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Authors
Garvin, Hugh L.
Green, Thomas M.
Lipworth, Edgar.

Publication Date
1958-02-17

UCRL 8182

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Contract No. W-7405-eng-48

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Printed for the U. S. Atomic Energy Commission

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ABSTRACT

The spins of I^{123}, I^{124} and I^{131} have been measured by the method of atomic beams; the spins are 5/2, 2, and 7/2 respectively. The result I = 7/2 for I^{131} confirms an earlier measurement by a different method.

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INTRODUCTION

This paper reports the first results of a program to determine the nuclear spins, magnetic moments, and quadrupole moments of some of the available radioactive halogens by the method of atomic beams. These quantities are of interest because an extension of measurements in a region of the periodic table where collective effects are not expected to dominate will serve to broaden the experimental basis of the single-particle shell model and lead to further tests of the model itself. In addition to the results of spin determinations of three iodine nuclei, this paper contains a description of an atomic-beam apparatus that appears particularly suited to the study of radioactive substances.

METHOD

The method used, an atomic-beam "flop-in" type of experiment, was first proposed by Zacharias et al.[1,2] In recent years there has been considerable application of this technique to the measurement of the spins and moments of radioactive nuclides,[3,4] and only a brief description of the method is given here.

The ground state of all halogen atoms is $P_{3/2}$. Thus (with normal ordering of the F levels) there are two observable flop-in transitions at low frequency. For $I > 0$ these are

$(F = I + 3/2,\ M_F = -I + 1/2) \rightarrow (F = I + 3/2,\ M_F = -I - 1/2)$ and

$(F = I + 1/2,\ M_F = -I + 3/2) \rightarrow (F = I + 1/2,\ M_F = -I + 1/2)$, where F is the total angular-momentum quantum number of the atom, I the nuclear-spin quantum number, and M_F the projection of the total angular momentum along the direction of quantization. Figure 1 shows the relevant energy-level diagram for a halogen atom with $I = 5/2$. The two transitions are indicated by arrows; they will be referred to as (+) and (-) transitions respectively.

A measurement of the frequencies of either or both of these transitions in the linear Zeeman region is sufficient to determine the spin. If g_F is the g factor of the particular F level in which a transition is observed at frequency ν_X (where X is the isotope) in a magnetic field H, we have

$$\nu_X \cong g_F \frac{\mu_0 H}{h}, \tag{1}$$

where μ_0 is the Bohr magneton and h is Planck's constant. For a $P_{3/2}$ electronic state the g factors are

$$g_F \cong \frac{4}{2I+3}, \quad (F = I+3/2), \tag{2}$$

$$g_F \cong \frac{4}{3} \frac{2I+9}{(2I+1)(2I+3)}, \quad (F = I+1/2).$$

Equations (1) and (2) are derived under the assumptions of pure Russell-Saunders coupling and of vanishing nuclear magnetic moment. The magnetic field H is measured by observation of a transition in an isotope of known spin; in this experiment a beam of Cs^{133} is employed. For Cs^{133}, I is 7/2, and (at moderate fields) for the transition

$(F = 4,\ M_F = -3) \rightarrow (F = 4,\ M_F = -4)$, we have

$$\nu_{Cs} \cong \frac{1}{4} \frac{\mu_0 H}{h}. \tag{3}$$

Hence at a given field H, the ratio of ν_X to ν_{Cs} is

$$\nu_X/\nu_{Cs} \cong 4\, g_F, \tag{4}$$

and it is necessary only to search for a signal due to the isotope of unknown spin at the discrete frequencies determined by Eq. (4). Once a signal has been observed, a resonance is traced out for both transitions at a number of different fields to establish the spin unequivocally. The particular isotope involved is identified by its decay half life. Measurements of the two transition frequencies at high field where electronic and nuclear moments are partially decoupled are necessary to determine the magnetic and quadrupole interaction constants; the exact procedure adopted will be more fully discussed in subsequent papers.

SAMPLE PREPARATIONS

Radioactive I^{123} and I^{124} were produced by the bombardment of powdered antimony metal with 48-Mev alpha particles in the Crocker 60-in. cyclotron. The 13-hr I^{123} is produced by the reaction $Sb^{121}(\alpha, 2n)I^{123}$ and the 4-day I^{124} by the reactions $Sb^{121}(\alpha, n)I^{124}$ and $Sb^{123}(\alpha, 3n)I^{124}$. The 8-day I^{131} was obtained in 30-mC lots from Oak Ridge National Laboratories in the form of NaI in sodium sulphite solution. Iodine is easily precipitated from NaI by mild oxidizing agents in acid solution. Carrier iodine (20 mg) in the form of NaI was added to the Oak Ridge sample and iodine precipitated by the addition of $NaNO_2$ and dilute sulphuric acid. The iodine was extracted into carbon disulphide and added to a flask containing approximately 200 mg of elemental iodine. The mixture was well shaken and the carbon disulphide evaporated off under vacuum. The dry flask was attached directly to a discharge tube, which is necessary to dissociate the iodine molecules into iodine atoms. The vapor pressure of iodine at room temperature is adequate for the maintenance of the discharge.

Several attempts were made to separate the iodine from the antimony metal by distillation under vacuum and condensation of the iodine on a cold surface, but yields were only moderate (approximately 50%) and difficulties arose because the antimony tended to volatilize with the iodine. A simple and efficient chemical separation procedure was devised which produces yields of 80 to 90%. The antimony powder is dissolved in concentrated HCl by the addition of a few drops of hydrogen peroxide and then a few milligrams of NaI carrier is added. When the solution is adjusted just past the neutral point with NaOH, the antimony precipitates as SbOCl which can be filtered off. The precipitate is washed with a few milliliters of NaOH containing NaI, and the iodine precipitated from the filtrate and extracted by use of the procedure already outlined.

For the production of I^{123} the antimony targets were bombarded for 3 hours with 48-Mev alpha particles at a beam current of 15 to 20 μamp. Though some information was obtained on I^{124} from these bombardments, for the specific study of I^{124} the bombarding time was increased to 9 hours and the I^{123} allowed to decay away for several half-lives.

APPARATUS

The atomic-beam machine used for these experiments is, in principle, of the standard "flop-in" type, but differs in many constructional details from those previously described in the literature. A sketch of the machine is shown in Fig. 2. The three magnets are mounted externally to the vacuum system. The A and B magnet gaps are surrounded by manifolds, with the A manifold being pumped by its own diffusion pump (MCF 300),[5] and the B manifold via a connection to the detector can D. The C-field can is pumped separately with another MCF 300 pump. With the exception of the A-magnet pump, all pumps are hung from the cans, and valves are included in all the pumping paths above the liquid nitrogen traps.[5] The valves allow the main system to be brought up to atmospheric pressure without cooling the pumps and warming the traps. The system can be pumped from atmospheric pressure to 3×10^{-6} mm Hg in about 5 minutes. The oven and detector pumps are MCF 700 oil-diffusion pumps. The oven, detector, and C-field cans together with the magnets are mounted upon heavy plates which sit astride Dural runners. Provision is made for independent horizontal and vertical motion of each plate. The magnet-energizing coils are external to the vacuum system and a single turn of water-cooled copper tubing on the coil formers of the A and B magnets keeps the temperature at a reasonable value.

