N$ [23].

V_{zz} [10^{21} V/m^2]	fA_2	fA_4	Δ_M [mK]	B_{hf} [T]
+20	0.356(2)	−0.083(3)	147.3(21)	343
+5	0.358(2)	−0.084(3)	164.2(21)	383
0	0.359(2)	−0.085(3)	169.8(21)	396
−5	0.360(2)	−0.085(3)	175.7(21)	409
−20	0.361(2)	−0.086(3)	193.4(21)	449

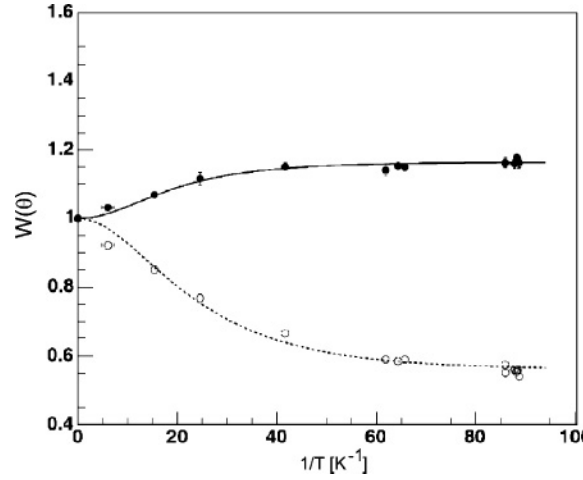

FIG. 5. Simultaneous fit of the $W(0°)$ (dashed line) and $W(90°)$ (solid line) anisotropies for the $2^+ \rightarrow 0^+$ pure $E2$ 1031 keV γ transition in the decay of ^{250}Bk.

Assuming the electric field gradient to be zero [15,16], using the magnetic moment $\mu = +4.10(7)\ \mu_N$ for ^{253}Es [23] and neglecting the small external magnetic field $B_{\mathrm{ext}} = 0.1$ T that was used to polarize the Fe host foil, one finds $|B_{hf}(\mathrm{Es\underline{Fe}})| = (396 \pm 8_{\mathrm{stat}})$ T for the magnetic hyperfine field of Es impurities in an iron host lattice. Increasing the statistical error by $\sqrt{\chi^2/\nu} = 2.6$ to take into account the not being unity of the χ^2 per degree of freedom and further including systematic errors that take into account the precision with which the detection geometry was known ("syst1") and that allow for $|V_{zz}| \leqslant 5 \times 10^{21}$ V/m^2 ("syst2", see Table I) yields

$$|B_{hf}(\mathrm{Es\underline{Fe}})| = [396 \pm 21_{\mathrm{stat}} \pm 20_{\mathrm{syst1}} \pm 13_{\mathrm{syst2}}]\ \mathrm{T}, \qquad (3)$$

which can be written as

$$|B_{hf}(\mathrm{Es\underline{Fe}})| = 396(32)\ \mathrm{T}. \qquad (4)$$

No sign is given as LTNO with detection of γ rays or α particles is not sensitive to the sign of the hyperfine interaction μB.

Note, finally, that the values of fA_2 and fA_4 for $V_{zz} = 0$, corrected for the fraction at good sites $f = 0.67(10)$ obtained from a fit of the anisotropy of the 1031 keV γ transition in the decay of ^{250}Bk [16], have yielded the values for the A_2 and A_4 angular distribution coefficients for the 6633 keV α transition of ^{253}Es that were reported in [16].

For the isotopes ^{254}Es, ^{255}Fm, and ^{250}Bk the fit results are summarized in Table II. For ^{254}Es, a simultaneous fit of the six anisotropy curves for the α decay of the $I^\pi = 7^+$ ground state of this isotope is shown in Fig. 3. With the same free parameters as before, i.e., fA_2, fA_4, and Δ_M, this fit yielded $\Delta_M = 90.1(44)$ mK (taking into account the value of χ^2/ν). Using the above mentioned value for the hyperfine field of einsteinium in iron, the magnetic moment of the ground state of ^{254}Es is then found to be

$$|\mu(^{254}\mathrm{Es})| = 4.35(41)\ \mu_N. \qquad (5)$$

This adds a new experimental value to the scarce amount of magnetic moment data for the actinides. Indeed, magnetic

TABLE II. Results from the fits of the α anisotropies for ^{254}Es (6429 keV α transition) and ^{255}Fm (7022 keV α transition), and the 1032 keV γ ray anisotropy in the decay of ^{250}Bk.

