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HYPERFINE STRUCTURE IN TRIVALENT PLUTONIUM

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ABSTRACT

The hyperfine structure constants of Pu^{3+} in CaF_2 , SrF_2 , and BaF_2 are analyzed and the values $\langle 1/r^3 \rangle = 7.57 \pm 0.57 \text{ au}$ and $a_{c_{\text{Pu}^{3+}}} = -(556 \pm 42) (g_J - 1) \mu_N / \text{I Mc/sec}$ are derived. Relativistic Dirac-Slater and Dirac-Fock wave functions give a value for $\langle 1/r^3 \rangle$ about 9% lower than the empirical value. The core polarization effect in the actinide ion Pu^{3+} is much larger than in the lanthanide series.

I. INTRODUCTION

Values of the nuclear dipole moment of ^{239}Pu derived from different types of measurements varied widely when the data were first analyzed. Subsequent analyses showed these results could be brought into agreement by a complete theoretical treatment including intermediate coupling and core polarization effects.¹ Attempts to explain the sign of the core polarization term with basis wave functions obtained from the central field model of atomic structure proved inadequate until admixtures of continuum states were included.¹ Unfortunately this type of calculation is very difficult. Exchange polarized Hartree-Fock calculations were able to account for the sign and approximate magnitude of core polarization effects² in the lanthanide series.

The electron paramagnetic resonance spectra of Pu^{3+} , $5f^5$, have recently been measured in cubic symmetry sites in CaF_2 , SrF_2 , and BaF_2 . The Zeeman splitting factors (g-values) have been interpreted by calculating the crystalline field mixing of the first excited $J = 7/2$ state with the ground $J = 5/2$ state.³ In this paper we analyze the hyperfine structure data for Pu^{3+} in a similar manner, and use the known nuclear magnetic moment⁴ of ^{239}Pu to derive values for the core polarization effect and $\langle 1/r^3 \rangle$. The latter quantity is compared with various theoretical calculations.

II. THEORY AND RESULTS

A. Theoretical Summary

The nonrelativistic hyperfine Hamiltonian may be written as

$$\mathcal{H} = \frac{2\beta\beta_N\mu_N}{I} \left[\sum_i \frac{\tilde{N}_i \cdot \tilde{I}}{r_i^3} + \frac{8\pi}{3} \sum_i \delta(\tilde{r}_i) \tilde{s}_i \cdot \tilde{I} \right], \quad (1)$$

where β and β_N are the Bohr magneton and the nuclear magneton respectively, μ_N is the nuclear moment, I is the nuclear spin, $\tilde{r}_i$ is the radius vector for the i th electron, and $\delta(\tilde{r}_i)$ is the Dirac delta function which is non-zero only for s electrons. The operator $\tilde{N}_i$ for the i th electron is written in terms of tensor operators as

$$\tilde{N}_i = \tilde{\ell}_i - (10)^{1/2} (\tilde{s}_i^{(1)} \tilde{C}_i^{(2)})^{(1)} \quad (2)$$

where $\tilde{\ell}_i$ and $\tilde{s}_i$ are the orbital and spin angular momentum vectors and $\tilde{C}_i^{(2)}$ is a second rank tensor.⁵ The operator $\tilde{N}_i$ has non-zero matrix elements only for electrons with $\ell > 0$. In order to account for the effect of core polarization which produces a net unpairing of s electrons the second term is included in Eq. (1). The angular transformation properties of this term are proportional to the operator $\tilde{s}_i$. Relativistic effects also transform as $\tilde{s}_i$ and are not distinguishable from core polarization in our present experiments.

For convenience of calculation, we follow the procedure of Bordarier et al.,⁶ and introduce the coupled double tensor operator $\tilde{w}^{(\kappa,k)K}$. For a configuration of equivalent electrons, $\tilde{\ell}^N$, the hyperfine operators are

defined as

$$\begin{aligned} \tilde{l} &= \left[\frac{2l(l+1)(2l+1)}{3} \right]^{1/2} \tilde{w}^{(01)1} \\ -10^{1/2} (\tilde{s}^{(1)} \tilde{c}^{(2)})^{(1)} &= \left[\frac{l(l+1)(2l+1)}{(2l-1)(2l+3)} \right]^{1/2} \tilde{w}^{(12)1} \\ \tilde{s} &= \left[\frac{(2l+1)}{2} \right]^{1/2} \tilde{w}^{(10)1} \end{aligned} \quad (3)$$

A general computer program has been written to evaluate matrix elements $\tilde{w}^{(\kappa,k)K}$ operators. The procedure followed is to calculate the angular matrix elements, parameterize the necessary radial integrals and core polarization effects, and evaluate the parameters from the measured hyperfine data.