The present mode of construction has certain mechanical advantages: (1) apparatus line-up is easily performed, (2) a change from a "flop-out" to a "flop-in" system can be simply achieved by reversing the B magnet, (3) the length of the C field can be readily changed, and different rf loops incorporated, (4) the system of valves allows easy and rapid access to the cans, and rapid attainment of a working vacuum; (5) component parts can readily be decontaminated or, if necessary, replaced (this is a major advantage, particularly for work with long-lived radioactive isotopes.)

During the early stages of construction, attempts were made to pass a potassium beam down thin-walled stainless steel tubes supported between the magnet gaps. The diameters of the A and B magnet tubes were 0.080 in. and 0.375 in. respectively. With this arrangement, only extremely weak beams were recorded at the detector. When the A tube was replaced by a 0.375-in.-diameter tube of the same length, a beam of approximately half the expected intensity was recorded. Attempts to improve the transmission by prolonged outgassing of the tubes in situ at 300 to 400°C gave no improvement, but subsequent cooling of the tubes with liquid nitrogen led to some improvement in transmission. After the tubes had been outgassed for 3 hours and a beam observed, the beam reduced to zero 10 seconds after application of power to the heating coils and reappeared as the tubes cooled. At this point the A- and B-magnet vacuum manifolds were incorporated.

The electronics associated with the apparatus is to a large extent conventional with the exception of the AB-magnet power supply. The magnets were wound to operate at 300 v at 5 amp and are energized by an electronically regulated supply that is stable to one part in 30,000; consequently changes in the C field due to time-dependent fringing effects are largely eliminated.

The calibrating oven F is loaded with cesium metal in a dry nitrogen atmosphere and is raised and lowered by a motor. The radioactive beam is collected upon buttons coated with evaporated silver which are introduced and removed from the machine via a vacuum lock. The button loader is shown at G. Buttons are stored in a vacuum bottle to prevent poisoning of the silver surface. Exposure of the buttons to air for one or two days reduces the collection efficiency markedly, presumably by the formation of a surface coating of silver sulphide. The collection efficiency of freshly silvered surfaces for iodine (both atomic and molecular) is high and reproducible.

The discharge tube is driven by an rf oscillator at 450 kc and is mounted on a movable plate as part of the oven can E; it is shown in Fig. 3 together with a regulating circuit that has been found particularly useful. The tube is quartz, and good capacitative coupling between the electrodes and discharge is achieved by painting the surface under the electrodes and in the neighborhood of the slit with Aquadag. The latter step was found to be essential to obtain an efficient discharge. The dissociation efficiency of this discharge is high for iodine and bromine (80 to 100%). The slit width and height are approximately 0.005 and 0.040 inch respectively. The discharge regulator was devised originally by Steinmetz in 1890,[6] and has the interesting property that at resonance the current through R is independent of R. An extremely stable discharge results if the circuit is turned to operate slightly off-resonance.

A General Radio type 805-C oscillator is employed to power the rf loop, which consists of a single U-shaped turn of 1/16-in.-diam. copper wire whose plane is oriented parallel to the C field. With this arrangement cesium resonance half-widths are about 130 kc. The oscillator frequency is measured with a Hewlett-Packard type 524 B frequency meter. During exposure of a button to the iodine beam, normally for 5 to 10 min., the frequency remains constant within a few hundred cycles. Little or no trouble has been experienced with C-field drift during the exposure time.

The I^{123} and I^{124} are detected by counting the K x-rays (~30 kev)[7] emitted after electron capture. Thin-crystal scintillation counters with a background counting rate of approximately 0.5 cpm are employed for this purpose; these counters have been described previously.[8]

The I^{131} is detected by counting the decay beta particles in small-volume gas-flow proportional counters. The buttons are inserted directly into the counters which, because of their small surface area, have the unusually low background of 2 to 3 cpm. These counters will be described elsewhere.

RESULTS

Iodine-131. The two resonances shown in Figs. 4 and 5 were each obtained with approximately 20 mC of I^{131} in the oven vial mixed with 200 mg of natural iodine carrier. The cesium resonance frequency was 5 Mc/sec corresponding to a C-field setting of 19.93 gauss. Similar resonances have been obtained at 7.99 gauss (cesium resonance frequency = 2 Mc/sec). The 7/2 (+) resonance buttons were each exposed for 10-min. periods, but the exposure time was reduced to 5 min for the 7/2 (-) resonance. The direct-beam counting rate was approximately 250 cpm for a 5-min exposure with the AB magnets turned off. When the magnets were turned on, the counting rate dropped to 40 cpm. With 200 mg iodine carrier, the discharge running time was approximately 3 hr. In Figs. 4 and 5, the small difference between the frequency of the resonance peaks and the corresponding values of $g_F \frac{\mu_0 H}{h}$ is due to partial decoupling of the nuclear and electronic magnetic moments by the C field.

It will be noted that the background signal level in the absence of rf power is appreciable. This large apparatus background has been encountered in work with bromine as well as iodine isotopes and is due to the high volatility of these substances. Attemps to reduce it by surrounding the buttons with a liquid-nitrogen-cooled shield have not produced much improvement; possibly what is required is a silver coating on the inner walls of the magnets and vacuum systems.

The observed resonances allow an unambiguous assignment of 7/2 for the nuclear spin of I^{131}, a result which verifies an earlier measurement by a different method.[9]

Iodine-123 and Iodine-124. Figure 6 shows the results of a spin search on cyclotron-produced iodine at a field of 11.97 gauss. Strong peaks will be noted at frequencies corresponding to (+) and (-) transitions in a spin 5/2 isotope, and a smaller (+) peak indicating the presence of an isotope with spin 2. The decay of a direct-beam button (Fig. 7) shows the presence of two components with half-lives of 15 hrs and 5 days respectively, and on the basis of the published half-lives,[10, 11, 12, 13, 14] these have been identified as I^{123} and I^{124}. Figures 8 and 9 show examples of 5/2 (+) and 5/2 (-) resonances at a field of 39.70 gauss. Similar resonances have been obtained at fields of 11.97 and 23.89 gauss. A decay curve of the 5/2 (+) resonance-peak button at 23.89 gauss is shown in Fig. 10. The presence of I^{123} is well illustrated. The long 4.2-day I^{124} tail is due to resonance overlap; at the fields employed, the I^{124}(2-) resonance occurs at approximately the same frequency as the 5/2 (+) resonance. The 5/2 (-) resonance-peak button decay (not shown) is uncontaminated by overlap. Because of this overlap, the antimony targets were allowed to stand for several half-lives of I^{123} prior to processing and searching for I^{124} resonances. Figure 11 shows a 2 (+) resonance obtained at a field of 39.70 gauss, and Fig. 12 the decay of the two resonance-peak buttons. Figure 13 exhibits a 2 (-) resonance observed at the same field, and Fig. 14 the result of decaying the three peak buttons.

On the basis of these results the spin of I^{123} is 5/2 and that of I^{124} is 2.

