Re-Examination of the Paramagnetic Resonance of Np^{239}†

M. Abraham, C. D. Jeffries, R. W. Kedzie, and J. C. Wallmann
Radiation Laboratory and Physics Department, University of California, Berkeley, California

(Received June 27, 1958)

The microwave paramagnetic resonance spectrum previously reported for 2.3-day Np^{239} activity incorporated into a crystal of $UO_2Rb(NO_3)_3$ has been re-examined and determined to be due, not to Np^{239}, but to radiation-induced resonance centers. Experiments tentatively suggest that these resonance centers may be Pu^{239} in an unusual oxidation state. An experiment in which Np^{239} nuclei were aligned by static hfs interactions (Pound and Bleaney methods) showed an anisotropy of the γ-ray pattern; this proves that the nuclear spin $I(Np^{239})>\frac{1}{2}$, in contradiction to our earlier result.

IN a previous paper,[1] we reported the observation of a pair of hyperfine lines in the microwave paramagnetic resonance spectrum of a curie of 2.3-day Np^{239} activity incorporated as NpO_2^{2+} ions in a single crystal of $(Np,U)O_2Rb(NO_3)_3$. Since the observed electronic spectroscopic splitting factors, $g_{||}\sim 3.2$ and $g_\perp\sim 0$, were, within estimated errors of measurement, the same as those observed earlier by Bleaney, Llewellyn, Pryce, and Hall[2] for long-lived Np^{237} in this same crystal, we interpreted the lines as a hyperfine doublet of Np^{239} showing, apparently, that the nuclear spin $I(Np^{239})=\frac{1}{2}$. We were not able to establish that these resonance lines were definitely associated with a lifetime of 2.3 days, because radiation damage appeared to destroy them in a shorter time than this. The decay product Pu^{239}, for which $I=\frac{1}{2}$, was certainly present in the crystal; however, the only oxidation state for which paramagnetic resonance has been seen, PuO_2^{2+}, has a spectroscopic splitting factor $g_{||}\sim 5.3$, clearly different from our observed value. We considered that there was only a remote possibility that the Pu might exist in an unusual oxidation state with, fortuitously, the g factor of NpO_2^{2+}. However, more recent experiments indicate that this is perhaps the case and that the pair of hyperfine lines are not due to Np^{239}.

These experiments are as follows. Paramagnetic resonance at 4.3°K of a single crystal of $(Pu,U)O_2Rb(NO_3)_3$ weighing about 200 mg and containing about 1 mg of Pu^{239} as PuO_2^{2+} was observed at 9200 Mc/sec. Two lines were seen with $g_{||}\sim 5.3$, in agreement with the original work of Bleaney *et al.*[2] Then this crystal was irradiated at room temperature with a Co^{60} γ source for 67 hours for a total dosage of 1.3×10^8 roentgens. The paramagnetic resonance was re-examined, and showed, in addition to the two lines at $g_{||}\sim 5.3$, a pair of new lines at $g_{||}\sim 3.2$ with a signal-to-noise ratio of about 7:1. The crystal was rotated about an axis perpendicular both to its z axis and to the magnetic field H; the lines were found to be in agreement with the spin Hamiltonian,

$$\mathcal{H}=g_{||}\beta H_z S_z+g_\perp\beta(H_x S_x+H_y S_y)+AS_zI_z+B(I_xS_x+I_yS_y),$$

with $S=\frac{1}{2}$, $I=\frac{1}{2}$, $A=0.0504\pm 0.001$ cm^{-1}, $g_{||}=3.18\pm 0.1$, $g_\perp\sim 0$, and $B\sim 0$. The values previously found[1] for a curie of Np^{239} in a similar crystal were $S=\frac{1}{2}$, $I=\frac{1}{2}$, $A=0.0503\pm 0.0005$ cm^{-1}, $g_\perp\sim 0$, $B\sim 0$, and $g_{||}=3.22/\cos\alpha$, where α is the fixed minimum angle between z and H and should be zero for perfect alignment of the crystal; α was estimated to be as large as 19° for the very active crystal. Thus, within experimental error, the two lines previously found in the very active $(Np^{239},U)O_2Rb(NO_3)_3$ crystal are identical to those appearing upon γ irradiation of the $(Pu^{239},U)O_2Rb(NO_3)_3$ crystal. Consequently we now believe that in the former crystal the lines are not the hfs lines of Np^{239}, but are the result of the same process producing them in the latter crystal. Our previous result that $I(Np^{239})=\frac{1}{2}$ is thereby incorrect. In an attempt to find the origin of the lines, a pure $UO_2Rb(NO_3)_3$ crystal was irradiated in a similar fashion for a dosage of 1.3×10^8 roentgens and its paramagnetic resonance spectrum was examined; no resonance lines were seen in the vicinity of $g_{||}\sim 3.3$. This tentatively suggests that the lines are perhaps the hfs lines of Pu^{239} in some unusual oxidation state, say Pu(IV) or Pu(VII), produced by irradiation; the evidence is not conclusive because of low signal-to-noise ratio and also because the $(Pu,U)O_2Rb(NO_3)_3$ crystal and the $UO_2Rb(NO_3)_3$ crystal were not grown under identical conditions of acidity, etc.

Finally, we have shown that $I(Np^{239})>\frac{1}{2}$ by producing a detectable anisotropy in the γ-ray angular distribution of Np^{239} aligned in zero magnetic field by static hyperfine interaction, as suggested by Pound[3] and by Bleaney.[4] Approximately 5 mC of 2.3-day Np^{239} activity was grown into a single crystal of $(Np,U)O_2Rb(NO_3)_3$ which was mounted in a cryostat with counters along and perpendicular to, respectively, the crystalline z axis. The difference in counting rate, $C(\pi/2)-C(0)$, was observed on an automatic recorder as the temperature

† Supported in part by the U. S. Atomic Energy Commission.