The tensor operators $\tilde{w}^{(\kappa,k)K}$ arise naturally in the relativistic hyperfine structure formalism developed by Sandars and Beck.⁷ They defined an effective operator representing the relativistic hyperfine Hamiltonian as

$$\mathcal{H}_{\text{eff}} = a(10) \tilde{w}^{(10)1} + a(01) \tilde{w}^{(01)1} + a(12) \tilde{w}^{(12)1} \quad (4)$$

The expressions for the $a(\kappa,k)$ are given in Ref. 6 and define the necessary combinations of radial integrals, which may then be calculated from relativistic wave functions. Such a calculation will be described in a later section. For purposes of fitting our data we combine Eq. (3) and (4) into the most convenient form to parameterize the experimental results,⁸

$$\mathcal{H}_{\text{eff}} = \frac{2\beta\beta_N \mu_N}{I} \sum_i \left[\tilde{L}_i \langle 1/r^3 \rangle_{01} - 10^{1/2} (\tilde{s}_i^{(1)} \tilde{C}_i^{(2)})_i^{(1)} \langle 1/r^3 \rangle_{12} + \tilde{s}_i \langle 1/r^3 \rangle_{10} \right] \quad (5)$$

In the nonrelativistic approximation (assuming no effects due to core polarization), $\langle 1/r^3 \rangle_{01} = \langle 1/r^3 \rangle_{12}$ and $\langle 1/r^3 \rangle_{10} = 0$. For our calculations we shall assume the nonrelativistic approximation $\langle 1/r^3 \rangle_{01} = \langle 1/r^3 \rangle_{12}$, but because core polarization and relativistic effects transform as $\tilde{s}$ we parameterize this operator.

B. Application to Pu³⁺

The electronic configuration of Pu³⁺ outside the closed shells is 5f⁵. The optical spectrum was originally measured by Lämmermann and Conway⁹ and the data further refined by Conway and Rajnak.¹⁰ We use the intermediate coupled wave functions given by Conway and Rajnak for our calculations. In the alkaline earth fluorides the ground state $J = 5/2$ and the first excited state $J = 7/2$ are strongly mixed by the crystalline field interaction. We assume the effects of higher J levels are negligible. We obtain the amount of J mixing between the 5/2 and 7/2 states from the previous analysis of the Zeeman splitting factors (g -values).³ The crystalline field wave function is written as

$$\Gamma_7^1 = \cos \phi |J=5/2, \Gamma_7^+\rangle - \sin \phi |J=7/2, \Gamma_7^{+'}\rangle,$$

where the crystalline field wave functions Γ_7^+ and $\Gamma_7^{+'}$ are the same as

defined previously. In Table I we list the pertinent results taken from our earlier work.

The hyperfine Hamiltonian can be evaluated by summing matrix elements of the type

$$\langle \Gamma_7^1 | \tilde{w}^{(\kappa, k)K} | \Gamma_7^1 \rangle ,$$

and multiplying by the appropriate factors given in Eq. (3). For the $J = 5/2$ level the complete intermediate coupled wave function consists of a linear combination of 28 LS states and the $J = 7/2$ level consists of a linear combination of 30 LS states. In calculating matrix elements of the Zeeman operator for our earlier paper, we obtained matrix elements of the operators $\Sigma \tilde{l}_i$ and $\Sigma \tilde{s}_i$ for the complete basis sets. The complexity of the operator $(\tilde{s}^{(1)} \tilde{c}^{(2)})^{(1)}$ obliged us to work with a truncated basis set in computing its matrix elements. The operators $\Sigma \tilde{l}_i$ and $\Sigma \tilde{s}_i$ were recomputed with the truncated basis sets in order to provide some estimate of the effects of truncation. The truncated basis sets are given in Table II. The notation is from Nielsen and Koster.¹¹