DISCUSSION

The spins of the stable iodine isotopes I^{127} and I^{129} are known to be 5/2 and 7/2 respectively. They indicate that the $d_{5/2}$ and $g_{7/2}$ single-particle shell-model levels are both readily available to the odd proton. The spins of 5/2 and 7/2 for I^{123} and I^{131} fit nicely into this picture. I^{124} possesses an odd number (71) of neutrons. Odd-A nuclei with neutron numbers between 65 and 75 consistently exhibit spin 1/2 because of the filling of the $h_{11/2}$ level in pairs. If it is assumed that the neutron part of I^{124} also possesses spin 1/2, the consequences of Nordheim's Rules can be investigated.[15] If the odd proton is assumed to be $g_{7/2}$, Nordheim's strong rule applies, and the expected spin is 3; if the proton is $d_{5/2}$, the weak rule applies, and a spin near but not necessarily equal to 3 is implied. As the observed spin is 2, it is likely that the odd proton of I^{124} is in a $d_{5/2}$ level.

ACKNOWLEDGMENTS

The authors wish to thank Professor William A. Nierenberg for support and advice, and to acknowledge the able assistance of Mr. Larry M. Cohen and Mr. Douglas MacDonald. Members of the Radiation Laboratory chemistry department and health chemistry group have made substantial contributions to the success of this work. This work was performed under the auspices of the U. S. Atomic Energy Commission.

REFERENCES

1. J. R. Zacharias, Phys. Rev. 61, 270 (1942).
2. Davis, Nagle, and Zacharias, Phys. Rev. 76, 1068 (1949).
3. William A. Nierenberg, Ann. Rev. Nuclear Sci. 7, 349 (1957).
4. Kenneth F. Smith, Progr. in Nuclear Phys. 6, 52 (1957).
5. The MCF series of pumps is manufactured by Consolidated Vacuum Corp., Rochester, N. Y. The valves employed are Temescal valves produced by the same company.
6. See Albert C. Candler, Modern Interferometers (Hilger and Watts, London, 1951), p. 32 for a description of this circuit and some early references.
7. A. H. Compton and S. K. Allison, X-Rays in Theory and Experiment (Van Nostrand, New York, 1935).
8. J. C. Hubbs and W. A. Nierenberg, Scintillation Detection of Very Low-Energy Gamma Rays, UCRL-3434, June 1956.
9. Livingston, Benjamin, Cox, and Gordy, Phys. Rev. 92, 1271 (1953).
10. Mitchell, Mei, Maunchien, and Peacock, Phys. Rev. 76, 1450 (1949).
11. L. Marquez and I. Perlman, Phys. Rev. 78, 189 (1950).
12. H. B. Mathur and E. K. Hyde, Phys. Rev. 96, 126 (1954).
13. J. L. Meem and F. Maienschien, Phys. Rev. 76, 328 (1949).
14. Aagaard, Andersson, Burgman, and Pappas, J. Inorg. & Nuclear Chem. 5, 105 (1957).
15. M. G. Mayer and J. H. J. Jensen, Elementary Theory of Nuclear Shell Structure (Wiley London, 1955).

Captions

Fig. 1. Energy-level diagram of $^2P_{3/2}$ state with I = 5/2. The two permissable "flop-in" transitions are indicated by arrows.

Fig. 2. Schematic view of atomic-beam apparatus

A, B, C - magnets
D - detector can
E - oven can
F - calibrating oven
G - button loader
H - stopwire assembly
I - liquid-nitrogen traps
J - valves

Fig. 3. Discharge tube and discharge-stablizing circuit.

Fig. 4. 7/2(+) resonance in I^{131}. The arrow indicates the expected position of the resonance peak in the absence of quadratic and higher-order shifts.

Fig. 5. 7/2(-) resonance in I^{131}.

Fig. 6. Spin search on cyclotron-produced I^{123} and I^{124}.

Fig. 7. Decay of direct-beam buttons containing-cyclotron produced I^{123} and I^{124}.

Fig. 8. 5/2(+) resonance in cyclotron produced I^{123}.

Fig. 9. 5/2(-) resonance in cyclotron produced I^{123}.

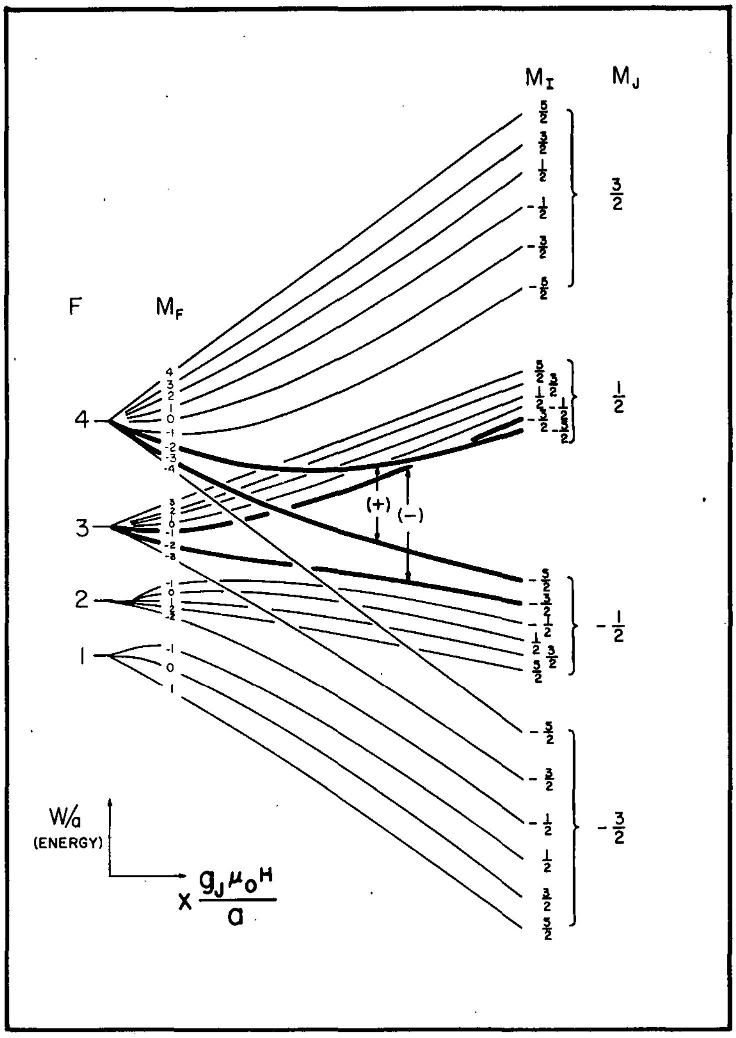
Fig. 10. Decay of 5/2(+) resonance-peak buttons in cyclotron-produced iodine. The short-lived component is due to I^{123} with spin 5/2; the long-lived component arises from overlap from the I^{124} 2(-) resonance.

Fig. 11. 2(+) resonance in cyclotron produced I^{124}.

Fig. 12. Decay of I^{124}. The two resonance - peak buttons of Fig. 11 were used to obtain this decay curve.

Fig. 13. 2(-) resonance in cyclotron-produced I^{124}.

Fig. 14. Decay of I^{124}. The three resonance-peak buttons of Fig. 13 were used to obtain this decay curve.



MU-14767

Fig. 1.

