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Ernest O. Lawrence Radiation Laboratory

ELECTRONIC ANGULAR MOMENTUM OF SOME LOW-LYING STATES OF CERIUM; NUCLEAR SPIN OF CERIUM-143

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Isaac Maleh

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ABSTRACT

The electronic angular momentum of some low-lying states of cerium were determined by means of the atomic-beam magnetic resonance technique. The work was done on the 32-d Ce^{141} isotope using both the reported nuclear spin result of Ce^{141} ($I = 7/2$) and the g_J states as determined on Ce^{140}. The results are: $g_J = -1.0772$, $J = 3$; $g_J = -0.9454$, $J = 4$; $g_J = -0.7651$, $J = 4$. Configuration mixing is required to interpret these results.

By the same experimental method the nuclear spin of Ce^{143} was determined. The result ($I = 3/2$) is consistent with angular correlation data and decay schemes of this isotope.

I. INTRODUCTION

By means of the atomic-beam magnetic resonance technique ground-state nuclear spins have been determined for those atoms whose electronic angular momentum states were known. A slight modification of the technique will yield the electronic angular momentum (J) if the nuclear spin is known. A very brief summary of the theory is presented; for a more thorough analysis, see Ref. 1.

An atom with electronic angular momentum J and nuclear spin I can have total angular momenta $F = |I + J|, |I + J - 1|, \ldots, |I - J|$. Each F state has a (2F+1)-fold degeneracy which can be removed by a magnetic field. At relatively low field ($H < \approx 50$ G), the hyperfine structure (hfs) Hamiltonian can be written

$$\mathcal{H}_{hfs} = A\underset{\sim}{I}\cdot\underset{\sim}{J} + B\underset{\sim}{Q}(I,J) - g_F\mu_o m_F H \qquad (1)$$

where

$$g_F \cong g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} \quad . \qquad (2)$$

For a known g_J, a given J (or I) and an assumed I (or J), g_F can be computed for the various F levels allowed. Energy differences as a function of magnetic field for $\Delta F = 0$, $\Delta m_F = \pm 1$ transitions are calculated and after the H field is set using a resonance of K^{39}, predicted transitions are looked for in the atomic beam machine. An energy level diagram for one of the states is shown in Fig. 1 with the transitions indicated.[2]

Making use of the results on Ce^{140} which yielded the g_J's of cerium[3] and the paramagnetic resonance result[4] on Ce^{141} yielding the spin of Ce^{141}, the J values corresponding to the given g_J's were

measured. Also with one of these g_J, J sets the nuclear spin of Ce^{143} was measured.

II. CERIUM-141

A. Experiment

The isotope was produced by bombarding approximately 1 gm of stable Ce metal for 10 days at the MTR at Arco, Idaho. The flux used was approximately 2×10^{14} neutrons/cm^2-sec. The reaction is:

$$Ce^{140}(n,\gamma)Ce^{141} \qquad (\tau \approx 32 \text{ days}).^5$$

The material was delivered 4-7 days later allowing time for the 33-h Ce^{143} and 9-h Ce^{137} to decay. A negligible amount of Ce^{139} (τ = 140 d) was produced. Figure 2 shows the decay of a direct beam proving that most of the activity is indeed the 32-d Ce^{141}. The main experimental difficulty lay in obtaining a steady beam since cerium is known to react with the more common oven materials. Smith and Spalding reported the use of a thorea crucible[3] as well as a tungsten crucible.[6] In earlier stages of the work a thorea crucible inside a Ta oven was used and afterwards some of the results confirmed with a tungsten oven and crucible.[7] The beam was detected using one-mil platinum foils to collect the atoms and continuous-flow beta counters to measure the activity.

As mentioned above[3] three states were observed in the beam of Ce^{140} having $g_J = -1.0772$, $g_J = -0.9454$, and $g_J = -0.7651$, respectively. A "J" search was conducted for each of these states using the paramagnetic resonance result on Ce^{141} ($I = 7/2$).[4]

B. Results

1. $g_J = -1.0772(2)$

For two values of the magnetic field (H = 5.566 G and H = 8.247 G), all the $J = 3$ predictions were verified. Figure 3 shows the results at H = 8.247 G, and Fig. 4 shows a decay of one of the resonance foils ($F = 13/2$). The frequency at the very high point also corresponds closely to that predicted for $g_J = -0.7651$, $J = 4$, $F = 15/2$, as well as to the $F = 7/2$ state of $g_J = -1.0772$. A thorea crucible was used throughout this investigation.

2. $g_J = -0.9454(1)$

Again at two settings of the magnetic field (5.566 G and 8.247 G) and using a thorea crucible a J search was performed yielding $J = 4$ consistently. Figure 5 illustrates the data at 8.247 G.

3. $g_J = -0.7651(1)$

Using a thorea crucible and a magnetic field of 8.247 G, a search resulted in either $J = 4$ or $J = 6$ (see Fig. 6). The $J = 6$, $F = 19/2$, and 17/2 resonances also correspond closely to those predicted for $g_J = -1.0772$, $J = 3$, $F = 11/2$ and $g_J = -0.9454$, $J = 4$, $F = 15/2$, respectively; and another search at 10.865 G confirmed $J = 4$ for four different F states while the $J = 6$, $F = 15/2$ prediction was low. Table I is a listing of the predicted resonances at 8.247 G.

Recent optical spectroscopy results[8] indicated five low-lying levels for cerium which include $g_J = -1.077$, $J = 3$; $g_J = -0.945$, $J = 4$; and a $g_J = -0.765$, $J = 2$ (also $g_J = -0.886$, $J = 4$;

$g_J = -0.735$, $J = 3$). Because of this, an attempt was made to see the remaining two states in the beam as well as verify the author's $g_J = -0.7651$, $J = 4$ assignment. A tungsten oven and crucible was used and at two different fields (H = 5.567 G and H = 10.865 G) predicgions for $J = 4$ and $J = 2$ were tested. Figures 7 and 8 clearly show that state seen in the beam is $g_J = -0.7651$, $J = 4$. Experimental difficulties (intensity mostly) prevent any comments concerning the existence of the other two states in the beam. Table II summarizes the results.