For each operator $\tilde{O}$ in the hyperfine Hamiltonian, the matrix elements are

$$\begin{aligned} \langle \Gamma_7^1 | \Sigma \tilde{O}_i | \Gamma_7^1 \rangle &= \langle J=5/2, \Gamma_7^+ | \Sigma \tilde{O}_i | J=5/2, \Gamma_7^+ \rangle \cos^2 \phi \\ &\quad - 2 \langle J=5/2, \Gamma_7^+ | \Sigma \tilde{O}_i | J=7/2, \Gamma_7^{+'} \rangle \sin \phi \cos \phi \\ &\quad + \langle J=7/2, \Gamma_7^{+'} | \Sigma \tilde{O}_i | J=7/2, \Gamma_7^{+'} \rangle \sin^2 \phi , \end{aligned} \quad (6)$$

where 0_i is ℓ_i , $-10^{1/2}(\tilde{s}^{(1)}\tilde{c}^{(2)})_i^{(1)}$ or s_i . The angle ϕ is obtained by the previous fit to the g-values. In Table III we give the values for the matrix elements shown in Eq. (6). We then evaluate Eq. (6) for the values of ϕ given in Table I with the results shown in Table IV.

The hyperfine structure constant may now be written (in the non-relativistic approximation) as

$$A = A_f + A_c$$

where

$$A_f = \theta [\langle \Gamma_7^1 | \Sigma \ell_i | \Gamma_7^1 \rangle + \langle \Gamma_7^1 | \Sigma -10^{1/2}(\tilde{s}^{(1)}\tilde{c}^{(2)})_i^{(1)} | \Gamma_7^1 \rangle]$$

$$A_c = C \langle \Gamma_7^1 | \Sigma s_i | \Gamma_7^1 \rangle$$
(7)

and

$$\theta = \frac{4\beta\beta_N\mu_N}{I} \langle 1/r^3 \rangle ;$$

C is a parameter which is proportional to μ_N/I and gives the contribution due to core polarization. We have used the CaF_2 and SrF_2 matrix elements in Table IV to obtain the parameters and then used the BaF_2 result as a check because the accuracy of the measurements in the first two crystals is greater than for BaF_2 . The results are $\theta = 0.00981 \text{ cm}^{-1}$, $C = -0.0147 \text{ cm}^{-1}$. The signs of the experimental values of A are unknown. In order to obtain consistent results they are chosen as negative. This choice is necessary because μ_N of ^{239}Pu is positive and thus θ must be positive.

Because we wish to compare our values with those from atomic beam and optical spectroscopy experiments we now calculate the hyperfine parameters for a pure $J = 5/2$ and a $J = 7/2$ state. We use the matrix elements given in Table III for the pure states and the parameters just evaluated. The results are given in Table V. In order to obtain the data for a free ion we have made use of the relationship

$$\frac{g_J}{a} = \frac{g}{A} ,$$

where g_J and a are the Landé g -value and hyperfine coupling constant for a free ion.¹² The hyperfine coupling constant for a free ion is also the sum of two terms similar to Eq. (7);

$$a = a_f + a_c .$$

We also list in Table V values for a_c and a_f .

The core polarization term may be written¹³

$$a_c = (g_J - 1) K \frac{\mu_N}{I} , \quad \text{where} \quad K = C \frac{I}{\mu_N} . \quad (8)$$

The term $(g_J - 1)$ is the value of the angular matrix element of the operator $\sum_i s_i$ for a pure J state. From our values of a_c we find

$$a_c = - (556 \pm 42) (g_J - 1) \frac{\mu_N}{I} \text{ Mc/sec} \quad (9)$$

for the Pu^{3+} free ion. From θ we calculate

$$\langle 1/r^3 \rangle_{\text{Pu}^{3+}} = 7.57 \pm 0.57 \text{ au}.$$

The errors in our parameters are estimated in the following way.

From the uncertainty in the experimental data the maximum error is 2%. There is also a 2% uncertainty in the value of μ_N of ^{239}Pu . If we assume the error due to truncation of the basis set for the operator $\Sigma - 10^{1/2} (\tilde{s}^{(1)} \tilde{c}^{(2)})_i^{(1)}$ is the same as for the operator $\Sigma \tilde{\ell}_i$, then considering the relative magnitudes of the matrix elements, the truncation error is about 0.7% of the total value of the matrix element of the operator $\Sigma \tilde{N}_i$. In the next section we show the error due to the assumption $\langle 1/r^3 \rangle_{01} = \langle 1/r^3 \rangle_{12}$ is negligible. The largest error probably is due to the neglect of higher J levels mixed in by the crystalline field. We have given the parameters an error of $\pm 7.5\%$ which we believe is a conservative estimate.

