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Slow Neutron Velocity Spectrometer Transmission Studies of Pu

by

W. W. Havens, Jr., E. Melkonian, L. J. Rainwater, M. Levin

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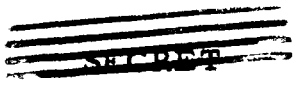

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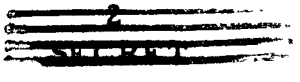

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SLOW NEUTRON VELOCITY SPECTROMETER TRANSMISSION STUDIES OF Pu

W. W. Havens, Jr., E. Melkonian, L. J. Rainwater, M. Levin

Introduction

The slow neutron transmission of several samples of plutonium is being investigated with the Columbia Neutron Velocity Spectrometer. The spectrometer, which has been described previously (Rainwater et al., Phys. Rev. 71, 65 (1947)), was recently improved, recalibrated, and the operational factor reevaluated (CUD-59). Results reported here were obtained by the procedure described in that report.

Results

The results of transmission measurements on a sample containing 0.479 gm/cm^2 of Pu using broad resolution are shown in Fig. 1. The large dip in transmission with a minimum near 133 microsec/m is caused by the strong resonance in Pu^{239} at 0.298 ev which had been previously observed (LA-266).

In order to obtain more information about this level, the sample was reinvestigated with several different resolution widths. The results of these measurements in the vicinity of this level are shown in Fig. 2a and 2b. The resolution function of the apparatus is somewhat uncertain

in this energy region, as the neutron intensity is composed of two parts: (a) those neutrons which are in the cascade part of the spectrum and have had few collisions and whose slowing down time is therefore small compared to the resolution width; (b) those neutrons which are part of the Maxwellian distribution. This distribution is in approximate equilibrium with the thermal motion of the H atoms in the source slab and the neutrons coming from this distribution have had many collisions. For the source used these neutrons are emitted on the average 30 microsec after the fast neutrons are incident on the source. Because of this effect the resolution function is neither triangular nor constant over this energy level, but is some complicated function of the neutron intensity distribution in this region. However the resolution function has very little effect on the transmission when the curvature of the transmission curve is small, and therefore the measured cross section is correct only at some distance from the resonance.

In the region from 30-82 microsec/meter the series of measurements shown in the following table were made:

Sample (gm/cm^2)	Region (microsec/meter)	Sum of arc and detector "on times" (microsec/m) (approximate resolution)
16.6085	30-73	6
7.039	30-73	3
0.479	39-81	6
0.479	63-80	2

The results, given in Fig. 3, show a strong dip with a minimum at about 70 microsec/m, which had not been observed in previous investigations.

In the higher energy region, samples containing 16.6085 gm/cm^2 and 7.039 gm/cm^2 were investigated with the results shown in Fig. 4. Neutron resonance levels are present at 7.5 ev, 10.9 ev, 14.3 ev, 17.4 ev, 22 ev, and 25.5 ev. There are other pronounced dips in transmission which have minima at 57, 84, 230, and 500 ev, which are probably caused by more than one resonance level, since for these dips the resolution width is well above the average spacing of the levels.

Analysis of the Data

These data may be most conveniently studied in two groups, those covering the energy region from 0 to about 6 ev, and those covering the region above 6 ev. Below 6 ev the resolution is relatively good, and a detailed study of the cross section variation is attempted. Above 6 ev, one must be content with locating levels and obtaining a rough idea of their strengths.

Energy Region 0 - 6 ev

This energy region contains a number of overlapping effects, making direct analysis of each effect difficult. A method of successive approximations was therefore used. Figure 5 shows the data plotted in the form $\sigma\sqrt{E}$ vs. E. The following terms have been found necessary

to explain the observed cross section variation in this region:

a) Scattering cross section. It is difficult to determine this accurately, but it seems reasonable to assume that it is about $4\pi r^2 \approx 10$ barns. A value of 9 barns resulted from the method of successive approximations to obtain a best fit.

b) A "1/v" term. This probably arises from the numerous levels at higher energies. A best match was obtained setting this term equal to 25 barns at 1 ev, i.e., $\sigma = 25/\sqrt{E}$.

c) A level at 1.06 ev. This level was not observed in the earlier measurements by Anderson et al., and must be due to an isotope which was not present in their samples, for, with the resolution used by them and the strength of this level, it could not have been missed. This resonance was finally attributed to absorption in Pu^{240}. Since Anderson et al. had used early samples of Pu which came from rods which were irradiated for a relatively short period of time, the Pu^{240} isotope could not have been built up to a very high concentration. However, since the samples used in these investigations came from fuel which had been irradiated for a much longer period of time, the Pu^{240} was present to a much greater extent. The analysis of the entire 0 - 6 ev region immediately under consideration does not distinguish between the various isotopes involved, so that the results will be given as though

the level were in Pu^{239} (as in Fig. 5). Later, in the analysis of this level itself, the actual isotopic constitution will be taken into account.

Since this level has not been completely resolved, the value of $\sigma_o \Gamma^2$ can best be obtained from a study of the region slightly off resonance where the resolution used is adequate. The value of Γ can then be determined by the usual type of fit at the level itself. The level parameters determined from studying the data immediately off resonance are:

$$E_o = 1.06 \text{ ev}$$

$$\sigma_o \Gamma^2 = 4.28 \text{ barns ev}^2 \text{ (gives } \sigma_o \Gamma^2 = 292 \text{ for } Pu^{240})$$

d) A level at 0.298 ev. This level is at an energy where high resolution work is readily performed. However, the peak cross section is so large that the thinnest sample available for the present work (0.479 gm/cm^2) was too thick to give a transmission significantly above zero to give a reliable measurement of the peak cross section. The level parameters were determined by a study of the sides of the level where the transmission of the sample was satisfactory. This method is subject to some uncertainty because of the next effect to be discussed. The best values of the level parameters appear to be:

$$E_o = 0.298 \text{ ev}$$

$$\sigma_o \Gamma^2 = 51.4 \text{ barns (ev)}^2$$

$$\Gamma = 0.098 \text{ ev}$$

from which

$$\sigma_o = 5360 \text{ barns.}$$

The Los Alamos group had adequately thin samples to work with, and hence could measure σ_o and Γ directly. Their values were:

$$\sigma_o = 5600 \pm 250 \text{ barns}$$

$$\Gamma = 0.097 \text{ ev}$$

which agree with the present results.

e) A level at negative energy. Both the present data and the Los Alamos data show that the four terms described above do not fit the data below 0.2 ev. The discrepancy is of the nature of excess cross section which increases with decreasing energy. The assumption of a level at negative energy appears satisfactory to explain the apparent discrepancy. The required level parameters are:

$$E_o = 0.23 \text{ ev}$$

$$\sigma_o \Gamma^2 = 27.5$$

Γ = completely undetermined.