C. Interpretation

Cerium has atomic number 58 and since it is very early in the rare earth series, it has usually been assigned the configuration $(4f)^2(6s)^2$ or $4f5d(6s)^2$. Table III shows the g_J values for all possible states using Russell-Saunders coupling. This table is for two unpaired electrons in the ground state. It is not possible to reconcile the experimental results without admixing a configuration containing four unpaired electrons into the ground state. For example, consider $g_J = -0.7651$, $J = 4$; with two unpaired electrons one has singlet or triplet states; to get $J = 4$ the terms needed are ^1G or ^3F, ^3G, ^3H. The ground state is either one of these pure R-S states or a mixture of these four, but each has a $g_J = -0.800$ or less while a $g_J = -0.7651$ is needed. If however a configuration such as $4f(5d)^2 6s$ is mixed into the ground state, it has a $^5I_4$ term with a $g_J = -0.600$ and the same parity; it would be possible to get $g_J = -0.7651$, $J = 4$. The ground state is probably a mixture of two or more configurations. The problem of fitting this data is difficult since the electronic wave functions

(needed for spin-orbit corrections)[9] as well as the amount of admixing are unknown. If there is an unpaired s electron, the hfs structure should be large; this measurement might yield information as to the amount and type of admixing.

III. CERIUM-143

A. Experiment

This isotope was produced by neutron bombardment for 48 hours at the G.E. reactor in Vallecitos, California. A flux of about 9×10^{13} neutrons/cm^2-sec produced just enough activity to make the experiment feasible. The reaction is:

$$Ce^{142}(n,\gamma)Ce^{143} \qquad (\tau = 33 \text{ h}).^{10}$$

Due to a smaller cross section or unfavorable lifetimes, no appreciable amount of the other cerium isotopes was produced. A tungsten oven and crucible was used throughout and beam detection was identical to that for Ce^{141}.

B. Result and Interpretation

Using the $g_J = -0.9454$, $J = 4$ state, a spin search was made at 6.915 G and 13.422 G. Both investigations, as shown in Figs. 9 and 10, yield $I = 3/2$.

Cerium-143 has 58 protons and 85 neutrons in its nucleus. Each of the three neutrons outside the magic number 82 is in the (f 7/2) state according to shell model theory. Three identical particles in the same j state usually couple to $J = j - 1$.[11] This would yield $J = 5/2$ which is not observed. If long range Majorana Forces were in operation, spin 3/2 would correspond to the lowest-lying state.[11]

The decay scheme for Ce^{143} is given in Fig. 11. The ground state spin assignment for Pr^{143} was determined by atomic beams[12] and those of the first two excited states by studying angular correlation results and nuclear alignment data.[13,14,15] The fact that Ce^{143} decays into the spin 3/2, 351-keV level and spin 5/2, 57-keV level makes improbable a spin greater than 5/2 for Ce^{143}. The atomic beam data indicates $I = 3/2$ which would also be consistent with the fact that no decay to the ground state of Pr^{143} ($I = 7/2$) is observed.[13]

The cerium atom with its mixed ground state configuration and this isotope with its anomalous spin make the element one of the more interesting ones in the periodic table.

FOOTNOTES AND REFERENCES

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

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7. For details on the thorea crucible, see Isaac Maleh, Electronic Angular Momentum of Some Low-Lying States of Cerium; Nuclear Spin of Cerium-143, Lawrence Radiation Laboratory Report UCRL-11794, December 1964 (Appendix).
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13. Richard M. Levy, Perturbed Angular Correlation Measurements on Ce^{141} and Pr^{143} (Ph.D. Thesis), Lawrence Radiation Laboratory Report UCRL-11663, September 1964 (unpublished).

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Table I. Cerium-141 predicted resonances at 8.247 G.

Frequency (Mc/sec)	State (g_J, J, F)
4.710*	c, 4, α
4.737*	a, 3, δ
4.801	c, 4, β
4.941	c, 4, γ
5.175	c, 4, δ
5.276	a, 3, γ
5.567	a, 3, β
5.739	a, 3, α
5.821	b, 4, α
5.933	b, 4, β
6.106	b, 4, γ
6.394	b, 4, δ

a $\longleftrightarrow g_J = -1.0772(2)$	$\alpha \longleftrightarrow F = F_{max} = \lvert I + J \rvert$; $I = 7/2$
b $\longleftrightarrow g_J = -0.9454(1)$	$\beta \longleftrightarrow F = F_{max} - 1$
c $\longleftrightarrow g_J = -0.7651(1)$	$\gamma \longleftrightarrow F = F_{max} - 2$
	$\delta \longleftrightarrow F = F_{max} - 3$

* These frequencies are very likely to have overlapping lines.

Table II. States of cerium seen by the method of atomic beams.

g_J	J
- 1.0772(2)	3
- 0.9454(1)	4
- 0.7651(1)	4

Table III. Possible configurations for cerium, and -g (for R-S coupling) for the given levels.

Configuration	Term	J = 6	J = 5	J = 4	J = 3	J = 2	J = 1
$4f^2\ 6s^2$	^3H	1.167	1.033	0.800			
	^3F			1.250	1.083	0.667	
	^3P					1.500	1.500
	^1I	1.000					
	^1G			1.000			
	^1D					1.000	
	^1S						
$4f^1\ 5d^1\ 6s^2$	^3H	1.167	1.033	0.800			
	^3G		1.200	1.050	0.750		
	^3F			1.250	1.083	0.667	
	^3D				1.333	1.167	0.500
	^3P					1.500	1.500
	^1H		1.000				
	^1G			1.000			
	^1F				1.000		
	^1D					1.000	
	^1P						1.000
$5d^2\ 6s^2$	^3F			1.250	1.083	0.667	
	^3P					1.500	1.500
	^1G			1.000			
	^1D					1.000	

FIGURE CAPTIONS

Fig. 1. Energy level schematic for one of the Ce^{141} states.

Fig. 2. Decay of a direct beam of Ce^{141}.

Fig. 3. A "J" search at 8.247 G for Ce^{141}, $g_J = -1.0772$.

Fig. 4. Decay of a resonance foil of Ce^{141}.

Fig. 5. A "J" search at 8.247 G for Ce^{141}, $g_J = -0.9454$.

Fig. 6. A "J" search at 8.247 G for Ce^{141}, $g_J = -0.7651$.

Fig. 7. Comparison of $J = 4$ and $J = 2$ predictions at 5.566 G; $g_J = -0.7651$ (using a tungsten oven and crucible).

Fig. 8. Comparison of $J = 4$ and $J = 2$ predictions at 10.865 G; $g_J = -0.7651$ (using a tungsten oven and crucible).

Fig. 9. Ce^{143} spin search at 6.915 G.

Fig. 10. Ce^{143} spin search at 13.422 G.

Fig. 11. Decay scheme of Ce^{143} according to R. M. Levy (Ref. 13).