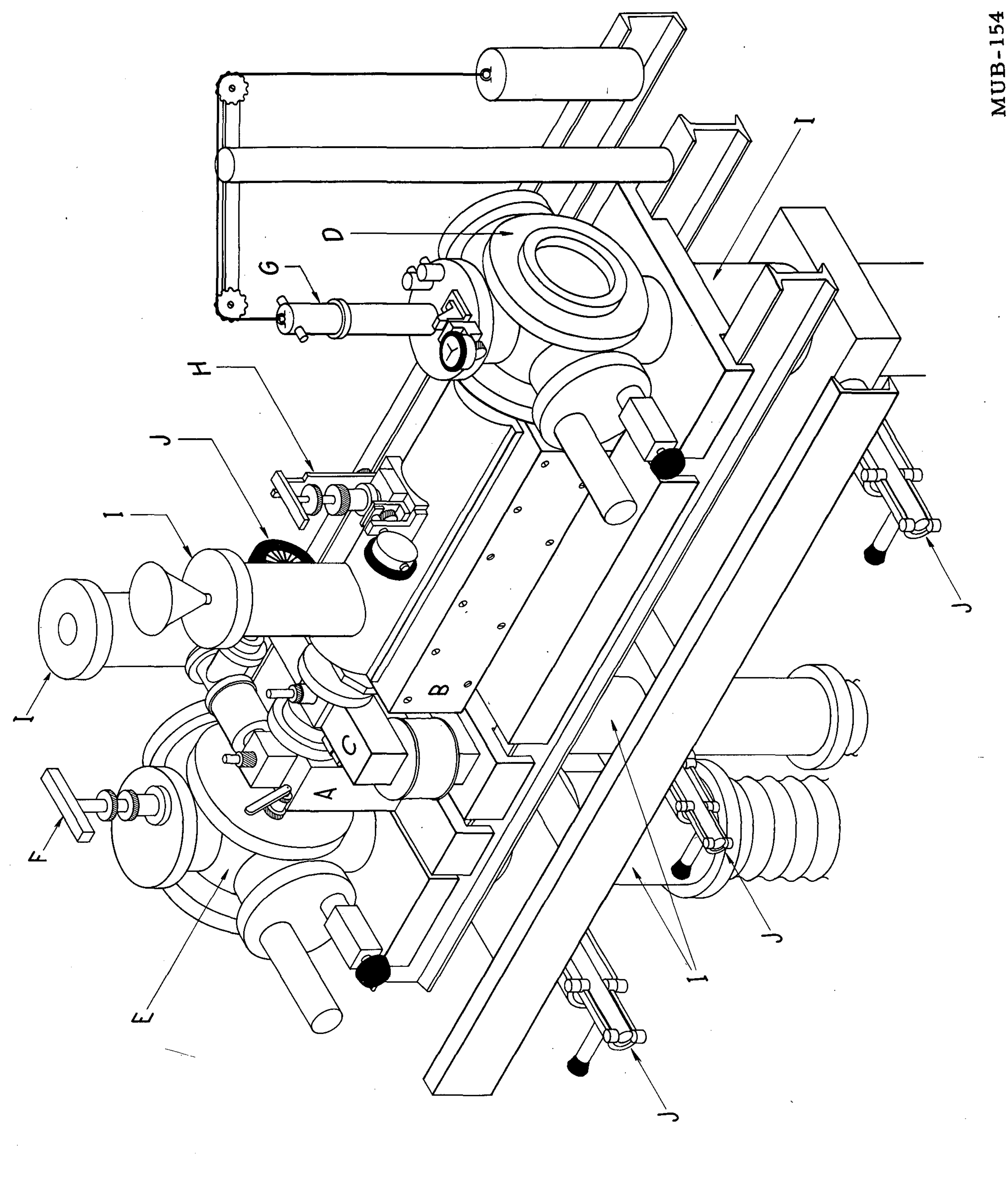
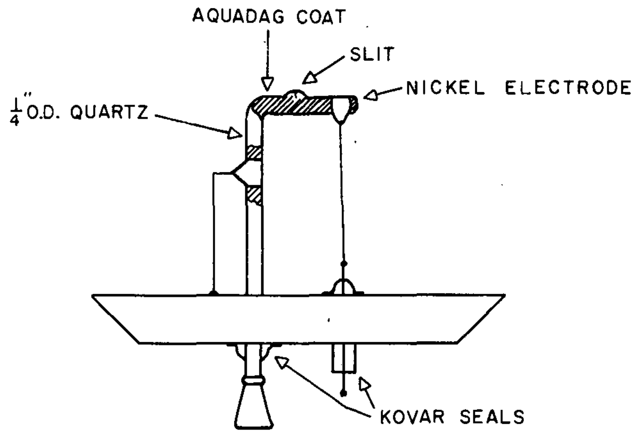
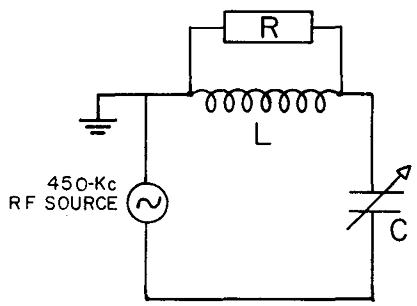


Fig. 2.



RF DISCHARGE TUBE



DISCHARGE STABILIZER

MU-14243

Fig. 3.