DISCUSSION

Recently there have been relativistic calculations for the elements and some ions through the actinide series. Dr. J. Mann from Los Alamos Scientific Laboratory has sent us the wave functions for the 5f electrons of PuI and PuIV (spectroscopic notation) obtained from his Dirac-Fock calculations¹⁴ and Dr. D. T. Cromer, also from LASL, has sent us the wave functions obtained from Dirac-Slater calculations.¹⁵ We have integrated these wave functions according to the definitions given by Bourdaries et al.⁶ and the results are given in Table VI. In order to compare our experimental result with their wave functions we define

$$\Delta = \langle 1/r^3 \rangle_{12} - \langle 1/r^3 \rangle_{01} \quad (10)$$

Then we may write

$$A_f = \frac{4\beta\beta_{NN}}{I} \langle 1/r^3 \rangle_{01} \langle \Gamma_7^1 | \Sigma \ell_i | \Gamma_7^1 \rangle \\ + \frac{4\beta\beta_{NN}}{I} (\Delta + \langle 1/r^3 \rangle_{01}) \langle \Gamma_7^1 | \Sigma -10^{1/2} (s^{(1)} c^{(2)})_i | \Gamma_7^1 \rangle$$

The percentage correction to the $\langle 1/r^3 \rangle_{01}$ of Table VI by our assumption that $\langle 1/r^3 \rangle_{01} = \langle 1/r^3 \rangle_{12}$ in our analysis is

$$\frac{\langle \Gamma_7^1 | \Sigma -10^{1/2} (s^{(1)} c^{(2)})_i | \Gamma_7^1 \rangle}{\langle 1/r^3 \rangle_{01} \langle \Gamma_7^1 | \Sigma \ell_i | \Gamma_7^1 \rangle} \times 100 \quad (11)$$

$\sim 0.3\%$ using data from Tables IV and VI. The corrected values are also given in Table VI. Comparison of the theoretical $\langle 1/r^3 \rangle_{01}$ and the experimental $\langle 1/r^3 \rangle$ gives a discrepancy of 9%. There is essentially no difference between the Dirac-Fock and the Dirac-Slater calculations for the $\langle 1/r^3 \rangle$ integrals.

The magnitude of the core polarization effect is much larger than found for rare earth ions. Bleaney¹⁶ has shown that for the Eu^{2+}

$$a_{c_{\text{Eu}^{2+}}} = - (63 \pm 10)(g_J - 1) \frac{\mu_N}{I} \text{ Mc/sec} \quad (12)$$

He assumed that this equation was valid throughout the 4f transition series but recent experimental and theoretical work questions this assumption.^{2,12} The core polarization effect in Pu^{3+} , Eq. (9) is almost an order of magnitude larger than found for Eu^{2+} in the lanthanide series. Our value for Pu^{3+} is slightly larger than the core polarization effect derived by Easley¹⁷ for Am^{2+} ,

$$a_{c_{\text{Am}^{2+}}} = - (420 \pm 42)(g_J - 1) \frac{\mu_N}{I} \text{ Mc/sec} \quad (13)$$

but corroborates the magnitude of the effect.

Bauche and Judd¹ and Armstrong and Marrus¹⁸ have shown that the equation

$$a_{c_{\text{atoms}}} = - (70 \pm 20)(g_J - 1) \frac{\mu_N}{I} \text{ Mc/sec} \quad (14)$$

is consistent with the interpretation of the data for $\text{PuI} (5f^6 7s^2)$ and AmI

(Sf^7s^2). From the values of $\langle 1/r^3 \rangle_{10}$ given in Table VI the equation

$$a_{\text{rel}} = - (67 \pm 10) \frac{\mu_N}{I} (g_J - 1) \text{ Mc/sec} \quad (15)$$

may be obtained. These calculations show relativistic effects for the atoms and ions are approximately equal and agree well with the empirical equation for the actinide atoms, PuI and AmI. The core polarization effects in the ions PuIV and AmIII are about seven times larger than relativistic effects in actinide atoms. Because of the agreement found for the calculated $\langle 1/r^3 \rangle_{01}$ with the experimental value the relativistic Dirac-Fock and Dirac-Slater calculations probably give the right magnitude for relativistic effects in the heavy atoms. Therefore it appears that a_{atoms} (Eq. (14)) is mostly due to relativistic effects; and that in the ions where there are no 7s electrons there are very large core polarization effects.

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Table I. Spin Hamiltonian parameters for trivalent Pu.

