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THE SPINS OF THALLIUM-197, -198m, -199, AND -204, AND THE HYPERFINE-STRUCTURE SPLITTING OF THALLIUM-204

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ABSTRACT

The nuclear angular momenta of four isotopes of thallium and the hyperfine-structure splitting of Tl^{204} in the $^2P_{1/2}$ state have been measured by the atomic-beam magnetic resonance method. The results are:

For 2.7-hr Tl^{197} $I = 1/2$,

1.8-hr Tl^{198m} $I = 7$,

7.2-hr Tl^{199} $I = 1/2$,

4.1-yr Tl^{204} $I = 2$, $\Delta\nu(^2P_{1/2}) = 732 \pm 5$ Mc.

The neutron-deficient isotopes are produced by alpha-particle bombardment of gold in the Berkeley 60-inch cyclotron; Tl^{204} is produced by neutron activation of metallic thallium samples.

THE SPINS OF THALLIUM-197, -198m, -199, AND -204,[1] AND THE HYPERFINE-STRUCTURE SPLITTING OF THALLIUM-204*

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INTRODUCTION

The atomic-beam method, as adapted to measurements on radioisotopes, has been used to determine all presently available unmeasured spins and moments in the thallium series. This research, dealing with an element near magic numbers 82 and 126, is part of a general program of the Berkeley atomic-beam group to determine as far as possible the unknown spins and nuclear moments of one or more elements in regions of special significance from the point of view of nuclear theory. In this frame of reference the question asked in this research is whether deformabilities or spheroidal effects are important for a proton system different by one from magic, as in Tl; the not entirely unexpected answer obtained is that at least where ground-state spins are concerned the effects in question are not important for neutron numbers as far as ten from magic. Measurements of the nuclear magnetic dipole and electric quadrupole moments, which are far more sensitive to such phenomena, will have to await improvements in the experimental technique that will permit determinations in the excited $^2P_{3/2}$ state.

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APPARATUS AND EXPERIMENTAL DETAILS

The research has been conducted with a new atomic-beam apparatus. The principal changes in the new machine are in the direction of increased flexibility and protection against radiation hazards. Under the first heading the A field has rotatable pole tips so that changes between A- and B-magnet parallel and antiparallel field gradients can be made externally. A and B fields are further obtained from high-impedance coils fed by an electronic regulated-current power supply. Radiation hazard is minimized by so designing the unit that the vacuum envelope need never be opened save for rare mechanical failures, and by providing a glove box at the side of the can for loading sources.

Aside from the aspects discussed above, the apparatus is, like that of Reference 2, a tapered system with a transmission of 10^{-5} and, as used for this research, a Zacharias-type machine utilizing the transition ($F = I + 1/2$, $|m_F| = I + 1/2 \leftrightarrow |m_F| = I - 1/2$) for which there is a change in sign of the high-field magnetic moment. Rejection of the full beam is between $10^3/1$ and $10^4/1$, depending upon the material under investigation and upon the source and can conditions, while a resonance in the $^2P_{1/2}$ state of thallium is on the order of 1% of the full beam for a single component.

Under production conditions that hold for the Tl isotopes, there is no clear-cut separation of the activities because of the more or less monotonic ordering of half lives; nevertheless, the apparatus background is sufficiently low that a resonance is clearly discernible above background.

C-field homogeneity is good (Fig. 1), even for the narrow resonance in thallium caused by apparatus velocity selection as a result of the small g_J value relative to the beam aperture. The maintenance of a constant C field to a small fraction of this line width for several hours is difficult. Fortunately the reliability of the regulated-current power supply is such that this condition is met in most runs. In case a serious shift in C field is found, all data taken from the preceding field measurement on are ignored.

ISOTOPE PRODUCTION AND SEPARATION

The neutron-deficient isotopes are produced by alpha-particle bombardment of gold with the evaporation of four, three, or two neutrons from the compound nucleus. Typically a bombardment is made for a period of 2 hours at an average current of 35 μa. Tl^{204} is made by neutron activation of metallic thallium samples; the ratio of thermal-activation cross sections[3] for Tl^{203} and Tl^{205} is 80/1, and the half lives of Tl^{204} and Tl^{206} are 4.1 yr and 4.3 m respectively. The sample so formed is therefore pure by activity soon after bombardment.

The first method for separation of thallium from gold targets was chemical. The gold target is dissolved in aqua regia to which has been added carrier thallium. Gold is precipitated as the metal by the addition of sodium bisulphite, and thallium is then precipitated as the iodide by subsequent addition of potassium iodide. Thallous iodide is dried and put in the oven with a large excess of potassium metal that serves to reduce the product to thallium at several hundred degrees centigrade. The oven is then heated to 700°-800° C to produce a beam of thallium. This chemistry gives yields of 50%, and can be performed in less than 2 hr, but it was discarded after several run failures because potassium hydroxide from partially reacted potassium metal sometimes causes rapid deterioration of source slits.

A successful and very rapid procedure is to separate thallium from the target by evaporation, for the ratio of thallium and gold vapor pressures at the melting point of gold is approximately $10^5/1$. The evaporator system illustrated in Fig. 2 is a water-cooled collector cup and a nearby evaporation crucible containing the target and carrier thallium. The target is cut into small pieces and placed in the crucible, the unit pumped down to a pressure of 10^{-5} mm Hg, and the crucible temperature rapidly raised to the melting point of gold as determined by the sag of a gold sample on the cool side of the crucible. Transfer takes place almost instantaneously. The collector is then removed from the evaporator and placed in the oven. Typical separation time is 30 to 45 minutes and typical yield is 75%.

PROCEDURE AND IDENTIFICATION

C-field measurements and apparatus adjustments are made from a cesium beam coming from the thallium oven. This cesium is the product of decomposition of cesium oxide, which in turn is the product of decomposition of cesium nitrate at a lower temperature. The decomposition vapor pressure of cesium over the melt is approximately 10 μ at 800° C. This procedure is used because of the well-known difficulties in the surface-ionization detection of thallium, and because the quantity of carrier may then be adjusted to a value convenient with respect to chemistry and oven temperature.

Spin measurements are typically made at a C field of 20 gauss. All measurements are, however, checked at lower fields to insure that decoupling is small. A spin search is made by collecting on a sulphur surface[2] samples of the beam taken at values of radio-frequency calculated from the magnetic field as determined by the frequency of the cesium resonance. Exposures are counted in scintillation counters identical to those described in Reference 2. Counter background of K-X-ray detection of the neutron-deficient isotopes is about 3/4 count per minute, and for beta detection of Tl^{204} is about 5 counts per minute.

