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NEUTRON CROSS SECTIONS OF U^{235} AND Pu^{239} AT LOW ENERGIES

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ABSTRACT

The ratio of capture to fission in U^{235} and Pu^{239} at low energies is discussed. It is probable that the capture to fission ratio is zero in Pu^{239} for energies of more than 1 ev. This fact could be established by a number of experiments which are suggested in this paper.

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I. Capture-to-Fission Ratio in U^{235} and Pu^{239}

The ratio α of the capture cross section and the fission cross section at low energies is of great significance for the construction of piles. If an energy region could be found in which this ratio is zero, piles could be made to operate in this region and avoid neutron losses.

The following cross sections are relevant in the low-energy region: The fission cross section σ_f,

The (elastic) scattering cross section σ_s,

The radiative capture cross section σ_r,

The total cross section $\sigma_t = \sigma_f + \sigma_s + \sigma_r$.

The scattering at the energies considered here can only be elastic and isotropic, since the wavelength of the neutron is large compared to the dimensions of the nucleus. The capture process is followed by the emission of γ-rays and leads to a radioactive nucleus.

The only cross sections that have been measured so far as a function of the neutron energy are the fission cross section and the total cross section σ_t. Other processes can not be measured easily with the velocity selector method. The fission cross section is measured by determining the amount of fission processes per incident neutron; the total cross section is measured by determining the attenuation of a neutron beam. It is in the nature of the velocity-selector method that cross sections can only be measured of processes where an observable effect follows immediately the capture of a neutron. This observable effect is the

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attenuation of the beam in the case of the measurement of the total cross section and the fission in the case of the fission cross section. The scattering cross section could be measured by the velocity-selector method if the scattered neutrons could be picked up by a detector which has the required "on" and "off" time periods to detect the scattering of neutrons with a given energy. It has not been done so far, probably because of intensity difficulties which make it hard to distinguish this scattering from stray scattering in other objects. The capture cross section can be measured by velocity selector only if the capture γ-rays could be detected which seems questionable.

All three cross sections can be measured with suitable methods, however, if monochromatic neutrons are produced with a crystal spectrometer.

Elementary laws of neutron absorption show that σ_f and σ_r contain a factor $1/\sqrt{E}$ at low energies, whereas σ_s approaches a finite value for $E \to 0$. The finite value of σ_s for low energies is of the order of $4 \pi R^2$ where R is the radius of the nuclei. This is about 10 b for heavy nuclei and we can therefore neglect σ_s for energies at which the other cross sections are large compared to 10 b. This is the case for energies below about 0.5b. We will refer to this region as the "low-energy region".

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II. The Plutonium Cross Sections

The cross sections σ_t and σ_f of plutonium have been measured by B. D. McDaniel and co-workers in LA - 266. The low-energy region is dominated by the well-known strong resonance at 0.3 ev. Neither σ_t nor σ_f can be represented, however, by a pure Breit-Wigner formula:

$$\sigma = \sqrt{\frac{E_o}{E}}\;\frac{\sigma_o}{1 + 4(E - E_o)^2/\Gamma^2}$$

The constants can be adjusted so that the Breit-Wigner expression describes well the region around the resonance within one half width. These constants are:

$$\sigma_{ot} = 5600 \pm 250 \text{ b}$$
$$\sigma_{of} = 3000 \pm 250 \text{ b}$$
$$E_o = 0.30 \text{ ev}, \quad \Gamma = 0.097 \text{ ev}.$$

E_o and Γ are identical for both cross sections as expected by theory. These Breit-Wigner expressions do not fit outside one half width. A background cross section σ_B must be added to it and McDaniel has shown that this background cross section σ_B can be considered the same within the experimental accuracy for both the total and the fission cross section. The function $\sqrt{E}\;\sigma_B$ is a slowly varying function of the energy. It is plotted in Fig. 1 in units of barns · meter/μ s.

The ratio α of capture to fission:

$$\alpha = \sigma_r/\sigma_f$$

can also be determined in the low energy region from the measured cross

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sections σ_t and σ_f, by neglecting σ_s:

$$\alpha = (\sigma_t/\sigma_f) - 1.$$

McDaniel finds this magnitude varying with energy. It has the value 0.4 at thermal energies and increases to 0.85 at 0.3 ev and falls off again at higher energies as indicated in Fig. 1. Although the experimental accuracy is not too good, it seems that this variation is outside the experimental error.

