Title
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ABSTRACT

The atomic-beam magnetic-resonance method has been used to investigate 2.36-day Np^{239} in the low-field or Zeeman region of hyperfine structure. The spin of this nuclide is found to be 5/2, in agreement with the conclusions of Hollander, Smith, and Mihelich from beta and gamma spectroscopy and with the predictions of the Bohr-Mottelson model, but apparently in conflict with measurements by the methods of optical and paramagnetic-resonance spectroscopy. The principal observations have been made in a low-lying electronic state with measured $J = 11/2$, $g_J = 0.6551 \pm 0.0006$, which is probably the ground state of the electronic configuration $(5f)^4 (6d)^1 (7s)^2$.

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The atomic-beam magnetic-resonance method has been used to investigate 2.36-day Np^{239} in the low-field or Zeeman region of hyperfine structure. The spin of this nuclide is found to be 5/2 in agreement with the conclusions of Hollander, Smith, and Mihelich from beta and gamma spectroscopy[1] and with the predictions of the Bohr-Mottelson model, but apparently in conflict with measurements by the methods of optical[2] and paramagnetic-resonance[3] spectroscopy. The principal observations have been made in a low-lying electronic state with measured $J = 11/2$, $g_J = 0.6551 \pm 0.0006$, which is probably the ground state of the electronic configuration $(5f)^4 (6d)^1 (7s)^2$.

The material is produced in curie amounts by neutron activation of depleted (0.4% U^{235}) uranium. A beam is detected in one of two ways:
(a) the beam is collected on a sulfur surface and counted in scintillation counters with low beta-detection efficiency but with high efficiency for gamma rays between 20 and 200 kev, or (b) the beam is collected on a flamed platinum foil and detected in flow proportional counters sensitive to beta particles above about 5 kev.

An initial attempt to form a beam of neptunium was made by vaporizing the material directly from the uranium. Although the relative vapor pressures are suitable, this technique failed as a result of uranium creep. At beam temperatures the uranium interacts with the tantalum oven slits to form a low-melting-point alloy. The resulting destruction of the slits invariably leads to an intolerable background count at the detector.

A beam of neptunium is, however, successfully made by a high-temperature decomposition of neptunium carbide, which is in turn formed by an intermediate-temperature reduction of neptunium oxide by carbon. The gross target material is oxidized in air, mixed with a large excess of graphite powder, and placed in

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*This work was done under the auspices of the U.S. Atomic Energy Commission.

a tantalum oven. The reduction stage is signaled by the liberation of large quantities of CO, starting at a temperature of about 1000°C. When this stage is completed, the oven temperature is raised to 1800 to 2500°C to obtain a beam. The temperature dependence of neptunium effusion rate in this region is typically a factor of 4 per 100° temperature rise.

Low-field runs covering a considerable range of g values (Fig. 1) indicate that prominent resonances arise from the system $J = 11/2$, $I = 5/2$ and that there are probably other electronic states in the beam. The accuracy in the assignment of g values from these data is, however, too low to convincingly establish this assignment. Therefore each of the five prominent resonances has been followed to a magnetic field of 25 gauss. All observations of the six resonances associated with $J = 11/2$ are given in Table I. The sixth resonance associated with the state $F = 3$ is very weak relative to the other five because of apparatus discrimination, and is included for completeness only; the reliability of observations on this state is probably no better than five to one.

The essential conclusions that may be drawn from Table I are that, to the accuracy of measurement, the system is in the Zeeman region of hfs; that relative and absolute resonance intensities are consistent with the assumption that $J = 11/2$, $I = 5/2$ comprises a major fraction of the beam; and that, to an accuracy of one part in a thousand, all six transitions fit the system $J = 11/2$, $I = 5/2$, $g_J = 0.6551$. With the electronic angular momentum and g value thus established, a search was made for the spin 1/2 previously reported for this nucleus. When detection system (b) is used, and at a time 5 days after production of the sample, the product of relative decay rate and detection efficiency for spin 1/2 is found to be conservatively less than 5% of that for the spin-5/2 state.