There is a large body of information regarding the decay modes and half lives of the neutron-deficient thallium isotopes,[4-12] including mass-spectrometric assignments[7] for isotopes 199, 198m, and 198. Tl^{198m} has been assigned a half life of 1.8 hr, and a decay branching of 40% to the ground state and 60% to electron capture. Isotopes 197, 198, 199, and 200 have been assigned half lives of 2.7, 5.3, 7.2, and 27 hr respectively, and decay principally by electron capture. Tl^{204} decays by beta emission with a half life of approximately 4.1 yr.[13-17] Isotope identification for the neutron-deficient isotopes consists of observing the decay of a sample collected at the appropriate spin position. Tl^{204} is, by arguments previously given, pure by activity when run.

EXPERIMENTAL RESULTS

A. Neutron-Deficient Isotopes

Thallium-199, -198m, and -198 investigations are made from bombardments of a 10-mil gold foil, which completely degrades the 48-Mev alpha beam. The decay and decay analysis of the full beam from such a run is shown in Fig. 3. The prominent activities are those of Tl^{199}, Tl^{198m}, and Tl^{198}. The untreated (i.e., nonnormalized) results of a spin search are shown in Fig. 4. Only spins 1/2 and 7 show an appreciable net effect. It has been shown that the decay of apparatus background samples is substantially identical to that of the full beam; therefore the decay curves of Figs. 5 and 6 are the observed decay rate less a quantity proportional to the direct beam but equal in magnitude at the time of exposure to the nearest (in time and frequency) apparatus background. No appreciable effect for Tl^{198} is seen in this or any other spin search. The reason for our failure to find this nuclide is not clear.

No measurable quantity of Tl^{197} is found for the bombardment procedure described above, presumably because of a high alpha-4n threshold or selective loss of this material by evaporation from the target. That the latter phenomenon does occur has been shown on other grounds. Therefore this isotope is produced selectively by bombardment of a thin gold foil at a lower average current than used for other runs. A full beam from such a bombardment is shown in Fig. 7--most of the activity is indeed due to Tl^{197}. The decay of an exposure at spin 1/2 is shown in Fig. 8.

The $^2P_{1/2}$ state hyperfine-structure splittings of the neutron-deficient isotopes are probably too large to be decoupled by the A and B fields, as shown by preliminary high-field measurements on Tl^{199}, and the observed absence of appreciable decoupling at lower fields. Therefore measurements of the nuclear moments of these isotopes will have to await techniques permitting determinations in the $^2P_{3/2}$ state.

B. Thallium-204

Untreated results of a spin search in Tl^{204} are shown in Fig. 9. Only spin 2 gives a large signal. No attempt has been made to obtain the half life of this material, but it has been shown that the half life is more than 6 months.

A resonance curve taken after the initial spin search (Fig. 10) and at a field of 14.237 gauss shows an inordinate shift in the position of the line and a width far in excess of that ordinarily observed. Therefore it was immediately suspected that this isotope has (as was later shown) an extremely small magnetic moment. Resonance curves taken at progressively larger fields are shown in Fig. 11, giving a hyperfine-structure splitting of 732 Mc with a quoted uncertainty of 5 Mc. The corresponding nuclear magnetic moment is $\pm 0.0894 \pm .002$ nuclear magnetons, and is too small to permit a reliable sign determination by the method of consistency.

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FIGURE LEGENDS

Fig. 1. Typical resonance in natural thallium.

Fig. 2. Evaporation unit used to separate radiothallium from gold targets.

Fig. 3. Decay and decay analysis of direct beam from 10-mil gold target.

Fig. 4. Results of one spin search with products of 10-mil target.

Fig. 5. Decay of sample taken at spin 1/2. Thick target.

Fig. 6. Decay of sample taken at spin 7. Thick target.

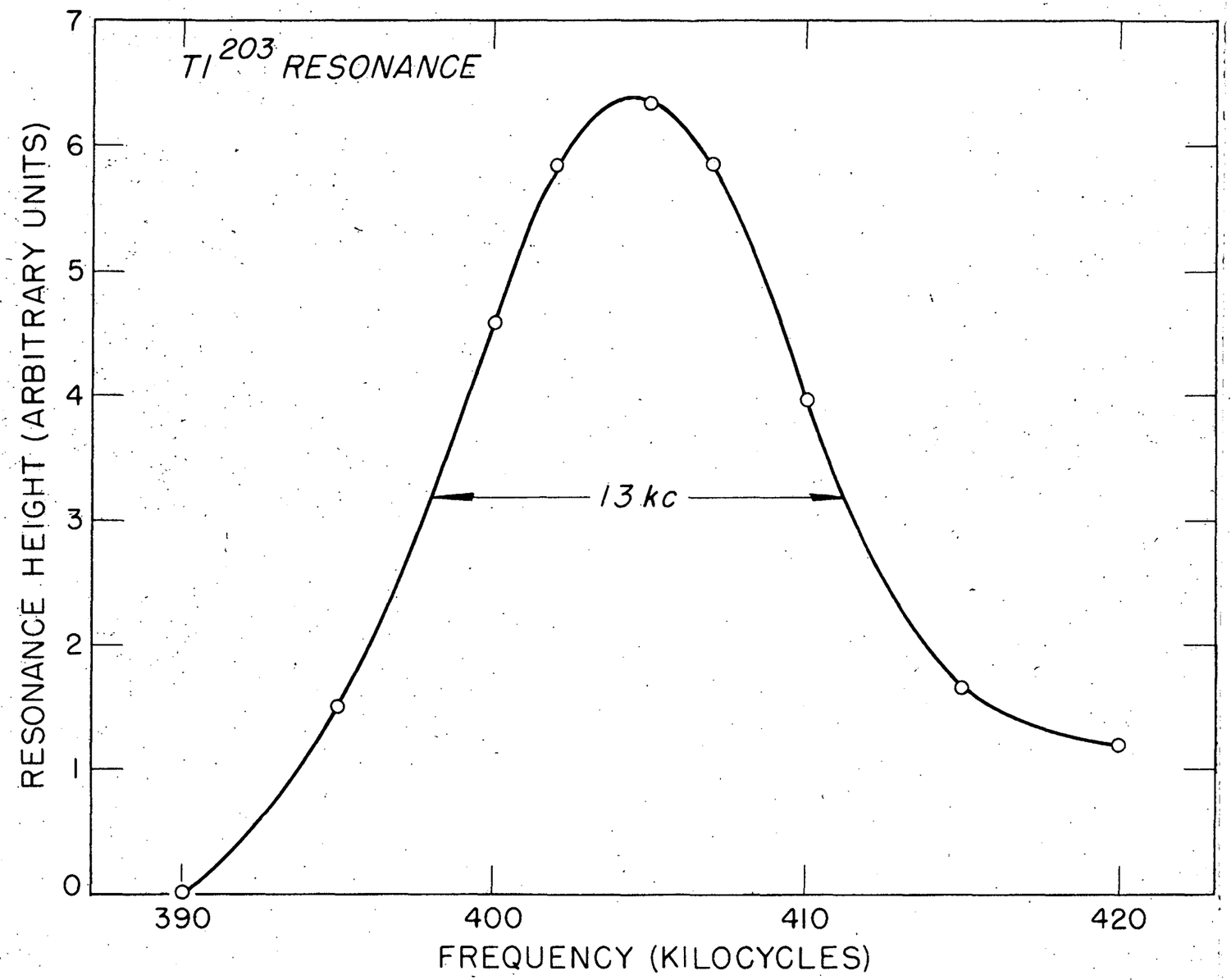
Fig. 7. Decay and decay analysis of direct beam from thin gold target.

