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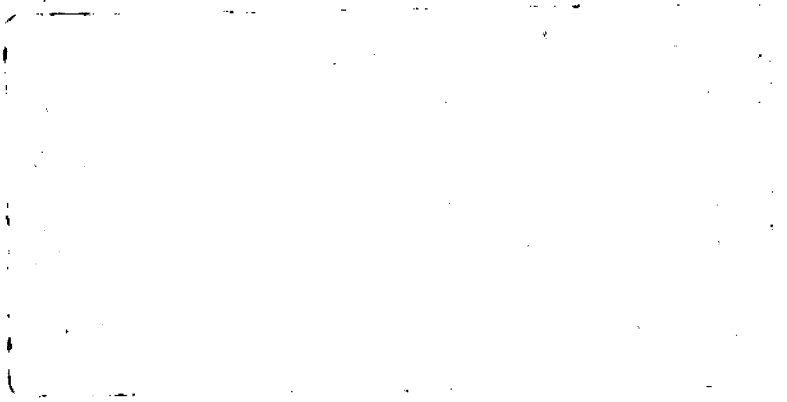

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ABSTRACT

The atomic-beam magnetic-resonance method has been used to investigate 163-day Cm^{242}. The spin of this even-even nuclide is found to be zero. Four low-lying electronic energy levels are found and the Lande g-factors are measured to be $g_{J_2} = 2.561 \pm .003$, $g_{J_3} = 2.000 \pm .003$, $g_{J_4} = 1.776 \pm .002$ and $g_{J_5} = 1.671 \pm .003$. No direct measurement can be made of the angular momenta of these levels, but other considerations contained in the text make highly probable the J values indicated in subscripts and arising from the electronic configuration $(5f)^7 (6d)^1 (7s)^2$.

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INTRODUCTION

The research reported here is a portion of a general program in the heavy-element region leading to nuclear moments and electronic properties. Since, as expected, the spin of Cm^{242} is zero, the principal contributions of this research are in the field of electronic structure. Electronic ground states have previously been identified from ^{92}U to ^{95}Am.[1] This research extends such knowledge to ^{96}Cm.

EXPERIMENTAL DETAIL

The experiment has been performed by the use of a conventional Zacharias apparatus. The apparatus has been described elsewhere.[2] Curium-242 is obtained essentially pure by activity, as established from observation of the alpha spectrum. The material is detected by allowing the thermal, neutral beam to fall on freshly flamed platinum foils which are introduced into and removed from the apparatus via a vacuum lock and are then placed in low-background ($\approx$ 0.5 counts/minute) proportional counters. Typical resonance counting rates are 2 to 25 counts/minute. Resonances are normalized by direct beams taken between each resonance exposure and the C field is calibrated with an auxiliary beam of K^{39} issuing from a movable source in the buffer chamber.

A beam of curium atoms is obtained by the same high-temperature procedure used in the neptunium research.[3] The curium, used in quantities of a few micrograms at a time, is mixed with several milligrams of uranium for carrier purposes, oxidized, and placed in a small tantalum oven with a large excess of carbon. The oxides are reduced to carbides at a temperature

of about 1200°C, and the oven is raised to a temperature of about 1800°C, where a beam of curium atoms is made by thermal decomposition of the carbides. No difficulties were encountered in this phase of the research—an atomic beam was obtained on the first try, and there were no subsequent failures of the procedure.

EXPERIMENTAL RESULTS

An initial search was made at a C field of 0.7 gauss over a range of frequency covering g values between 1 and 3. The search was later extended down to g = 0. The results indicate four prominent resonances in this interval. The four resonances were followed to a field of 27 gauss—the data obtained is given in Table I and the resonances are illustrated in Fig. 1. The essential results are that there are four resonances of comparable intensity which, to the accuracy of measurement, occur at a frequency that is directly proportional to the external magnetic field. The system is therefore in the Zeeman region of hfs or in the Paschen-Back region of hfs (Zeeman region of optical spectroscopy) with a total hfs width less than the observed line width, 100 kc/sec. Because the magnetic-dipole hfs constant for 5f and 6d electrons is about 500 g_I Mc/sec, the latter possibility forces a nuclear magnetic moment less than about 2×10^{-4} nm. On the other hand no integer values of I and J can fit the observed g values if the system is assumed to be in the Zeeman region of hfs where $\vec{F} = \vec{I} + \vec{J}$ is a good quantum number. Thus the spin of Cm^{242} is almost certainly zero, and four low-lying electronic energy levels have been found. This makes possible a crude measurement of the relative positions of the four energy levels by a careful measurement of relative transition intensities in the four states. This has been done according to the procedure outlined in the section on curium energy levels. The results of these measurements are shown in Table II and show a monotonic decrease in transition intensity toward the state of highest g value.