Samples have been shown in several ways to be Np^{239}. First, aliquot fractions of the target and intense direct beam exposures have been shown to have a gamma spectrum essentially identical to that reported for this isotope,[4] and, secondly, half-lives have been taken on the target and on a direct beam and two resonances (Fig. 2).

If we assume that the ground-state configuration of neptunium contains only 5f and 6d electrons,[5] only the configurations $(5f)^4 (6d)^1$ and $(5f)^2 (6d)^3$ have, from Hund's rule, ground-state angular momenta $J = 11/2$. The configuration of uranium has been found[6] to be $(5f)^3 (6d)^1$ and that of plutonium is[7] probably $(5f)^6$. It is therefore highly probable that the ground-state configuration of neptunium is $(5f)^4 (6d)^1$. The g value of this state in pure Russell-Saunders coupling is 0.615; however, results of optical spectroscopic investigations in this region clearly show that this coupling scheme is inadequate to describe configurations involving unpaired 5f and 6d electrons. A much better approximation that has been found to give considerable success in interpreting the g values of uranium is that the electrostatic coupling between 5f and 6d electrons is small in comparison to the fine-structure coupling in each shell. In this approximation the two shells are individually in R-S coupling, and the electrostatic interaction between 5f and 6d electrons removes the degeneracy in the total angular momentum. Thus under this approximation the ground-state wave function is $({}^2D_{3/2}\text{-}{}^5I_4)_{11/2}$, giving a g value of 0.6547 if diamagnetic corrections and the relativistic breakdown of R-S coupling are neglected.

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LEGENDS

Fig. 1. Low-field search in neptunium. Resonances from the states $F = 8$ through $F = 4$ are shown.

Fig. 2. Decay of a direct beam and of resonances from the states $F = 8$ and $F = 6$.

Table I

Summary of data

	Magnetic field $\left(\frac{\mu_0 H}{h}\right)$	Total angular momentum					
		F=8	F=7	F=6	F=5	F=4	F=3
Experimental observations (g_F)	1.443 Mc/sec					0.76 ±0.02	1.04 ±0.02
	1.985 Mc/sec	0.43 ±0.015	0.479 ±0.015	0.534 ±0.015	0.635 ±0.015		
	2.969 Mc/sec	0.451 ±0.010	0.485 ±0.010	0.543 ±0.010	0.624 ±0.010	0.788 ±0.010	
	5.880 Mc/sec	0.451 ±0.005		0.540 ±0.005			
	11.544 Mc/sec	0.449 ±0.003	0.484 ±0.003				
	18.786 Mc/sec		0.4841 ±0.0015				
	27.386 Mc/sec			0.5379 ±0.0010	0.6222 ±0.0010	0.7697 ±0.0010	1.0649 ±0.0010
	35.535 Mc/sec	0.4505 ±0.0008	0.4856 ±0.0008	0.5379 ±0.0008	0.6223 ±0.0008	0.7686 ±0.0008	
Mean experimental value (g_F)		0.4504 ±0.0008	0.4853 ±0.0007	0.5379 ±0.0006	0.6223 ±0.0006	0.7692 ±0.0006	1.065 ±0.0010
Calculated g_F values; $J=11/2$, $I=5/2$, $g_J = 0.6551$, $g_I = 0$		0.4504	0.4855	0.5381	0.6223	0.7697	1.0645
Mean observed resonance intensity in percent direct beam		0.3%	0.3%	0.25%	0.15%	0.15%	0.04%
Calculated intensity for only $J=11/2$, $I=5/2$ in beam		0.47%	0.47%	0.47%	0.45%	0.36%	0.12%