Isotope	I^π	A_2	A_4	Δ_M [mK]	f
^{254}Es[a]	7^+	0.47(7)	−0.030(14)	90.1(44)	–
^{255}Fm[a]	$7/2^+$	0.65(10)	0.04(9)	23.3(18)	–
^{250}Bk	2^-	–[b]	–[b]	57.2(42)	0.67(10)

[a]To extract A_2 and A_4 the value $f = 0.67(10)$ obtained from the 1032 keV γ ray in the decay of ^{250}Bk was used.
[b]For the 1032 keV $2^+ \rightarrow 0^+$ pure $E2$ γ transition with $A_2 = -0.5976$ and $A_4 = -1.0690$, the $L = 0$, $L = 1$, and $L = 2$ contributions in the first forbidden $2^- \rightarrow 2^+$ β transition preceding this γ transition were fitted via the U_k coefficients yielding, respectively, $a_0 = 0.60(8)$, $a_1 = 0.40(8)$, and $a_2 = 0.00(5)$.

moments were determined for only about 20 of the more than 200 ground and isomeric states of the actinides [24].

The magnetic hyperfine interaction strength μB in Fe has also been obtained for two other isotopes, viz. ^{255}Fm and ^{250}Bk. From the α anisotropies observed for ^{255}Fm (with $I^\pi = 7/2^+$; Fig. 4), the fit resulted in $|\mu B(^{255}\text{FmFe})| = 223(17)$ μ_NT (including the χ^2/ν correction as well as corrections for the precision with which the detection geometry was known). From the anisotropy for the 1031 keV γ ray in the decay of ^{250}Bk (with $I^\pi = 2^-$; Fig. 5), $|\mu B(^{250}\text{BkFe})| = 313(23)$ μ_NT was obtained (with similar conditions for the error determination). Unfortunately, however, neither the hyperfine field of fermium or berkelium in iron, nor the magnetic moment of the ground states of ^{255}Fm or ^{250}Bk are known, so that no further information can be derived from these results.

IV. DISCUSSION

Our result for the hyperfine magnetic field of einsteinium in Fe has been extensively discussed in Ref. [15] and seems to indicate a large shift of the delocalization-localization transition for the $5f$ electrons. In Ref. [15] it is stated that since the LTNO is an integrated method and provides average hyperfine fields, the values obtained are often lower than the true ones. This is typically an issue when the fraction f is very low (such that in the two-site model that is used a large fraction of the impurity isotopes are supposed to feel no magnetic field at all) and/or the nuclear orientation of the isotope studied is not saturated. If the fraction f is very low the validity of the two-site model can indeed be questioned, while if the nuclear orientation is not saturated the results of two-parameter fits are subject to a correlation between the temperature dependent nuclear orientation parameters B_k, that determine the shape of the observed anisotropy curve $W(\theta)$, and the temperature independent parameters (usually the fraction f or the A_k directional distribution coefficients) that determine the amplitude of the anisotropy.

When the orientation is saturated or a large anisotropy and good statistics are obtained the temperature dependent and temperature independent parameters in Eq. (1) are (almost)

TABLE III. Magnetic moments and corresponding fractions f at good lattice sites from LTNO results with large or saturated anisotropy, compared with the magnetic moment values from NMR/ON experiments.

Isotope	μ(LTNO) [μ_N]	f	μ(NMR/ON) [μ_N]	Refs.
^{69}As	1.58(16)	0.90(5)	1.6229(16)	[25–28]
^{106}In	4.87(15)	0.945(2)	4.921(13)	[29]
^{108}In	4.53(10)[a]	0.71(3), 0.77(2)	4.561(3)	[30]
^{189m}Au	6.22(20)	0.78(1)	6.17(15)	[31,32]

[a]Average of two values.

not correlated anymore, rendering a higher reliability to the hyperfine interaction parameters obtained in a two-parameter fit. This was demonstrated in the past for several impurity isotopes in iron for which large or fully saturated anisotropies were observed and perfect correspondence was found between the magnetic moment values obtained with LTNO and with NMR/ON (see Table III). Since the fraction f in our case is rather large and in the same range as the values listed in Table III, the deviation of the hyperfine field value of Es in Fe obtained here [i.e., $B_{hf}(\underline{\text{Es}}\text{Fe}) = 396(32)$ T] from its real value should thus be reasonably small.

Given the experimental error, together with the fact that the 'theoretical' error bar on the values calculated by Torumba *et al.* [15] is of the order of 50 T (see, e.g., Fig. 9 in Ref. [15] and [33]), our experimental result is in reasonable agreement with the value of +323 T that is calculated in Ref. [15] for the ferromagnetic LDA (local density approximation) scenario. The value of −417 T that Torumba *et al.* calculated for the ferrimagnetic LDA scenario is not considered as these authors showed that beyond Bk, and thus also for Es, the ferromagnetic LDA solution has the lowest energy (Sec. IV A and Fig. 1 in Ref. [15]). Further, the value of −741 T that is calculated in Ref. [15] for trivalent ferrimagnetic Es in Fe using B3PW91-like hybrid functionals (containing a mixture of Hartree-Fock and LDA exchange) deviates more than 300 T from our result. This scenario is therefore to be regarded as much less probable. Note that if the ferromagnetic LDA scenario is to be preferred, this implies that the delocalization-localization transition in the actinide series does not happen until einsteinium is reached. As mentioned already in Ref. [15], hard experimental evidence could be provided if a more accurate value for the hyperfine field as well as its sign could be determined, which would require, e.g., a field-shift NMR/ON experiment.