The measurements in the energy region above 0.5 ev are firstly, less accurately known and, secondly, harder to interpret because of the fact that the scattering cross section can no longer be neglected. The total as well as the fission cross section show a very broad resonance at 12 ev and some more at higher energies. The resonance at 12 ev may well be a group of unresolved sharp resonances.

The value of α can only be determined if one knows the value for σ_s. The only measurement available is a thermal value of $\sigma_s = 9.8$ b by Fermi (private communication contained in the Los Alamos Handbook, LA - 140). Although it is theoretically to be expected that the scattering cross section between resonances (potential scattering) is a very slowly varying function of the energy, it is not safe to assume that Fermi's value also holds in the region above 1 ev. The influence of the resonance at 0.3 ev may be large enough to contribute appreciably to the thermal scattering cross section as the analysis in section V shows. If σ_s is chosen to be 10 b throughout, one obtains a curve for α which could be interpreted to converge to zero above 1 ev (see Fig. 1). It is therefore at present not in disagreement with experiments to assume $\alpha = 0$ in plutonium for energies at which the strong resonance has no influence.

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III. Uranium (235) Cross Sections

The measurements of the total and fission cross section in U^{235} were performed by McDaniel and co-workers in LA - 158 with more accuracy than the plutonium measurements because of the absence of the strong α-activity. U^{235} shows a great number of resonances, both in absorption and in fission. The ratio α of capture to fission could be determined directly in the low-energy region to be $\alpha = 0.17$ independent of the energy, within the accuracy of the measurement. At higher energy, the scattering cross section was assumed to be equal to the one of U^{238} which is 9.2 b. On this basis, it was found that α could be assumed constant over the complete region of observation (0-200 ev) with a few remarkable exceptions. They consist of two resonance levels at 2 ev and 4.8 ev. These levels do not occur at all in the fission cross section but only in the absorption cross section. McDaniels shows in LA - 158 that these levels really belong to U^{235}. The absorption of a neutron in these resonances leads only to capture and not at all to fission. It is not excluded that similar levels exist at higher energies too. The lack of resolutions would make the discovery impossible.

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IV. Theoretical Interpretation

In order to explain some of the features described before, especially the behavior of the capture-to-fission ratio, we propose the following assumption: In general, neutron resonance levels show approximately equal properties in respect to fission and capture. The value of the fission breadth Γ_f and the capture breadth Γ_r is a slowly varying function of the excitation energy of the compound nucleus, as Bohr and Wheeler have assumed. The ratio is expected than to be almost the same for all levels within the energy region discussed in this report (0 to 200 ev neutron energy). In contrast to this, however, we have to assume in view of the experimental evidence in U^{235} that there exists a number of "exceptional" levels whose fission width is exceptionally low.

Since α in U^{235} is normally 0.17, the normal ratio of $\Gamma_f : \Gamma_r$ in U^{235} is about 6:1, which gives rise to a total width $\Gamma = \Gamma_f + \Gamma_r$ of about $7\Gamma_r$. If Γ_r is assumed to be of the order of 0.1 ev one gets a total width of 0.7 ev which is somewhat higher than the observed widths in resolved levels of U^{235}. However, a value $\Gamma_r \sim 0.05$ is not excessively small compared to results from other elements and the subsequent value $\Gamma \sim 0.35$ ev is in line with the observations. The two exceptional levels at 2 ev and 4.8 ev must have a fission width Γ_f at least fifty times smaller in order to explain the failure of these resonances to appear in the fission cross section curve.

We now apply those assumptions to the case of plutonium. From the work of Bohr and Wheeler, one should conclude that the compound nucleus Pu^{240} created in the neutron absorption of plutonium should have a lower

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fission threshold than U^{236} created after neutron absorption in U^{235}. The fission threshold of U^{236} should be just a little under the excitation energy E_B which is delivered by slow-neutron capture in U^{235}. (E_B is the binding energy of the neutron.) This is so because of the fact that the fission width for low-lying levels is not very much larger than the radiation width. This unusually small value of the fission width indicates proximity to the fission threshold. Pu^{240} differs from U^{236} by one α-particle. Its relative proton-neutron ratio is therefore slightly larger; this should cause a larger readiness to undergo fission. We, therefore, should expect a larger fission width for equal energies.