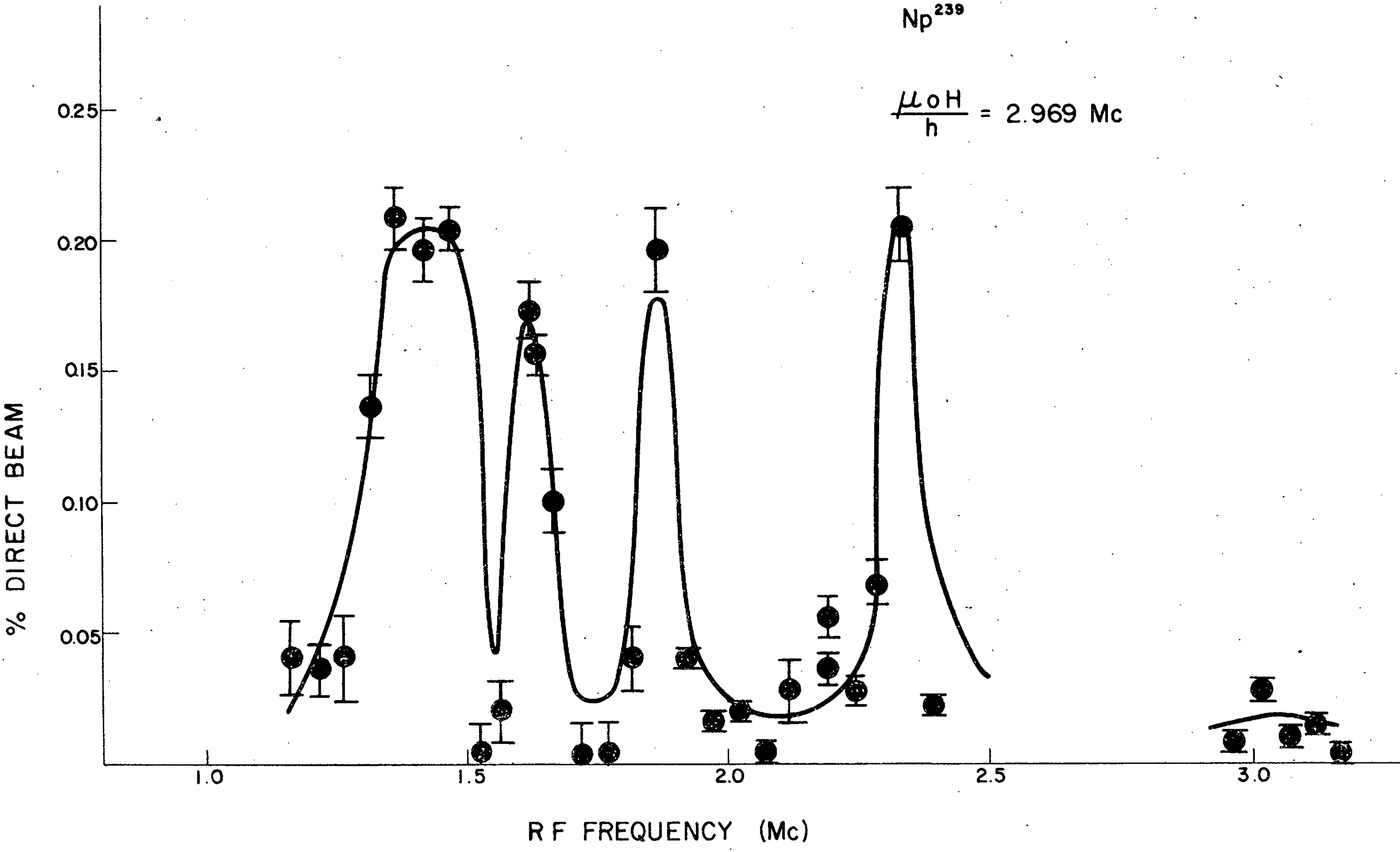
Np^{239}
$\frac{\mu_o H}{h} = 2.969$ Mc
0.25
0.20
0.15
0.10
0.05
% DIRECT BEAM
1.0
1.5
2.0
2.5
3.0
R F FREQUENCY (Mc)