All of the above terms grouped together give the following equation for the cross section in the 0 - 6 ev region.

$$\sigma\sqrt{E} = \underset{(a)}{9\sqrt{E}} + \underset{(b)}{25} + \underset{(c)}{\frac{1.10}{0.0005 + (E-1.06)^2}} + \underset{(d)}{\frac{7.0}{0.0024 + (E-0.298)^2}} + \underset{(e)}{\frac{3.3}{(E+0.23)^2}}$$

The solid line in Fig. 5 is the above equation and demonstrates the extent of fit to the data. The agreement between the solid curve and the experimental results is poor in the region of the 1.06 ev level because of the finite resolution of the apparatus.

The detailed methods used for analyzing the observed cross section to give terms stated above are as follows. In studying a portion of the energy interval considered, it is possible to determine conveniently only two parameters at a time. However, it is fortunately possible to choose intervals where most of the effects, except the ones considered, have small contributions which may be estimated from only approximate knowledge of their characteristics.

The energy interval 1.2 - 2 ev is useful for determining the $1/\sqrt{E}$ term and $\sigma_0\Gamma^2$ for the 1.06 ev level, since the constant term is relatively small and varies slowly (used as $\sigma_{scatt}\sqrt{E}$), and terms (d) and (e) contribute very little. Figure 6 shows a plot of

$$\left[(\sigma - 9)\sqrt{E} - \frac{7.0}{(E - 0.298)^2} - \frac{3.3}{(E + 0.23)^2}\right] \text{ vs. } \frac{1}{(E - 1.06)^2}.$$

If the present interpretation is correct, the data when plotted in this way should give a straight line with the slope giving $1/4\ \sigma_0\Gamma^2\sqrt{1.06}$ for the 1.06 ev level, and the intercept giving the coefficient of the $1/\sqrt{E}$ term. The plot shows a reasonably good straight line, and yields the

the parameters given above.

In the energy interval 2 - 6 ev, the effect of the 1.06 ev level is small, while the constant term shows greater variation (as $\sigma_{scatt}\sqrt{E}$). A plot of

$$\sigma\sqrt{E} - \frac{1.10}{(E - 1.06)^2} - \frac{7.0}{(E - 0.298)^2} - \frac{3.3}{(E + 0.23)^2} \quad \text{vs.} \quad \sqrt{E}$$

gives the scattering term for the value of the slope, and at the same time checks the consistency of the $1/\sqrt{E}$ from the intercept. This is shown in Fig. 7.

Determination of the level parameters for the 0.298 ev level is somewhat difficult because of the uncertain contribution of the negative level, and because some of the data appear unreliable, particularly on the high energy side of the level where there is as much as 20% disagreement with the Los Alamos data. If a thin enough sample had been available, it would have been simplest to obtain σ_o and Γ directly. If a Breit-Wigner formula were strictly obeyed, a plot of $1/\sigma\sqrt{E}$ vs. $(E - E_o)^2$ would give a unique straight line with the slope giving $\left[1/4\,\sigma_o\Gamma^2\sqrt{E_o}\right]^{-1}$, and the intercept giving $\left[\sigma_o\sqrt{E_o}\right]^{-1}$. To determine the deviation from a Breit-Wigner formula, a plot of

$$\frac{1}{\left[(\sigma - 9)\sqrt{E} - 25 - \dfrac{1.10}{(E - 1.06)^2}\right]} \quad \text{vs.} \quad (E - 0.298)^2$$

was made (Fig. 8). The deviation from a straight line is marked and is the basis for the assumption of a negative level. The straight line finally chosen was the one that gave the best Breit-Wigner fit to the residual cross section on the assumption of a negative level. This residual cross section

$$\sigma' \sqrt{E} \equiv (\sigma - 9)\sqrt{E} - 25 - \frac{1.10}{(E - 1.06)^2} - \frac{7.0}{0.0024 + (E - 0.298)^2}$$

plotted in the form $\frac{1}{\sqrt{\sigma' \sqrt{E}}}$ vs. E is shown in Fig. 9. The results show an approximate straight line as expected, with the slope giving $(1/4\ \sigma_o \Gamma^2 \sqrt{E_o})^{-1/2}$, and the intercept giving $E_o (1/4\ \sigma_o \Gamma^2 \sqrt{E_o})^{-1/2}$. Combining these gives E_o and $\sigma_o \Gamma^2$.

The 1.06 ev level has thus far been studied only by its effects in the region off resonance. A study of the level itself can now give a value for Γ as well as confirm the value of $\sigma_o \Gamma^2$.

In order to investigate this level more thoroughly, the transmission of the thinnest sample was measured in the vicinity of the level (Fig. 3) and attempts were made to obtain the Breit-Wigner parameters for this level by fitting theoretical curves to the experimental results, calculated in approximately the same manner as that previously described (Havens and Rainwater, Phys. Rev. 70, 154 (1946)).

The analysis used here is more complicated than that previously

described because the resonance levels are so close together that the "background cross section", which can usually be considered constant over the resonance level, varies with energy; and in order to determine the Breit-Wigner parameters this variation in background cross section must be taken into consideration.

The background cross section was taken as:

$$\sigma = 9 + \frac{25}{\sqrt{E}} + \frac{7.0}{\sqrt{E}(E - 0.298)^2} + \frac{3.3}{\sqrt{E}(E + 0.23)^2} \text{ barns.}$$

The transmission due to this background cross section was calculated from the formula $T = e^{-n\sigma}$ assuming that all the effective background was caused by the Pu^{239}. The experimental transmission was divided by this background transmission, and the residual transmission attributed to the level in Pu^{240}.

Theoretical transmission curves were calculated by integrating Breit-Wigner cross section curves over the resolution function of the apparatus.

There are three parameters which affect the fit of the theoretical curve to the experimental results. These are the resolution width of the apparatus, the value of Γ chosen, and the value of $\sigma_o \Gamma^2$.

The effect of the variation of the resolution width is shown in Fig. 10, where three curves are presented in which $\sigma_o \Gamma^2$ and Γ re-

mained constant and the base of the triangular resolution function was changed from 2 to 3.3 microsec/m. The change in the curve is not large, except at the point of minimum transmission. Since the resolution width is known to better than 0.3 microsec/m, the uncertainty due to this cause can be almost neglected, compared with the other uncertainties involved.

The effect of varying $\sigma_o\Gamma^2$ and Γ must be investigated simultaneously, for as one increases both $\sigma_o\Gamma^2$ and Γ the transmission at any point can be kept constant but the curve will broaden out. The variation in both $\sigma_o\Gamma^2$ and Γ are shown in Fig. 11. The solid curve is on the average wider than one would expect from the experimental data and the dashed curve is narrower than one would expect from the experimental data.