The experimental evidence of the well known strong resonance in Pu seems to contradict this. Its fission width is smaller than the average fission width in U^{235}. The latter one is about equal to the total widths of the U^{235}-levels which is about 0.3 to 0.5 ev. The fission width of the Pu-resonance level is $1/(1 + \alpha)$-times its total width which amounts to about 0.05 ev.

We propose to explain this discrepancy by assuming that the strong resonance level in plutonium is one of its "exceptional" levels with an unusually small fission width. It is supposed to be of the same nature as the two levels at 2 ev and 4.8 ev in U^{235}. The "regular" fission width in Pu for slow neutrons is assumed to be many times larger than the radiation width as claimed by the theory. We should therefore expect for any normal level a value of α very near to zero. If we assume the same ratio between the fission widths of regular and exceptional levels (about 50) one would expect a total width of about 2 to 3 ev for the regular levels.

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The present experimental results on plutonium are not in disagreement with this assumption. Since the resonance at 12 ev and higher are assumed to be regular ones, they should be broad and have an $\alpha = 0$. The background cross section σ_B of the Breit-Wigner shape of the 0.3-ev resonance is supposed to be the tail-effect of broad regular levels and should therefore also show $\alpha = 0$. The fact that σ_B is about equal for σ_t and σ_f supports this. (In the low-energy region $\sigma = (\sigma_t/\sigma_f) - 1$.) The observed value of α in the low-energy region is the combined effect of the resonance level and the background. One can separate the effects of background and resonance by assuming that the α pertaining to the resonance is equal to a value α_r and that α is zero for the background. We then get for the observed α:

$$\alpha_{obs} = \alpha_r \frac{\sigma_f - \sigma_B}{\sigma_f} = \alpha_r - \frac{\sigma_B}{\sigma_f} \qquad (1)$$

One can obtain α_r by fitting this expression to the value at the center of resonance where α is almost equal to α_r and one obtains $\alpha_r = 0.90$. The points obtained from (1) are shown in Fig. 1. The fit is as good as one can expect from the experimental accuracy. Thus, the proposed explanation is not disproved by the observations.

We will not attempt to give an explanation to the existence of these exceptional levels. We restrict ourselves to the fact that they have been observed. There may be some type of excitation of the nucleus which does not lead easily to fission. It was sometimes proposed to distinguish two groups of levels according to the way the spin of the neutron adds to

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the spin of the bombarded nucleus. If the spin of the bombarded nucleus is I, the levels created by slow neutron absorption fall into two groups: the ones with a spin I + 1/2 and the ones with I - 1/2. One may be tempted to say that the fission width Γ_f depends critically on the angular momentum so that Γ_f could have a very different value in the two groups. It seems, however, from the U^{235} evidence that the "exceptional" levels are very few in numbers whereas the ratio of levels with I + 1/2 to the ones with I - 1/2 should be of the order (I + 1)/(I - 1).

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V. Suggested Experiments

Because of the significance of the capture-to-fission ratio in plutonium, experiments should be made to prove or disprove the assumptions made in this paper. The following investigations suggest themselves:

1.) Refinement of the velocity-selector method could settle the question whether the 12-ev maximum in the Pu cross section is one broad resonance or can be resolved into a number of narrow ones. According to the hypothesis of this paper the widths of "regular" levels in Pu should be at least a few ev.

2.) More accurate measurements of fission and absorption cross sections in the neighborhood of the maximum of the 12-ev resonance should be made to obtain a reliable value of α at that energy. The value of the cross section at that energy is as high as 120 b so that the error due to the unknown scattering cross section is rather small. The measurements of McDaniel (LA-158) are only of exploratory nature and could be improved considerably, especially by a choice of thickness of absorber suitable for that energy region.