Fig. 8. Decay of sample taken at spin 1/2. Thin target.

Fig. 9. Results of spin search in Tl^{204}.

Fig. 10. Thallium-204 resonance. The vertical line indicates the approximate center for large hyperfine structure.

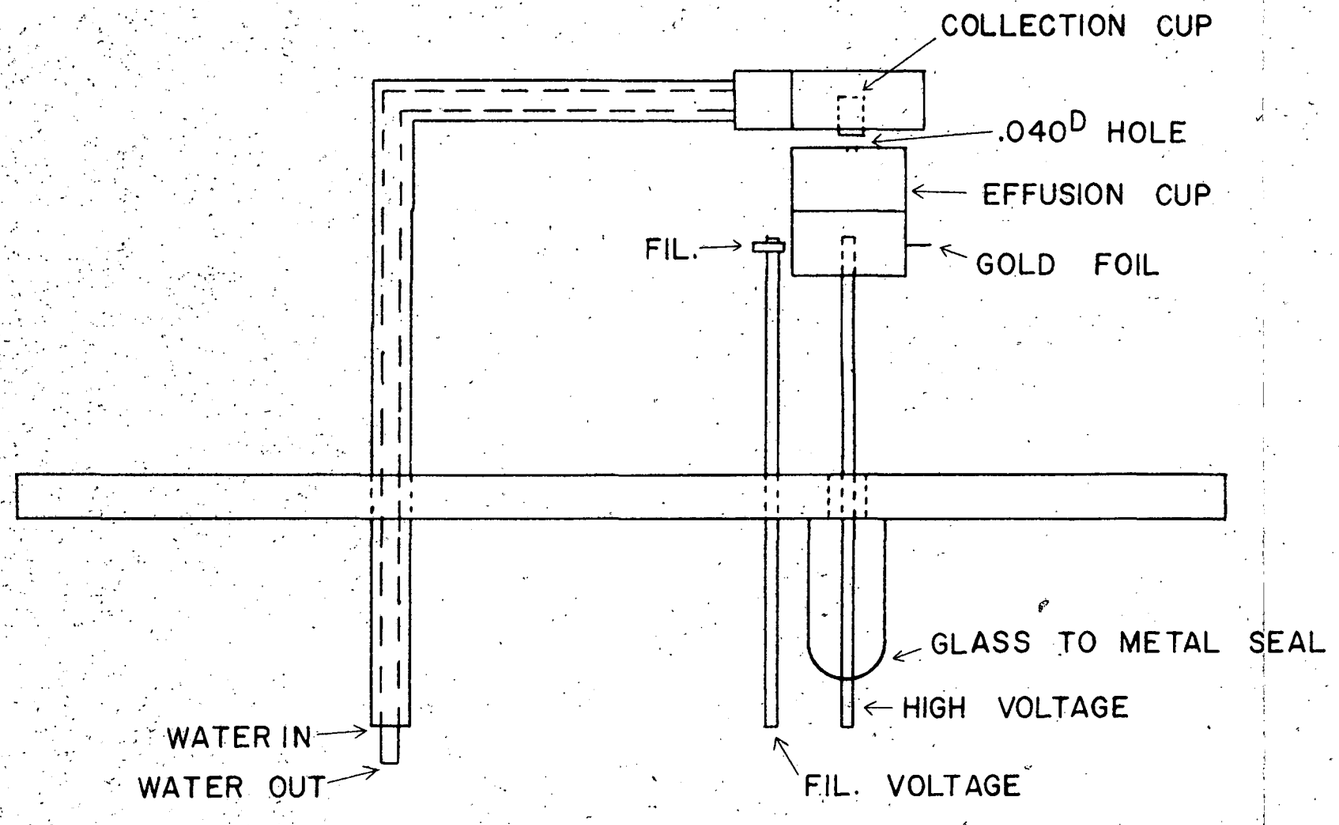
Fig. 11. Thallium-204 hyperfine structure determination at four values of C-field. At the top is indicated a hyperfine structure scale which ignores the nuclear magnetic moment term. The effect of this term is in all cases considerably less than a resonance half-width.