Ion	Matrix	ϕ	$ g $ Measured	$ A \text{ cm}^{-1} \times 10^3$
Pu^{3+}	CaF_2	-15.1°	1.297 ± 0.002	6.72 ± 0.06
Pu^{3+}	SrF_2	-13.2°	1.250 ± 0.002	8.46 ± 0.1
Pu^{3+}	BaF_2	-11.0°	1.187 ± 0.004	10.2 ± 0.3

Table II. Truncated basis sets for the $J = 5/2$ and $J = 7/2$ levels.

(Notation from Nielsen and Koster.¹¹)

	$J = 5/2$		$J = 7/2$	
	Coef.	Coef. ² × 100	Coef.	Coef. ² × 100
6_F	0.1003	1.01	0.1037	1.07
6_H	0.8121	65.95	0.8951	80.12
4_{F3}	-0.1283	1.65	-0.1018	1.04
4_{G1}	-0.3097	9.59	-0.2451	6.01
4_{G3}	-0.1106	1.22	0.0677	0.45
4_{G4}	-0.3775	14.25	0.3053	9.33
	Total	93.67	Total	98.02

Table III. Numerical values of the general hyperfine matrix elements for trivalent Pu in cubic symmetry.

$\Sigma \tilde{O}_i$	$\Sigma \tilde{L}_i$		$\Sigma -10^{1/2} (\tilde{s}^{(1)} \tilde{C}^{(2)})_i^{(1)}$		$\Sigma \tilde{S}_i$	
	Truncated	Complete	Truncated		Truncated	Complete
$\langle J=5/2, \Gamma_7^+ \Sigma \tilde{O}_i J=5/2, \Gamma_7^+ \rangle$	-1.259	-1.317	0.090		0.479	0.483
$2 \langle J=5/2, \Gamma_7^+ \Sigma \tilde{O}_i J=7/2, \Gamma_7^{+'} \rangle$	1.603	+1.627	-0.068		-1.603	-1.627
$\langle J=7/2, \Gamma_7^{+'} \Sigma \tilde{O}_i J=7/2, \Gamma_7^{+'} \rangle$	1.674	+1.703	-0.116		-0.204	-0.203

Table IV. Numerical values of the hyperfine matrix elements of trivalent Pu for the alkaline earth fluorides.

ϕ		$\Sigma \tilde{l}_i$		$\Sigma -10^{1/2} (\tilde{s}^{(1)} \tilde{c}^{(2)})_i^{(1)}$	$\Sigma \tilde{s}_i$		$2\Sigma (\tilde{l}_i + 2\tilde{s}_i)$
		Trunc.	Compl.		Trunc.	Compl.	Compl.
CaF ₂	-15.1°	-0.657	-0.703	0.059	0.030	0.028	-1.294
SrF ₂	-13.2°	-0.750	-0.798	0.064	0.087	0.086	-1.252
BaF ₂	-11.0°	-0.852	-0.902	0.070	0.154	0.154	-1.188

$$\Sigma \tilde{N}_i = \Sigma (\tilde{l}_i(\text{comp}) - 10^{1/2} (\tilde{s}^{(1)} \tilde{c}^{(2)})_i^{(1)})$$

CaF ₂	-0.644
SrF ₂	-0.733
BaF ₂	-0.832

Table V. Extrapolated hyperfine parameters for pure $J = 5/2$ and $J = 7/2$ states of Pu^{3+} in a cubic crystalline field and as a free ion.

	<u>A</u>	<u>A_f</u>	<u>A_c</u>	<u>a</u>	<u>a_f</u>	<u>a_c</u>	<u>g_J</u>
	$(\text{cm}^{-1} \times 10^{-3})$			$(\text{cm}^{-1} \times 10^3)$			
$J = 5/2$	-19.19	-12.02	-7.16	11.51	7.21	4.30	0.420
$J = 7/2$	18.55	15.55	3.00	6.18	5.18	1.00	0.865

Table VI. Calculated values for the radial parameters using relativistic wave functions.

	$\langle 1/r^3 \rangle_{01}$ (au)	$\langle 1/r^3 \rangle_{01}(\text{corr.})$ (au)	$\langle 1/r^3 \rangle_{12}$ (au)	$\langle 1/r^3 \rangle_{10}$ (au)
$f_{s^2}^6$ PuI DS	6.092		7.594	-0.706
f^5 PuIV DS	6.879	6.86	8.166	-0.595
$f_{s^2}^6$ PuI DF	6.184		7.849	-0.788
f^5 PuIV DF	6.936	6.92	8.308	-0.639

FOOTNOTES AND REFERENCES

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