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Publication Date

1955-01-27

UCRL ~~2852~~
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Contract No. W-7405-eng-48

BETA EMITTER Np²³⁸ I. Beta Spectroscopy

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Hilding Slätis
Thomas O. Passell

January 27, 1955

Berkeley, California

Beta Emitter Np²³⁸

I. Beta Spectroscopy

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ABSTRACT

The beta decay of Np²³⁸ has been studied using several beta spectrometers. In addition to the known conversion lines corresponding to transitions of energies now determined as 44.0, 102.2, 986, and 1029 kev, new conversion lines, corresponding to transitions of 942 and 927 kev were found. These transitions indicate the presence of closely spaced levels at higher excitation energies. Intensities of conversion lines and beta groups were determined and compared with previous work. Fermi-Kurie plots confirm the allowed shape of the hard (1.25 Mev) beta group and give indication of complexity in the soft beta group (0.27 Mev). The energy deviations of the ground rotational band levels from the simple rotational energy formula are discussed.

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INTRODUCTION

Early investigations¹⁻⁴ of the decay of Np²³⁸ have shown a half life of 2.1 days, a complex beta spectrum, two highly converted transitions of 43- and 103-kev energy and two gamma rays of 983- and 1030-kev energy. The beta components were reported to have an allowed shape, 47 percent of the

*Present address: Phillips Petroleum Company, Idaho Falls, Idaho.

1. G. T. Seaborg, A. C. Wahl, and J. W. Kennedy, The Transuranium Elements: Research Papers (McGraw-Hill Book Co., Inc., New York, 1949), National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, p. 13.
2. A. H. Jaffey and L. B. Magnusson, The Transuranium Elements: Research Papers (McGraw-Hill Book Co., Inc., New York, 1949), National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, p. 978.
3. M. S. Freedman, A. H. Jaffey, and F. Wagner, Jr., Phys. Rev. 79, 410 (1950).
4. J. W. Mihelich, Phys. Rev. 87, 646 (1952).

disintegrations belonging to the hard component, the maximum energy of which is 1272 kev, and 53 percent to the soft component of 258-kev energy.

The present paper sets forth and interprets material gained from further beta spectroscopic work on Np^{238} , some of which has been partially reported in prior notes.⁵⁻⁷

EXPERIMENTAL PROCEDURE AND RESULTS

For the Stockholm work the Np^{238} was obtained by bombarding natural uranium metal with protons of 12.5-Mev energy in the 225-cm cyclotron of the Nobel Institute of Physics,* and for the Berkeley work, by irradiation of Np^{237} with neutrons in the MTR reactor, Reactor Testing Station, Arco, Idaho.**

The neptunium activity was chemically purified by various combinations of the procedures given by Magnusson, Thompson, and Seaborg.⁸

The following types of measurements were made:

1. Beta spectroscopy with the Stockholm intermediate image, long lens spectrometer.⁹ (At a resolution of ~2 percent.) (Referred to as SS.)

*We are greatly indebted to Dr. Hugo Atterling, who developed and tested the special target holder for the uranium metal and who carried out the three bombardments.

**We express appreciation to Drs. W. B. Lewis, Richard Smith, and others at the Reactor Test Station for facilitating the irradiation.

5. Slätis, Rasmussen, and Atterling, Phys. Rev. 93, 646 (1954).
6. Rasmussen, Passell, and Stephens, University of California Radiation Laboratory Unclassified Report UCRL-2585, 1954 (unpublished).
7. T. O. Passell, Ph.D. Thesis, University of California Radiation Laboratory Unclassified Report UCRL-2528, 1954 (unpublished).
8. Magnusson, Thompson, and Seaborg, Phys. Rev. 78, 363 (1950).

2. Beta spectroscopy with the Stockholm double-focusing spectrometer.¹⁰
(At resolution of 0.5 percent.) (Referred to as SD.)
3. Beta spectroscopy with the Berkeley double-focusing spectrometer.¹¹
(At resolution of 1.2 percent.) (Referred to as BD.)
4. Electron spectroscopy of photo and Compton electrons from a (0.0005-inch) uranium metal radiator with the Berkeley ring-focusing solenoidal spectrometer.¹² (At resolution of ~2 percent.) (Referred to as BS.)

Where duplicate information was obtained, we present the best or the average of the best determinations. In general, regarding beta spectroscopic results, the energy measurements on the SD spectrometer are taken as standard, since this instrument is specifically designed for precision energy comparisons. However, the energy differences of the high-energy conversion lines determined on the BD spectrometer are preferred by virtue of the much greater source strength and consequent better counting statistics in the Berkeley work. The BD measurements were not as useful as SS and SD for the continuum and lines below 0.7 Mev (disintegration energy of Np²³⁹) because of the larger admixture of the troublesome Np²³⁹. The SS measurements form the basis for spectrum shape studies of the hard beta

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9. H. Slätis and K. Siegbahn, Arkiv för Fysik 1, 339 (1949).
 10. A. Hedgran, Arkiv för Fysik 5 (1) (1952).
 11. G. D. O'Kelley, Ph.D. Thesis, University of California Radiation Laboratory Unclassified Report UCRL-1243 (1951), unpublished.
 12. G. D. O'Kelley, California Research and Development Company Unclassified Report MTA-38 (May 1954), unpublished.

particles, as the background due to scattered electrons entering the counter is minimized by the intermediate image slit system.