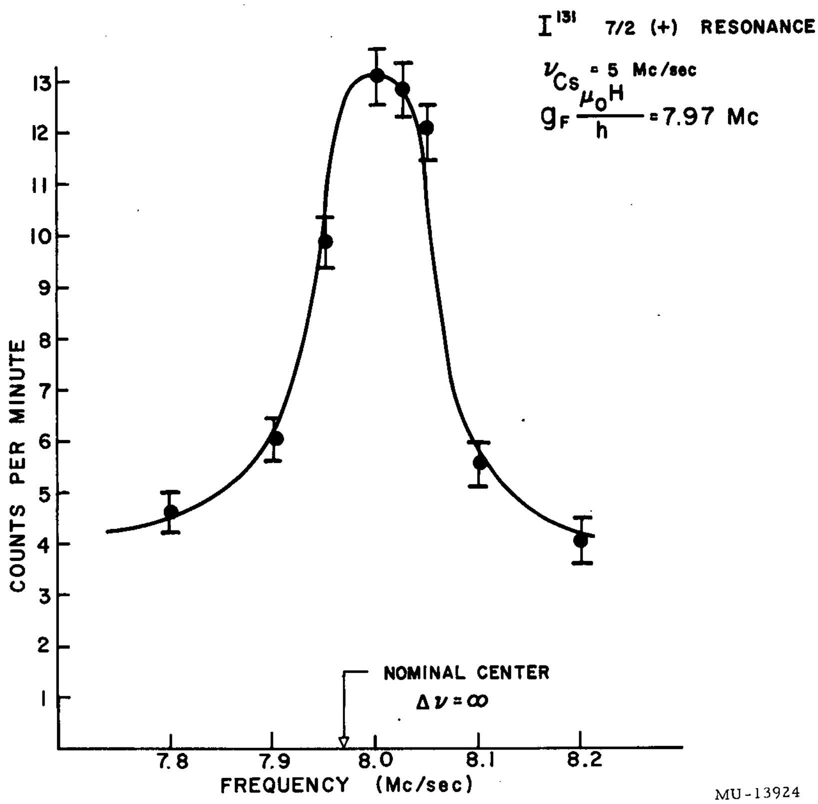
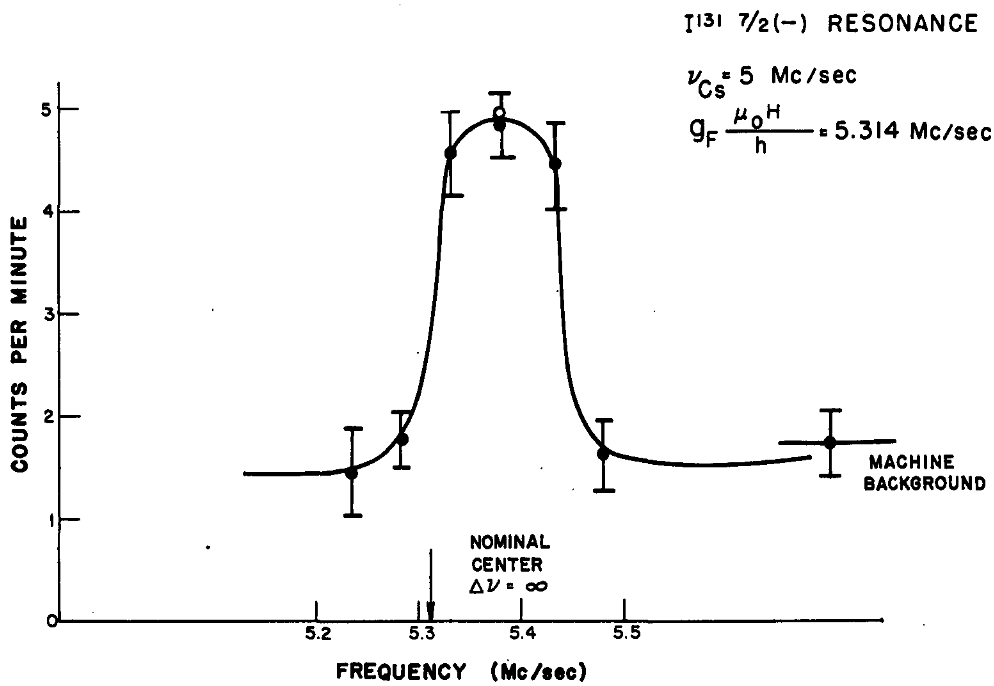
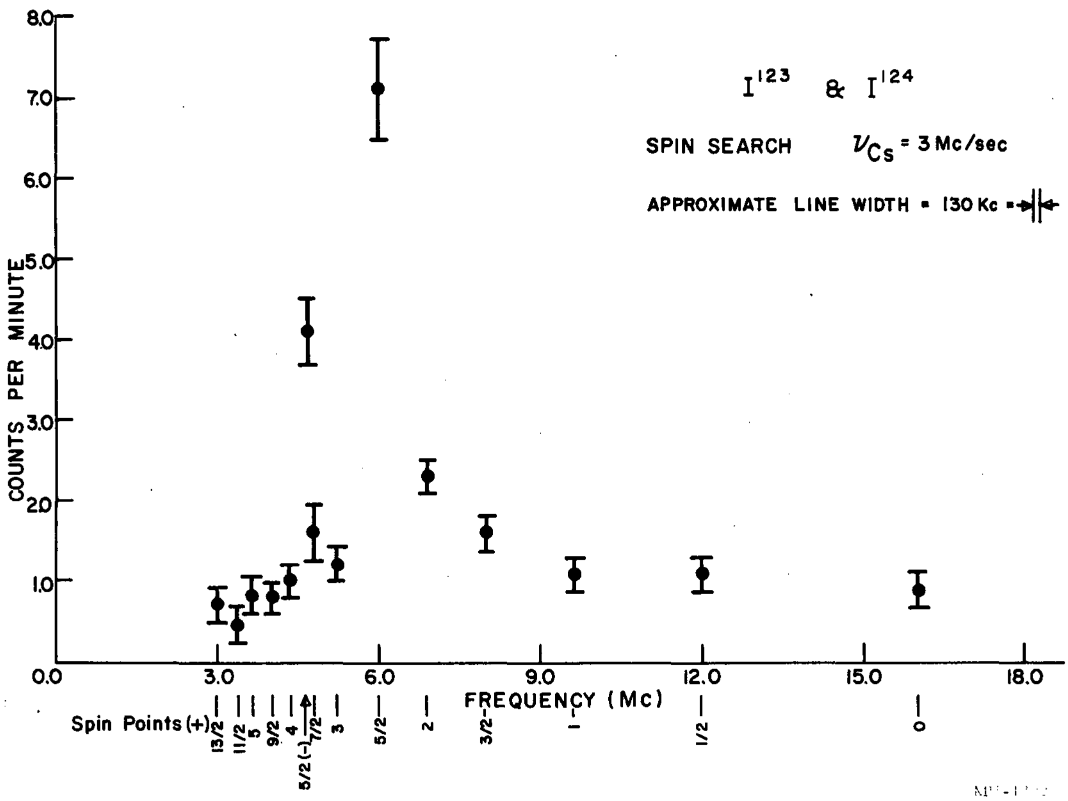

I131 7/2 (+) RESONANCE
νCs = 5 Mc/sec
gF μ0H/h = 7.97 Mc
COUNTS PER MINUTE
13
12
11
10
9
8
7
6
5
4
3
2
1
NOMINAL CENTER
Δν = ∞
7.8
7.9
8.0
8.1
8.2
FREQUENCY (Mc/sec)
MU-13924

Fig. 4.



MU-13923

Fig. 5.



I^{123} & I^{124}
SPIN SEARCH ν_{Cs} = 3 Mc/sec
APPROXIMATE LINE WIDTH = 130 Kc
COUNTS PER MINUTE
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0.0
3.0
6.0
9.0
12.0
15.0
18.0
FREQUENCY (Mc)
Spin Points (+)
13/2
11/2
5
9/2
4
5/2(-)
7/2
3
5/2
2
3/2
1
1/2
0

Fig. 6.