3.) Measurements of the scattering cross section of Pu would greatly help to interpret the present measurements. Only the thermal scattering cross section is known and it is not justified to assume that the scattering cross section in the region above 1 ev is equal to the thermal one. This is because the influence of the resonance on the scattering is still too large to be neglected as can be seen in the following analysis: the scattering cross section is given by the following expression:

$$\sigma_s = \pi\lambda\!\!\!^{-2} \left\{ f \left| \frac{\Gamma_n}{E - E_r + i(\Gamma/2)} + p_1 \right|^2 + (1 - f) \left| p_2 \right|^2 \right\}$$

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Here f is the statistical factor

$$f = \frac{1}{2}\left(1 \pm \frac{1}{2I + 1} \right) \qquad (2)$$

where I is the spin of the bombarded nucleus and the upper or lower sign is valid when the spin of the compound state made at resonance is $I + \frac{1}{2}$ or $I - \frac{1}{2}$ respectively. Γ_n and Γ are the neutron and the total width respectively; Γ_n is proportional to E, p_1 and p_2 are the potential scattering amplitudes for the two possible spin orientations. p_1 belongs to the spin orientation which gives rise to the resonance at 0.3 ev. There is good theoretical reason to assume that the p_i are real and proportional to $E^{1/2}$ over an energy region large compared to the level spacing. p_1 and p_2 should be almost equal.

Far from resonance the scattering cross section should assume the value $\sigma_s^{(p)}$ (potential scattering cross section):

$$\sigma_s^{(p)} = \pi\bar{\lambda}^2 \left\{ f\,|p_1|^2 + (1 - f)\,|p_2|^2 \right\} \approx \pi\bar{\lambda}^2\,|p_1|^2 \qquad (3)$$

Theory and experiment indicate that $\sigma_s^{(p)}$ is near $4\pi R^2$ where R is the radius of the nucleus. This gives for plutonium a value of $\sigma_s^{(p)}$ 12 b.

The absorption cross section $\sigma_a = \sigma_f + \sigma_r$ in the neighborhood of resonance is given by

$$\sigma_a = \sigma_f + \sigma_r = \pi\bar{\lambda}^2 f \, \frac{\Gamma_n \Gamma}{(E - E_r)^2 + (\Gamma/2)^2} + \sigma_B \qquad (4)$$

where σ_B is the background contribution which is negligible near resonance. We can use these formulas to express the thermal scattering cross section in terms of the resonance value σ_0 of σ_a (neglecting Γ compared to $E - E_r$ in

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the denominator, and assuming $p_1 = p_2 = 2R/\lambda\!\!\!^{-})$:

$$\sigma_s = 4\pi R^2 \,(1 - 2C + C^2/f) \qquad (5)$$

where

$$C = \frac{\sigma_0}{8\pi R^2}\,\frac{R}{\lambda\!\!\!^{-}_r}\,\frac{\Gamma}{E_r} \approx \frac{1}{10} \qquad (6)$$

by using for σ_0 the maximum value of σ_t since the scattering can be neglected at resonance. The first order correction to the potential scattering value does not depend on f and is about 20 . If one compares this result with the experimental value of 9.8 b as measured by Fermi, one would conclude that $\sigma_s(p) = 4\pi R^2$ should be 12 b, which checks with our assumption of $R = 10^{-12}$ cm. Formula (6) also shows that the influence of the 12-ev level would be very much smaller mainly because of the smaller value of σ_0. This computation is not good enough to conclude that the scattering cross section further away from resonance should be 12 b as it would follow from the calculation. It only shows that it would not be reasonable to use the thermal value of 9.8 b as the scattering cross section above 1 ev.

Measurements of the scattering cross section in the region above 1/2 ev would therefore be of great importance for the direct determination of α. If these values are available the capture to fission ratio could be determined directly, without any reference to theory, from accurate values of σ_f and σ_t:

$$\alpha = \frac{\sigma_t - \sigma_s}{\sigma_f} - 1 \,.$$

Scattering cross section measurements could be made with the velocity-selector method but, so far, have never been attempted.

