THE GIANT RESONANCE OF PHOTODISINTEGRATION OF TANTALUM

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Summary

The neutron yield from tantalum has been measured as a function of bremsstrahlung energy, and from this the shape of the cross section for the nuclear absorption of photons by tantalum has been deduced. This cross section shows definite evidence that it is made up of two component parts. The result is in agreement with the theoretical predictions of Danos (1958) for this nucleus, both regarding the existence of the two components and the energy separation of the two peaks. Using the Danos theory, the intrinsic electric quadrupole moment for tantalum is calculated to be $6 \cdot 1$ barns ± 10 per cent.

I. Introduction

The theories put forward to account for the nuclear giant dipole resonance fall into two general groups, characterized by the particular type of nuclear model considered.

The independent particle model has been the subject of investigations by Levinger and Bethe (1950), Burkhardt (1953), and Levinger and Kent (1954). Also, Wilkinson (1956) has considered the absorption of radiation by transitions between single-particle states in the shell model. His results are, in general, roughly compatible with observations of the main features of the giant resonance (maximum cross section σ_m, resonance energy E_m, integrated cross section $\int \sigma(E)\mathrm{d}E$).

On the other hand, the model assuming long-range correlations between nucleons in the excited state, or hydrodynamic model, has been the basis of work by Goldhaber and Teller (1948), Steinwedel, Jensen, and Jensen (1950), Danos (1952), and Araujo (1954). These theories consist essentially in variations of the original ideas of Goldhaber and Teller, in which vibrations were set up within the nucleus by the action of the radiation. The model they considered in most detail assumed that a spherical region of " proton fluid " vibrated in opposition to a similar, nearly coincident sphere of " neutron fluid ", this vibration having a natural frequency corresponding to the energy of the giant resonance. Steinwedel, Jensen, and Jensen (1950) have considered the case where the target nucleus is spherical, and remains spherical even under excitation by γ-rays. Motion of the nucleons within the nucleus causes density changes inside a rigid spherical surface. The total nuclear density is assumed to be constant, while allowing local complementary periodic changes in neutron or proton density.

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Using calculations for the hydrodynamics of two fluids in a sphere, the results obtained by Steinwedel, Jensen, and Jensen predict that:

$$E_m \text{ should be proportional to } A^{-\frac{1}{3}},$$
$$\int \sigma(E)\mathrm{d}E \text{ should be proportional to } A,$$

where $\sigma(E)$ is the absorption cross section at energy E, and A is the mass number of the nucleus in question. Experimental work so far does not definitely distinguish between this predicted dependence of E_m on A and the Goldhaber-Teller prediction of an $A^{-\frac{1}{6}}$ variation.

II. The Resonance Width

A feature of the half-width, Γ MeV, of the giant resonance which was examined by Wilkinson (1956) is its variation from one nucleus to another. Nathans and Halpern (1953) have observed that for nuclei with closed shell structure the resonance widths are unusually small, and Wilkinson has given a qualitative account of factors which could contribute to this effect. As one of the factors, he has suggested that rare earth nuclei not having closed shells may have a broader resonance, because of a breakdown of the j–j coupling model of shell structure considered in his model. However, he has not offered a quantitative explanation.

Recently, Danos (1956, 1958) pointed out that by considering a hydrodynamic model, it is possible to predict quantitatively that the giant resonance in deformed nuclei is, in fact, the sum of two resonances at different energies. This would lead one to expect that the observed resonance in these nuclei would be broadened or double-peaked, depending on the shapes and energy separation of the two components. The resonance width for nuclei with closed or nearly closed shells (and therefore of spherical shape) would not exhibit this effect. The detailed shape of the resonance in deformed nuclei is thus of particular interest, as it provides one experimental test of the ability of the hydrodynamic model to describe the behaviour of nuclear matter. The other principal features of the giant resonance are not sensitive to the theory used to predict them. Therefore a test of any model cannot be based upon their measurement (Montalbetti, Katz, and Goldemberg 1953).

Danos (1956, 1958) has applied to the model of Steinwedel, Jensen, and Jensen (1950) boundary conditions representing a spheroidal nucleus having semi-axes a and b, where a is directed along the axis of rotational symmetry. In the solution of the eigenvalue equations in ρ_p, the proton density, to find the frequencies ω_a and ω_b of the lowest vibration modes, it is found that the eigenvalues ω depend on the orientation of the vibration considered, and also on the deformation parameter ε, which is defined by

$$\varepsilon=(a^2-b^2)/R^2,$$

where R is the radius of a sphere equal in volume to the spheroid. By computation over a range of ε-values, the following empirical formula has been shown to give accurately the splitting of the eigenvalues (Danos 1958),

$$\omega_b/\omega_a=0\cdot 911a/b+0\cdot 089.$$

ω_a here represents the resonance frequency for absorption of polarized γ-rays by a nucleus whose a-axis coincides with the direction of polarization, and similarly for ω_b. Curves of absorption cross section for these polarized beams would then have the usual resonance shape, peaked at ω_a and ω_b respectively, and having cross sections $\sigma_a(E)$ and $\sigma_b(E)$. For randomly oriented nuclei, the effective total cross section at energy E is expected to be