The values of the parameters for the solid curve are:
E_o = 1.06 ev; Γ = 0.065 ev; $\sigma_o\Gamma^2$ = 458, which gives σ_o = 110,000 barns; and for the dashed curve we have E_o = 1.06 ev; Γ = 0.030 ev; $\sigma_o\Gamma^2$ = 254, which gives σ_o = 280,000 barns.

Taking all known errors into consideration, the final results for this level can be given as:

$$E_o = 1.06 \pm 0.01 \text{ ev}$$
$$0.03 < \Gamma < 0.065 \text{ ev} \quad (\Gamma \sim 0.042 \text{ ev})$$
$$250 < \sigma_o\Gamma^2 < 460 \text{ barns (ev)}^2 \quad (\sigma_o\Gamma^2 \sim 350 \text{ barns (ev)}^2)$$
$$60,000 < \sigma_o < 500,000 \quad (\sigma_o \sim 200,000 \text{ barns})$$

The value of $\sigma_o\Gamma^2$ obtained in this way agrees approximately with that obtained by studying the region off resonance given previously ($\sigma_o\Gamma^2 \pm 292$).

Energy Region above 6 ev

This region consists of a group of levels between 6 ev and 35 ev, which appear to be distinguishable and for which values of $\sigma_o\Gamma^2$ may be estimated, and numerous levels above 35 ev which cannot be distinguished and for which no calculations may be done.

Before analysis of the levels in the 6 to 35 ev region can be attempted, the background cross section must be determined and accounted for. Using the results from the study of the 0 to 6 ev region, this background cross section has been taken as

$$\sigma = 9 + \frac{25}{\sqrt{E}} ,$$

the remaining terms being negligible in this region. The $1/\sqrt{E}$ term appears to be due predominantly to the levels above 35 ev, so that the coefficient of this term is taken to be constant over the levels for which $\sigma_o\Gamma^2$ has been obtained.

The levels at 7.5, 10.9, 14.3, 17.4, 22, and 25.5 ev have been analyzed for $\sigma_o\Gamma^2$ in accordance with the procedure described in CUD-59, assuming that the background cross section was given by the above formula. The results are:

E_o	$\sigma_o \Gamma^2$
7.5 ev	10
10.9	30
14.4	100
17.4	10
22.0	20
25.5	20

It should be pointed out that the accuracy of $\sigma_o \Gamma^2$ for these levels is extremely poor as the closeness of the level spacing and the complexity of the results make interpretation extremely difficult. However, it was thought interesting to obtain as much information as possible from the data, although the method of analysis admittedly places great stress on both the theory and the experimental data.

It is possible to obtain better data on the resonances by using higher resolution and varying the sample thickness; however, the time involved in making these measurements increases rapidly as the resolution is improved and the sample thickness decreased. Therefore, it is questionable as to whether or not further measurements should be made for the purpose of better resolving higher energy levels.

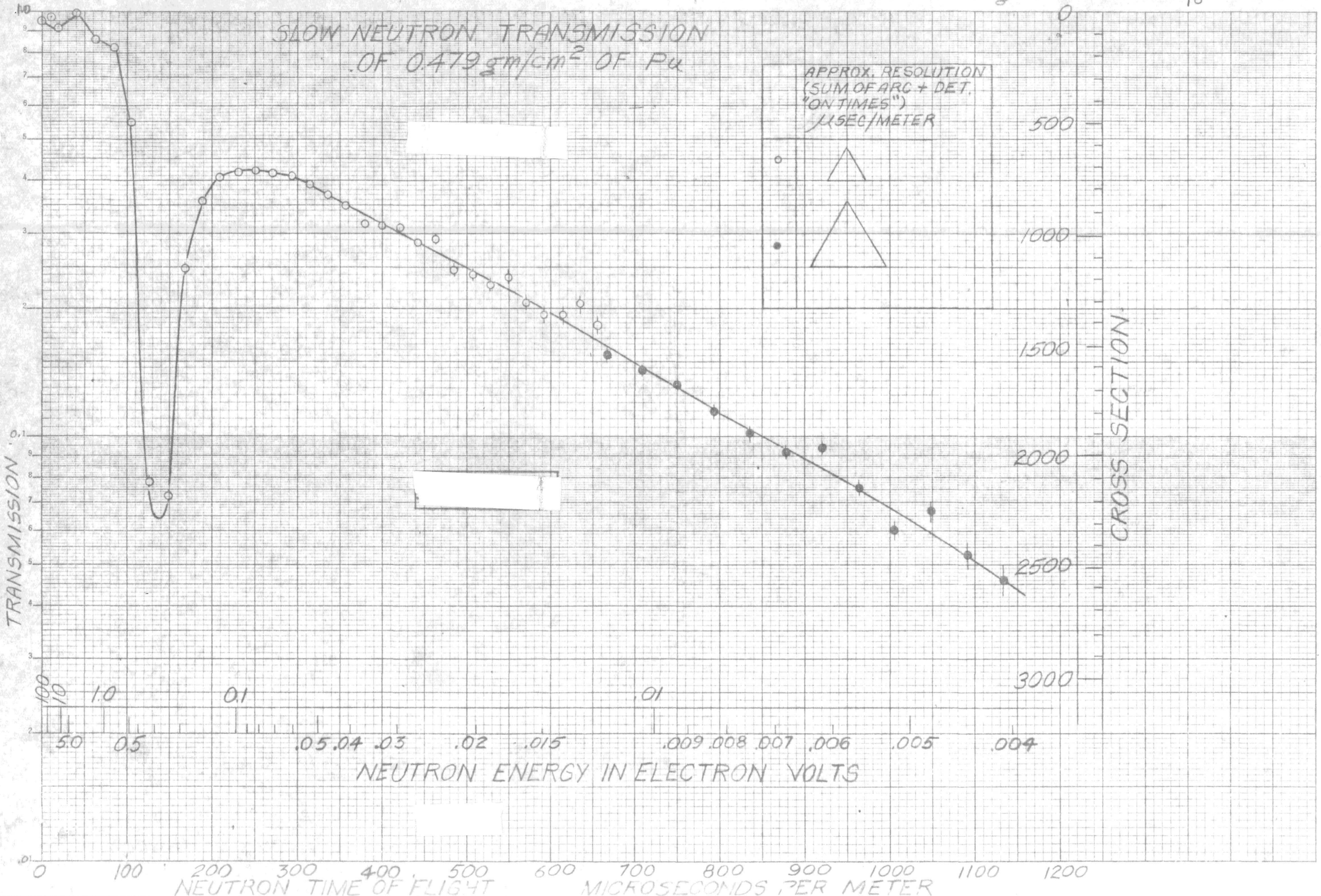
In conclusion, we do not believe that the results presented in this report are as clean cut as can be obtained. In particular, it is necessary to obtain better data between the 0.298 ev level and the 1.06 ev level, using a thicker sample, and to study the region of exact reso-

nance of the 0.298 ev level with a much thinner sample. Also, it would be desirable to study the transmission of other samples containing different amounts of Pu^{240} and Pu^{241} in order to determine the effects of these other isotopes. Further measurements are in progress, and when other samples already requested are delivered further information can be obtained about this problem.