Figure 1 shows the spectrum taken with the SD spectrometer in the region of the conversion lines of the 102.2-keV gamma ray. Note the lines assigned to Np^{239} present in small amount. By means of the well-known strong conversion lines of Np^{239} it was possible to make corrections for the small contamination of this activity when computing the intensities of the beta components.¹³

Figure 2 shows the spectrum taken with the BD spectrometer of the region above 3500 gauss cm. The K lines of the four high-energy gamma transitions are clearly evident. The arrows labeled for L and M lines each point to the calculated abscissa for the foot of the leading edge of the line (about 20 gauss cm to the right of the peak). The unlabeled arrow at 3887 gauss cm points to the calculated position of the K line of a transition 146 keV less energy than the most energetic gamma ray. (With the decay schemes proposed in the article¹⁴ following this, which we refer to as paper II, it is important to search for such a transition.) There is no evidence for this line, and we can from these data say that it is less than 10 percent of the intense line K1030.

The conversion line energy and intensity data are summarized in Table I. The energy figures are based on averages of the best Stockholm or Berkeley data. As the intensities are of considerable importance in constructing

13. Regarding the shape of the Np^{239} spectrum see H. Slätis, Arkiv för Mat. Astr. Fysik 35A (3) (1947).

14. Rasmussen, Stephens, Strominger, and Åström, Phys. Rev. (following paper).

the decay scheme and as there is some variation in intensities obtained in various studies, Table I includes three intensity columns: one for the work of Freedman et al.,³ one for Slätis et al.,⁵ and one for the BD work previously unreported.

There is fair agreement in the intensity figures between the different investigations except for some extremely weak high energy lines and except for the lowest energy 21.8-keV L_{II}^{44} line. At such low energies spectrometer measurements are subject to some uncertainty from counter window transmission losses. In the work of Slätis et al.,⁵ the low energy cut-off of the beta spectrum (window of 6 Formvar films, each of about $15 \mu\text{g cm}^{-2}$ thickness) was at about 200 gauss cm, and their intensities of L_{II}^{44} , L_{III}^{44} , and MNO^{44} listed in Table I contain corrections of 22, 13, and 3 percent, respectively. In view of the disagreement, the total intensity figure for conversion lines of the 44-keV transition has not been given much weight in the construction of a decay scheme.

It is of some interest to collect as in Table II yet other determinations of intensity of the low-energy conversion lines. Passell⁷ has measured on the BD spectrometer the relative intensities of these lines using both Np^{238} and Cm^{242} samples. A very thin counter window was used, and no transmission correction was made.

Dunlavy and Seaborg¹⁵ and Dunlavy¹⁶ have determined $L:(M+N+O)$ ratios with Cm^{242} samples by track counting in electron-sensitive photographic emulsions, and their results are presented in Table II also.

15. D. C. Dunlavy and G. T. Seaborg, Phys. Rev. 87, 165 (1952).

16. D. C. Dunlavy, unpublished results (1953) (110 electron coincidence events counted).

Table I

Conversion Lines Accompanying Beta Decay of Np^{238}

Electron momentum gauss cm*	Electron line energy*	Shell Converting	Electron binding energy (Pu) ¹⁷	Gamma energy	Abundance per 100 beta disintegrations		
					Freedman, Jaffey, Wagner ³	Slätis, Rasmussen, Atterling ⁵	Berkeley double- focusing spectro- meter (previously unreported)
503	21.8	L _{II}	22.2	44.0	38	28.7	-
549	25.9	L _{III}	18.1	44.0	20	21.0	-
671	38.2	M _{II}	5.6	43.8	44.0	14	**
678	39.0	M _{III}	4.6	43.6			
713	42.9	N	1.4	44.3	3.2	15.2	
720	43.7	O	0.3	44.0			
991	80.1	L _{II}	22.3	102.4	102.2	1.9	1.4
1017	84.1	L _{III}	18.1	102.2			
1097	96.7	M _{II,III}	~5.0	101.7	1.2	0.7	-
4046	805.3	K	121.7	927	-	0.05	0.03
4099	819.8	K	121.7	942	-	0.10	0.07
		L	-	-	-	0.06	-
4259	864.4	K	121.7	986	0.3	0.26	0.20
		L	-	-	-	0.13	0.04
4414	907.6	K	121.7	1029	0.3	0.22	0.20
		L	-	-	-	0.08	0.04
		M,N,O	-	-	-	0.06	0.02