Turning to ^{255}Fm, as this is an odd-neutron isotope one can safely estimate the magnetic moment to be smaller than $\mu = 1\,\mu_N$. The experimental result $|\mu B(^{255}\text{Fm}\underline{\text{Fe}})| = 223(17)\,\mu_N$T then yields a lower limit of about 230 T for the hyperfine field of Fm in Fe. This value is not far from the value of about 285 T which Torumba *et al.* (Fig. 9 in Ref. [15]) obtained for the LDA ferromagnetic solution for Fm in Fe and would imply that the delocalization-localization transition would even not happen until Fm. However, assuming $5f$ electrons in Fm to be localized they calculate a value of −787 T. If combined with our hyperfine interaction result this would imply a magnetic

moment value of $\pm 0.30\ \mu_N$, which in principle is also possible. No clear conclusion can thus be drawn in this case.

For berkelium, Torumba *et al.* [15] quote −78 T for the hyperfine field in iron for the ferrimagnetic LDA scenario (for Bk the ferrimagnetic LDA solution has the lowest energy) and −127 T from a calculation using B3PW91-like hybrid functionals with 40% of Hartree-Fock exchange. Combining these values with our experimental result $|\mu B(^{250}\mathrm{BkFe})| = 313(23)\ \mu_N$T, yields for the magnetic moment of ^{250}Bk a value of $|\mu| = 4.0(3)\ \mu_N$ for the lower hyperfine field value and $|\mu| = 2.5(2)\ \mu_N$ for the larger value. Note, however, that, as was mentioned already, the estimated uncertainty on the hyperfine field values for the actinides calculated by Torumba *et al.* is about 50 T, so that,unfortunately, no firm value for the magnetic moment of ^{250}Bk can be obtained.

Finally, we have calculated the magnetic moment of the odd-odd nucleus ^{254}Es in the deformed single-particle model using the numerical calculations described in Ref. [34]. For the configuration of the ground state with $I^\pi = 7^+$ we adopted $\pi_{7/2}[633]\nu_{7/2}[613]$ as suggested in Ref. [35]. A quadrupole deformation of $\epsilon_2 = 0.20$ was used in the calculations, very close to that calculated for ^{254}Es in Ref. [36]. No octupole deformation was assumed, i.e., $\epsilon_3 = 0$. With the parameters $g_s = 0.6 g_s^{\mathrm{free}}$, $g_l^p = 1$, $g_l^n = 0$, $g_R = Z/A = 0.4$ that were used before for ^{228,230}Pa [34] we find from Eq. (6) of Ref. [34] $\mu = 3.52\ \mu_N$ for the above given configuration. Alternatively, when we assume the neutron to be in a [624]7/2 orbit we calculate $\mu = 4.57\ \mu_N$ for the magnetic moment of ^{254}Es. The calculation is not sensitive to the special value of g_R used. Both values are in fair agreement with our experimental result $|\mu| = 4.35(41)\ \mu_N$ for the magnetic moment of ^{254}Es, thus strengthening the reliability of the hyperfine field value of 396(32) *T* for Es in Fe that was used to obtain this magnetic moment.

Summarizing, from the emission anisotropies of α particles from oriented ^{253,254}Es and ^{255}Fm and the 1031 keV γ ray of ^{250}Bk, the hyperfine magnetic field for Es impurities in Fe host has been determined, together with the magnetic moment of ^{254}Es and the hyperfine interactions μB for ^{255}Fm and ^{250}Bk in iron. The value obtained for the magnetic hyperfine field of einsteinium impurities in iron, i.e., $|B_{hf}(\mathrm{EsFe})| = 396(32)$ T, was compared to results from *ab initio* calculations and provides support for the delocalization-localization transition in the actinide series to happen not until einsteinium is reached. The magnetic moment value of $|\mu| = 4.35(41)\ \mu_N$ for ^{254}Es adds a new result to the scarce number of magnetic moments for the actinide isotopes available to date. Comparison with theoretical calculations for this moment supports the reliability of the hyperfine field obtained for Es in Fe.

ACKNOWLEDGMENTS

This work was supported by INTAS (Grant No. 00-00195), the Russian Foundation for Basic Research (Grant Nos. RFBR 03-02-16175 and 07-02-00034), the Fund for Scientific Research Flanders (FWO), and Grant No. LA08015 of the Ministry of Education of the Czech Republic.

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