[1] Abraham, Jeffries, Kedzie, and Wallmann, Phys. Rev. **106**, 1357 (1957).

[2] Bleaney, Llewellyn, Pryce, and Hall, Phil. Mag. **45**, 991, 992 (1954).

[3] R. V. Pound, Phys. Rev. **76**, 1410 (1949).

[4] R. Bleaney, Proc. Phys. Soc. (London) **A64**, 315 (1951).

of the crystal was periodically changed by a heat leak from 1.5°K to 7°K. A small, reversible, and reproducible anisotropy of about $\frac{1}{2}\%$ was observed, several times larger than background noise. The sign of the anisotropy reversed after the crystal was rotated through 90°, as expected; the sign did not change on rotation of 180°. From this experiment we can conclude definitely that $I(\mathrm{Np}^{239})>\frac{1}{2}$, since a γ-ray anisotropy is not possible for nuclei with $I=\frac{1}{2}$. This result is consistent with that of Hubbs and Marrus,[5] who find $I(\mathrm{Np}^{239})=\frac{5}{2}$ by radioactive atomic beam methods.

[5] J. C. Hubbs and R. Marrus, Phys. Rev. **110**, 287 (1958).

PHYSICAL REVIEW VOLUME 112, NUMBER 2 OCTOBER 15, 1958

Photoneutron Yields in the Rare-Earth Region*

E. G. Fuller, B. Petree, and M. S. Weiss†
National Bureau of Standards, Washington, D. C.

(Received June 16, 1958)

The total photoneutron yield curves for Sn, I, La, Ce, Sm, Tb, Ho, Er, Yb, Ta, Au, and Pb have been measured for x-ray energies from 7 to 40 Mev with about one percent statistical uncertainty. The Penfold-Leiss matrix was used to convert these yield curves to *integrated* neutron yield cross sections directly without smoothing the original activation curves. The cross sections derived from the integral curves were corrected for multiple neutron emission above the $(\gamma,2n)$ threshold using the statistical model. The widths found for the giant resonances for the closed-shell nuclei decreased from 5 Mev to 3.8 Mev in going from Sn to Pb. The widths for the elements having large nuclear deformations for most of their isotopes were considerably broader. These widths decreased slowly from 8.6 Mev for Sm to 6 Mev for Ta. These widths are consistent with the broadening of the giant resonance to be expected on the Danos model if values of the intrinsic quadrupole moment are taken from Coulomb excitation data. The neutron yield cross sections corrected for multiple neutron emission were integrated to 22 Mev. Using these integrals and defining f by $\int\sigma dE=0.06(NZ/A)f$, these data gave an average value of $f=1.34\pm0.21$.

INTRODUCTION

It has recently been suggested by both Okamoto[1] and Danos[2] independently that on a classical hydrodynamic model of the nucleus one might expect the giant resonance for a deformed unoriented nucleus to be split into two resonances. These two resonances would be associated with the major and minor axes of the nucleus. In the case of a nucleus with positive intrinsic quadrupole moment the "giant resonance" would be made up of a low-energy resonance shifted to an energy slightly lower than that expected for a spherical nucleus of the same mass number and a high-energy resonance shifted to a slightly higher energy. Danos[2] has pointed out that the area under the low-energy resonance would be one-half that under the higher energy resonance.

In a "poor resolution" measurement of the giant resonance the splitting resulting from the ground-state deformation would not be observable. The effect of the deformation might, however, show up as a statistically significant broadening in the giant resonance compared to that obtained for a nearby spherical nucleus. Consistent with this picture is the observation of the Pennsylvania group[3] that the giant resonances for closed-shell nuclei are narrower than those in general observed for other nuclei. It was the object of the present work to see if the large intrinsic quadrupole moments of the rare earth nuclei were reflected in the total width of the giant resonance for these nuclei. The total neutron yield curve was measured for a series of twelve nuclei ranging from tin through the rare earths to lead. Measurements were made from the (γ,n) threshold in each element up to about 40 Mev. The samples were all of normal isotopic abundance. The preliminary results of this work have been reported previously.[4]

EXPERIMENTAL APPARATUS AND PROCEDURES

The method used in this study was similar to that which has previously been used in studies of the total neutron yields from various elements.[3,5] The experiment consisted of measuring the number of neutrons emitted from a series of samples as a function of the peak betatron operating energy. The experimental arrangement is shown in Fig. 1. The neutron detector is of conventional design[5] containing eleven BF_3 counters connected in parallel. The amplified pulses from the

* This work supported by the U. S. Atomic Energy Commission.
† Presently on educational leave at the Massachusetts Institute of Technology, Cambridge, Massachusetts.
[1] K. Okamoto, Progr. Theoret. Phys. Japan **15**, 75 (1956).
[2] M. Danos, Bull. Am. Phys. Soc. Ser. II, **1**, 199 (1956); Nuclear Phys. **5**, 23 (1958).
[3] R. Nathans and J. Halpern, Phys. Rev. **73**, 437 (1954); R. Nathans and P. F. Yergin, Phys. Rev. **98**, 1296 (1955).
[4] Fuller, Petree, and Weiss, Chicago Photonuclear Conference, November 1956 (unpublished report); Petree, Weiss, and Fuller, Bull. Am. Phys. Soc. Ser. II, **2**, 16 (1957).
[5] Halpern, Mann, and Nathans, Rev. Sci. Instr. **23**, 678 (1952).