Table I

Summary of experimental observations

$\frac{\mu_0 H}{h}$ (Mc)	$g_J=2$	$g_J=3$	$g_J=4$	$g_J=5$
0.998	2.50±.05	1.95±.05	1.74±.04	1.63±.04
2.969	2.554±.016	2.00±.02	1.78±.02	1.67±.02
9.679	2.561±.012	2.000±.008	1.772±.009	———
18.786	2.561±.003	2.000±.003	1.776±.002	1.671±.003
Weighted mean	2.561±.003	2.000±.003	1.776±.002	1.671±.003

Table II

Resonance intensity in each of the four states at the rf current setting that maximizes the transition probability.

State	Observed intensity (%)
J = 2	1.45 ± .25
J = 3	1.08 ± .17
J = 4	0.83 ± .12
J = 5	0.43 ± .06

THE CONFIGURATION OF CURIUM

A large body of chemical information[4] and two investigations of CmIV by paramagnetic resonance absorption[5] have shown that the ground-state configuration of CmI should be either $(5f)^8 (7s)^2$ or $(5f)^7 (6d)^1 (7s)^2$, with a preference for the latter based largely on the fact that the equivalent configuration is found for GdI,[6] the rare-earth analogue of CmI. The f^7d configuration leads, in pure Russell-Saunders coupling, to the ground-state term ^9D, giving rise to levels $J = 2$, $g_J = 2.668$; $J = 3$, $g_J = 2.083$; $J = 4$, $g_J = 1.850$; $J = 5$, $g_J = 1.733$; and $J = 6$, $g_J = 1.667$. On the other hand the configuration f^8 leads, in pure R—S coupling, to the term ^7F with J values from 6 to 0, all having $g_J = 1.50$. It is found in Pu that the ^7F term is very near R—S coupling, the $^7F_1$ state having a g value of $1.4975 \pm .0010$. Therefore the observed g values strongly suggest that the configuration of CmI is indeed $(5f)^7 (6d)^1 (7s)^2$, but that pure R—S coupling is here, as in Np, a poor approximation for configurations involving 5f and 6d electrons. The observed g values for the ground states of UI and NpI indeed indicate that, to a fairly high degree of approximation, the electrostatic interaction between 5f and 6d electrons is small in comparison to the fine-structure splittings of either system.

A calculation has been made of the energies and g values of the configuration $(5f)^7 (6d)^1 (7s)^2$ in intermediate coupling, the only approximation being that the $^8S_{7/2}$ state is not perturbed by electrostatic terms between 5f and 6d electrons. The fit that can be made to the energy levels of Gd is quantitative and gives for the parameters of interest $24G_1 + 96G_3 (4f\text{-}5d) = 6500\ cm^{-1}$ and $a(5d) = 500\ cm^{-1}$. On the other hand the theory fits the g values of CmI only semiquantitatively. The question then arises: What are the essential differences between CmI and GdI? First, there is a striking disparity in the relative values of a and G between the two systems. While the Gd data fit very nearly at the R—S extreme (G much larger than a), the Cm data fit much better near the other limit for which G is small in comparison to a. This is, however, the approximation that gives the observed g values in Np and U and so must be taken at face value. We therefore now include the possibility that the g values of the two subshells are not those of pure R—S coupling, i.e., we allow configuration perturbations in the d and f shells. Then, in the limit G very small in comparison to a, the g_J values of the levels in Cm will be:

$$g_J = g_{J_1} \frac{J(J+1) + j_1(j_1+1) - j_2(j_2+1)}{2J(J+1)} + g_{J_2} \frac{J(J+1)+j_2(j_2+1)-j_1(j_1+1)}{2J(J+1)} .$$

