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THE DOPPLER COEFFICIENT FOR REACTORS CONTAINING THORIUM

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Abstract

The Doppler increase in the effective resonance integral has been calculated for a model absorber which approximates the resonance structure of Th^{232}. The results indicate that for rods of pure thorium the increase depends approximately on the square root of the temperature and very good agreement is obtained with experimental values of $\frac{1}{I}\frac{dI}{dT}$. The calculations also show that the addition of scattering material to the fuel rods will lessen the temperature dependence of the increase in the effective resonance integral.

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CONTENTS

1. INTRODUCTION

In the early days of the development of atomic power it was discovered that resonance capture of neutrons by U^{238} would not permit the establishment of a self–sustaining chain reaction in a natural uranium homogeneous reactor. To reduce the parasitic capture in the U^{238}, it was found necessary to isolate the fuel from the moderator. Apart from making the development of natural uranium thermal reactors practicable, the lumping of fuel into fuel elements has the definite advantage of isolating fission products and facilitating the replacement of spent fuel.

Neutron capture by fertile material is no longer regarded as parasitic, since in the process the fertile atoms become fissile and it is now appreciated that the world's mineral resources contain far more fertile than fissile material. Complete use of the resources of fertile material will not be possible unless the reactors of the future, while producing power from the burn–up of fissile nuclei convert at least an equal quantity of fertile into fissile material. In the development of such breeder reactors we are no longer interested in minimising resonance capture, although it is still desirable to isolate the fuel for ease of replacement.

The work of the present paper is concerned with the Th^{232}–U^{233} cycle, which appears to hold good possibilities for breeding. The ultimate aim is to design a reactor which produces $1 + \delta$ atoms of U^{233} for every atom lost by fission, where the quantity δ must be such as to balance the fission product poisoning and any other effects which reduce reactivity during the operation of the reactor. Such a design would not only make it possible to burn a small fraction of the fissile material as at present, but the fuel elements would not need replacing until a large percentage of the Th^{232} in the rods had been converted to U^{233} and burnt. The possible increase in fuel element lifetimes would be limited mainly by metallurgical problems.

The quantity δ introduced above must be known exactly. If it were too large the reactivity of the system would build up to a dangerous level; if it were too small the parasitic capture in the fission products would ultimately reduce the excess reactivity to an unworkable level.

Although reactivity changes with temperature are small, it is obvious that the delicate neutron balance envisaged for future reactors can be upset even by small changes in the operating temperature. Another reason for studying the variation of resonance absorption with temperature is that reactors with the dual role of power production and breeding are being designed to operate at high temperatures.

In this paper the Doppler increase in the effective resonance integral is calculated for each of the resolved resonances of Th^{232} and the contribution from the unresolved resonances is determined for a model absorber, the resonance structure of which approximates that of Th^{232}.

2. THEORETICAL CONSIDERATIONS

2.1 The Effective Resonance Integral

The effective resonance integral for a homogeneous mixture is given by

$$I = \int \frac{\sigma_a}{1 + \frac{\sigma_a + \sigma_s}{\sigma_m}} \frac{dE}{E} \qquad (1)$$

where σ_a, σ_s are the resonance absorption and scattering cross sections of the fertile atoms for neutron energy E, and σ_m is the constant potential scattering cross section of the mixture per fertile atom. The integration is to be extended over the whole resonance region.

For a heterogeneous lattice, it has been shown by Keane et. al. that the effective resonance integral is given approximately by (1), where now

$$\sigma_m = \frac{\sigma_t (1 + N\bar{l}\,\sigma_p)}{1 + N\bar{l}(\sigma_p + \sigma_t)} \qquad (2)$$

Here σ_p is the potential scattering cross section per fertile atom for the mixture in the fuel rods, $\bar{l}$ is the mean chord length of a fuel rod, N is the number of fertile atoms per cm^3 of the fuel rods and σ_t is the potential scattering cross section of the system per fertile atom, given by

$$\sigma_t = \sigma_p + \frac{V_m}{NV_f}\,\Sigma_s \qquad (3)$$

where $\frac{V_m}{V_f}$ is the volume ratio of moderator to fuel rods and Σ_s is the macroscopic scattering cross section of the moderator.