*Calibration for 44-kev gamma lines from Th B A line taken as $H_{\text{Pa}} = 534.11$ from the work of Meyer and Schmidt.¹⁸
 Calibration for 102-kev gamma lines from Th B F line taken as $H_{\text{Pb}} = 1388.56$ from the work of G. Lindström.¹⁹
 Calibration for higher energy lines from Cs¹³⁷ K line taken as $H_{\text{Pb}} = 3381.3$ from the work of G. Lindström, K. Siegbahn, and A. H. Wapstra.²⁰

The SD measurement of absolute energy of K₉₈₆ is used, but the energy differences to other high energy lines is taken from BD work.⁶

**Berkeley measurements give the M_{II}:M_{III}:N,O ratio as 1.85:1.35:1. (See Passell.⁷)

Table II

Intensities of Conversion Lines of the 44-Kev Transition

Ratio	Freedman <i>et al.</i> ³ Np ²³⁸	Slatis <i>et al.</i> ⁵ Np ²³⁸	Passell ⁷ Cm ²⁴²	Berkeley double focussing ⁷ Np ²³⁸	Dunlavey, Seaborg ¹⁵ Cm ²⁴²	Dunlavey re- determination by same method ¹⁶ Cm ²⁴²	Gellman <i>et al.</i> ²¹ (E2 theoreti- cal)
L _{II} :L _{III}	1.91	1.37	1.44	1.26	-	-	~1.2
(L _{II} +L _{III})/(M+N+O)	3.26	3.27	3.42	3.33	4.8	2.93 ± 0.64	-

The E2 nature of the 44.0 and 102.2-kev transitions is clearly confirmed by comparison of the L subshell conversion results with the theoretical calculations of Gellman *et al.*²¹

The hard beta spectrum was taken repeatedly in the intermediate image spectrometer in an effort to determine precisely the shape. The intermediate image spectrometer was felt to be especially suitable for spectrum shape studies, as it has a very low scattered electron background. The log ft values of the hard beta particles are in good agreement with a first forbidden $\Delta I = 2$, yes, assignment, although Freedman *et al.*³ reported an allowed spectrum shape. The reinvestigation was in part prompted by the availability of accurate tables²² for determination of spectrum shape correction factors.

17. Hill, Church, and Mihelich, *Rev. Sci. Instr.* 23, 523 (1952).

18. Meyer and Schmidt, *Phys. Rev.* 94, 927 (1954).

19. G. Lindström, *Arkiv för Fysik* 4 (1) (1951).

20. G. Lindström, K. Siegbahn, and A. H. Wapstra, *Proc. Phys. Soc. B* 66, 54 (1953).

21. Gellman, Griffith, and Stanley, *Phys. Rev.* 85, 944 (1952).

22. M. E. Rose, C. L. Perry, and N. M. Dismuke, Oak Ridge National Laboratory Report ORNL-1459 (1953); Tables for the Analysis of Allowed and Forbidden Beta Transitions.

These tables show that the correction factor for the $\Delta I = 2$, yes spectrum shape for high atomic numbers is considerably less pronounced than the $Z = 0$ approximation correction sometimes used. In the conventional Fermi-Kuri plot of Fig. 3 (where the Fermi function was obtained from the National Bureau of Standards tables²³) are shown a straight solid line and a dashed $\Delta I = 2$, yes, line calculated from the tables of Rose et al.²² The endpoint energies were chosen to give qualitative best fits to the experimental points in each case. Parts of the spectrum are obscured by soft beta rays or by high energy conversion lines. We are inclined to give most weight to the fit near the end of the spectrum. There the slopes of allowed and forbidden spectra are quite different, with the data agreeing much better with the allowed line. Very pertinent to the shape study is the question of possible complexity of the hard beta spectrum. If there were admixture of two groups with endpoints differing by only 44 kev, it would hardly be possible to determine their abundances by resolution of a Fermi-Kurie plot. Our belief that the hard beta spectrum consists of essentially a single group going to the 44-kev first-excited state is based on L-x-ray-beta coincidence work reported in paper II.¹⁴ (The contrary result previously reported⁶ was shown to be in error by later experiments using the same apparatus.) The fit for the lower energy points of Fig. 3 seems better for the forbidden shape, but this may be due to an incomplete subtraction of the Np^{239} spectrum, endpoints of which are indicated by arrows in Fig. 3.

Thus, we support the conclusion of Freedman et al.³ that the hard beta group has allowed shape, and we find an energy 1.25 Mev compared with their 1.272 Mev.

23. Tables for the Analysis of Beta Spectra, National Bureau of Standards Applied Mathematics Series 13, U.S. Government Printing Office, Washington, D.C. (1952).