Acknowledgments

We wish to thank Mrs. Janet Bendt, who assisted in numerous calculations involved in the paper, and the members of the cyclotron staff, who aided with the measurements.

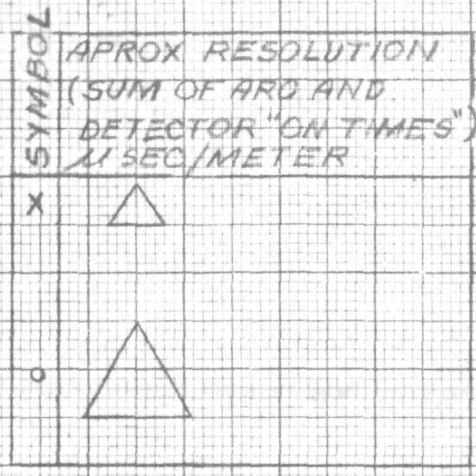
Fig. 1


SLOW NEUTRON TRANSMISSION
OF 0.479 gm/cm² OF Pu
APPROX. RESOLUTION
(SUM OF ARC + DET.
"ON TIMES")
μSEC/METER
TRANSMISSION
CROSS SECTION
NEUTRON ENERGY IN ELECTRON VOLTS
NEUTRON TIME OF FLIGHT
MICROSECONDS PER METER
100
10
1.0
0.1
.01
5.0
0.5
.05
.04
.03
.02
.015
.009
.008
.007
.006
.005
.004
0
500
1000
1500
2000
2500
3000
0
100
200
300
400
500
600
700
800
900
1000
1100
1200

19

Fig. 2a

SLOW NEUTRON TRANSMISSION OF 0.479 gm/cm² OF Pu



TRANSMISSION

1.0 0.9 0.8 0.7 0.6 0.5 0.4 0.3 0.2 0.1 0

0.8 0.7 0.6 0.5 0.4 0.3 0.25 0.20 0.15 0.13 0.12

NEUTRON ENERGY IN ELECTRON VOLTS

0.298 e.v.

CROSS SECTION

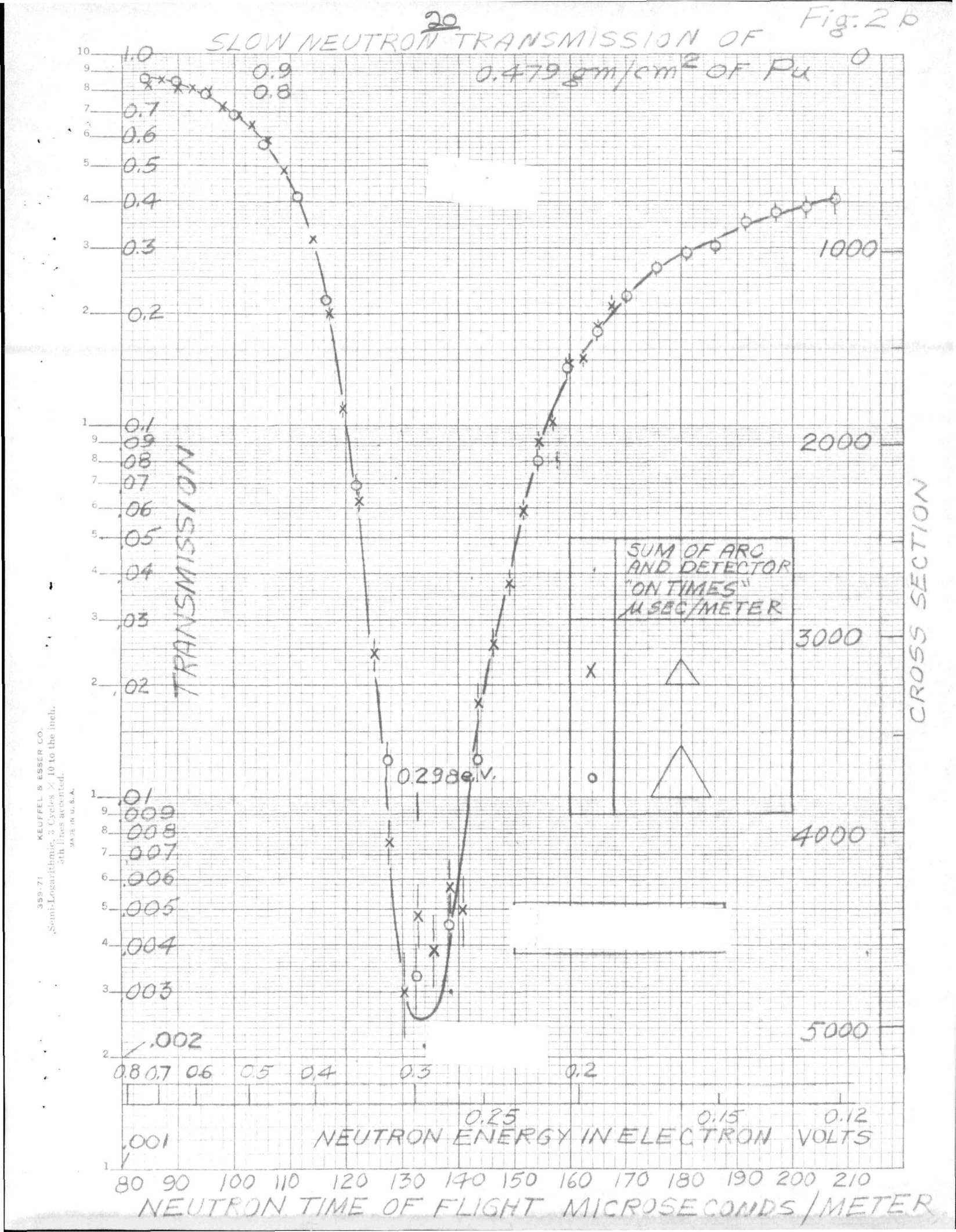
500 1,000 1500 2,000 2500 3000 3500

80 90 100 110 112 130 140 150 160 170 180 190 200 210

NEUTRON TIME OF FLIGHT MICROSECONDS/METER

359-12 KEUFFEL & ESSER CO.
10 × 10 to the half inch, 5th lines accented.
MADE IN U. S. A.