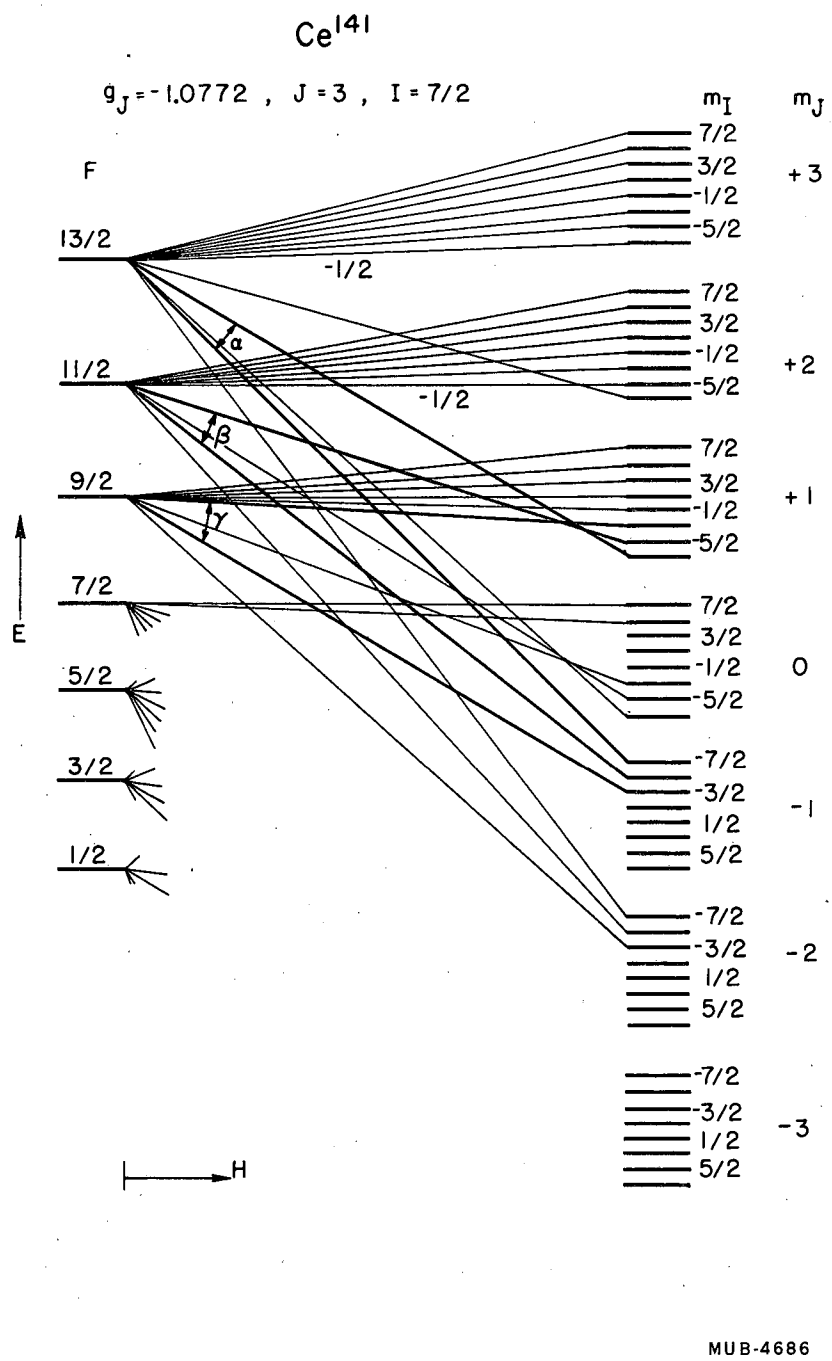

Ce141
gJ = -1.0772 , J = 3 , I = 7/2
F
13/2
11/2
9/2
7/2
5/2
3/2
1/2
E
H
α
β
γ
-1/2
-1/2
mI
mJ
7/2
3/2
-1/2
-5/2
+3
+2
+1
0
-1
-2
-3
-7/2
-3/2
1/2
5/2
MUB-4686

Fig. 1

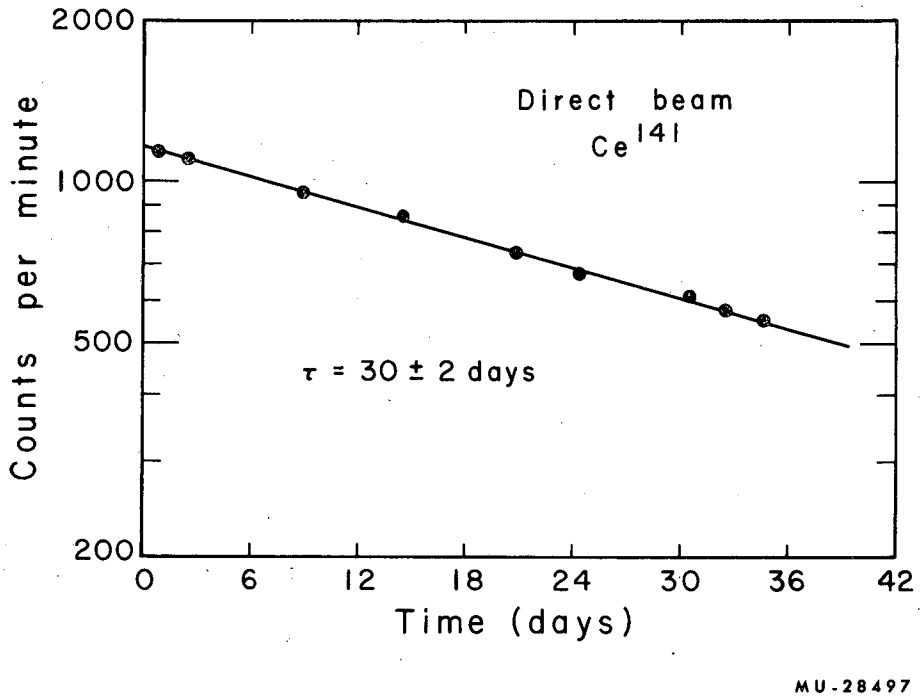

2000
1000
500
200
Counts per minute
Direct beam
Ce141
τ = 30 ± 2 days
0
6
12
18
24
30
36
42
Time (days)
MU-28497