2.2 Doppler Increase in the Effective Resonance Integral

Assuming the resonance contours to be of Breit–Wigner form, we have, at temperature T^oA for a resonance at energy E_r,

$$\sigma_a = \frac{\Gamma_\gamma}{\Gamma}\,\sigma_o\,\psi(x,\xi)$$

and $$\sigma_s = \frac{\Gamma_n}{\Gamma}\,\sigma_o\,\psi(x,\xi),$$

where σ_o is the microscopic cross section of the fertile material for both absorption and scattering at exact resonance energy; Γ_γ, Γ_n are respectively the radiation and neutron widths of the resonance;

$$\Gamma = \Gamma_\gamma + \Gamma_n$$

and

$$\psi(x,\xi) = \frac{\xi}{2\sqrt{\pi}}\int_{-\infty}^{\infty} e^{-\frac{1}{4}\xi^2(x-y)^2}\,\frac{dy}{1+y^2}$$

with $$x = \frac{2}{\Gamma}(E - E_r)$$

and $$\xi = \frac{\Gamma}{2}\left(\frac{m}{M}E_r kT\right)^{-\frac{1}{2}}.$$

m is the mass of a neutron, M the mass of a fertile atom and k Boltzmann's constant.

Hence, for a variation in temperature from 0^o to T^oA, the Doppler increase in the effective resonance integral (1), due to a single resonance at energy E_r, is

$$\sigma_m\,\frac{\Gamma_\gamma}{E_r}\left\{\int_o^\infty \frac{\psi}{\psi + \frac{\sigma_m}{\sigma_o}}\,dx - \int_o^\infty \frac{dx}{1 + \frac{\sigma_m}{\sigma_o}(1+x^2)}\right\} \qquad (4)$$

To obtain this result we have replaced $\frac{1}{E}$ by $\frac{1}{E_r}$ since E varies only slightly over the practical width of the resonance.

The expression (4) may be rewritten as

$$\sigma_m \frac{\Gamma_\gamma}{E_r} C\left(\xi, \frac{\sigma_m}{\sigma_o}\right)$$

where

$$C(\xi, a) = \int_o^\infty \frac{\psi}{\psi + a} dx - \frac{\pi}{2\sqrt{a(1+a)}} \tag{5}$$

Thus the total Doppler increase in the effective resonance integral due to all resonances is

$$\Delta I = \Sigma \sigma_m \frac{\Gamma_\gamma}{E_r} C\left(\xi, \frac{\sigma_m}{\sigma_o}\right) \tag{6}$$

the sum being over all the resonances of the fertile atoms.

3. NUMERICAL CALCULATION OF THE DOPPLER INCREASE

3.1 Resonance Data for Th^{232}

The contribution from each of the resolved resonances of Th^{232} is calculated from equation (4) using the values of the resonance parameters given by Sampson and Chernick (12).

For the unresolved resonances in the energy range 10^2 ev. to 10^4 ev. statistical information only is available and it seems reasonable to calculate the contribution of these resonances using a model absorber whose resonances have the average parameters of Th^{232} resonances.

The average values used in the numerical calculations are a constant spacing of 20 ev. between resonances the first of which is at 330 ev. (i.e. 20 ev. above the highest energy resolved resonance), a constant radiation width Γ_γ = 0.03 ev. and the neutron width for a resonance at E_r ev. given by $\Gamma_n = 0.0015 \sqrt{E_r}$ ev.

Neglect of absorption by other than s– states and of the statistical variation of the reduced neutron width gives a very small error in the value of the effective resonance integral, since the contributions from the two effects nearly cancel.