Fig: 2 b



359-71 KEUFFEL & ESSER CO.
Semi-Logarithmic, 3 Cycles X 10 to the inch.
5th lines accented.
MADE IN U.S.A.

SLOW NEUTRON TRANSMISSION OF Pu

APPROX RESOLUTION (SUM OF ARC DETECTOR AND DETECTOR "ON TIMES") μ SEC/METER	SAMPLE g/cm²	SYMBOL
△	0.479	○
△	7.039	□
△	0.479	×
△	16.608	●

TRANSMISSION

1.0 0.9 0.8 0.7 0.6 0.5 0.4 0.3 0.2 0.1

5.0 4.0 3.0 2.0 1.5 1.4 1.3 1.2 1.1 1.0 0.9 0.8

NEUTRON ENERGY IN ELECTRON VOLTS

30 32 34 36 38 40 42 44 46 48 50 52 54 56 58 60 62 64 66 68 70 72 74 76 78 80 82

NEUTRON TIME OF FLIGHT MICROSECONDS PER METER

1.06 e.v.

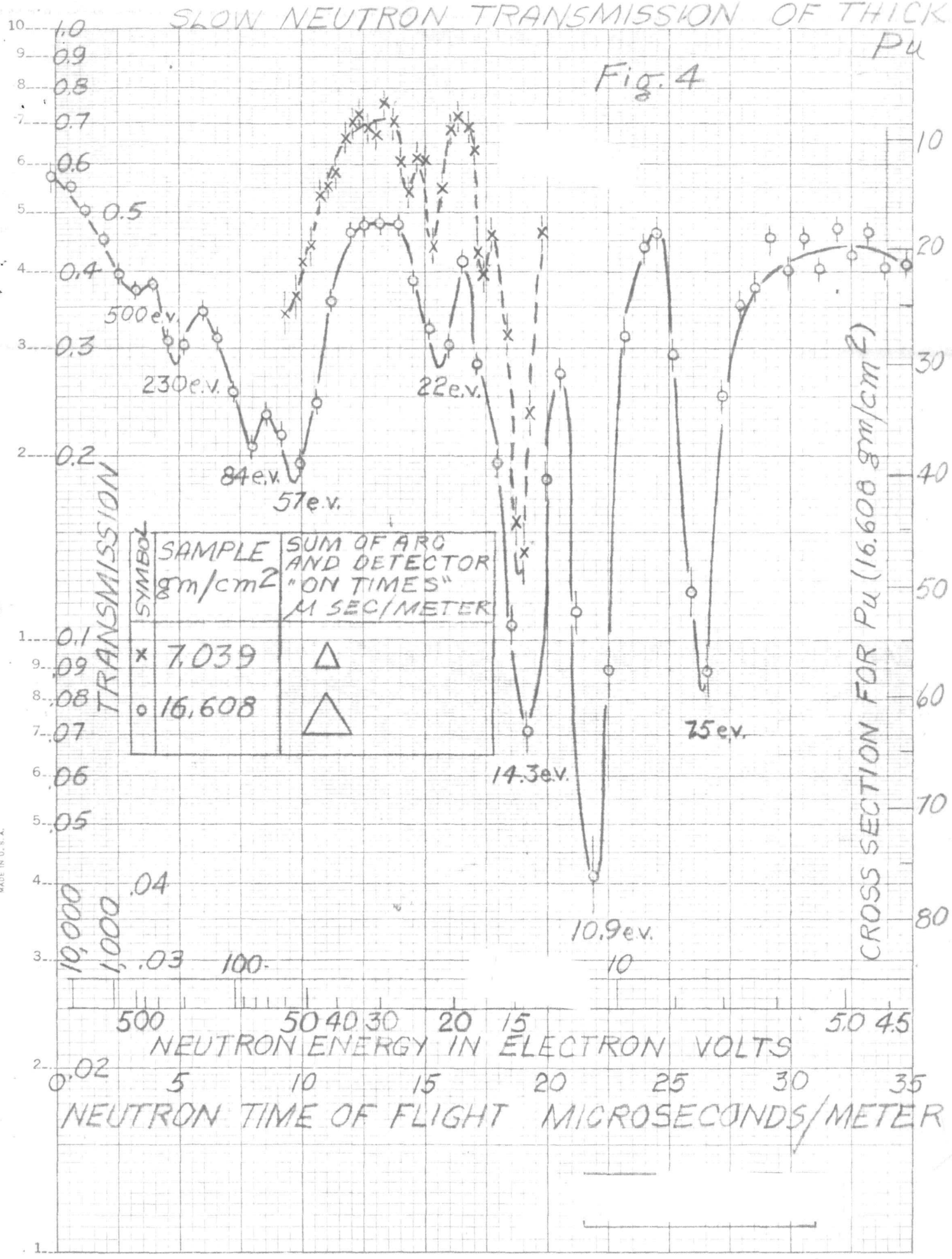
0 10,000 20,000 30,000 40,000 50,000 100,000 110,000 120,000

CROSS SECTION FOR 240 IN. 0.479 g/cm² SAMPLE (MEAN BACKGROUND CORRECTION USED)

359-11L KEUFFEL & ESSER CO.
10 × 10 to the ½ inch, 5th lines accented.
MADE IN U. S. A.

SLOW NEUTRON TRANSMISSION OF THICK Pu

Fig. 4



359-61 KEUFFEL & ESSER CO.
Semi-Logarithmic, 2 Cycles × 10 to the inch.
5th lines accented.
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$\sigma\sqrt{E}$ vs. E; $0 < E < 6$ e.v.

—— $\sigma\sqrt{E} = 25 + 9\sqrt{E} + \frac{3.3}{(E+0.23)^2} + \frac{7.0}{0.0024+(E-0.298)^2} + \frac{1.10}{0.0005+(E-1.06)^2}$

$\sigma\sqrt{E}$ (barns/ev)

10,000 9 8 7 6 5 4 3 2 1000 9 8 7 6 5 4 3 2 100 9 8 7 6 5 4 3 2

0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0 1.1 1.2 1.3 1.4 1.5

1 2 3 E (e.v.) 4 5 6

0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0 1.1 1.2 1.3 1.4 1.5

E (e.v.)

○ 0.479 gm/cm² SAMPLE

× 7.039 " "

● 16.61 " "

KEUFFEL & ESSER CO., N. Y. NO. 359-73L
Semi-Logarithmic, 3 Cycles × 10 to the inch
MADE IN U. S. A.