Figure 4 shows a Fermi-Kurie plot of the soft beta continuum from SS data after subtraction of hard beta and Np^{239} beta contributions. Parts of the spectrum are obscured by conversion lines. The Fermi-Kurie plot has a definite curvature, indicating the probable presence of more than one soft beta group, but it is not felt that the data are of sufficient accuracy to justify any resolution of the plot. The endpoint of the most energetic soft beta group seems to be about 0.27 Mev, in satisfactory agreement with the 0.258 Mev of Freedman et al.³

The intensity ratio between soft beta rays and hard beta groups was found to be 55:45 in this investigation. This ratio is in good agreement with the ratio 53:47 of Freedman et al.³

SPECTROSCOPIC MEASUREMENT OF URANIUM RADIATOR PHOTOELECTRONS

In order to study the relative intensities of the high energy gamma rays (unresolvable by scintillation spectrometer) the spectrum of electrons ejected from a 0.0005-inch thick uranium metal radiator was studied in the Berkeley thick-lens solenoidal beta spectrometer.¹²

The sample of Np^{238} was enclosed in a cylindrical aluminum shield of sufficient thickness to absorb the beta radiation of Np^{238} , and on the plane face of the shield in the normal position for the spectrometer source was attached a disk of uranium metal 0.0005-inch thick and 0.5-inch in diameter.

Figure 5 shows one of the several series of measurements. The two prominent peaks are due to photoejection of K electrons by the 1029- and 986-kev gamma rays of Np^{238} . Centers of the peaks lie about 10 kev lower in energy than calculated, but this is accounted for by the thickness of the uranium radiators. Diffuse peaks due to L shell conversion are seen at

higher energies. At energies just below the K_{986} peak the electrons due to Compton scattering events sharply rise. The Compton electrons unfortunately obscure the region of great interest where the K photopeaks of the weaker 941- and 925-keV gamma rays are expected. One can only say that these gamma rays are in somewhat lower intensity than the higher energy gamma rays.

The dotted lines of Fig. 5 indicate how the main peaks were resolved for determination of their relative intensities. The areas of the peaks are in the ratio $K_{1029}:K_{986} = 1:0.90$. For these energies the photoelectric absorption cross section varies nearly inversely as the square of the gamma energy.²⁴ Making such a correction we obtain the gamma intensity ratio $\gamma_{1029}/\gamma_{986} = 1/0.83$. An effect which is difficult in our case to estimate quantitatively arises from the change in angular distribution of photoelectrons with energy.²⁴ The angle of the accepted electron trajectories in the solenoidal beta spectrometer with respect to the axis is between 20° and 24° , so qualitatively the effect should give higher effective transmission for the K_{1029} as compared with the K_{986} electrons. The effect should not be large with our geometries and is therefore neglected. Our ratio is to be compared with that of 1.02:1 determined by Freedman *et al.*³

It might be possible to observe photopeaks from the weaker gamma rays if a radiator of lower atomic number than uranium were used (increasing the energy difference between the K photoelectron energy and the Compton electron maximum), but this experiment has not been done by us.

24. C. M. Davisson and R. D. Evans, *Revs. Modern Phys.* 24, 79 (1952).

DISCUSSION

It is obvious that the several hard gamma rays of similar energies signify close-lying levels in Pu²³⁸ in the vicinity of 1 Mev, and the possibility of their interpretation in terms of Bohr-Mottelson type rotational bands seems promising.

In attempting to construct a decay scheme from electron spectroscopic data, one seeks to find as many cases as possible where the energies of two or more transitions add to equal the energy of another transition. In this connection it may be pointed out that the energy differences between the high energy gamma K conversion lines are known much better than their absolute energies.* The possible sums are listed below.

$$\begin{array}{ll}
 E_{\text{hard beta}} - E_{\text{soft beta}} \approx 980 \text{ keV}; & E_{\gamma 986} = 986 \text{ keV} & (1) \\
 E_{\gamma 1029} - E_{\gamma 986} = 43.2 \text{ keV}; & E_{\gamma 44} = 44.0 \text{ keV} & (2) \\
 E_{\gamma 986} - E_{\gamma 942} = 44.6 \text{ keV}; & E_{\gamma 44} = 44.0 \text{ keV} & (3) \\
 E_{\gamma 1029} - E_{\gamma 927} = 102.3 \text{ keV}; & E_{\gamma 102} = 102.2 \text{ keV} & (4)
 \end{array}$$

It is difficult to decide which of the gamma sums are true and which are coincidental. From the above beta spectroscopic information a simple level scheme with levels at 986 and 1073 keV was proposed,⁶ accounting for all four energetic gamma rays through transitions to the known levels of 0, 44, and 146 keV. Scintillation counter coincidence studies (reported in paper II) later showed the proposed scheme to be incorrect in some respects. Hence, the decay scheme proposals and accompanying interpretation will be left to paper II.