3.2 Calculation of $C(\xi, a)$

For $a = \frac{\sigma_m}{\sigma_o} > 1$, $C(\xi, a)$ may be very easily evaluated by using the result

$$\int_o^\infty \frac{\psi}{\psi + a} dx = \sum_{r=1}^{\infty} (-1)^{r+1} \int_o^\infty \frac{\psi^r}{a^r} dx . \tag{7}$$

Values of the first few terms of this series have been tabulated by Keane and McKay (6) and the series converges very rapidly for $a > 1$. Equation (1) may also be used for $0.5 < a < 1$, although six or seven terms of the series may have to be taken in this case, but for $a < 0.5$ numerical integration using the tables of $\psi(x, \xi)$ given by Rose et al (11) is necessary. For the temperature range considered (up to 900°K) $C(\xi, a)$ may be determined approximately from the relation

$$C(\xi, a) = A(10\xi)^{-\nu} \qquad (8)$$

where A and ν are functions of a only. Figure 1 shows A and ν as functions of a for the range $.001 < a < 1$.

3.3 Evaluation of ΔI

The increase in the effective resonance integral was evaluated at temperatures 300°, 600° and 900° A for σ_m in the range 20 to 1000 barns.

The sum in equation (6) was taken as

$$\frac{\sigma_m \Gamma_\gamma}{20} \int_{320}^{10^5} C(\xi, \frac{\sigma_m}{\sigma_o}) \frac{dE}{E} \qquad (9)$$

added to the contributions from the resolved resonances. The value of (9) was determined by using Simpson's rule with intervals of 20 ev. for the range 320 to 400 ev, Weddle's rule with intervals of 100 ev. in the range 400 to 1000 ev. and, for m = 3 and 4, Weddle's rule with intervals of $1.5 \times 10^{m-1}$ in the range 10^m to 10^{m+1} ev.

Within about 2% the values of ΔI calculated for different temperatures and dilution fit the expression

$$\Delta I = (.065 + \frac{.115T}{300})\, C\, \sigma_m^{(.758 - \frac{.045T}{300})}$$

where $C = 1 - \frac{3\sigma_m + 300 - T}{6000}$

provided this factor is less than 1, otherwise C = 1. An alternative expression for the Doppler increase is given by

$$\Delta I = bT^n$$

where b and n depend on σ_m and are depicted graphically in Figure 2. It is seen from the figure that $n \approx 0.5$ for low values of σ_m and decreases to about 0.36 for $\sigma_m = 1000$.

4. THE EFFECTIVE RESONANCE INTEGRAL FOR Th^{232} AT 0°A

A numerical evaluation of the effective resonance integral for Th^{232} at 0°A, as a function of σ_m, was reported earlier by Keane et al (5). The results obtained for $100 < \sigma_m < 1000$b were found to fit the formula

$$I_o = 1.40\,(\sigma_m)^{0.45}$$

Slight curvature in the log-log plot of I_o against σ_m for $10 < \sigma_m < 100$b requires a correction factor so that for the range $10 < \sigma_m < 1000$b we obtain the formula

$$I_o = 1.40\, a\, (\sigma_m)^{0.45}$$

where $a = 1, \quad 100 < \sigma_m < 1000$b.

$a = [1 - 7(100 - \sigma_m)\, 10^{-4}], \quad 10 < \sigma_m < 100$b.

5. COMPARISON WITH EXPERIMENTAL RESULTS

A large amount of experimental data on the effective thorium resonance integral at 300°A is contained in articles by Macklin and Pomerance (7), Redman (9) and Dayton and Pettus (3).

The results quoted by Macklin and Pomerance (7) are doubtful since the abscissa should be $\sigma_m = \sigma_p + \frac{1}{NI}$ which is not the case for the pure metal with $\sigma_m \simeq 13$ b nor for the oxide at $\sigma_m \simeq 21$ barns. It would appear that the contribution $\frac{1}{NI}$, which depends on the size of the sample, has been neglected, so no comparison with the data given by Macklin and Pomerance will be attempted.

5.1 Redman's Results

For ThO_2 rods at 300°A, Redman (9) found that the effective resonance integral is given by the formula

$$I_{300} = 10.2 + 12.3 \frac{S}{M} .$$

where $\frac{S}{M}$ is the surface to mass ratio of the samples. The formula was obtained for the range of $\frac{S}{M}$ from 0.4 to 1.8 cm^2/gm. The 1/v contribution has been included in this value of I_{300}.