This approximation will fit the data essentially to the experimental accuracy, provided that one assumes the following values:

$$J_1 = 7/2, \; g_{J_1} = 2.002$$

$$J_2 = 3/2, \; g_{J_2} = 0.890.$$

CURIUM ENERGY LEVELS

The energy separations of the four observed states in curium are measurable from an observation of resonance intensities for a known rf current. The procedure is to first measure in the state of highest g value, the intensity at resonance for various values of the rf current (Fig. 2). The resonance intensity in the other states is measured at an rf field calculated from the previous observations in the state J = 2 and the Majorana formula.[7]

The ratio of these observed intensities (I_{J_1}/I_{J_2}) can be related to the energy separations ($E_{J_1} - E_{J_2}$) according to the formula

$$I_{J_1}/I_{J_2} = \exp\left(\frac{E_{j_1} - E_{j_2}}{kT}\right) \frac{\sum_{m=-J_1}^{J_1} \bar{P}_m(J_1)}{\sum_{m=-J_2}^{J_2} \bar{P}_m(J_2)} . \qquad (1)$$

Here $\bar{P}_m(J)$ is the probability for transition, averaged over the distribution of velocities in the beam, of an atom in the state J, m to the state J, -m under the action of the applied rf current. Explicitly it is written as

$$\bar{P}_m(J) = \left[(J-m)!\,(J+m)!\right]^2 \int_0^\infty \sin^{4J}\left(\frac{b}{v}\right) v^3 \frac{2}{a^4} \exp(-v^2/a^2)$$

$$\times \left(\sum_r \frac{(-1)^r \cot^{2r}\left(\frac{b}{v}\right)}{(J-m-r)!\,(J+m-r)!\,[r!]^2} \right)^2 dv , \qquad (2)$$

where b is proportional to the applied rf current amplitude, α is the most probable velocity in the Maxwell-Boltzmann distribution for the beam temperature, and the sum is over all values of r that keep the factorials positive. It is empirically determined that the apparatus does not preferentially select atoms of a given velocity to any measurable degree, hence $\overline{P}_m(J)$ does not include such a factor. These integrals were computed with the aid of an IBM-650 computer. The quantities $\sum_m \overline{P}_m(J)$ are plotted in Fig. 3 for the J states present in curium. The applicability of this theory to curium is demonstrated by comparing the results in the state J=2 to the experimentally determined intensity curve (Fig. 2).

By the use of the experimentally determined intensities and the computed probabilities at maximum, the energy level separations were computed from equation (1). The results are shown in Fig. 4.

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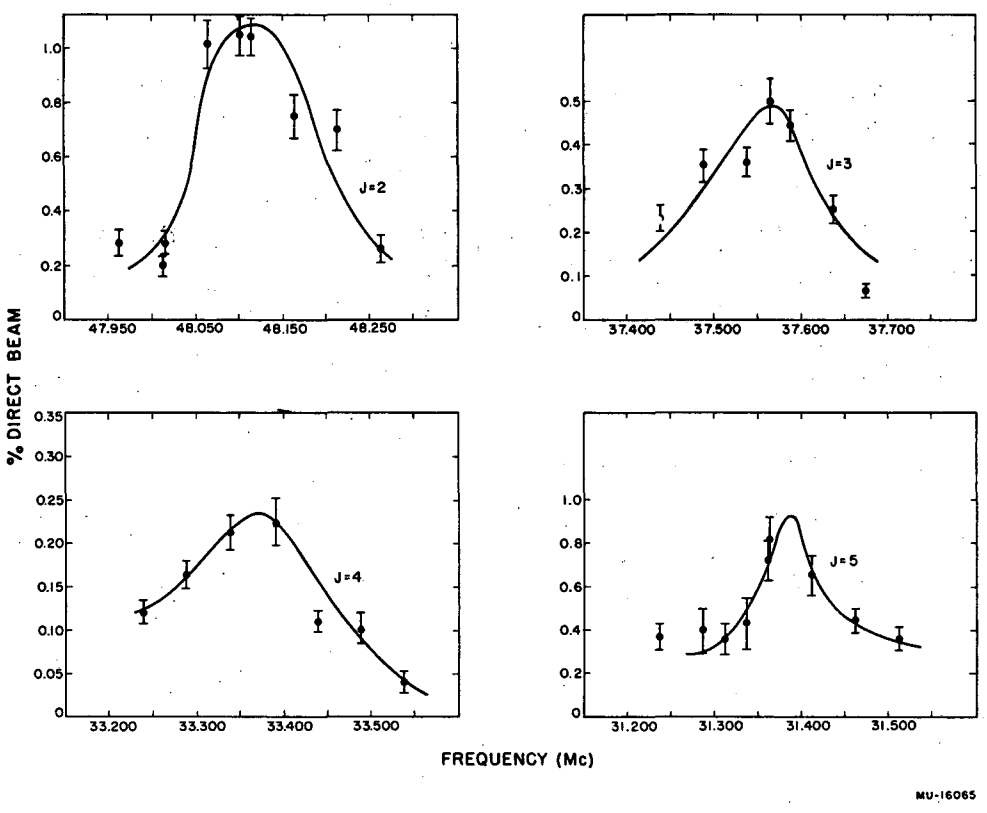