*For a more detailed discussion on the determination of these energy differences see Rasmussen, Passell and Stephens.⁶

It is of some interest to compare our values for the energies of the states of the ground rotational band with the predictions of the Bohr-Mottelson theory.²⁵ To a first approximation the energies should be given by $E = \frac{h^2}{8\pi^2 I} I(I + 1)$ with only even I values above a spin zero base state. From the first excited state energy 44.0 kev, the rotational quantum energy is 7.33 kev and by the simple formula the second excited state should lie at 146.7 kev. From our energy data it actually lies at 146.2 kev. The theory predicts such a deviation, and a "vibration rotation interaction" correction term of the form constant $\times I^2(I + 1)^2$ subtracts from the simple energy formula. In samples of Cm^{242} the gamma transition between the 6+ and 4+ rotational states of Pu^{238} has been observed²⁶ and its energy measured by scintillation spectroscopy as 157 ± 2 kev. Thus, we may check the theoretical correction term by calculating the constant using the energies of the 2+ and 4+ levels and independently using energies of the 2+ and 6+ levels. In the former case we calculate for the constant 0.0017 kev and in the latter, 0.0032 kev. The disagreement is probably real but may partly be due to experimental error, as the deviation term is so small and the above calculation consequently depends so sensitively on the energy. The constants agree, for example, if an energy of 44.2 kev were used for the 2+ state. More accurate measurements of all the low energy transition energies would be of great interest to check on this point. The disagreement, if real, would mean that the rotational energy level perturbations cannot be accounted for completely by a vibration rotation interaction treatment.

25. A. Bohr and B. R. Mottelson, Dan. Mat. Fys. Medd. 26 (14) (1952). cf. p. 33.

26. F. Asaro, S. G. Thompson, and I. Perlman, Phys. Rev. 92, 694 (1953).

The L subshell conversion coefficients clearly confirm the E2 character of the 44- and 102-kev transitions.

The resolution of M-subshell conversion lines of the 44.0-kev transition permits some comparison with the theoretical calculations of Church and Monahan.²⁷ Church's threshold conversion coefficients (non-relativistic) indicate that M shell conversion with an E2 transition occurs predominantly with the p electrons (M_{II} , M_{III}). Such is the experimental observation here. It is interesting to note that the $M_{II}:M_{III}$ ratio 1.37:1 is exactly equal to the $L_{II}:L_{III}$ ratio obtained by Slatis et al.⁵

ACKNOWLEDGMENTS

We wish to acknowledge the help of a number of persons in these investigations: Dr. Hugo Atterling and Fil Kand Mrs. Muusa Punnis in Stockholm and Peter Gray and Donald Strominger in Berkeley.

One of us (J.O.R.) wishes to acknowledge a fellowship from the Nobel Institute of Physics during part of this investigation.

This work was supported in part by the U.S. Atomic Energy Commission.

27. E. L. Church and J. E. Monahan, Talk abstracted in Bull. Am. Phys. Soc. 29, 25 (1954).

Figure Captions

- Fig. 1. Electron spectrum of Np^{238} (with some Np^{239}) in the region of the conversion lines of the 102.2-kev gamma ray. (Stockholm double-focusing spectrometer.)
- Fig. 2. The high energy part of the Np^{238} beta spectrum. (Berkeley double-focusing spectrometer.) For interpretation of the arrows see the text.
- Fig. 3. Fermi-Kurie plot of the hard beta spectrum of Np^{238} . (Stockholm long lens intermediate image spectrometer.)
- Fig. 4. Fermi-Kurie plot of the soft beta spectrum of Np^{238} (Stockholm long lens intermediate image spectrometer.) Contributions from the hard beta group and from the beta spectrum of Np^{239} contamination have been subtracted out.
- Fig. 5. Spectrum of electrons ejected from a thin uranium metal radiator by the gamma radiation of Np^{238} . (Berkeley thick-lens beta spectrometer.) Dotted lines show how the K photo lines for the two most intense gamma rays were resolved for the purpose of the relative intensity determination.

COUNTING RATE PER MOMENTUM
INTERVAL (ARBITRARY UNIT)