Table I compares the value of I_{300} calculated from the theory with the value obtained by Redman. The theoretical value includes a 1/v contribution of 3.5 barns, and is therefore denoted I^+_{300}.

The maximum difference in the two estimates of I_{300} is about 10% at the extremes of the range, which is rather good agreement.

TABLE I

$\frac{S}{M}$	σ_m	I_o	ΔI_{300}	I^+_{300} (theoretical)	I^+_{300} (Redman)
.4	64	9.0b	3.8 b	16.3 b	15.1 b
1.0	130	12.4 b	6.3 b	22.2 b	22.5 b
1.8	218	15.9 b	8.6 b	28.0 b	32.3 b

5.2 Results of Dayton and Pettus

Dayton and Pettus (3) have reported a variety of measurements of the effective resonance integral for thorium rods and slabs and for ThO_2 rods. A selection of their results is given in Table II, in which is included the theoretical value of I_{300} for comparison. All the values in Table II include the 1/v contribution.

TABLE II

	σ_m	I^+_{300} (Dayton and Pettus)	I^+_{300} (Theoretical)
	(22	10.7 b	10.6 b
Thorium	(28	12.5 b	11.6 b
Rods	(38	16.1 b	13.1 b
	(62	20.5 b	16.0 b
	(27	12.7 b	11.5 b
Thorium	(37	14.9 b	13.0 b
Slabs	(142	27.5 b	23.0 b
	(42	15.5 b	13.7 b
ThO_2	(52	16.1 b	14.7 b
rods	(72	26.6 b	17.2 b
	(81	25.7 b	18.0 b
	(139	34.3 b	22.9 b

Allowing for the errors quoted by Dayton and Pettus, in particular an error of 2.6 b in the value of I^+_{300} for a thorium rod with σ_m = 62, it is found that the experimental values for thorium metal are all higher with a discrepancy of up to 20%. For the ThO_2 rods the experimental values are all higher than the theoretical values being nearly 50% higher at σ_m = 140 b. Since the theoretical value of I^+_{300} for ThO_2 rods with σ_m = 130 b agrees almost exactly with Redman's experimental result, the results of Dayton and Pettus for ThO_2 at σ_m = 140 b are almost 50% higher than Redman's results.

6. COMPARISON WITH OTHER THEORY

6.1 Dresner's Calculation

The theoretical results of Dresner (4) cover the range of σ_m in which we are interested. However, when evaluating the contribution to the effective resonance integral for unresolved resonances, Dresner has taken the total level width equal to the neutron width , an assumption which will overestimate the effective resonance integral. Table III compares the present values with Dresner's calculated values of the effective resonance integral at 0°A and the Doppler increase to 300°A for σ_m = 20 and 200 b, when the 1/v contribution is neglected.

TABLE III

σ_m	I_o (Dresner)	ΔI_{300} (Dresner)	I_o (present)	ΔI_{300} (present)
20	5.9 b	2.1 b	5.1 b	1.7 b
200	18.5 b	9.0 b	15.2 b	8.2 b

There is a certain lack of agreement between Dresner's values and those obtained in the present paper. This is to be expected in view of the approximation made by Dresner.

6.2 Results of Blake and Hughes

In a theoretical calculation of the effective resonance integral for Th^{232}, Blake and Hughes (1) obtained a value of 109 barns for the infinitely dilute resonance integral. This implies a contribution of 25.9 b from the unresolved resonances whereas our model gives 11.2 b. Since the experimental value of the resonance integral is about 70 b, the value obtained by Blake and Hughes is unreasonably high. It follows that their values of the effective resonance integral will be too high.

For σ_m = 200 and 2000 b Blake and Hughes obtain values of the effective resonance integral which agree with the values calculated by Dresner. Since Blake and Hughes made the same approximation as Dresner this agreement is to be expected.