24937-2

FIG. 1

24900-2



EVAPORATOR

FIG. 2

THALLIUM

RUN No. 1, DIRECT BEAM

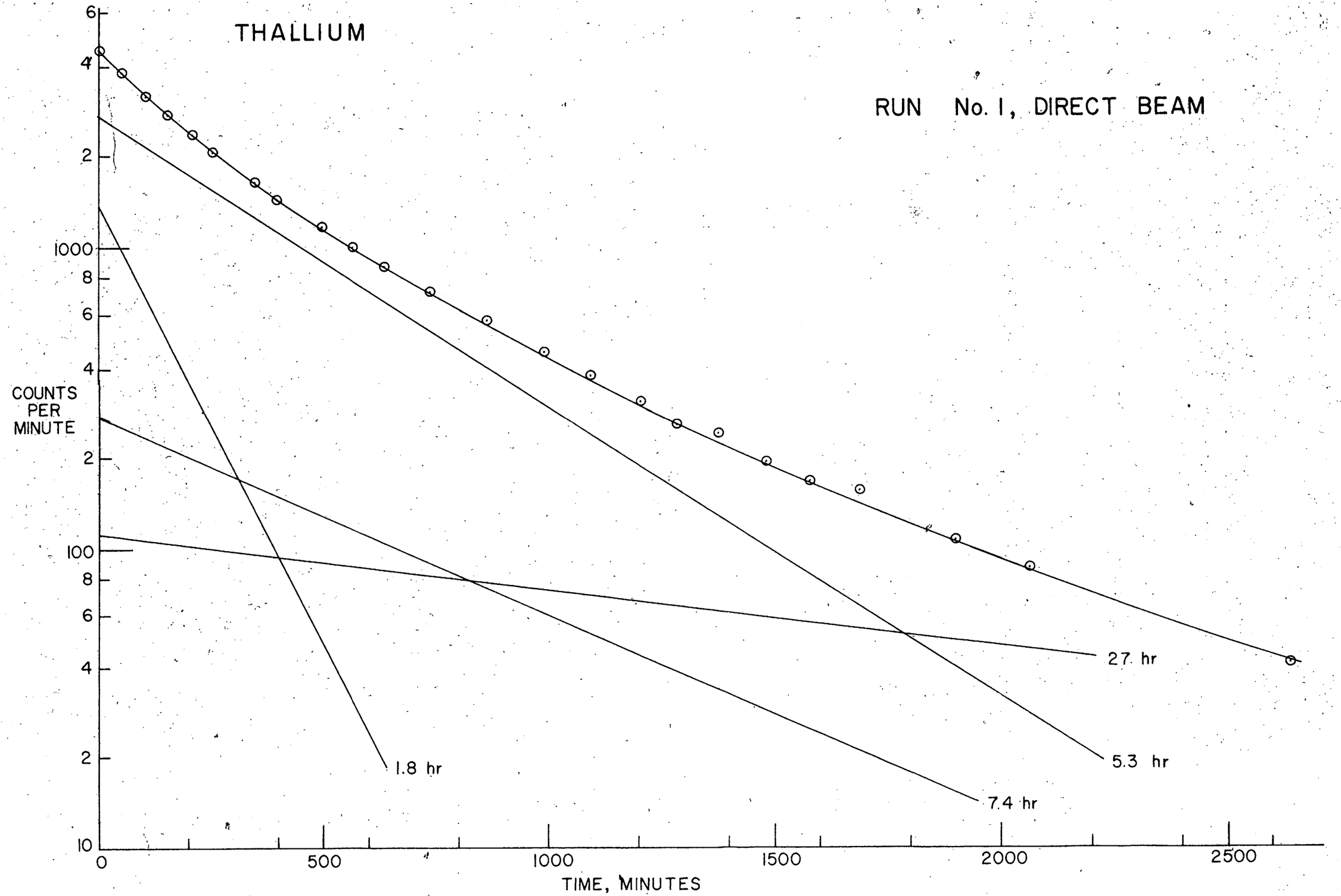
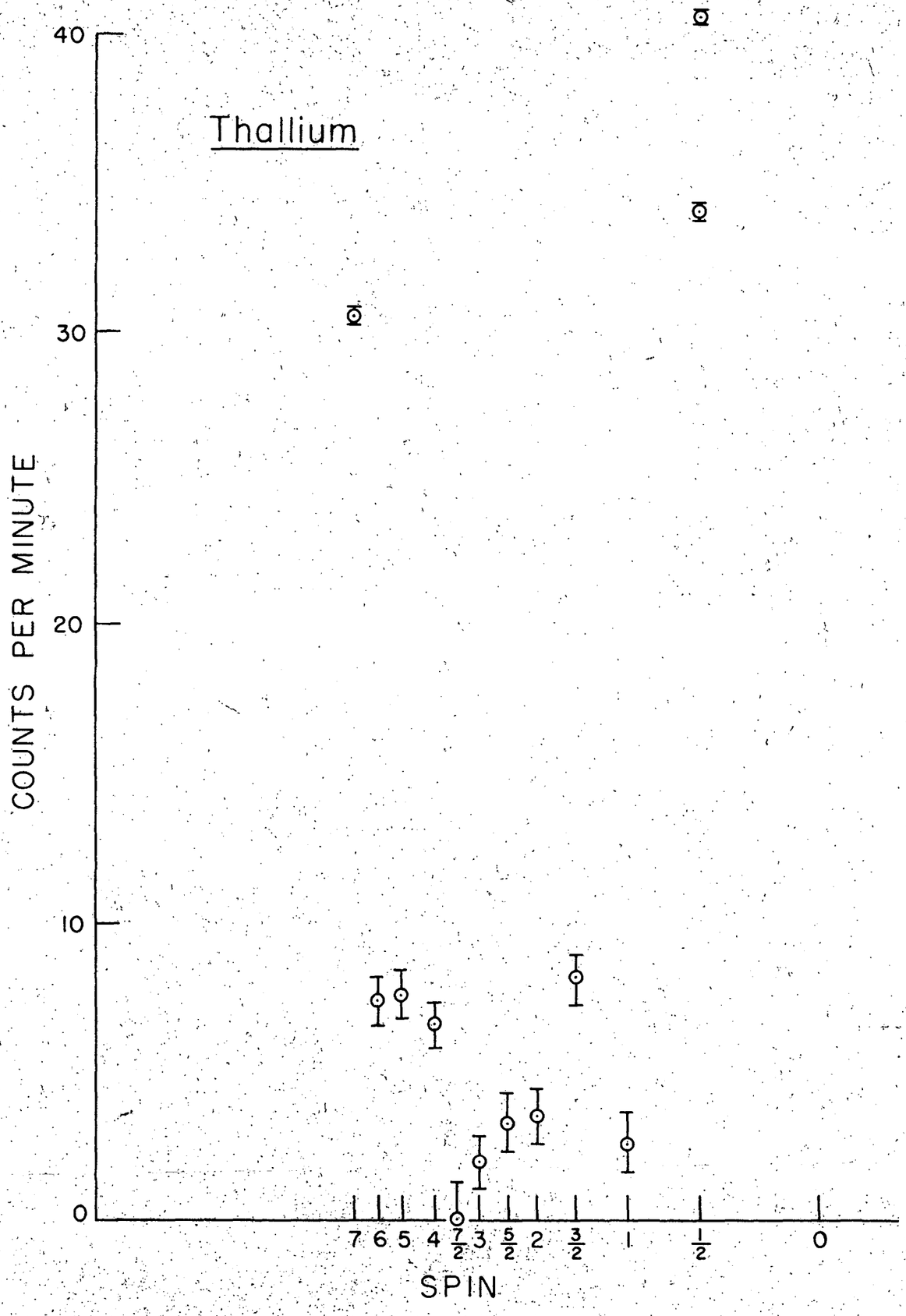