Fig. 2

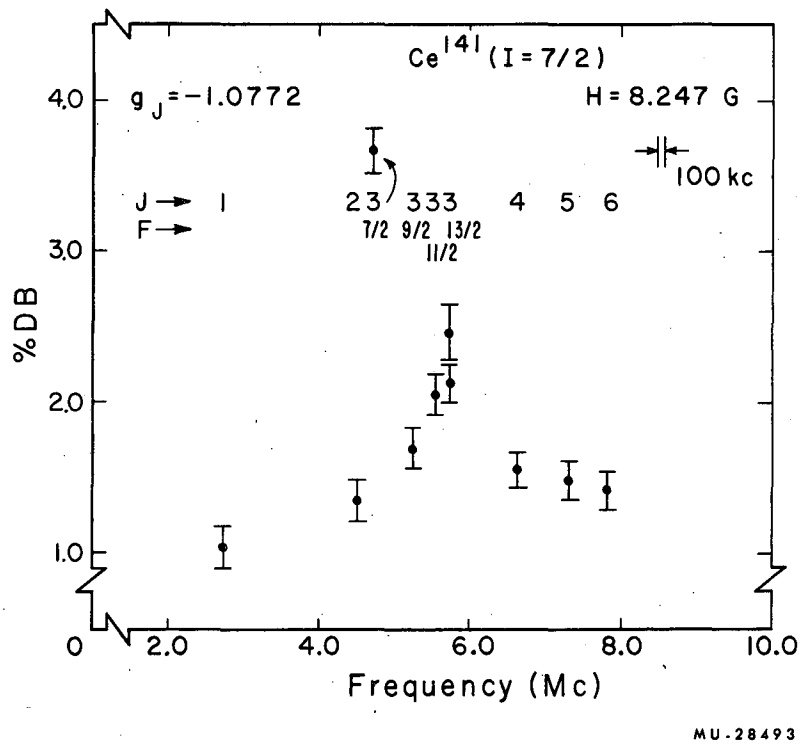

Ce141 (I = 7/2)
gJ = −1.0772
H = 8.247 G
100 kc
J → 1 2 3 3 3 3 4 5 6
F → 7/2 9/2 13/2 11/2
% DB
4.0
3.0
2.0
1.0
0
2.0
4.0
6.0
8.0
10.0
Frequency (Mc)
MU-28493

Fig. 3

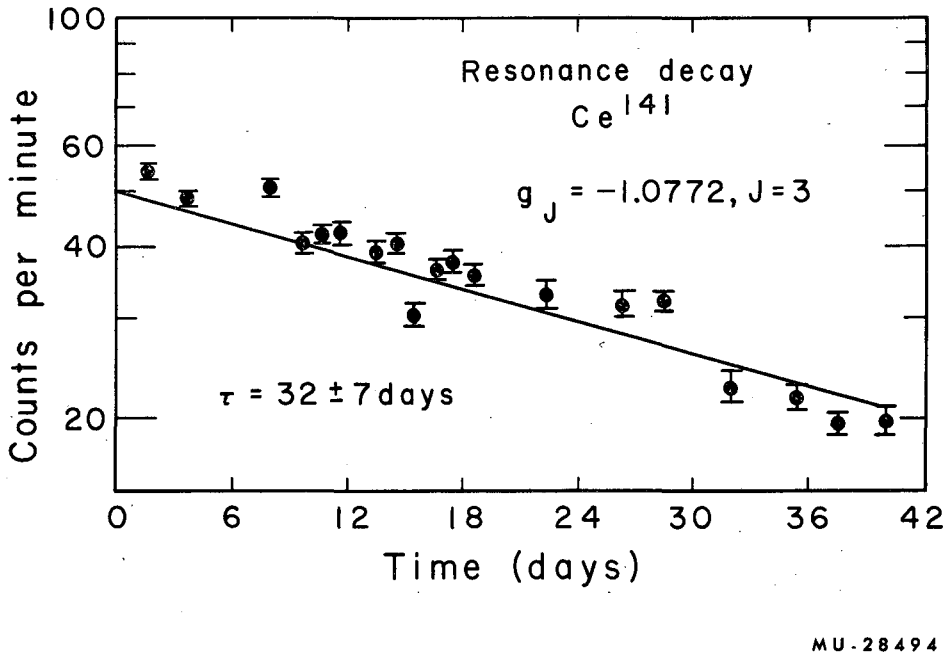

Resonance decay
Ce141
gJ = −1.0772, J=3
τ = 32 ± 7 days
Counts per minute
100
60
40
20
0
6
12
18
24
30
36
42
Time (days)
MU-28494

Fig. 4

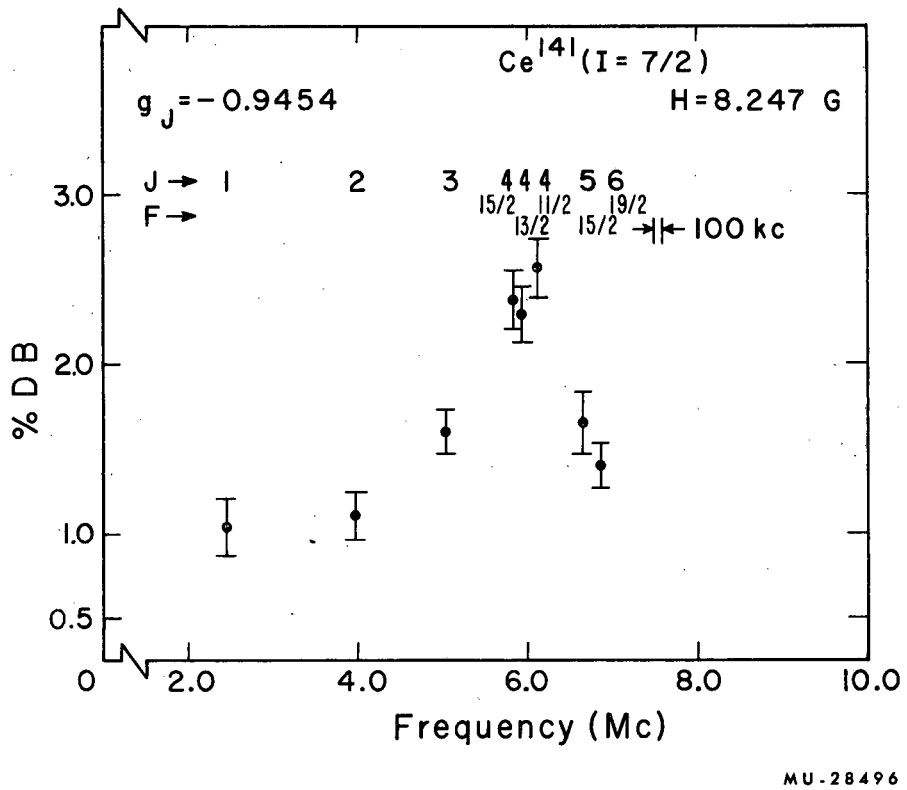

Ce141 (I = 7/2)
gJ = −0.9454
H = 8.247 G
J → 1 2 3 4 4 4 5 6
F → 15/2 11/2 19/2
13/2 15/2
100 kc
% D B
3.0
2.0
1.0
0.5
0
2.0
4.0
6.0
8.0
10.0
Frequency (Mc)
MU-28496