7. COMPARISON WITH EXPERIMENTAL VALUES OF THE DOPPLER COEFFICIENT

Measurements of the fractional increase of the effective resonance integral with temperature for Th^{232}, have been reported by Rodeback (10), Davis (2), Small (13) and Pearce (8). Table IV gives a comparison between the experimental values of $\frac{1}{I^+_{300}} \frac{\Delta I^+}{\Delta T}$ and those obtained from the present theoretical calculation.

TABLE IV

Author	σ_m	Experimental Value with 1/v included	Theoretical Values	Temperature Range
Rodeback	30b	$2.0 \pm .3 \times 10^{-4}/^oC$	2.1×10^{-4}	20 – 775°C
Davis	30–40b	$2.3 \pm .2 \times 10^{-4}/^oC$	2.1×10^{-4}	20 – 650°C
Small	36b	$3.0 \pm .3 \times 10^{-4}/^oC$	2.5×10^{-4}	20 – 270°C
"	73b	$2.2 \pm .5 \times 10^{-4}/^oC$	2.7×10^{-4}	20 – 270°C
Pearce	30b	$2.7 \pm .15 \times 10^{-4}/^oC$	2.5×10^{-4}	20 – 295°C

The theoretical values agree well with the experimental values for different temperatures. Until further results for σ_m outside the range 30 – 40b are available we have no check for variations of σ_m.

8. CONCLUDING REMARKS

Thorium metal rods of diameters 2 to 4 cms. correspond to σ_m in the range 20 to 30 barns. For such values the Doppler increase in the effective resonance integral for temperatures 0 to 900° A, as found in the present paper, fits the expression

$$\Delta I = (7.3 + \frac{12}{I}) \quad 10^{-2} \; T^{0.49}$$

within a few percent. If we take the density of thorium as 11.2 then alternatively we may write

$$\Delta I = (7.3 + 33.6 \frac{S}{M}) \, 10^{-2} \; T^{0.49}$$

When σ_m = 200 barns the Doppler increase in the effective resonance integral from 0 to 900°A, fits the formula

$$\Delta I = 0.74 \; T^{0.42}$$

Thus as the dilution increases in a homogeneous mixture or as the rod size decreases in a heterogeneous lattice, the Doppler increase in the effective resonance integral depends on a smaller power of the temperature. This is to be expected since the larger σ_m, the greater $a = \sigma_m/\sigma_o$ for each resonance, and so the lower the power of T on which each resonance depends. The variation of ΔI with T shown above agrees very well with Pearce's results.

Figure 3 shows the experimental data mentioned in section 5, together with the theoretical curves for the effective resonance integral at 300°A as calculated in the present paper and as determined by Dresner. The two theoretical curves are far more consistent than the experimental results, so it hardly seems worthwhile to try to improve the theory until better experimental results are available, or until some reason for the discrepancy between Redman's data and those of Dayton and Pettus can be found.

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REFERENCES

(1) Blake, J.P.H. and Hughes, G.A., AERE R/R 2506 (1958).

(2) Davis, M.V., J. App. Phys. 28, 714, (1957).

(3) Dayton, I.E. and Pettus, W.G., Nuc. Sci. & Eng. 3, 286, (1958).

(4) Dresner, L., Nuc. Sci. & Eng. 1, 68, (1956).

(5) Keane, A., McKay, M.H., and Cox, C., AAEC/E 43, (1959).

(6) Keane, A. and McKay, M.H., Aust. J. Appl. Sci. 11, No. 3 (1960).

(7) Macklin, R. and Pomerance, H., Paper P/934, Geneva Conference, (1955).

(8) Pearce, R.M., AERE R/R 2806, (1959).

(9) Redman, W.C., Paper P/600, Geneva Conference, (1958).

(10) Rodeback, G., NAA–SR–1641, (1956).

(11) Rose, M.E., Miranker, W., Leak, P., Rosenthal, L. and Hendrickson, J.K., WAPD–SR–506 (Vol. II), (1954).

(12) Sampson, J.B. and Chernick, J., Prog. Nuc. Eng., Series 1, Vol. 2, 223, (1958).

(13) Small, V.G., AERE R/R 2774 (1959).