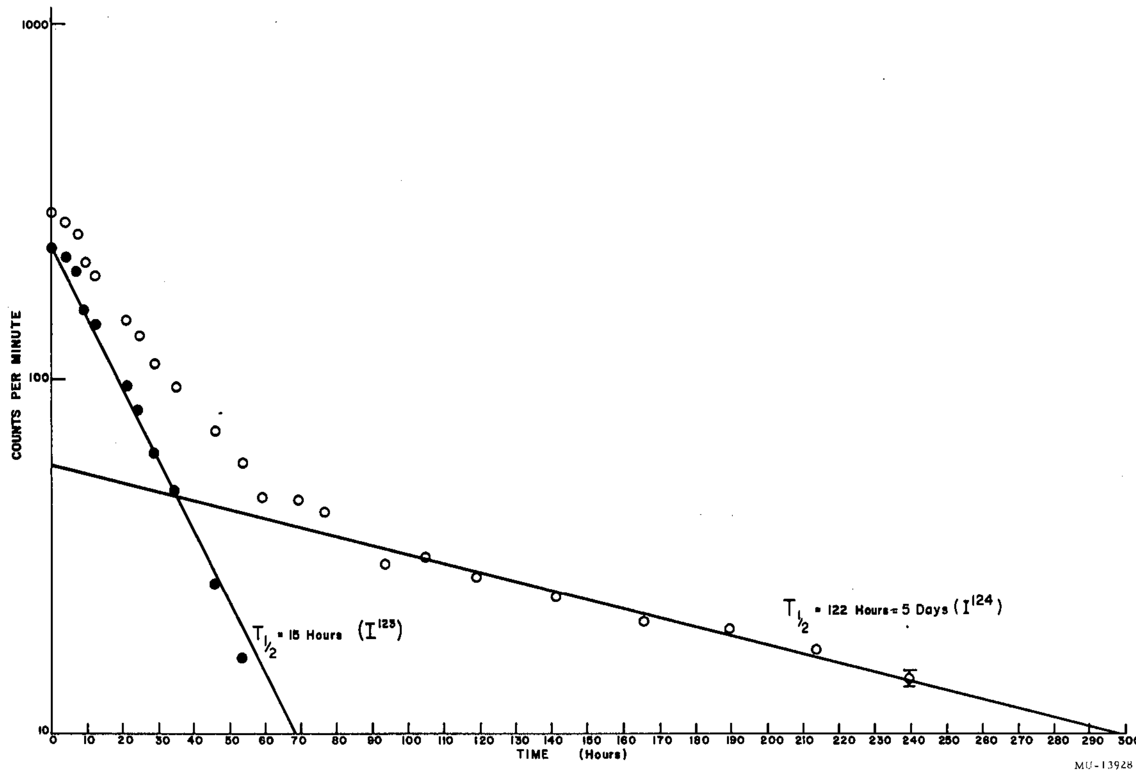

1000
100
10
COUNTS PER MINUTE
TIME (Hours)
$T_{1/2}$ = 15 Hours (I^{123})
$T_{1/2}$ = 122 Hours = 5 Days (I^{124})
0 10 20 30 40 50 60 70 80 90 100 110 120 130 140 150 160 170 180 190 200 210 220 230 240 250 260 270 280 290 300
MU-13928

Fig. 7.

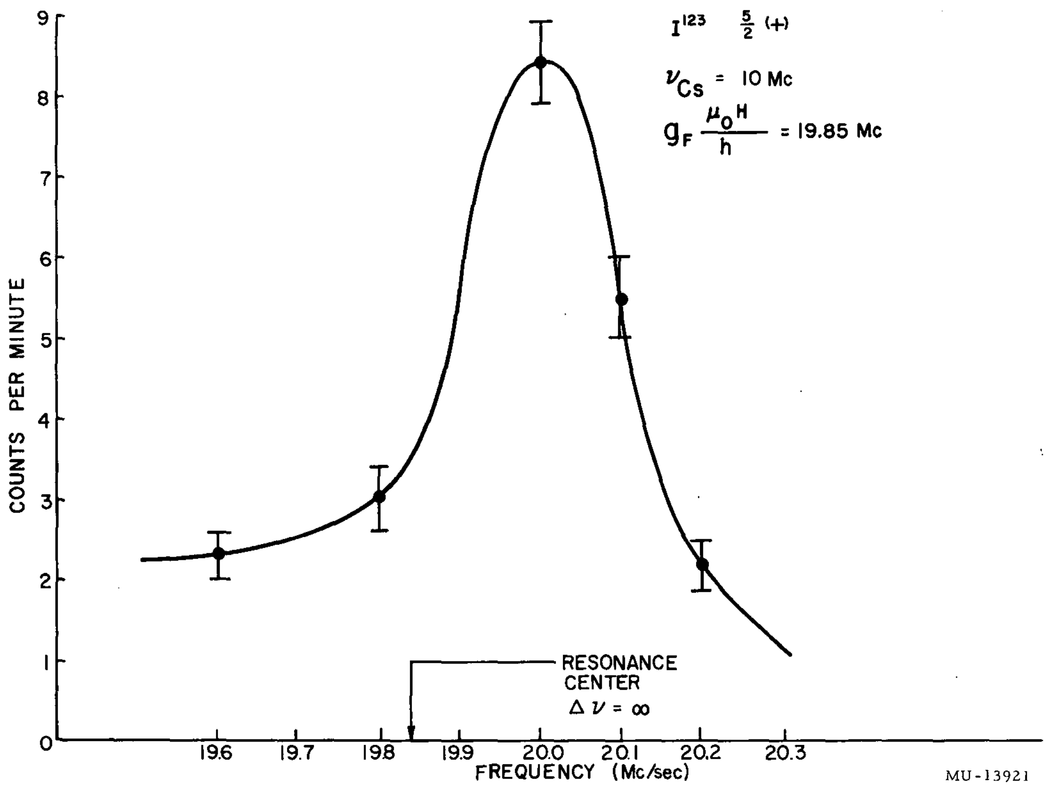

I123 5/2 (+)
νCs = 10 Mc
gF μ0H/h = 19.85 Mc
COUNTS PER MINUTE
0
1
2
3
4
5
6
7
8
9
RESONANCE
CENTER
Δ ν = ∞
19.6
19.7
19.8
19.9
20.0
20.1
20.2
20.3
FREQUENCY (Mc/sec)
MU-13921

Fig. 8.