Fig. 5

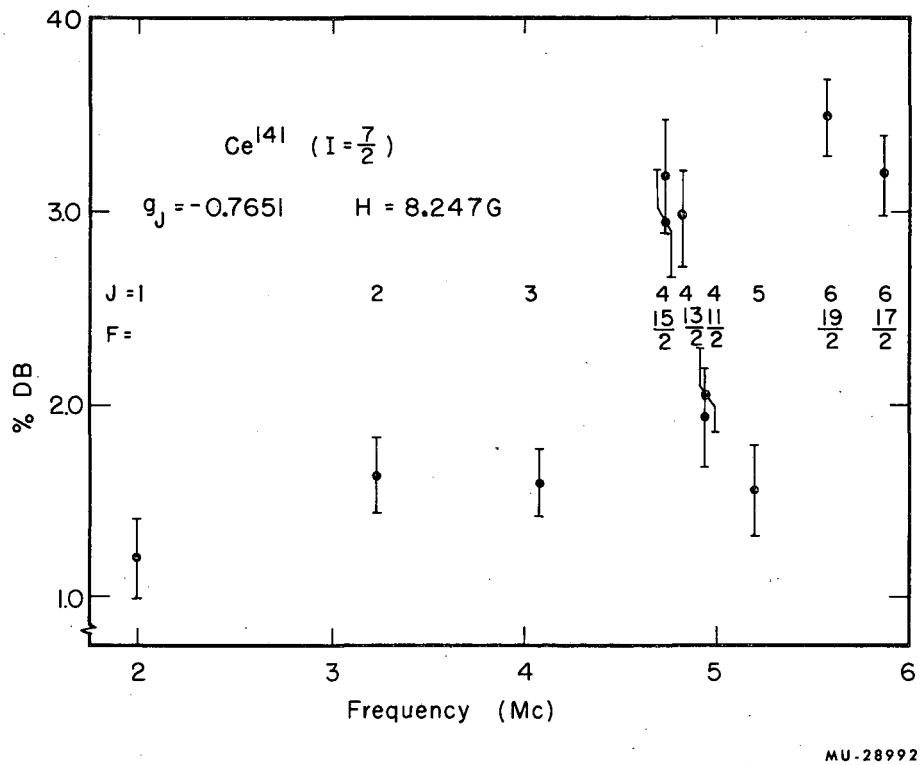
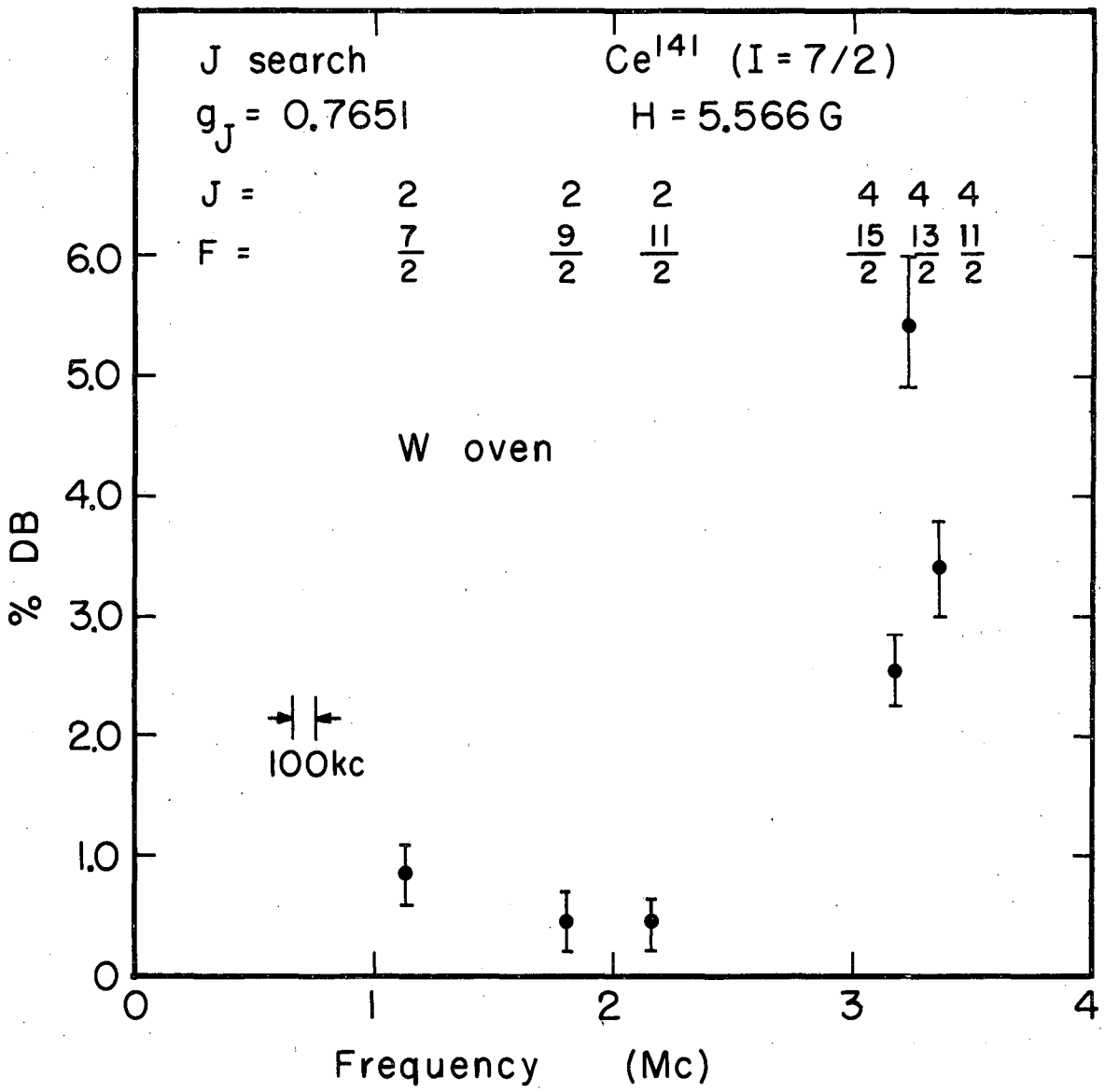

40
3.0
2.0
1.0
% DB
Ce141 (I = 7/2)
gJ = -0.7651
H = 8.247G
J = 1
2
3
4
4
4
5
6
6
F =
15/2
13/2
11/2
19/2
17/2
2
3
4
5
6
Frequency (Mc)
MU-28992