FIG. I. GRAPH OF 'A' AND 'ν' AS FUNCTIONS OF 'a'.

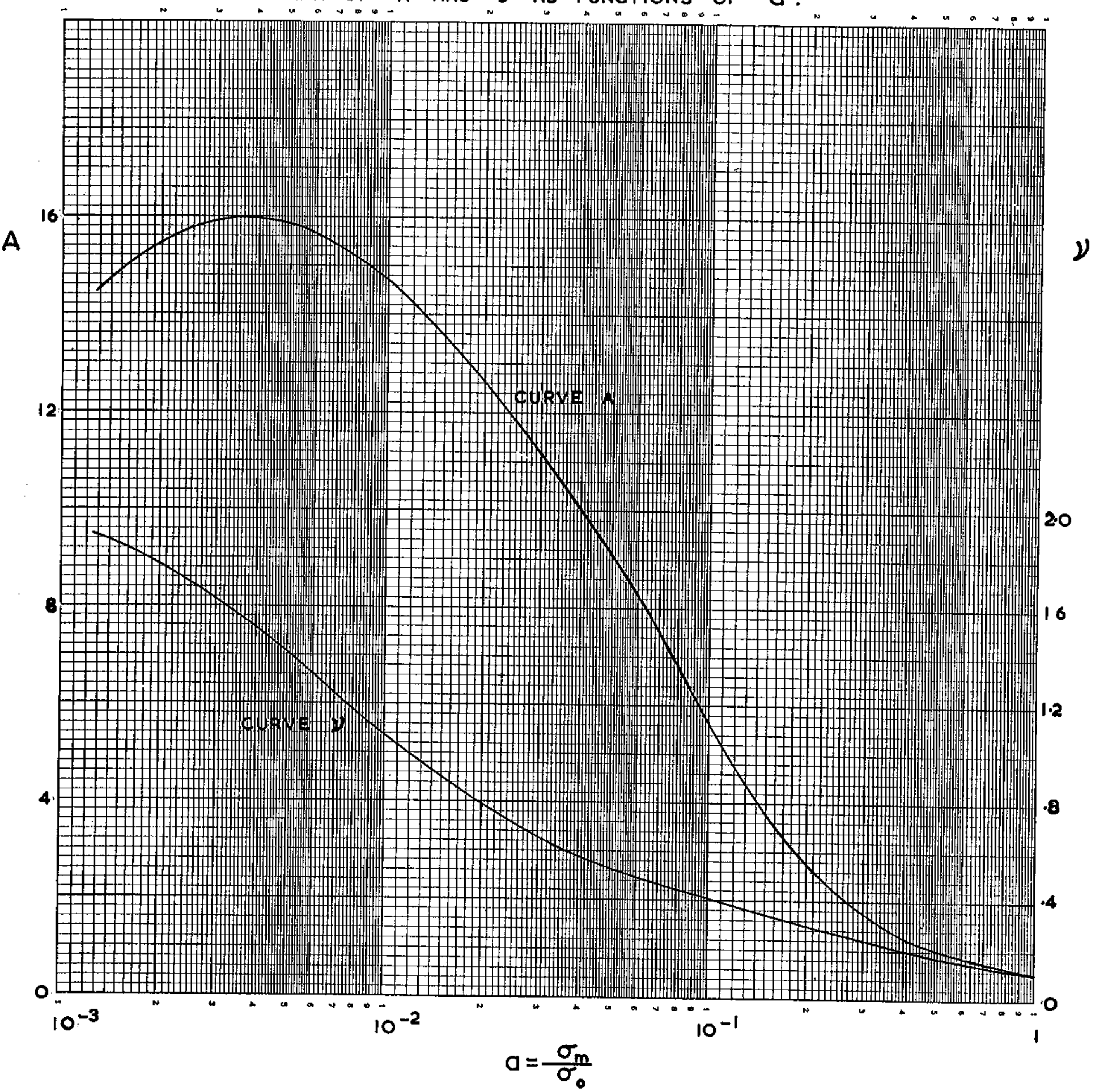

FIG. 2. GRAPH OF b AND n AGAINST σ_m.