Fig 1 -

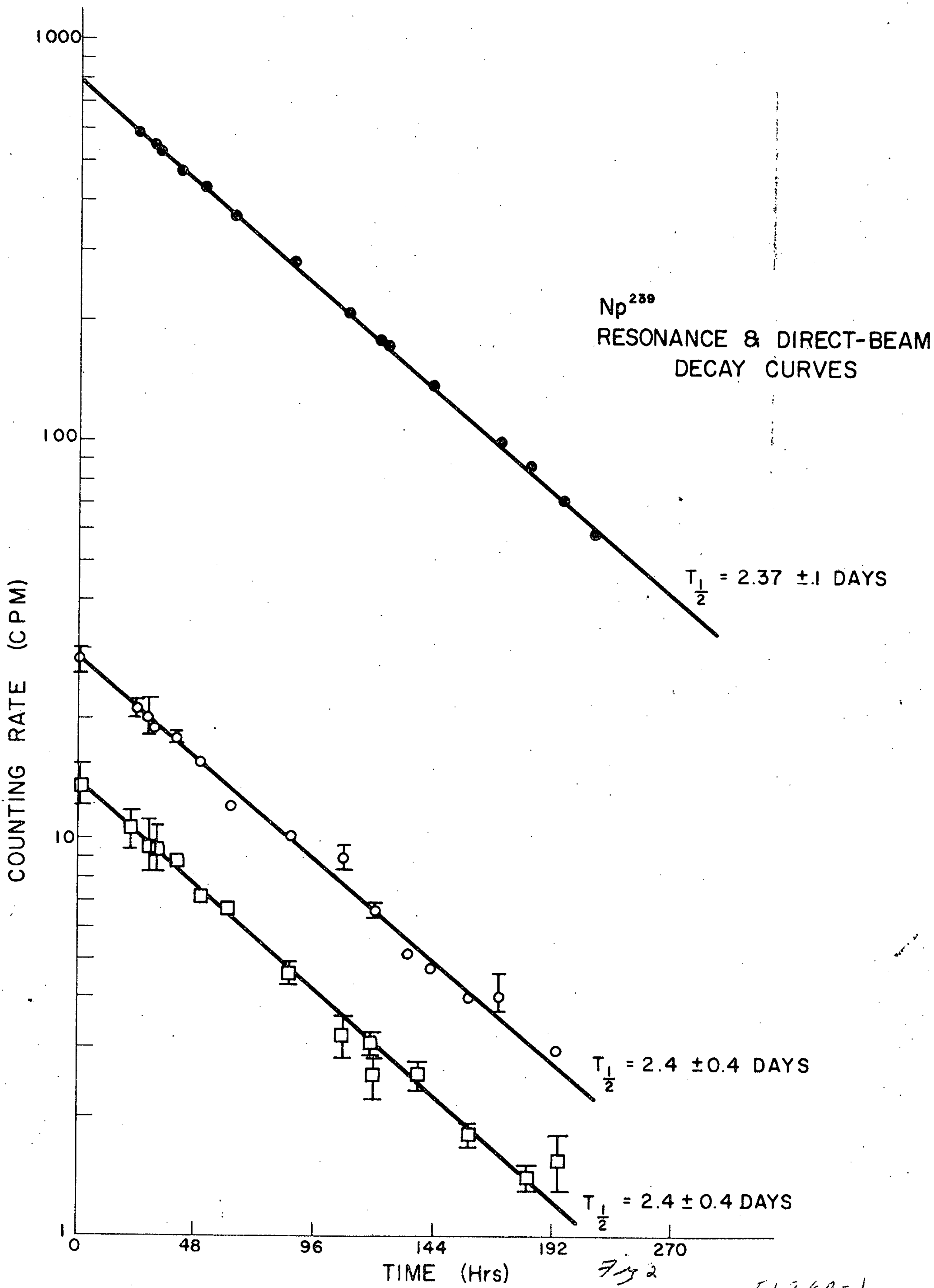
Np^{239}
RESONANCE & DIRECT-BEAM DECAY CURVES
$T_{\frac{1}{2}}$ = 2.37 ±.1 DAYS
$T_{\frac{1}{2}}$ = 2.4 ±0.4 DAYS
$T_{\frac{1}{2}}$ = 2.4 ± 0.4 DAYS
COUNTING RATE (CPM)
1000
100
10
1
TIME (Hrs)
0
48
96
144
192
270

Fig 2