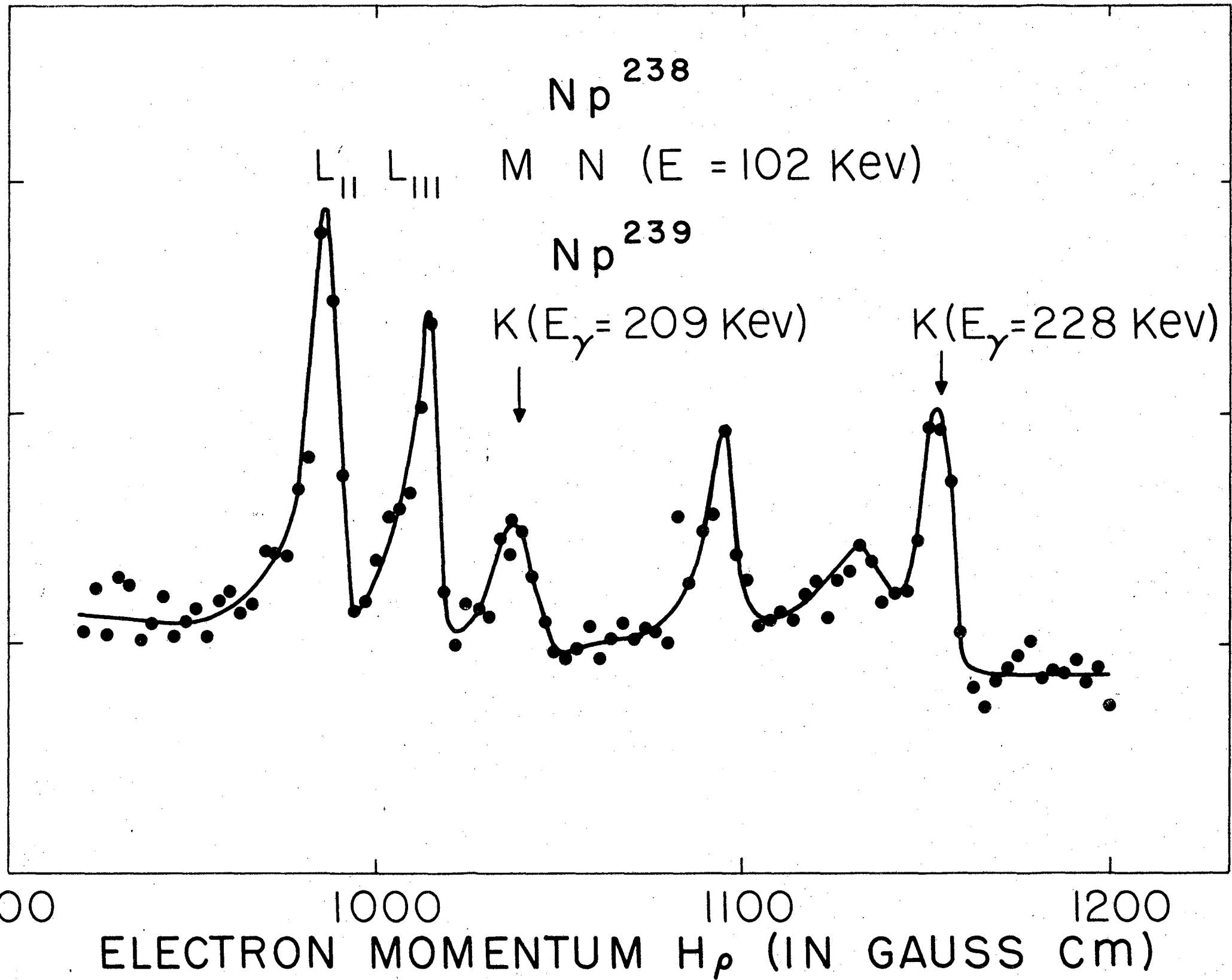
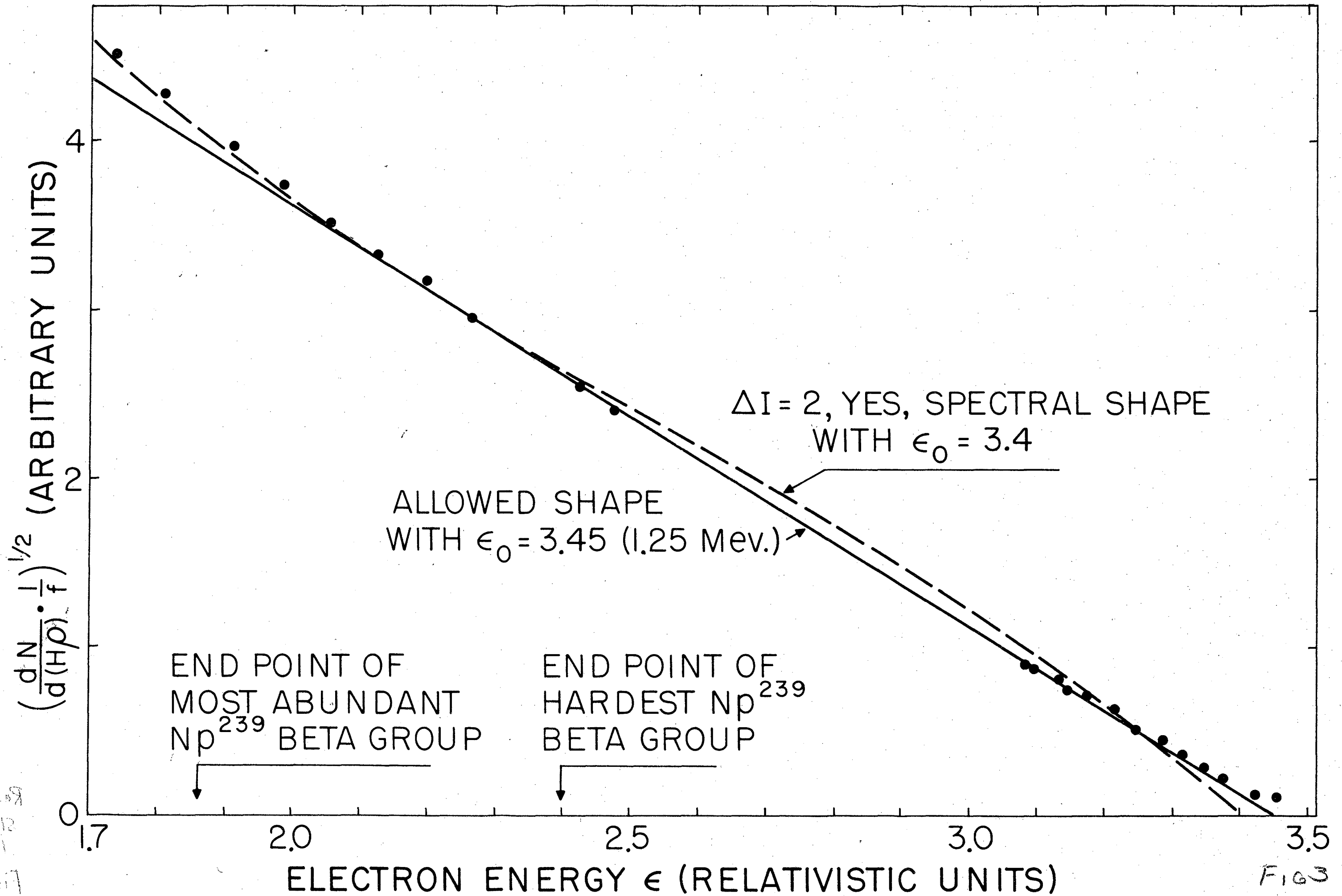
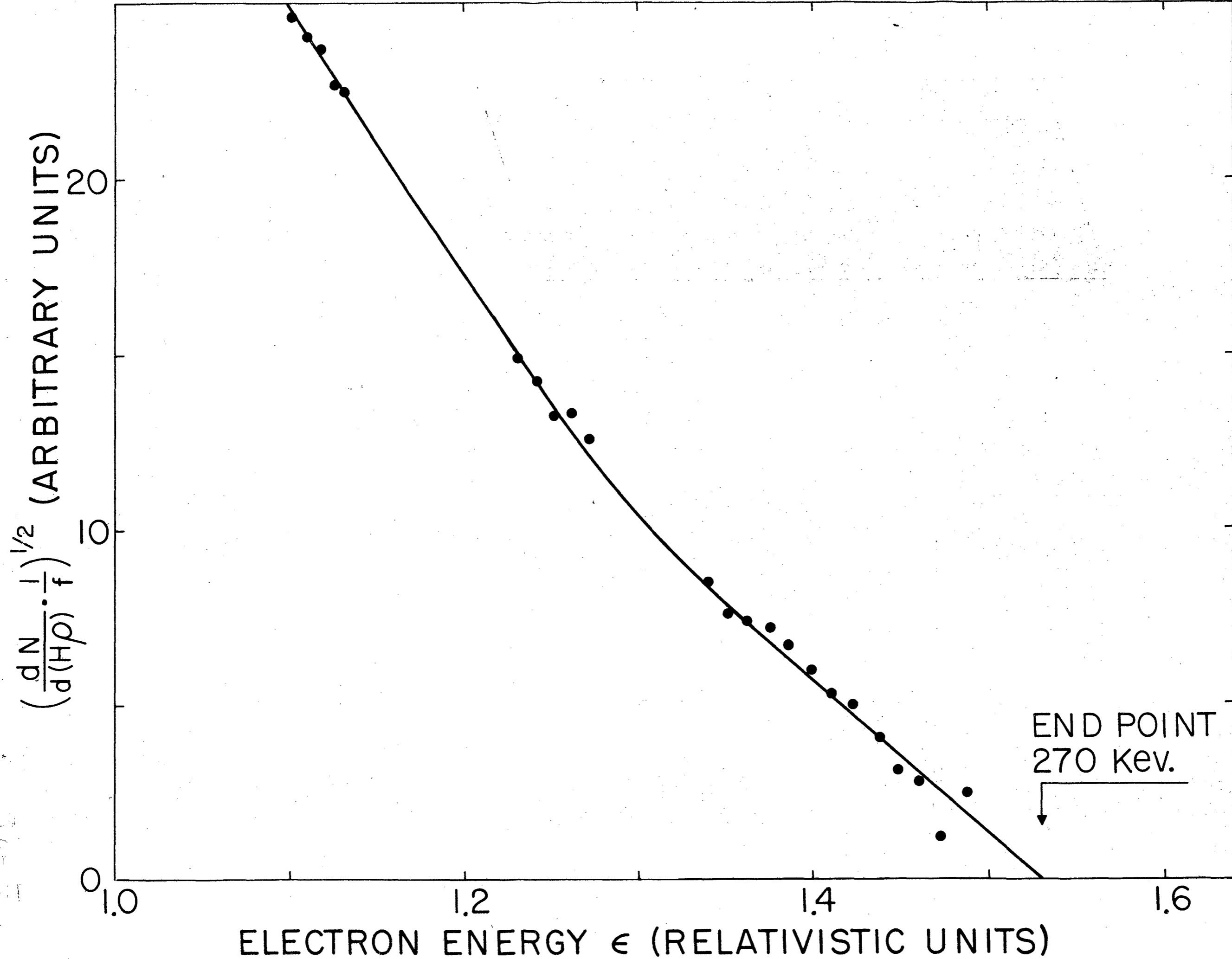
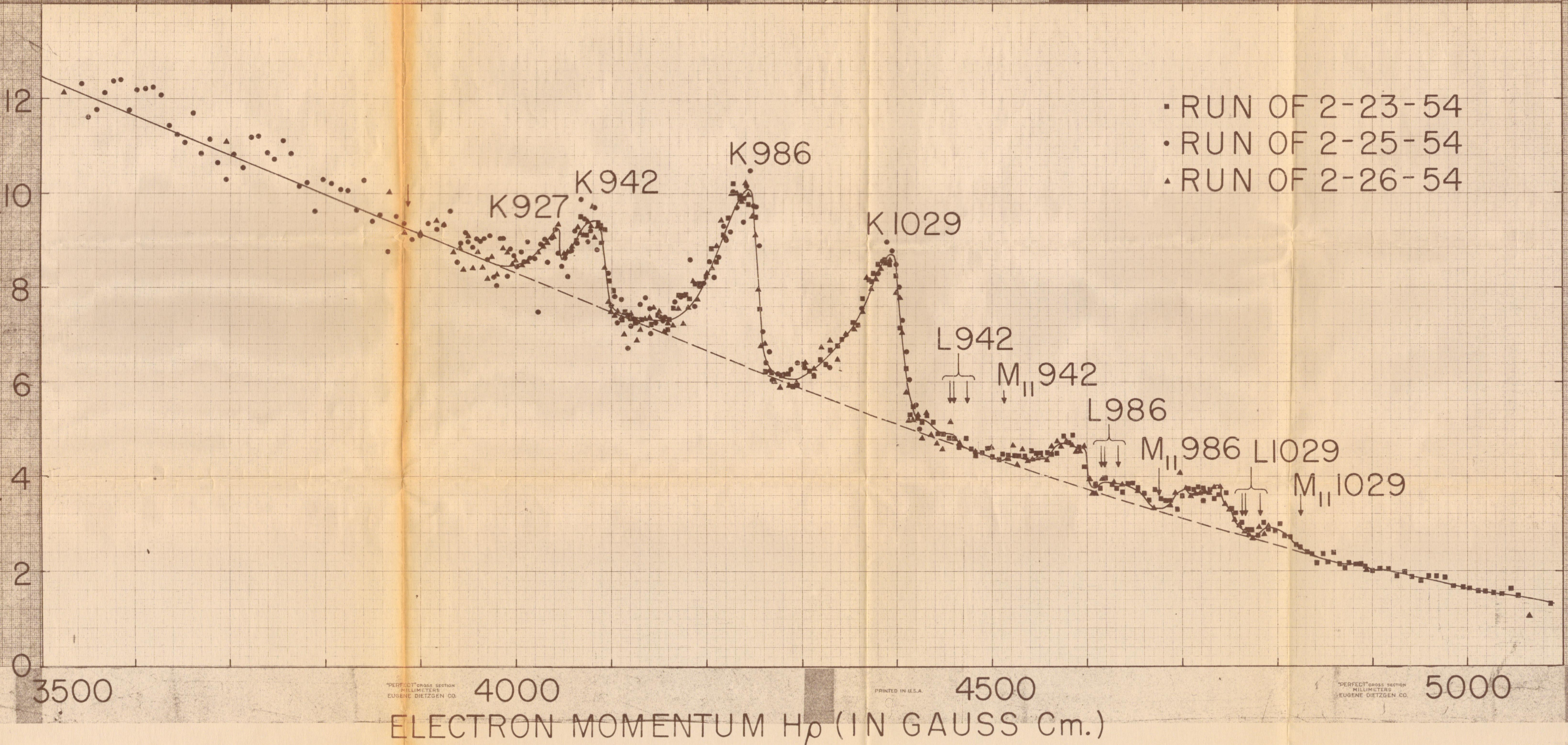


Fig. 1
2/12/54
Rosenbluth





COUNTING RATE PER MOMENTUM INTERVAL
(ARBITRARY UNITS)



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fig 2

COUNT RATE PER MOMENTUM INTERVAL

6

4

2

K 986

K 1029

4000

4200

4400

4600

4800

ELECTRON MOMENTUM $H\rho$ (IN GAUSS cm)

MU-8836

23031-3 Fig 5