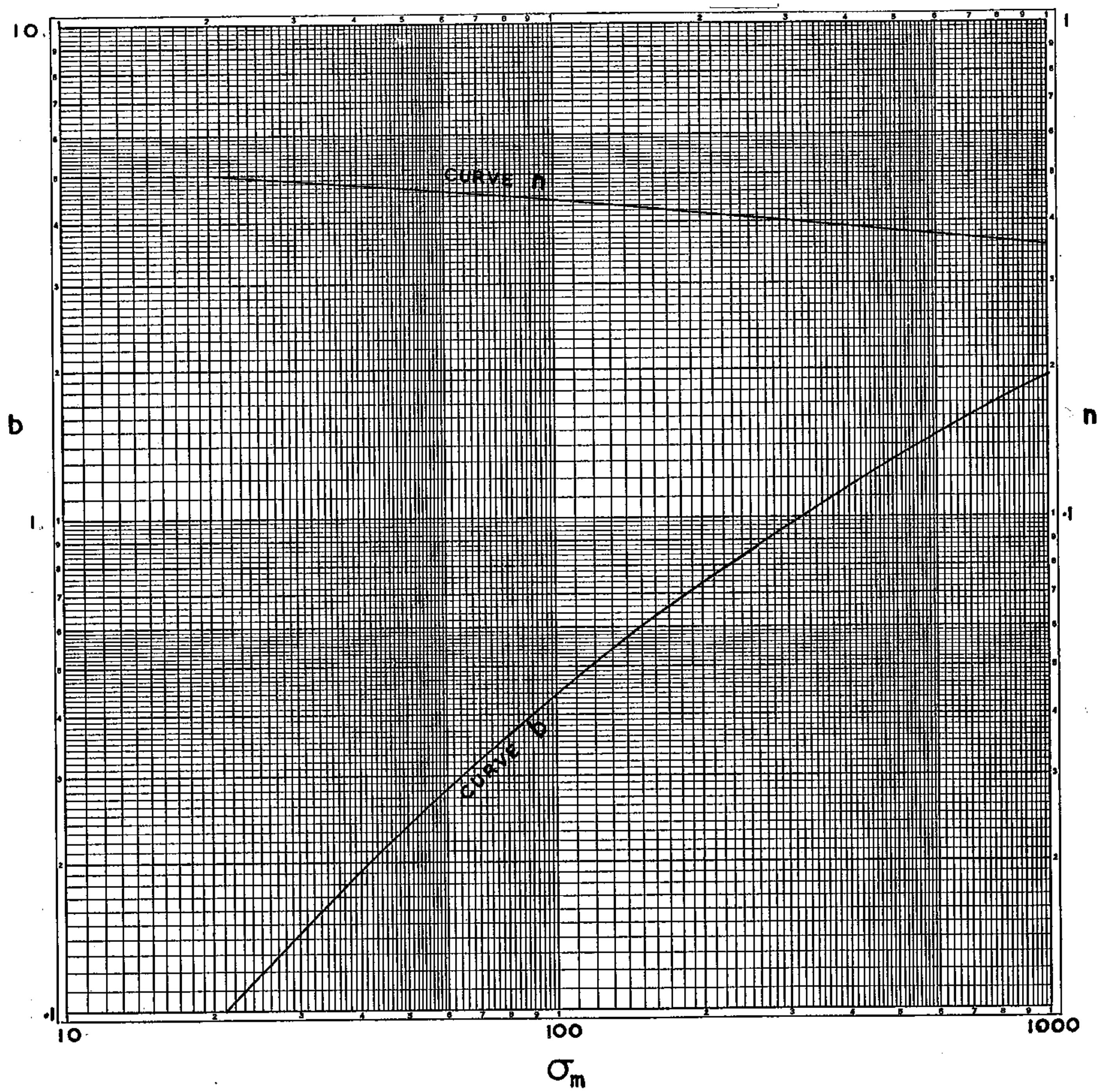

FIG. 3. COMPARISON OF THEORY WITH EXPERIMENT.