FIG. 3

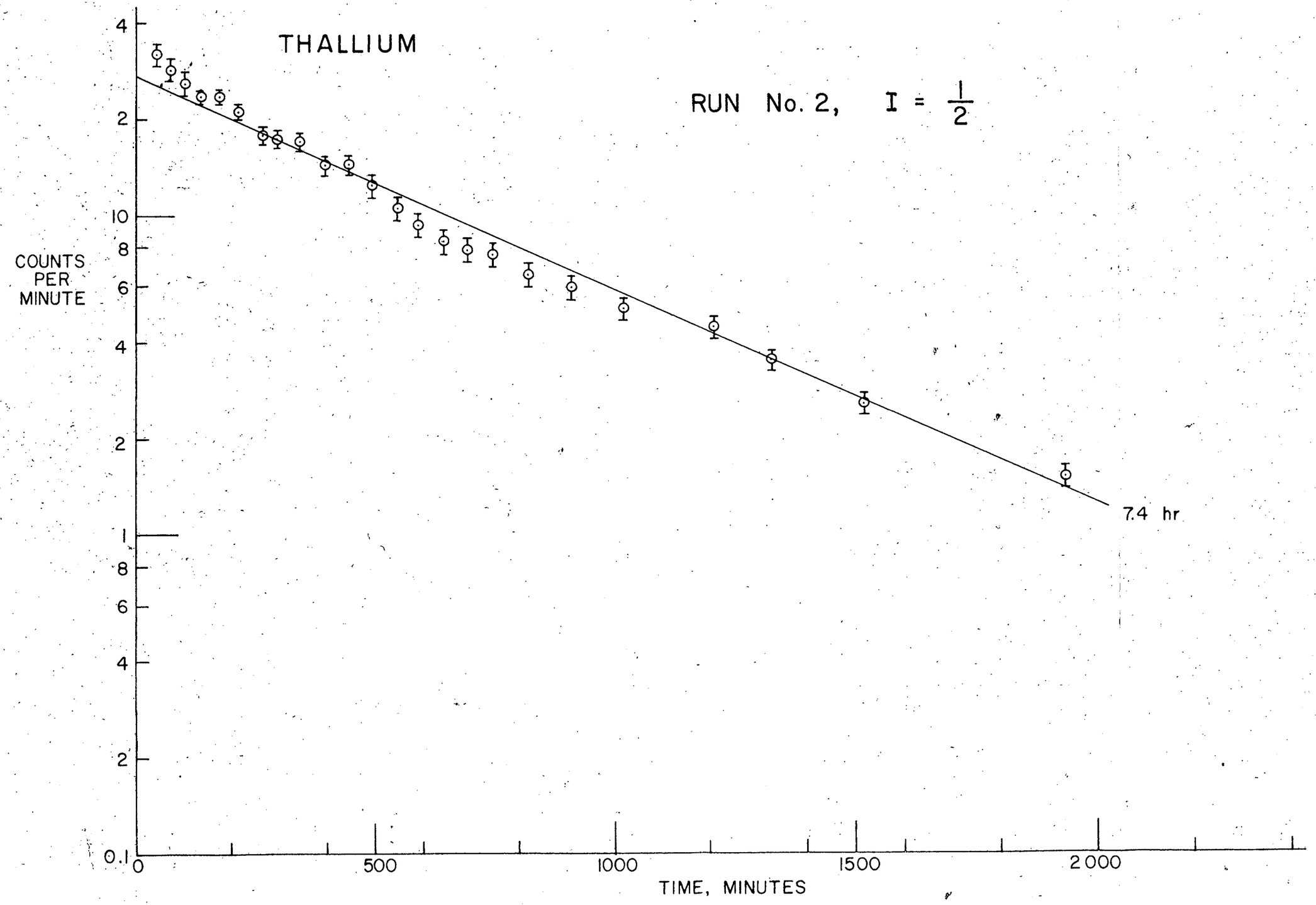
24401-2



24934-2

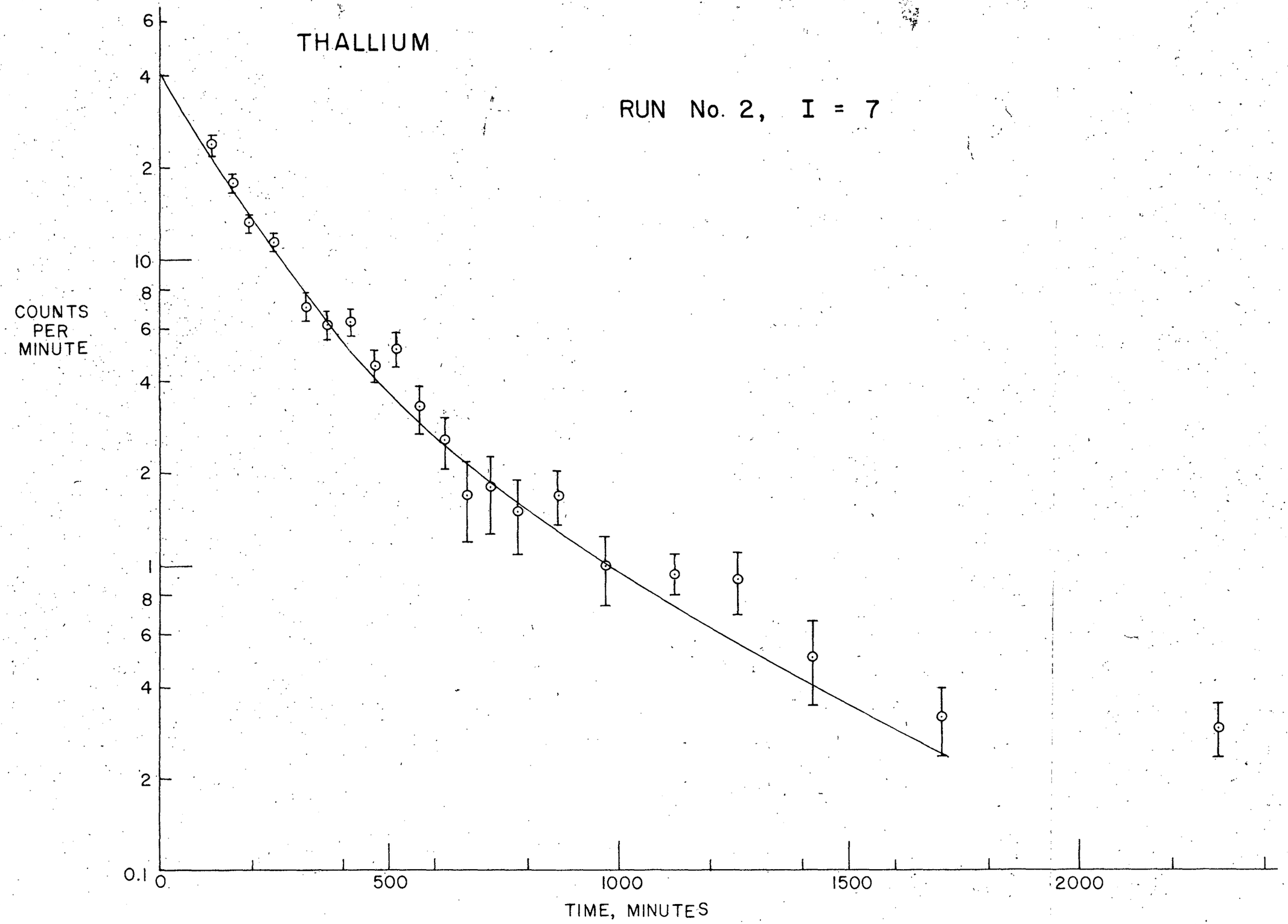
Fig. 4

24934-2



24905-2

FIG 5

