

UNIVERSITY OF  
CALIFORNIA

*Ernest O. Lawrence*

*Radiation  
Laboratory*

TWO-WEEK LOAN COPY

*This is a Library Circulating Copy  
which may be borrowed for two weeks.  
For a personal retention copy, call  
Tech. Info. Division, Ext. 5545*

BERKELEY, CALIFORNIA

## DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

For Pub. Phys. Rev. Letters

UCRL-9010

UNIVERSITY OF CALIFORNIA  
Lawrence Radiation Laboratory  
Berkeley, California  
Contract No. W-7405-eng-48

PARAMAGNETIC RESONANCE IN TETRAVALENT Pa<sup>231</sup>

John D. Axe, Ru-tao Kyi, and H. J. Stapleton

December 1959

Printed for the U. S. Atomic Energy Commission

Paramagnetic Resonance in Tetravalent Pa<sup>231</sup>\*

John D. Axe and Ru-tao Kyi

Lawrence Radiation Laboratory

University of California, Berkeley, California

and

H. J. Stapleton

Department of Physics

University of California, Berkeley, California

The paramagnetic-resonance spectrum of tetravalent Pa<sup>231</sup> has been observed in a single crystal of Cs<sub>2</sub>ZrCl<sub>6</sub>. A melt of Cs<sub>2</sub>ZrCl<sub>6</sub> was "doped" with ~500 μg of anhydrous Pa<sup>231</sup>Cl<sub>4</sub> and allowed to crystallize in an atmosphere of hydrogen by slow passage through a furnace. At 4.2°K and a frequency of 9457 MCS, the observed resonance pattern consisted of four widely separated hyperfine components. The spectrum was isotropic to within the accuracy of the field measurements (~ 1/2%). These features are interpreted as the usual  $\Delta M_S = \pm 1$ ,  $\Delta M_I = 0$  transitions between the eigenstates of the spin Hamiltonian

$$\mathcal{H} = g\beta H \cdot S + A I \cdot S,$$

with  $S = 1/2$ ,  $I = 3/2$ ,  $|g| = 1.15 (\pm 0.02)$ , and  $|A| = 0.0518 (\pm 0.001) \text{ cm}^{-1}$ . The large hyperfine interaction necessitates the use of exact solutions for the energy levels.<sup>2</sup> No resonance was detected at 77°K.

The nuclear-spin value of 3/2 is verified.<sup>3</sup> The paramagnetism can be most plausibly ascribed to a single 5f electron. A magnetically isotropic Kramers' doublet is expected to be the lowest lying as the result of the octahedral perturbation present at the zirconium site.<sup>4,5</sup> As an alternative, a 6d<sup>1</sup> configuration is expected to give rise to a fourfold degenerate magnetically anisotropic level if octahedral symmetry

\*Supported in part by the U. S. Atomic Energy Commission and the Office of Naval Research.

is preserved,<sup>6</sup> or an anisotropic doublet if distortion is present.<sup>7</sup> Further work on the optical absorption spectra of this system is in progress and should allow amplification of the above conclusions. An attempt to measure the nuclear magnetic moment of Pa<sup>231</sup> by a "double-resonance" technique is likewise in progress. We wish to thank Professors C. D. Jeffries and B. B. Cunningham for their invaluable advice.

### References

1. Prepared by the method of Sellers, Fried, Elson, and Zachariassen, J. Am. Chem. Soc. 76, 5935 (1954).
2. B. Bleaney, Phil. Mag. 42, 441 (1951).
3. H. Schüller and H. Gollnow, Naturwiss. 22, 511 (1934).
4. See for example J. S. Griffith and L. E. Orgel, J. Chem. Phys. 26, 988 (1956).
5. C. A. Hutchison and B. Weinstock, to be published in J. Chem. Phys. These authors discuss paramagnetic resonance in another f<sup>1</sup> octahedral system, NpF<sub>6</sub>.
6. B. Bleaney, Proc. Phys. Soc. (London) 73, 939 (1959).
7. Griffiths, Owen, and Ward, Proc. Royl Soc. (London) A219, 526 (1953).