Fig. 6



MUB-2830

Fig. 7

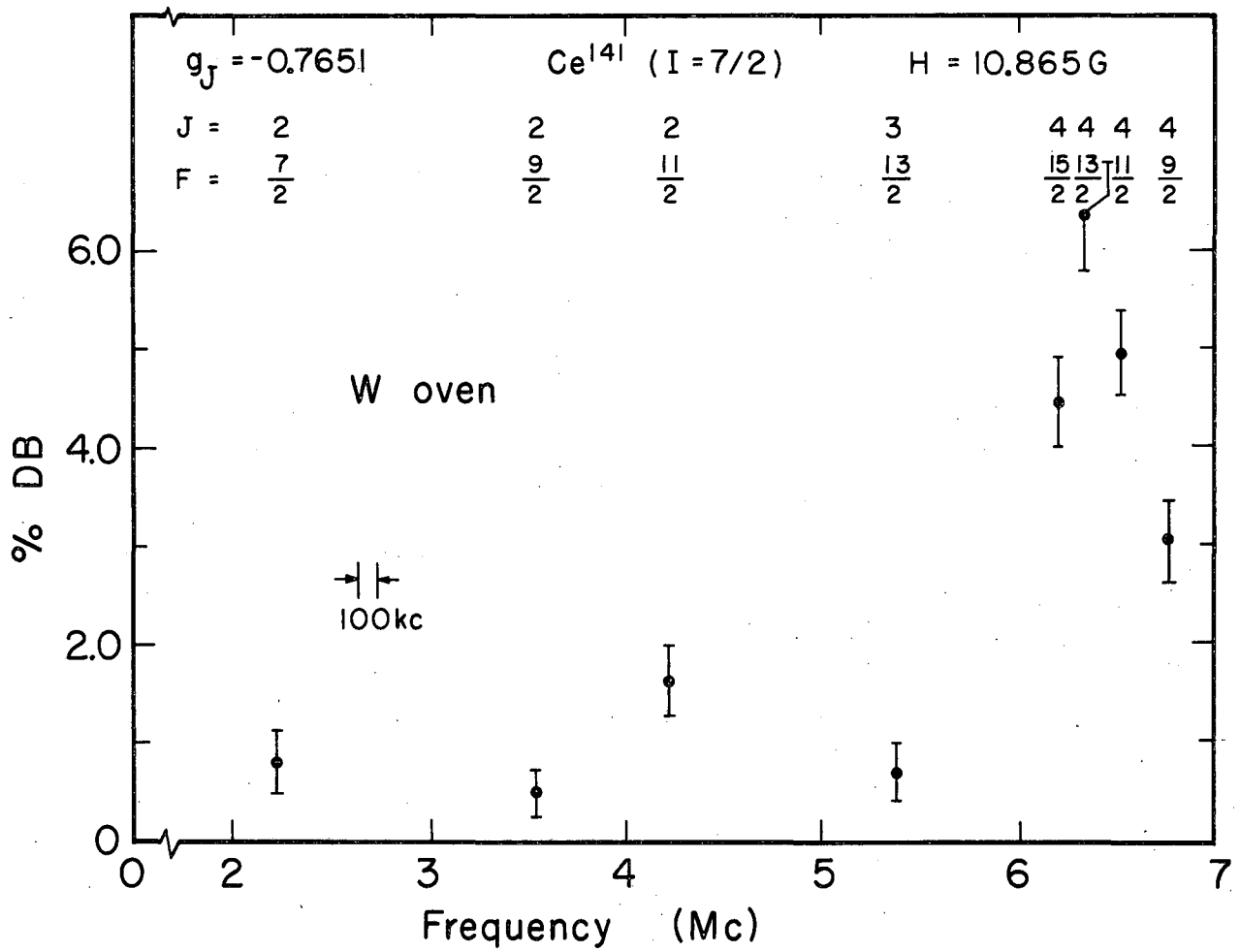


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Fig. 8

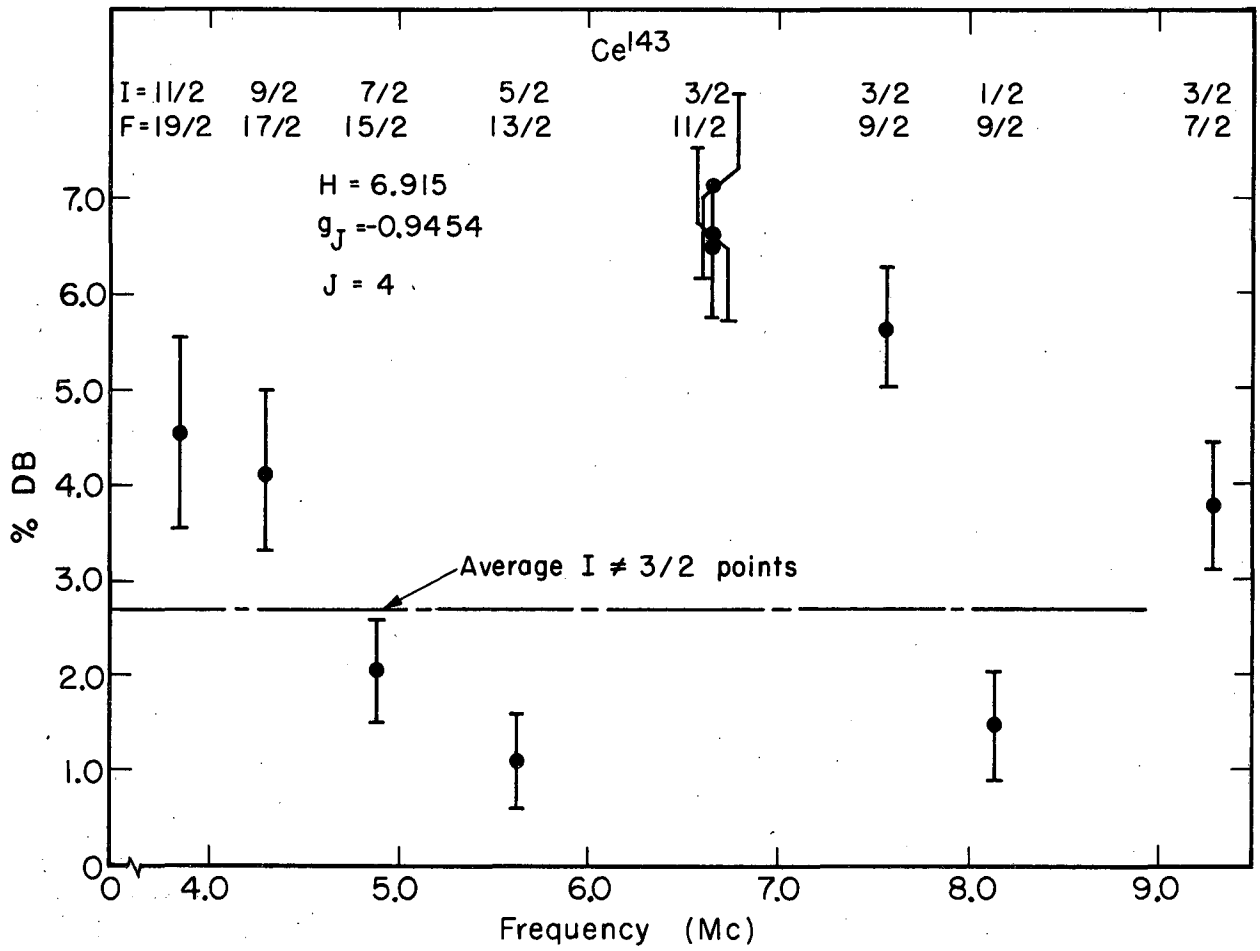

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Fig. 9

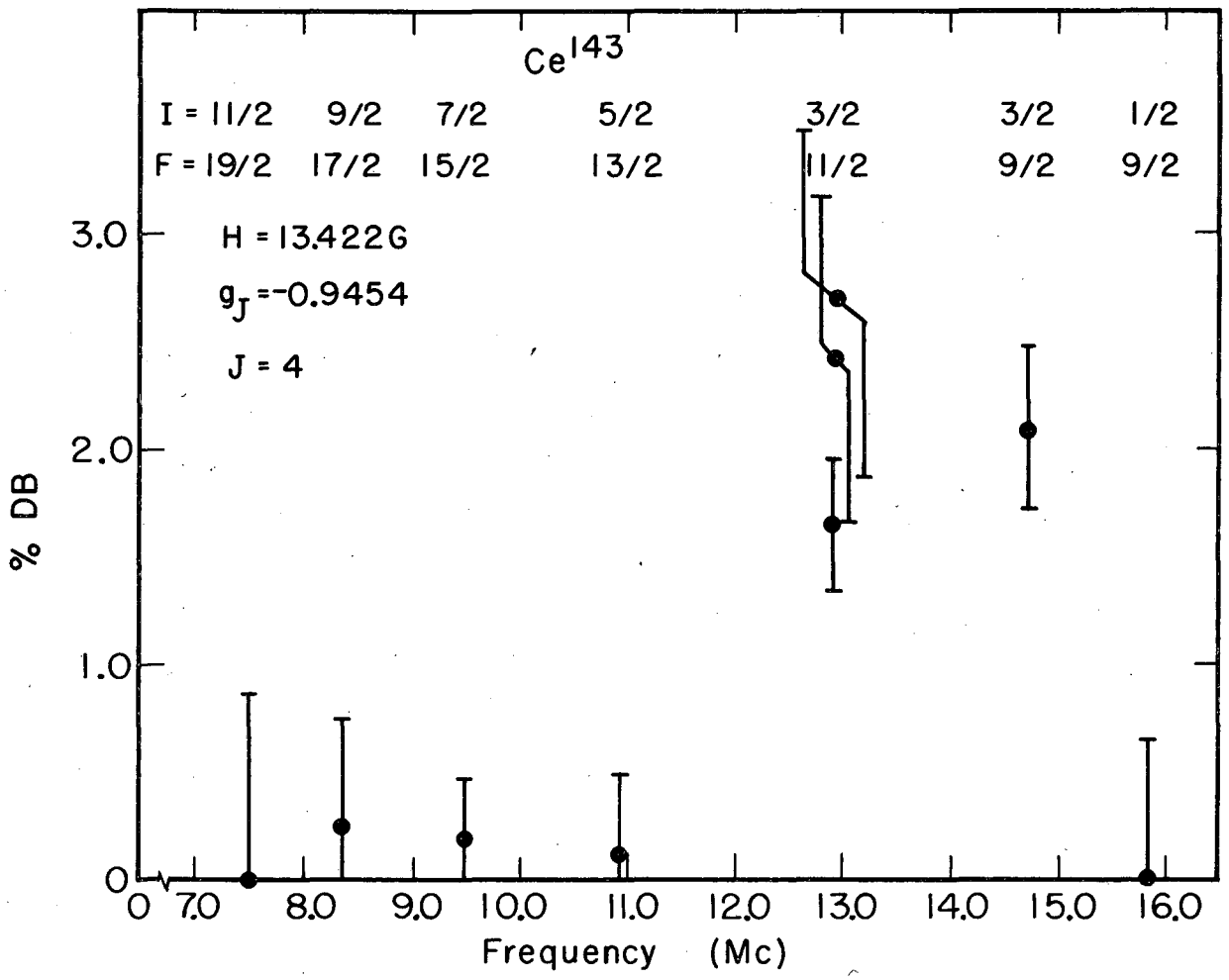

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Fig. 10

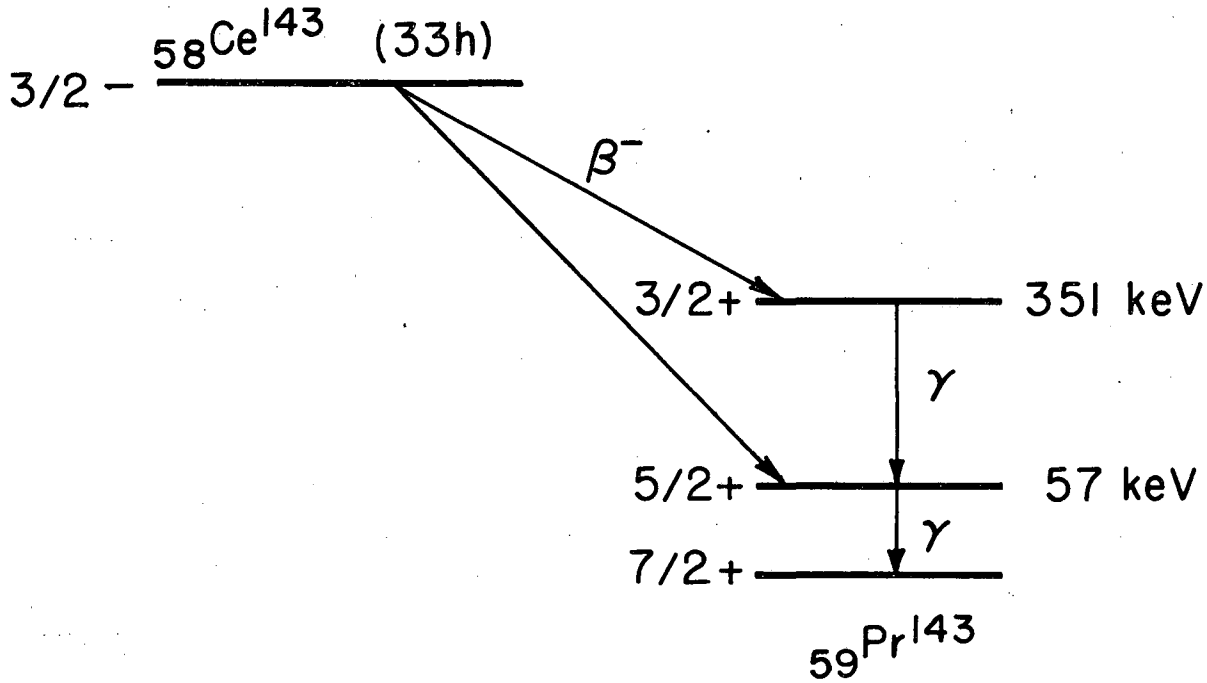

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Fig. 11

APPENDIX

Making of the Thorea Crucible

A small amount of water is mixed with $TH(NO_3)_4$ until a slightly viscous liquid is obtained. Then ThO_2 is added until a very viscous clay but not crystalline substance is present. A crucible is then made using a large stainless steel (s.s.) rod as a base, a drill guide to limit the diameter and a smaller s.s. rod to shape the cavity. If the slurry is too wet, the shape will not hold and if it is not wet enough, it is very