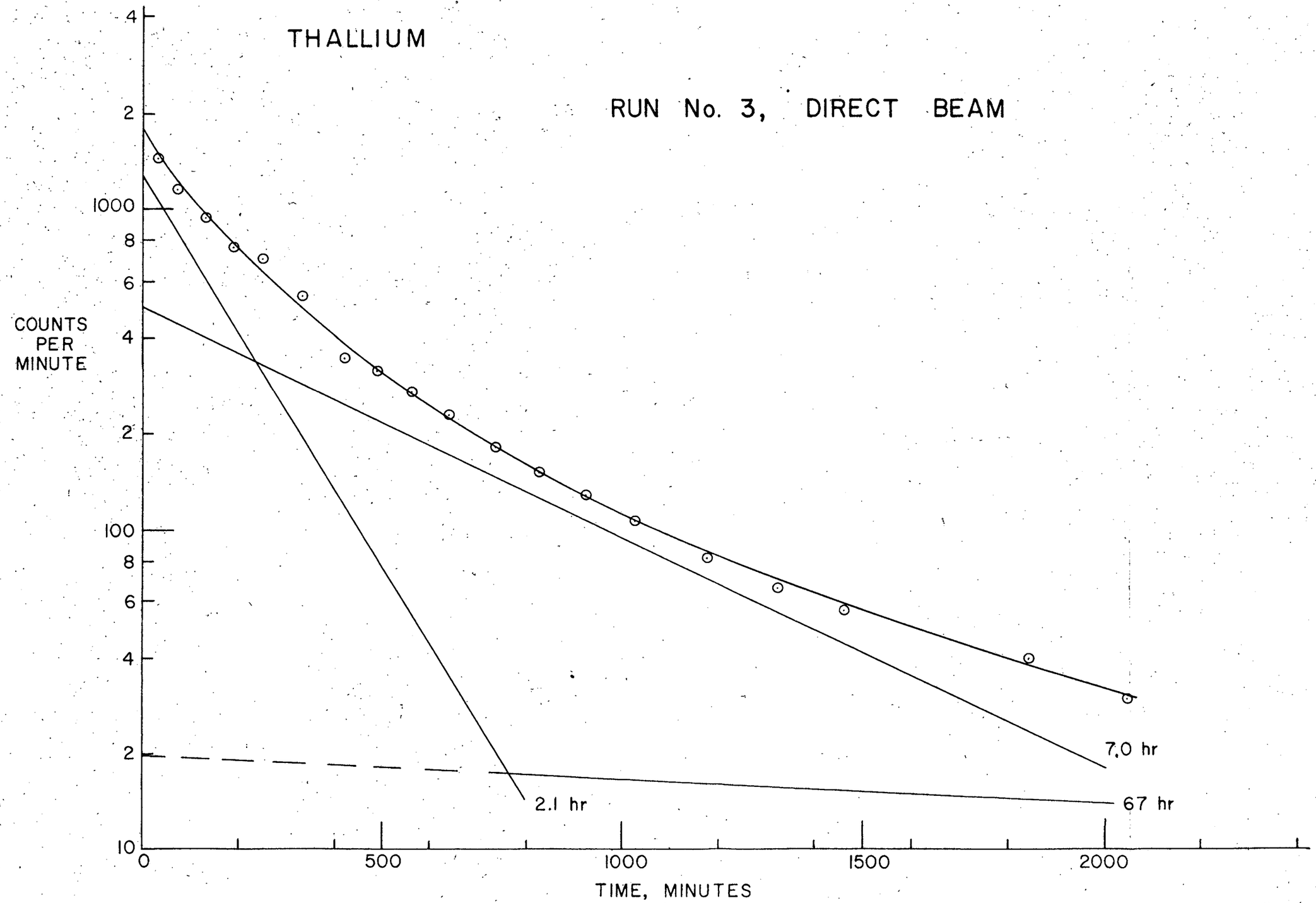


24904-2

24904-2

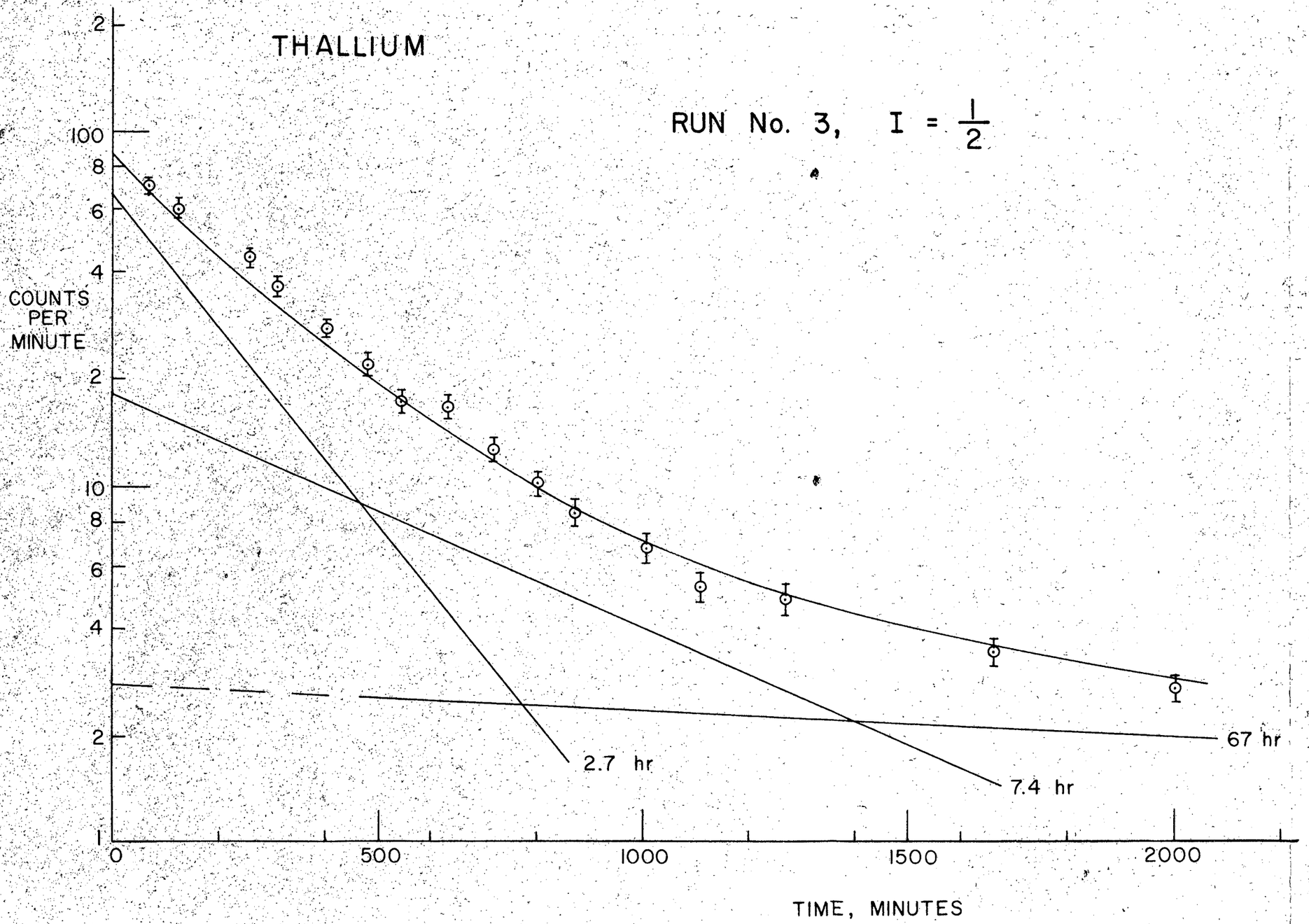
THALLIUM

RUN No. 3, DIRECT BEAM



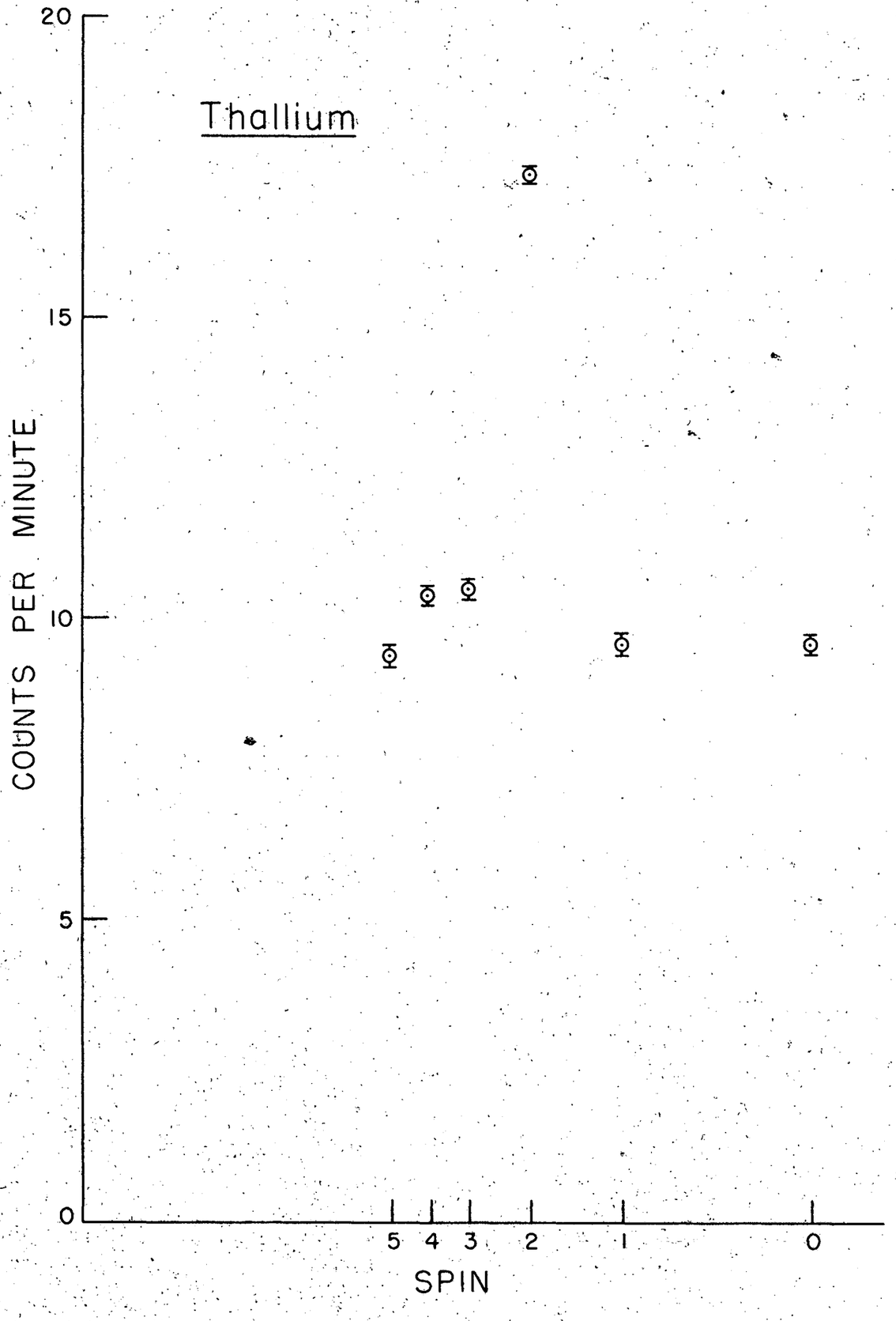
34906-2

FIG. 7


THALLIUM
RUN No. 3, I = 1/2
COUNTS PER MINUTE
2
100
8
6
4
2
10
8
6
4
2
1
0
500
1000
1500
2000
TIME, MINUTES
2.7 hr
7.4 hr
67 hr