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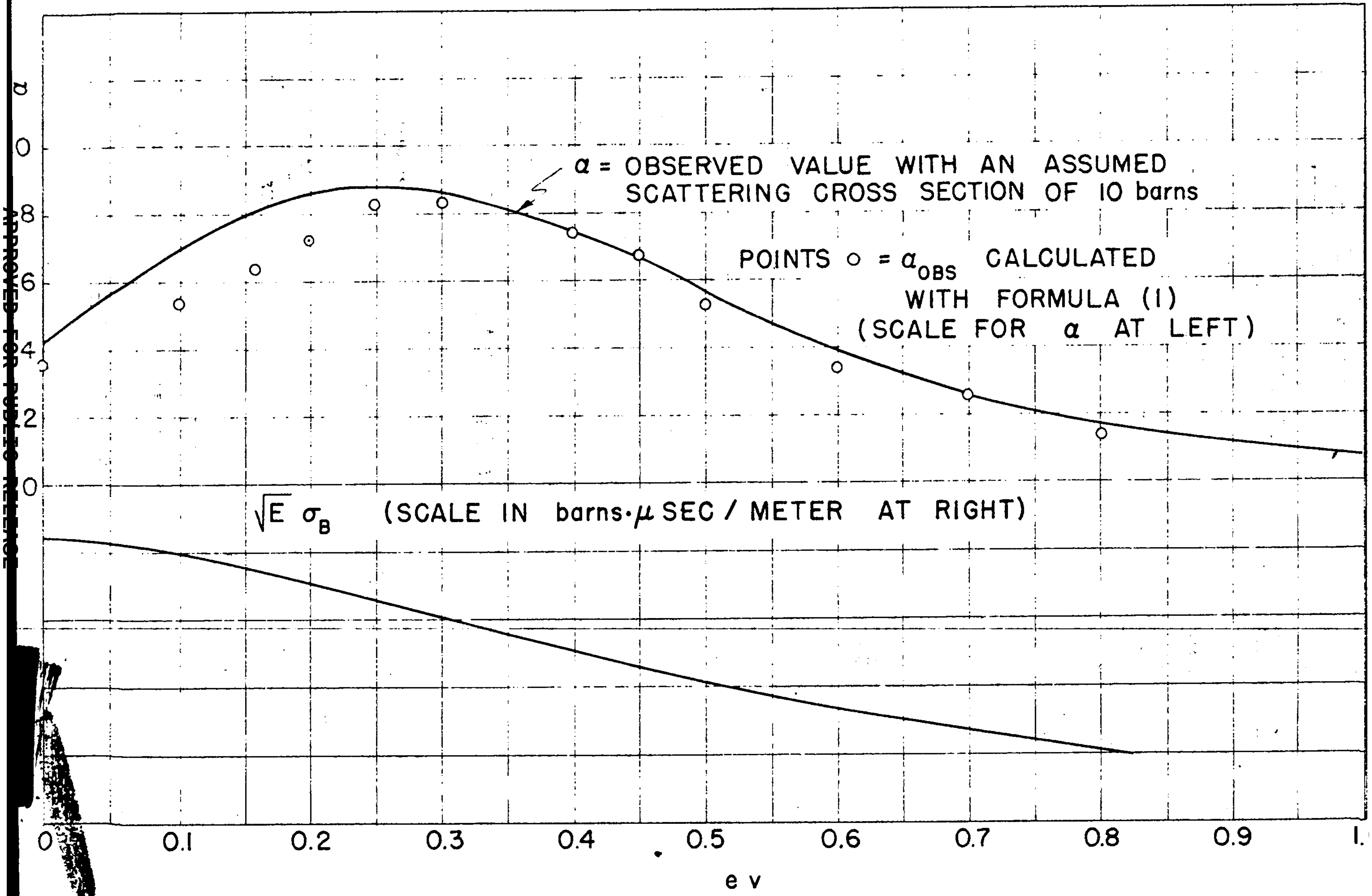
α
0
8
6
4
2
0
α = OBSERVED VALUE WITH AN ASSUMED SCATTERING CROSS SECTION OF 10 barns
POINTS ○ = α_{OBS} CALCULATED WITH FORMULA (1) (SCALE FOR α AT LEFT)
$\sqrt{E}\,\sigma_B$ (SCALE IN barns·μSEC / METER AT RIGHT)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.
e v