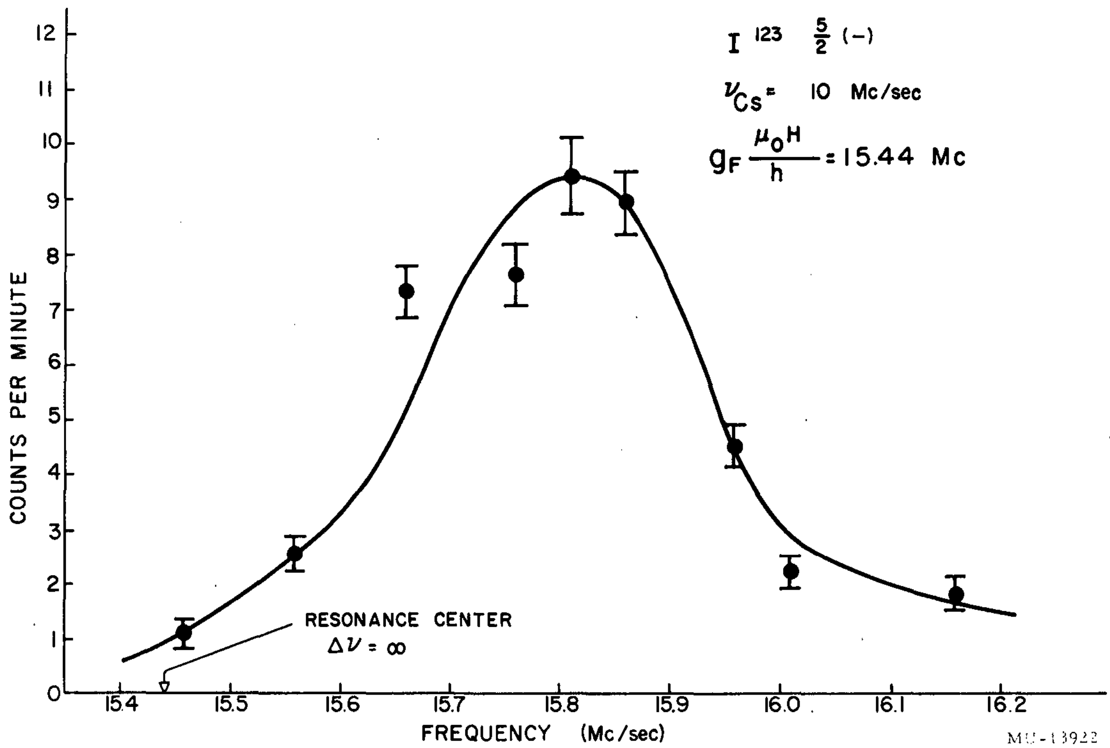
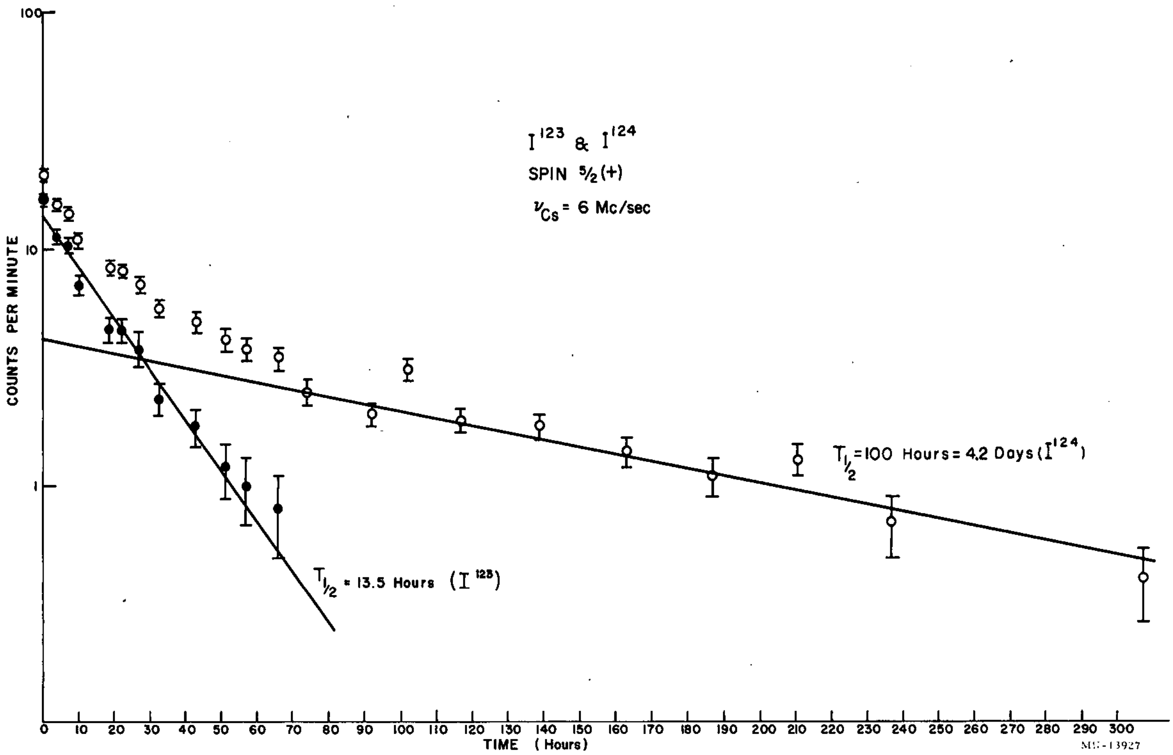
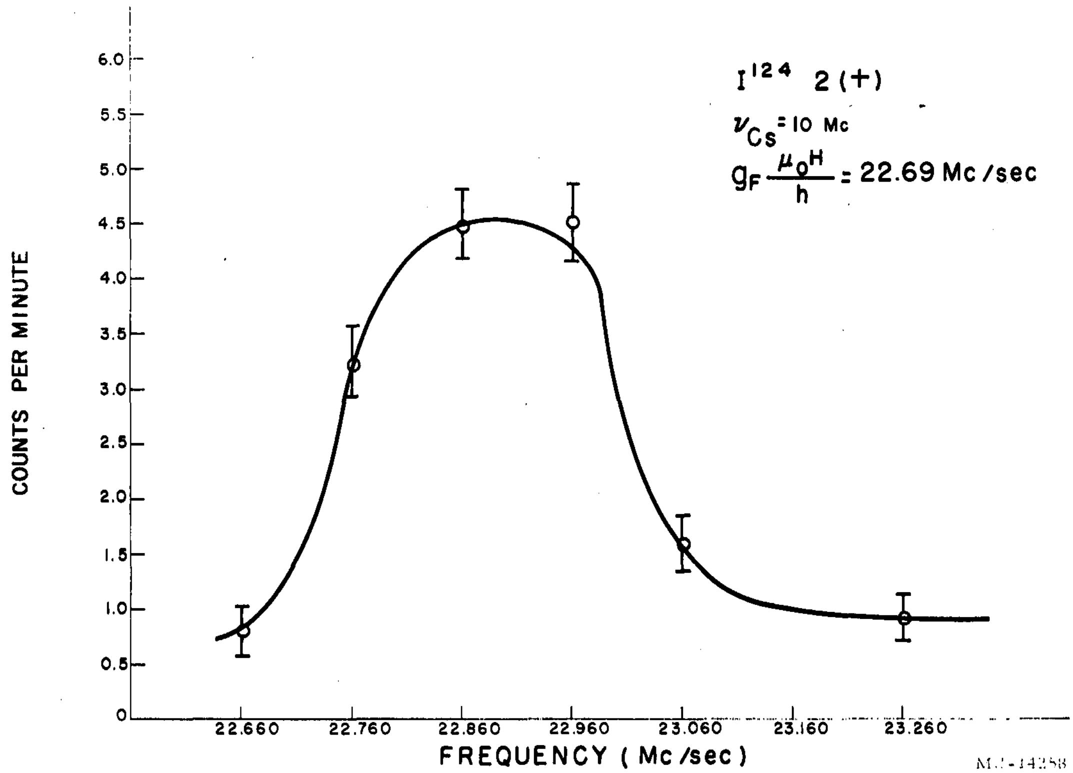

I 123 5/2 (-)
ν Cs = 10 Mc/sec
g F μ0H/h = 15.44 Mc
12
11
10
9
8
7
6
5
4
3
2
1
0
COUNTS PER MINUTE
RESONANCE CENTER
Δν = ∞
15.4
15.5
15.6
15.7
15.8
15.9
16.0
16.1
16.2
FREQUENCY (Mc/sec)

Fig. 9.