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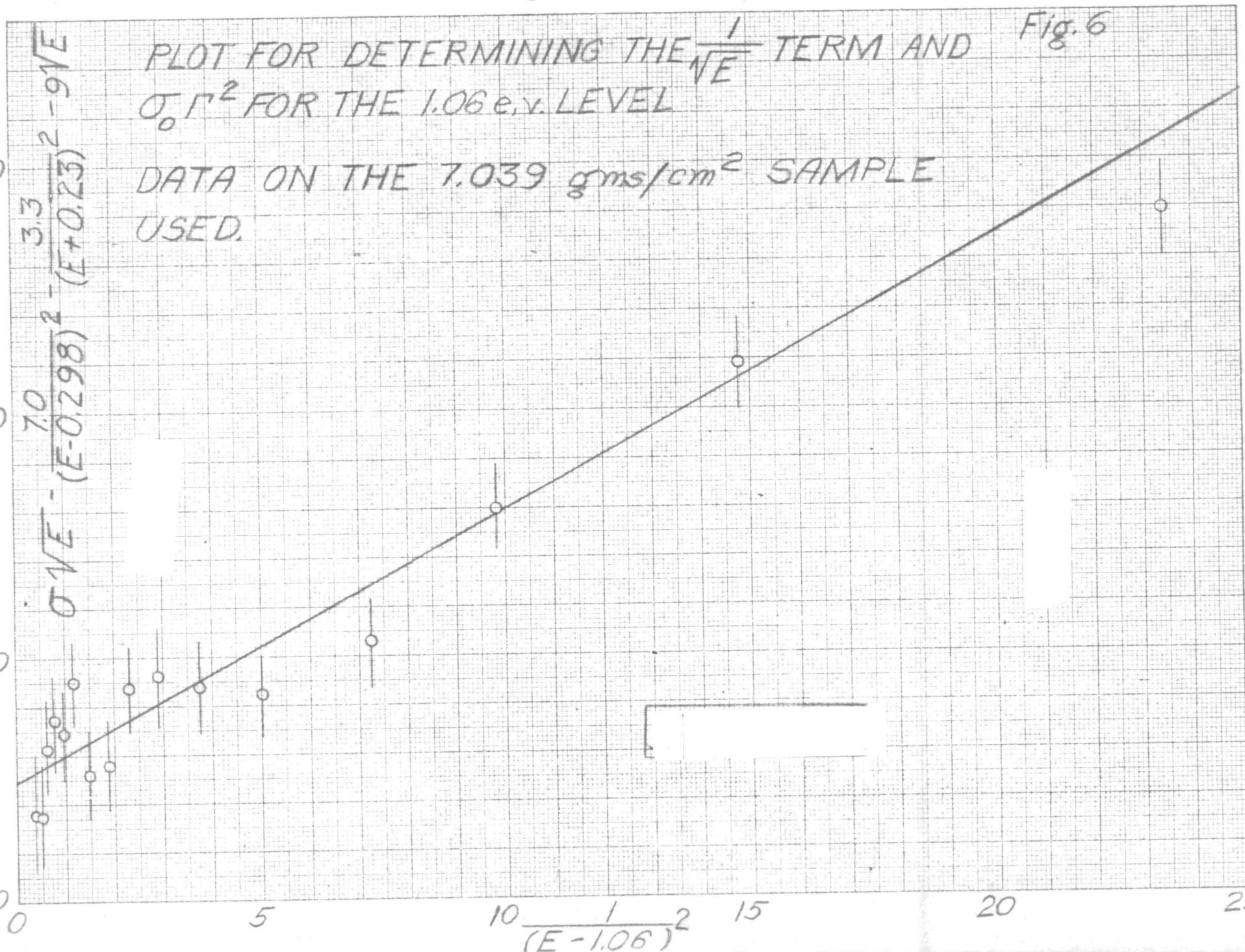
Millimeters, 10th lines heavy

Fig. 6

PLOT FOR DETERMINING THE $\frac{1}{\sqrt{E}}$ TERM AND $\sigma_0 \Gamma^2$ FOR THE 1.06 e.v. LEVEL

DATA ON THE 7.039 gms/cm^2 SAMPLE USED.



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Millimeters, 10th lines heavy
DATA SHEETS