difficult to shape at all so one usually ends up waiting for enough H_2O to evaporate to enable the mixture to hold its shape. The nitrate seems to help as a binder and the iron oxide coming from the s.s. tools also contribute to shape stability. (A plastic mold was made and did not give as good results.)

The ThO_2 crucibles (see Fig. A-1) are then left in a hood (all the work is done in a hood to confine the α-emitting thorium) for a day or two to dry. In order to get rid of the various thorium nitrate hydroxides and make the crucibles hard enough for final shaping, they are heated in air to about 600-700°F for 2-3 hours. Care has to be taken at $\approx 400°F$ since this seems to be when $Th(NO_3)_4 \cdot 4H_2O$ swells instead of simply decomposing.* This completely destroys the crucible shape and so the author usually keeps the crucibles at $\approx 380°F$ or so for many hours ($\approx$ 3-4) to prevent this rapid swelling and destruction. This initial heating process takes about one day in all. The crucibles are then shaped to fit a Ta oven with fine sandpaper and abrasive drills. They are then heated to 1000°C in air and afterwards brought up to about 2000°C in the atomic beam apparatus. The last step is of course under vacuum and part of an oven outgassing procedure.

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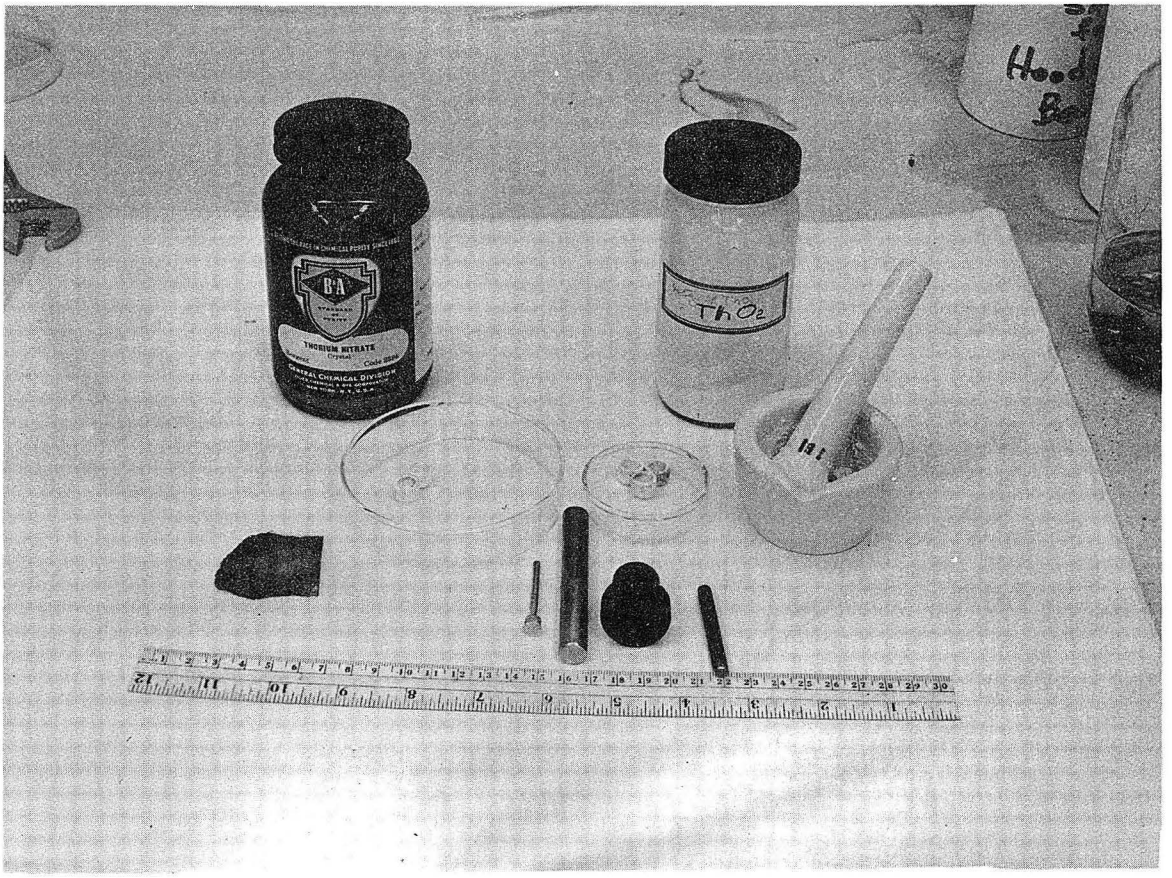
* Handbook of Physics and Chemistry, 44th Edition, p. 671.

APPENDIX FIGURE CAPTION

Fig. A-1. ThO_2 crucibles together with materials and tools used to shape them. Crucibles on left have been heated in an air furnace to 1000°C, those on right to approximately 600°F. The abrasive drill (at 15-cm mark) and sandpaper (at 4-cm mark) were used to achieve the final form.



ZN-4645

Fig. A-1

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