Fig. 1. Transitions in the four observable states of Cm^{242} at 13.4 gauss.

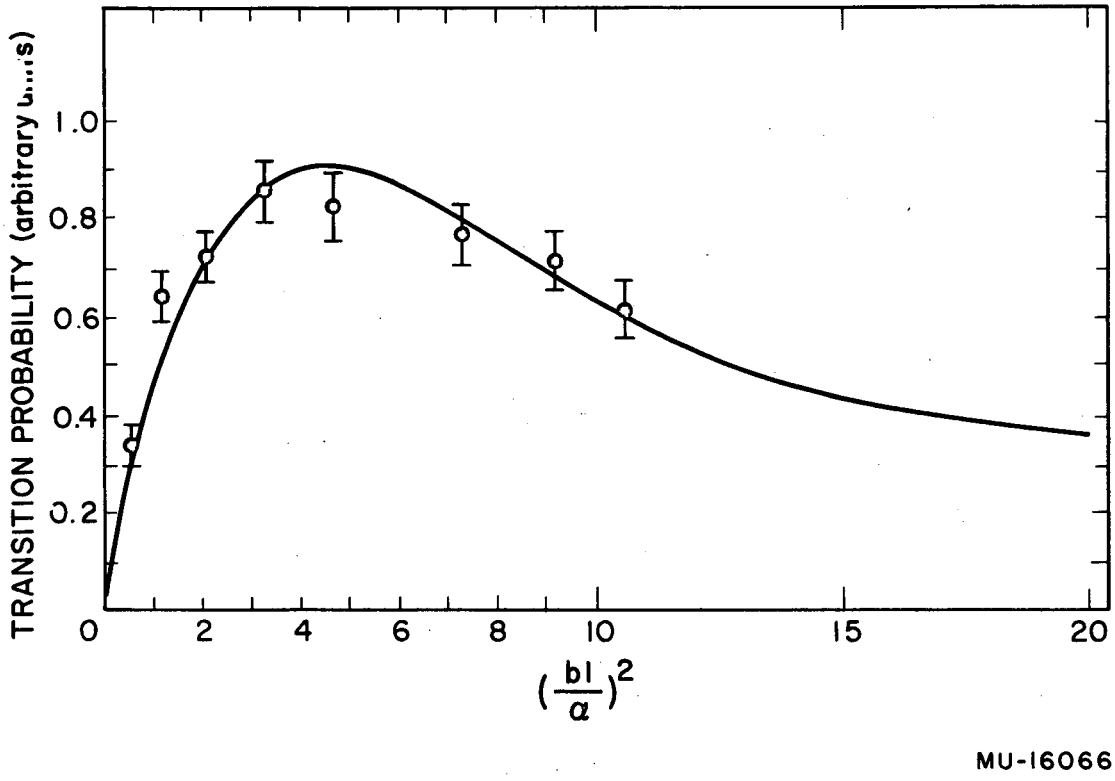

Fig. 2. Comparison of experimental observations and the theoretical transition probability in the J=2 state of curium.

