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September 1970

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THE HYPERFINE STRUCTURE ANOMALY OF  $^{241}\text{Pu}$  AND  $^{239}\text{Pu}$  AND THE  
NUCLEAR MOMENT OF  $^{241}\text{Pu}^*$

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The hfs anomaly of an s electron for the  $^{241,239}\text{Pu}$  isotopes is shown to be  $^{241}\Delta^{239} = -6.3 \pm 0.2\%$ . The value for  $\mu_N(^{241}\text{Pu})$  is found to be  $-0.718 \pm 0.017$  nm.

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The hyperfine structure (hfs) constants for a number of levels in the Pu I and Pu II optical spectra have been measured by Fabry-Perot interferometry for the isotopes  $^{241}\text{Pu}$  ( $I = 5/2$ ,  $Z = 94$ ,  $A = 147$ ) and  $^{239}\text{Pu}$  ( $I = 1/2$ ,  $A = 145$ ) [1]. The measured ratio of the hfs constants differed from that found earlier by Bleaney *et al.* [2] in electron paramagnetic resonance (epr) experiments of  $^{241,239}\text{PuO}_2^{++}$  (Pu VII) diluted in a crystal of  $\text{UO}_2\text{Rb}(\text{NO}_3)_3$ . In the course of ENDOR and epr measurements on  $^{241,239}\text{Pu}^{3+}$  (Pu IV) in  $\text{CaF}_2$  [3] this ratio has been remeasured. The data from the various measurements are collected in Table I.

In a recent analysis of the hfs of Pu IV in  $\text{CaF}_2$  [4] it was shown that approximately 5% of the total hyperfine interaction comes from unpaired s electrons through the mechanism of core polarization. The ground configuration of

\* Work performed under the auspices of the U. S. Atomic Energy Commission.

Pu II,  $f^6s$ , is fairly pure and can be described by coupling the two lowest levels of the  $f^6$  core ( ${}^7F_{0,1}$ ) with an  $s$  electron and then mixing these two  $J = 1/2$  states [5,6]. The hfs of the  $J = 1$  level of Pu I ( $f^6s^2$ ) has been shown to be extremely small [7], so almost all the hfs of the ground state of Pu II can be attributed to the  $s$  electron. Therefore the difference in the hfs ratios between the optical measurement of the lowest level of Pu II and the resonance measurements of Pu IV is attributed to the hfs anomaly  ${}^{241}\Delta^{239}$  of the  $s$  electron [8]. The hfs splittings of the excited states of Pu I and Pu II listed in Table I also are probably due to  $s$  and  $p_{1/2}$  electrons but since many close lying configurations interact further discussion is dependent upon the detailed analysis of the optical spectra [6]. Varying the percentage  $s$  electron contribution to the total hfs constant will cause changes in the hfs ratio of  ${}^{241}\text{Pu}$  to  ${}^{239}\text{Pu}$ . This may be the reason for the discrepancy in the ratio between Pu VII and Pu IV.

Since the  $s$  electron contribution to  $A(\text{Pu IV})$  is small

$$\frac{A({}^{241}\text{Pu IV})}{A({}^{239}\text{Pu IV})} \approx \frac{g_I({}^{241}\text{Pu})}{g_I({}^{239}\text{Pu})} \quad (1)$$

Then the hfs anomaly is

$${}^{241}\Delta^{239} = \frac{A({}^{241}\text{Pu II})}{A({}^{239}\text{Pu II})} \times \frac{A({}^{239}\text{Pu IV})}{A({}^{241}\text{Pu IV})} - 1 = -6.3 \pm 0.2\% \quad (2)$$

The Bohr-Weisskopf theory [8] allows an estimate of  ${}^{241}\Delta^{239}$  assuming  $g_s = g$  (neutron),  $g_l = \frac{Z}{A}$ , and the coefficient  $b \approx 5.5\%$  extrapolated from their table.

Then

$${}^{241}\Delta^{239} \approx 0.3b \frac{g_s g_l}{g_s - g_l} \left( \frac{1}{g_I({}^{241}\text{Pu})} - \frac{1}{g_I({}^{239}\text{Pu})} \right) = -5.4\% \quad (3)$$

The agreement between the theory and experiment is surprisingly good, which was not true for the stable odd-neutron isotopes of Yb( $Z = 70$ ,  $A = 101, 103$ ) [9].

A better value for  $\mu_N$  of  $^{241}\text{Pu}$  may be calculated from the hfs ratio, Eq. (1), and  $\mu_N(^{239}\text{Pu}) = +0.200 \pm 0.004 \text{ nm}$  [10]. The new value for  $\mu_N(^{241}\text{Pu}) = -0.718 \pm 0.017 \text{ nm}$  where the sign is obtained from the optical data.

I wish to thank J. Conway and M. Fred for many valuable discussions on the optical spectra of Pu I and Pu II and W. Kolbe and G. Kaindl for their help and advice.

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Table I. Hfs constants for various levels of Pu ions.

Ion	Level ( $\text{cm}^{-1}$ )	A ( $10^{-3} \text{ cm}^{-1}$ )	$\frac{A(^{241}\text{Pu})I(^{241}\text{Pu})}{A(^{239}\text{Pu})I(^{239}\text{Pu})}$	Reference
$^{241}\text{Pu}$ I	23766	$-58.25 \pm 0.2$	$-3.367 \pm 0.02$	1
$^{239}\text{Pu}$ I	23766	$86.5 \pm 0.2$		
$^{241}\text{Pu}$ II	0	$-167.3 \pm 0.5$	$-3.362 \pm 0.02$	1
$^{239}\text{Pu}$ II	0	$248.8 \pm 0.5$		
$^{241}\text{Pu}$ II	22038	$-32.7 \pm 1$	$-3.41 \pm 0.2$	1
$^{239}\text{Pu}$ II	22038	$48 \pm 1$		
$^{241}\text{Pu}$ IV	0	$ 4.807  \pm 0.010$	$ 3.590  \pm 0.010$	3
$^{239}\text{Pu}$ IV	0	$ 6.695  \pm 0.003$		
$^{241}\text{Pu}$ VII	0	$ 245  \pm 1 \text{ gauss}$	$ 3.53  \pm 0.02$	2
$^{239}\text{Pu}$ VII	0	$ 347  \pm 1 \text{ gauss}$		



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