Fig. 7

PLOT FOR DETERMINING THE CONSTANT AND THE $\frac{1}{\sqrt{E}}$ TERMS

DATA ON 16.61 gms/cm^2 SAMPLE USED

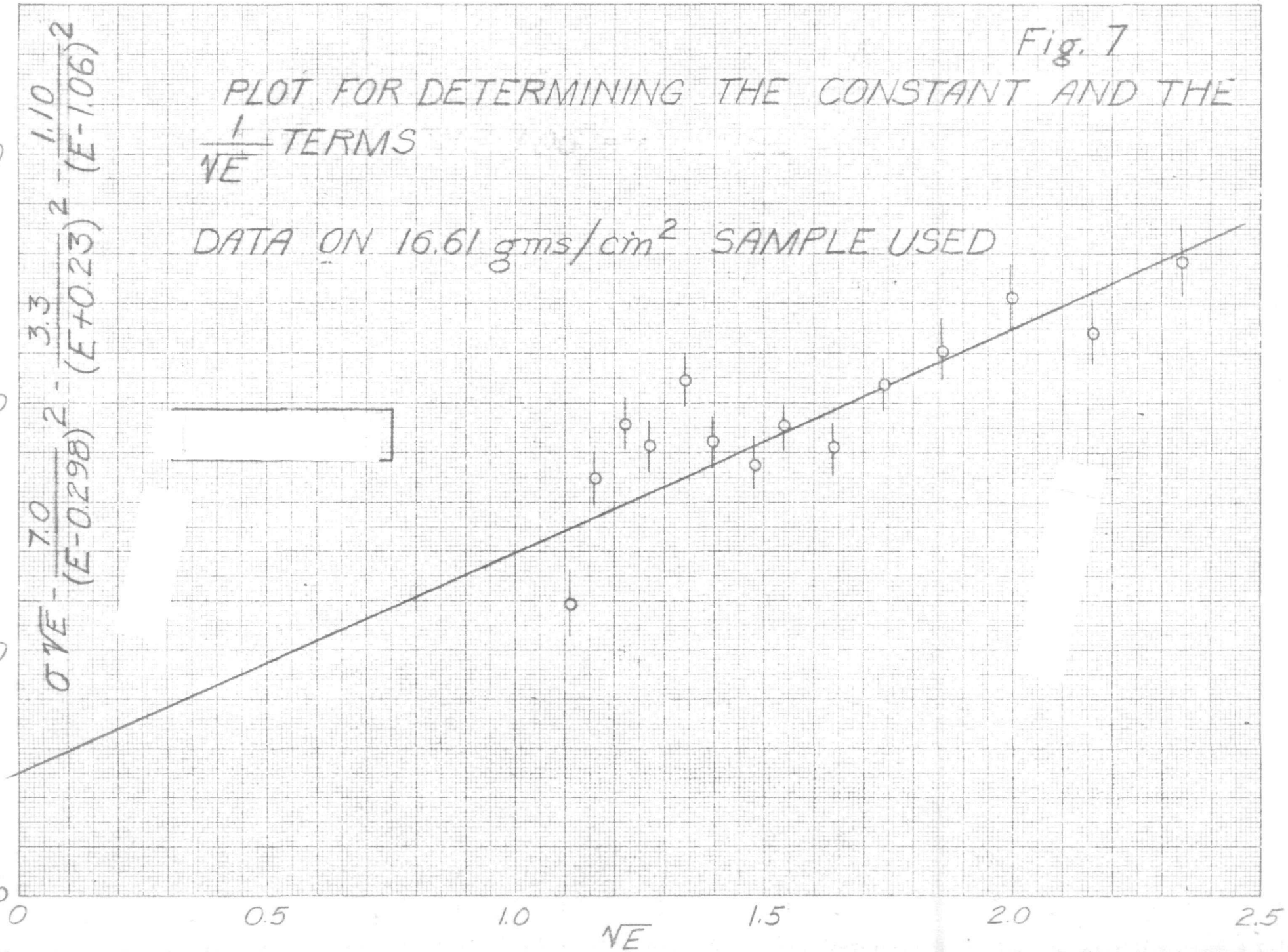
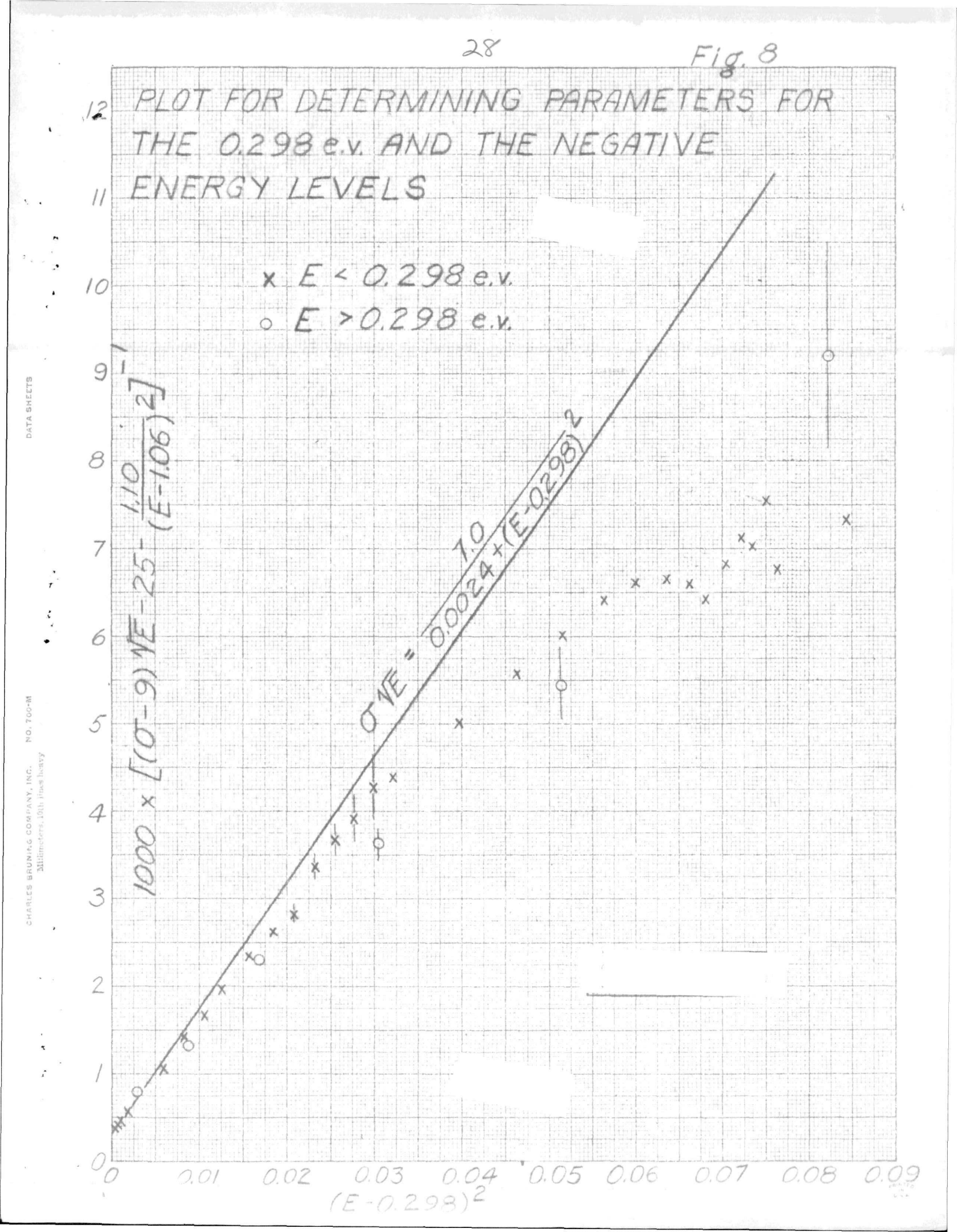


Fig. 8



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PLOT FOR DETERMINING THE PARAMETERS OF THE NEGATIVE LEVEL

Fig. 9

$$\left[(\sigma-9)\sqrt{E}-.25-\frac{1.10}{(E-1.06)^2}\right]^{-\frac{1}{2}}=\frac{7.0}{-0.0024+(E-.298)^2}$$