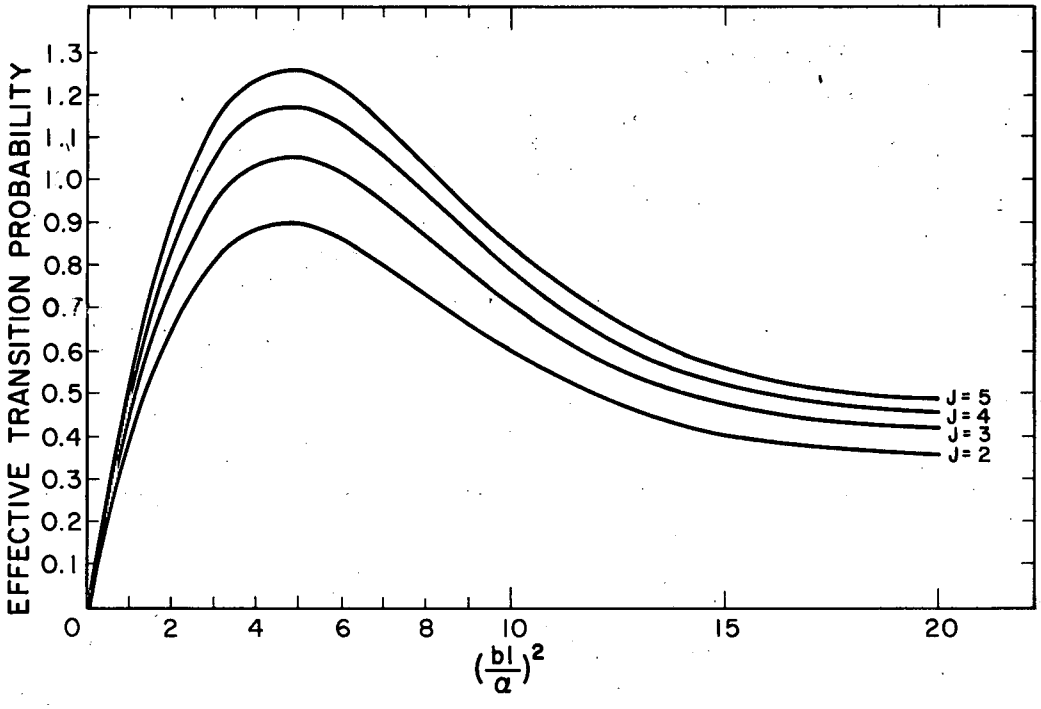

Fig. 3. Calculated transition probabilities for the four states of curium. The abscissa is a quantity which is proportional to the square of the applied rf current. The ordinate is the sum over all flop-in transitions in the particular J state, of the Majorana transition probabilities averaged over the atomic-beam velocity distribution.

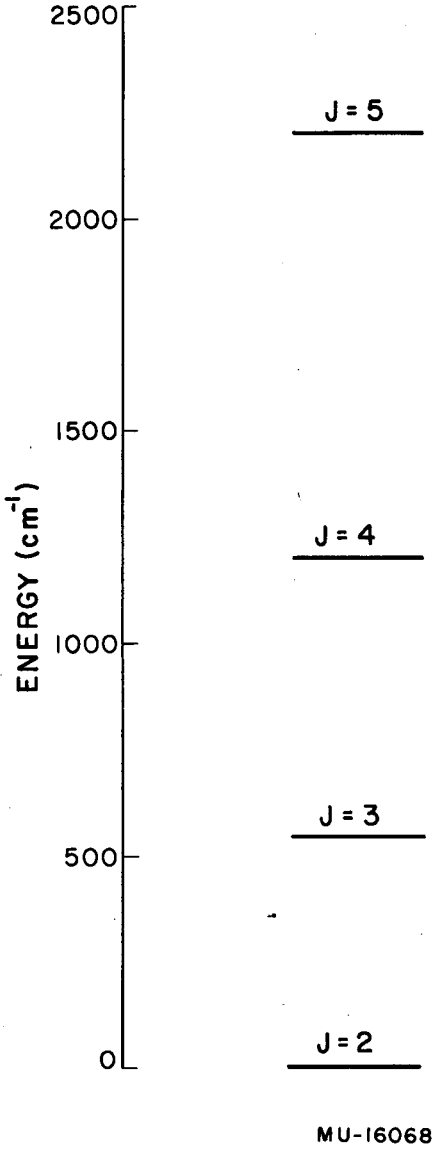

Fig. 4. Approximate curium energy levels as inferred from transition intensities.