24907-2

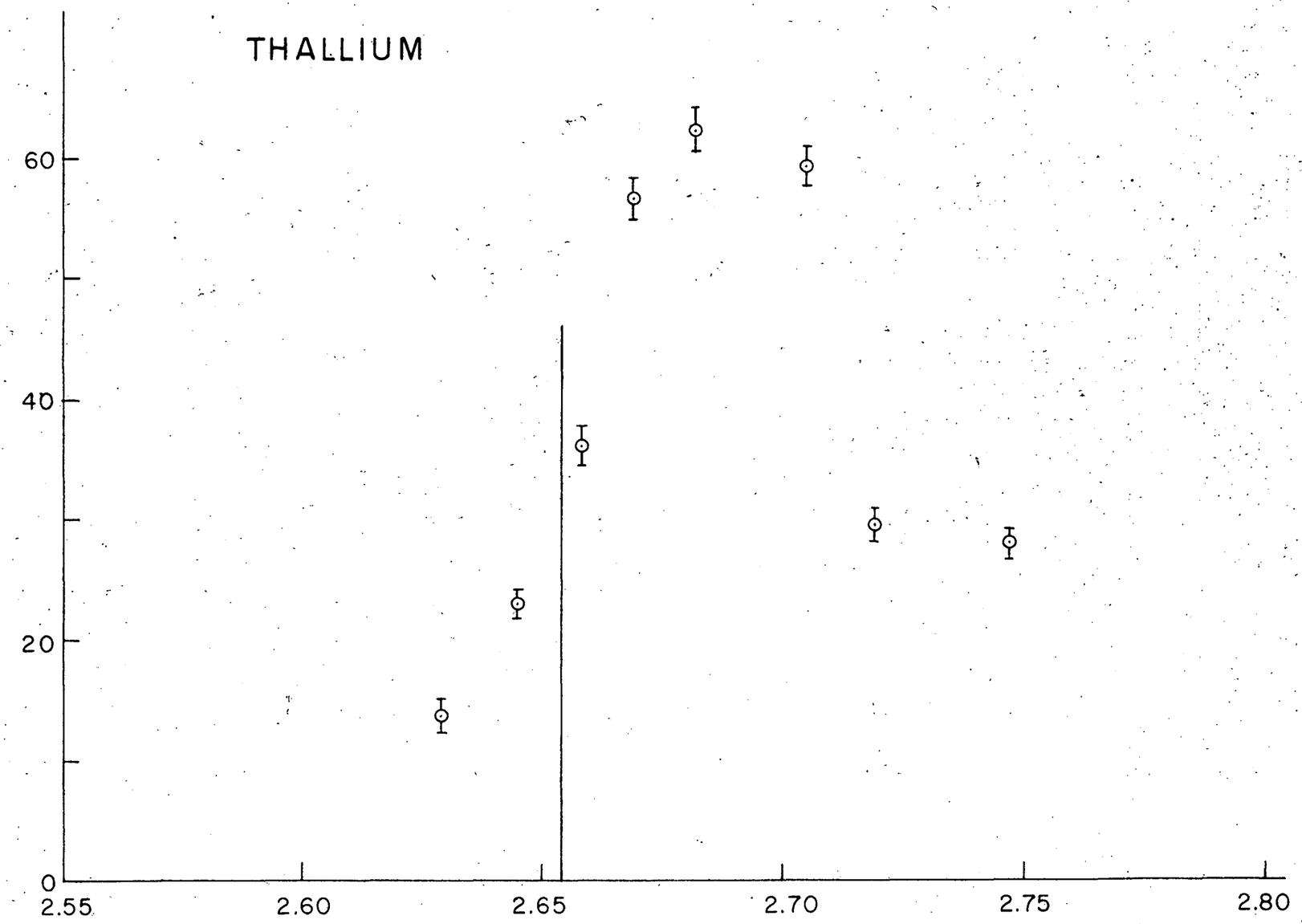
FIG. 8



24936-2

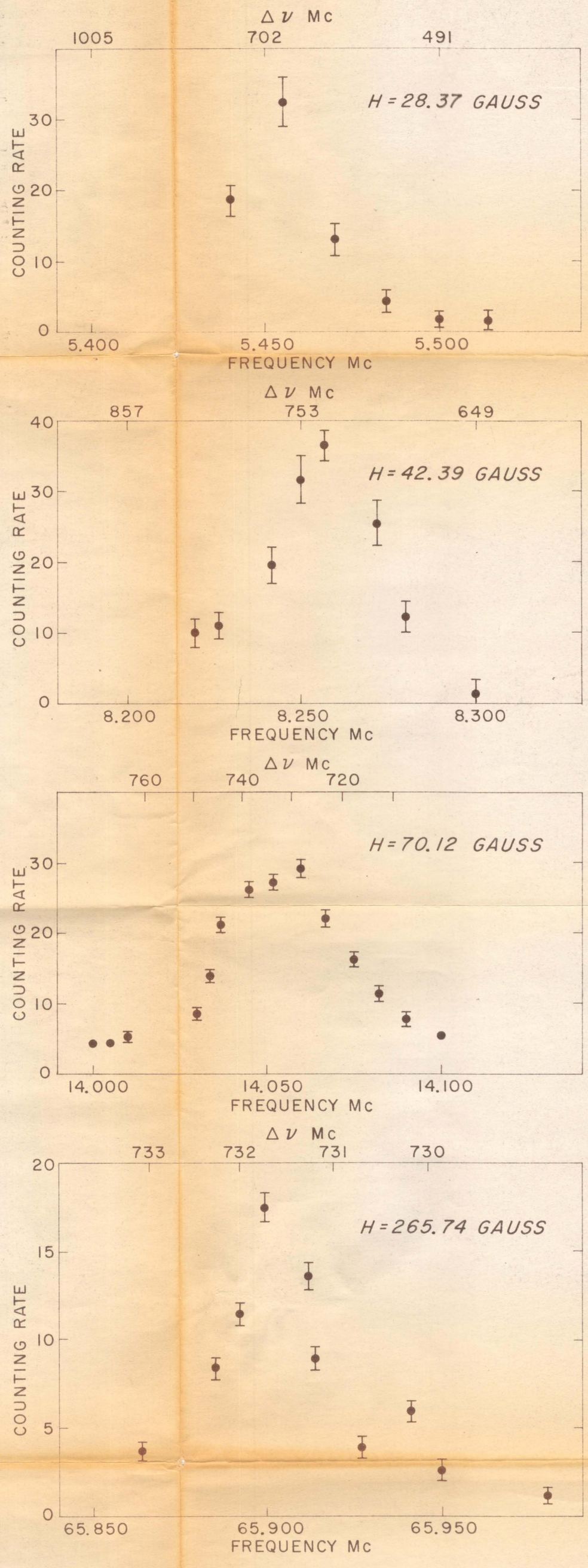
FIG-9

24936-2



RUN No. 5, I = 2

FIG. 10


Δν Mc
1005
702
491
H = 28.37 GAUSS
COUNTING RATE
30
20
10
0
5.400
5.450
5.500
FREQUENCY Mc
Δν Mc
857
753
649
H = 42.39 GAUSS
40
30
20
10
0
8.200
8.250
8.300
FREQUENCY Mc
Δν Mc
760
740
720
H = 70.12 GAUSS
30
20
10
0
14.000
14.050
14.100
FREQUENCY Mc
Δν Mc
733
732
731
730
H = 265.74 GAUSS
20
15
10
5
0
65.850
65.900
65.950
FREQUENCY Mc