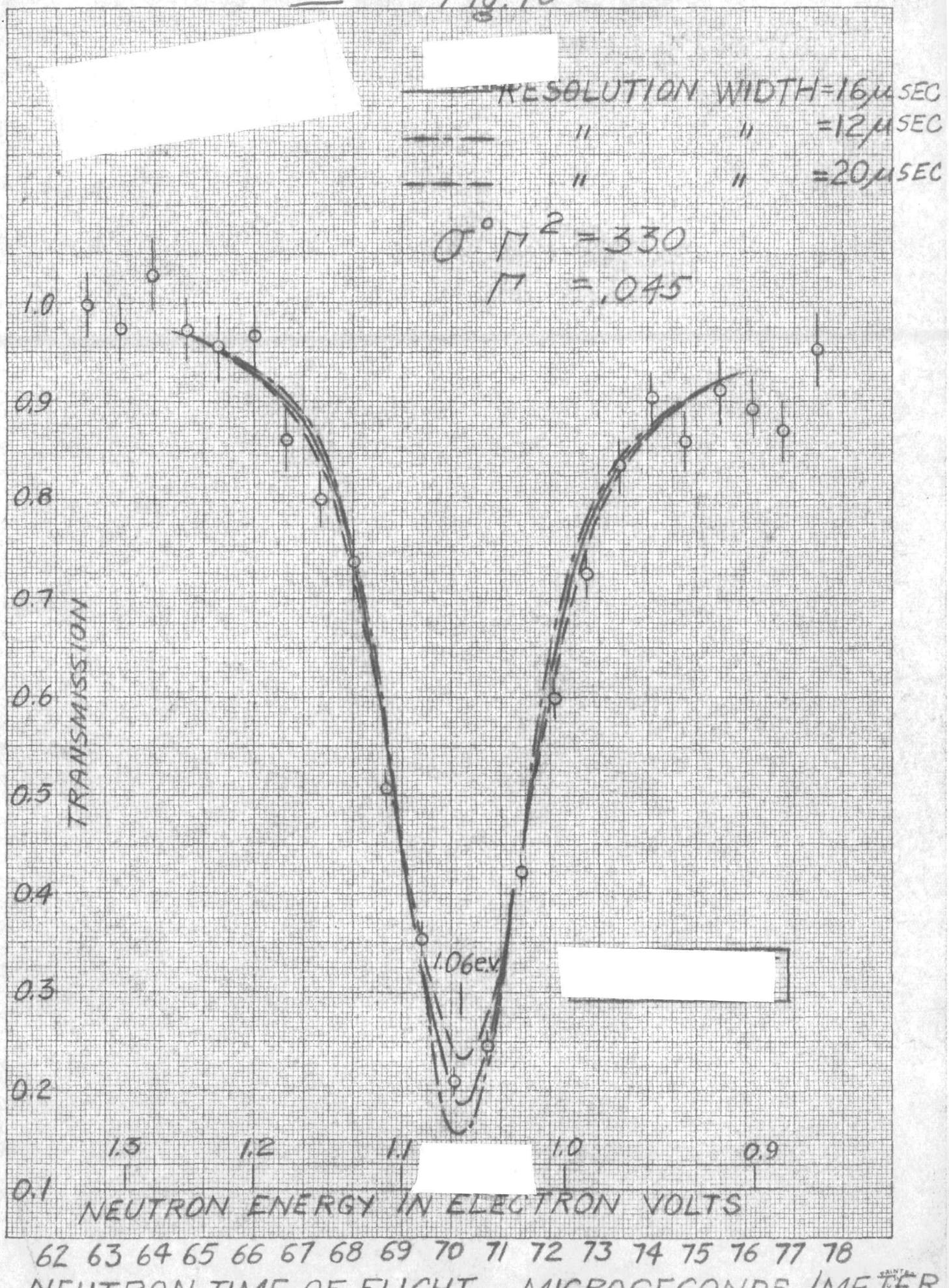
$$\sigma\sqrt{E}=\frac{3.3}{(E+0.23)^2}$$

.3
.2
.1
0

0 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12

E (e.v.)

Fig. 10



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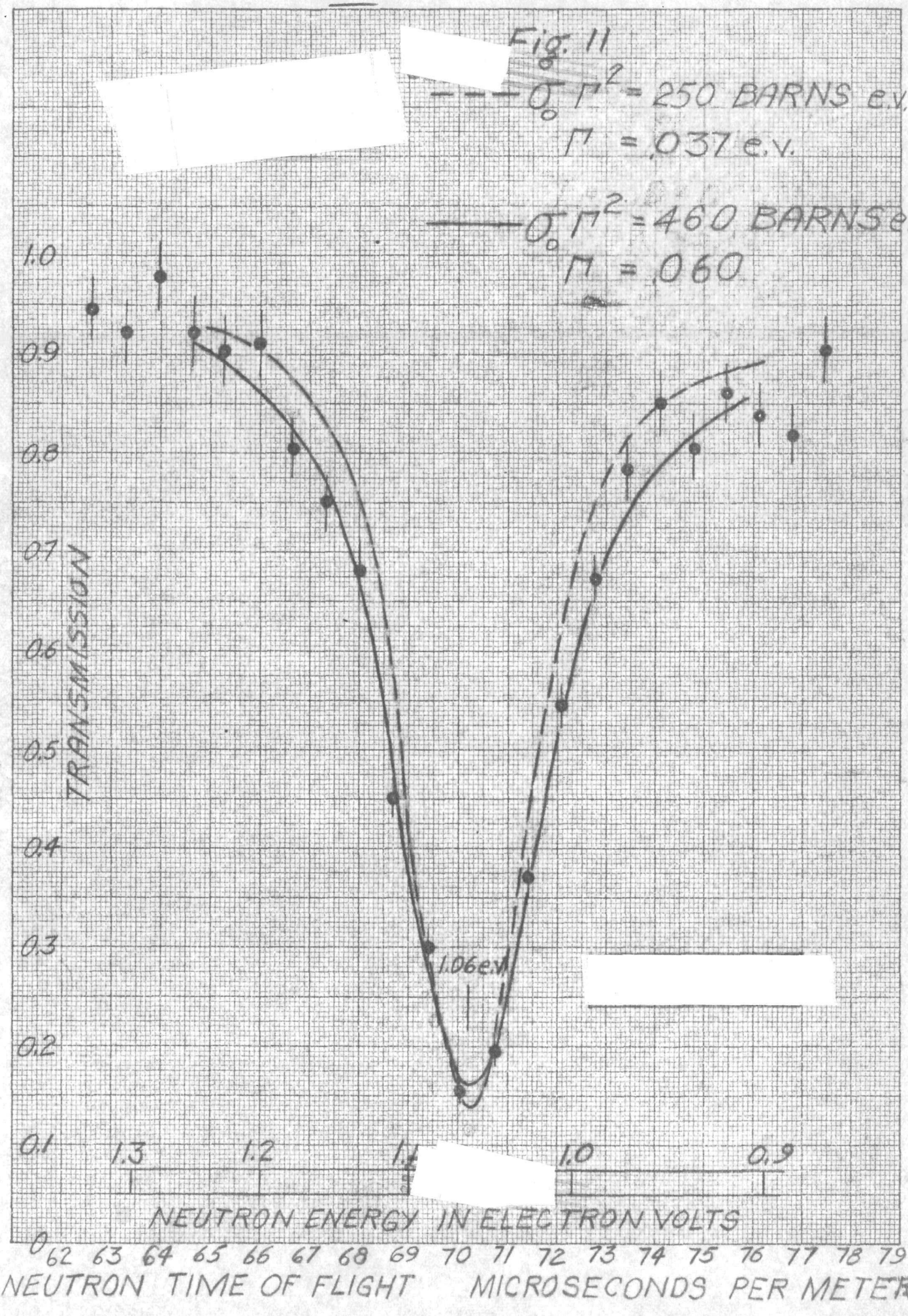
Millimeters, 10th lines heavy



DATA SHEETS
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Millimeters, 10th lines heavy