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PARAMAGNETIC RESONANCE IN TETRAVALENT Pa^{231}

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Paramagnetic Resonance in Tetravalent Pa^{231}*

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The paramagnetic-resonance spectrum of tetravalent Pa^{231} has been observed in a single crystal of Cs_2ZrCl_6. A melt of Cs_2ZrCl_6 was "doped" with ~500 μg of anhydrous Pa^{231} Cl_4 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 $S = 1/2$, $I = 3/2$, $|g| = 1.15$ (±.02), and $|A| = 0.0518$ (±.001) cm^{-1}. The large hyperfine interaction necessitates the use of exact solutions for the energy levels.[2] No resonance was detected at 77°K.

The nuclear-spin value of 3/2 is verified.[3] 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.[4,5] As an alternative, a $6d^1$ configuration is expected to give rise to a fourfold degenerate magnetically anisotropic level if octahedral symmetry

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is preserved,[6] or an anisotropic doublet if distortion is present.[7] 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^{231} 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.

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