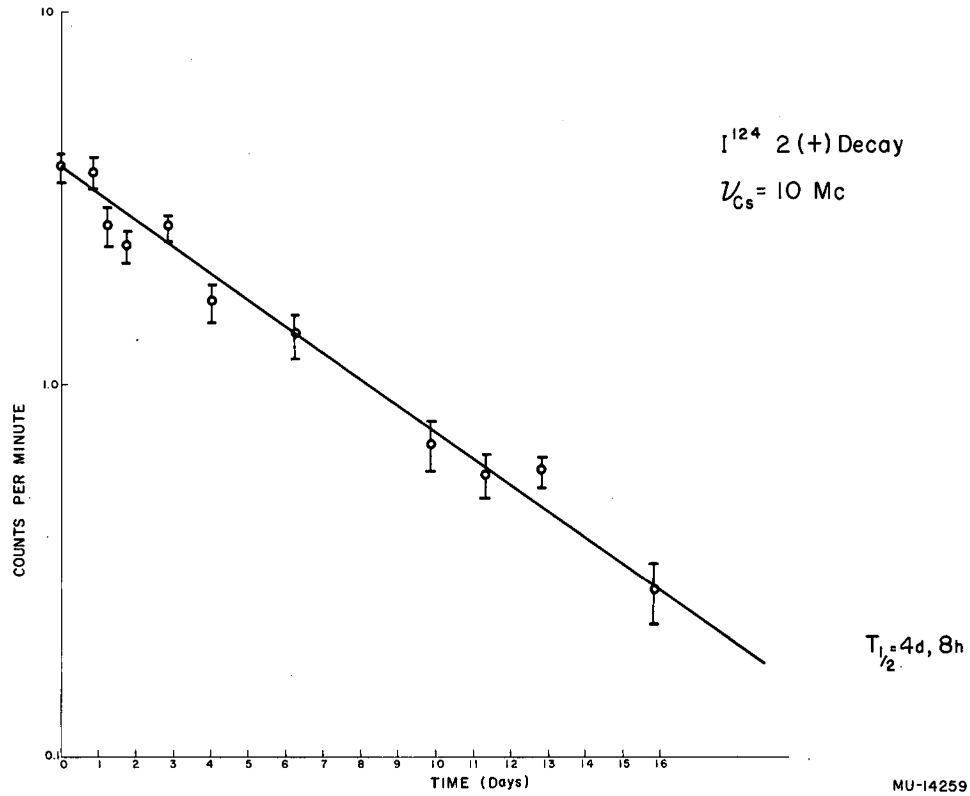
I^123 & I^124
SPIN 5/2 (+)
ν_Cs = 6 Mc/sec
COUNTS PER MINUTE
100
10
1
T½ = 100 Hours = 4.2 Days (I^124)
T½ = 13.5 Hours (I^123)
0 10 20 30 40 50 60 70 80 90 100 110 120 130 140 150 160 170 180 190 200 210 220 230 240 250 260 270 280 290 300
TIME (Hours)
MU-13927

Fig. 10.



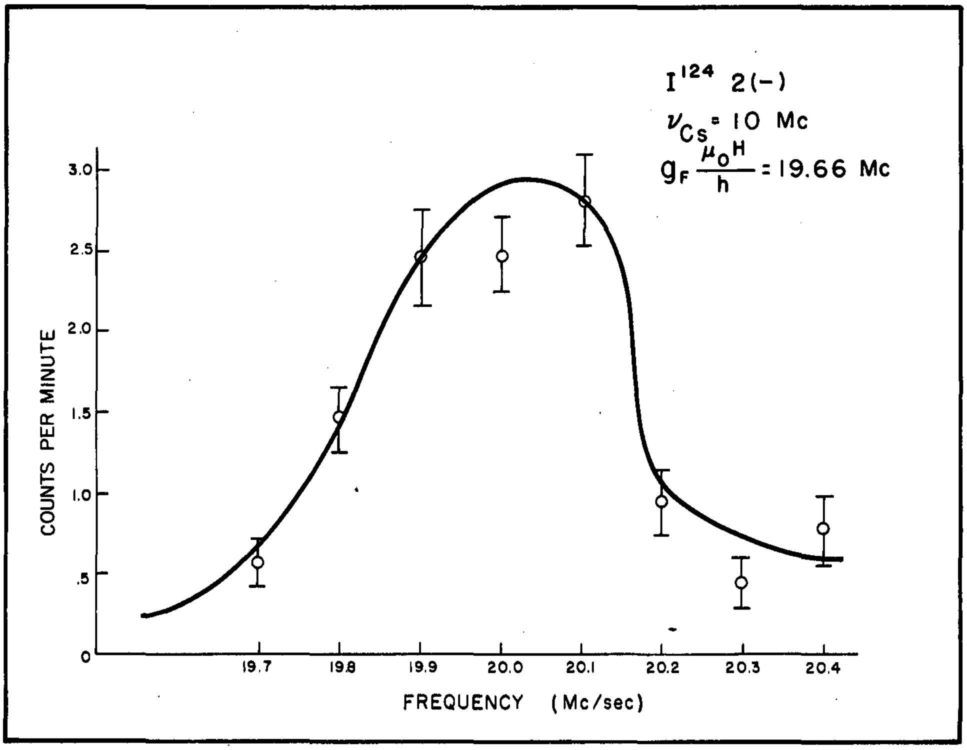
I^{124} 2(+)
ν_{Cs} = 10 Mc
$g_F \frac{\mu_0 H}{h}$ = 22.69 Mc/sec
COUNTS PER MINUTE
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0
22.660
22.760
22.860
22.960
23.060
23.160
23.260
FREQUENCY (Mc/sec)

Fig. 11.



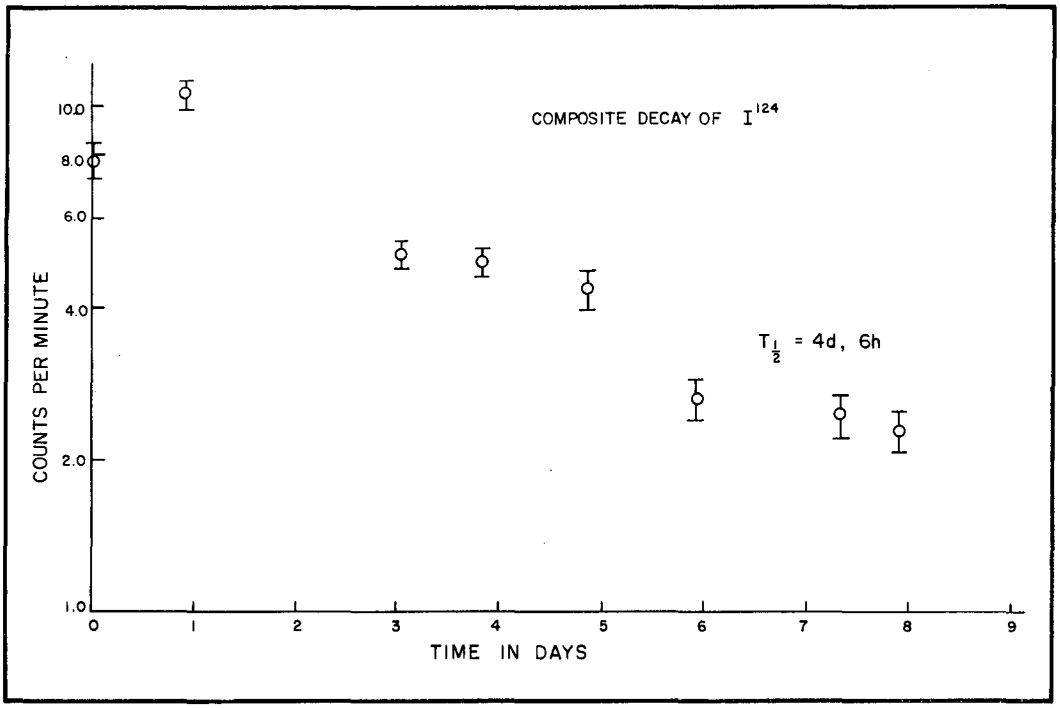
I^{124} 2 (+) Decay
ν_{Cs} = 10 Mc
$T_{1/2}$ = 4d, 8h
COUNTS PER MINUTE
TIME (Days)
10
1.0
0.1
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
MU-14259

Fig. 12.



MU-14768

Fig. 13.



MU-14769

Fig. 14.