$$\langle\sigma(E)\rangle=\{\tfrac{1}{3}\sigma_a(E)+\tfrac{2}{3}\sigma_b(E)\},$$

since there are two axes of length b and only one of length a. The total cross section should thus be the sum of two resonance curves, separated in energy, with one twice as high as the other if it is assumed that these resonance curves have the same width. More generally, the areas under these resonance curves should be related by the equation

$$\int\sigma_a(E)\mathrm{d}E\bigg/\int\sigma_b(E)\mathrm{d}E=\tfrac{1}{2}.$$

In a nucleus with ε greater than zero, the larger peak will occur at the higher energy.

In the present experiment, a careful study has been made of the absorption cross section of a nucleus having a large deformation in the ground state. Work involving experimental readings at small energy intervals is essential in order to be able to detect for certain the presence or absence of the broadening or double-peaking effect. In much other work of a similar nature, energy intervals of 1 MeV or larger have been used, and the effect could easily have been missed.

III. The Experiment

An examination of the resonance in photon absorption by a deformed nucleus has been undertaken. In a heavy nucleus (say Z greater than 50), the sum of $\sigma(\gamma,n)$ and $\sigma(\gamma,2n)$ gives a very good approximation to the absorption cross section, assuming that the contribution of competing reactions in which protons are emitted is small enough to be neglected. This is so for the heavy nuclei. For example, Toms and Stephens (1955) found that the yield of protons emitted from tantalum under the action of 23 MeV bremsstrahlung was less than 10^{-3} of the corresponding neutron yield.

Since the intrinsic quadrupole moment of a nucleus (Q_0) is directly proportional to ε, an element with a high quadrupole moment was used as target. In this case tantalum 181, a 100 per cent. natural isotope, was chosen. (Further tests are to be done on some rare earths, which are equally suitable.) Since the nucleus ^{181}Ta has Z (=73) and N (=108) well away from the magic number values of 50, 82, and 126, one would expect this nucleus to be deformed.

The spectroscopically measured electric quadrupole moment Q_s is related to the intrinsic moment Q_0 by the equation

$$Q_0=Q_s\frac{(I+1)(2I+3)}{I(2I-1)},$$

where I is the nuclear spin. Q_s has been determined for ^{181}Ta by Murakawa and Kamei (1957), who find Q_s to be $+2\cdot7\pm0\cdot3$ barns. Since for tantalum, I is 7/2,

Q_0 is calculated to be $+5\cdot 8$ barns. A determination of Q_0 from probabilities of γ-ray transitions found in Coulomb excitation of low-lying states has given a value of $+6\cdot 8$ barns (Alder *et al.* 1956).

Since in the ^{181}Ta nucleus Q_0 is positive, a is greater than b. Thus, if the predictions of Danos are correct, the absorption cross section resonance should be broadened or double-peaked, due to the two components, and the higher energy component should have the larger $\int \sigma(E)\mathrm{d}E$. For example, taking the low energy peak to be at 13 MeV, and Q_0 to be $+6\cdot 0$ barns, the theory indicated a separation in peaks of about $2\cdot 8$ MeV.

Bremsstrahlung from the Melbourne 18 MeV electron synchrotron was used for the irradiation of the tantalum target. Maximum X-ray energy was variable between 8 and 18 MeV by means of variation and precise measurement of the d.c. bias used in exciting the magnet. An energy calibration of the machine by means of (γ,n) threshold measurements on ^{31}P, ^{32}S, ^{55}Mn, ^{63}Cu, and ^{65}Cu had been completed one month prior to this work, and the effective X-ray energies were considered to be known absolutely within $\pm 0\cdot 2$ MeV, with relative energies defined considerably better. The X-ray pulse was obtained in each cycle by cutting off the r.f. accelerating field 100 μsec before the peak of the a.c. cycle. The electron beam would then spiral inwards onto a platinum target $0\cdot 005$ in. thick. The length of time (250 μsec) over which the bulk of the beam emerged in each cycle has allowed gating circuits to be omitted from the counting electronics, there being no significant pile-up of electron pulses in the counters.

The arrangement of target, neutron counters, and paraffin and concrete "house" was similar to that described by Nathans, Halpern, and Mann (1952). The target consisted of a block of better than $99\cdot 9$ per cent. pure tantalum, 1 in. square and $0\cdot 14$ in. long in the direction of the beam. In the analysis of results, correction was made for the absorption of X-rays of different energies over the length of this block.

A pair of ^{10}B-enriched boron trifluoride proportional counters was used to count neutrons thermalized by the 13 cm of paraffin separating the counters from the target. At this distance, according to Rossi and Staub (1949), the counting efficiency will be energy independent. The counter pulses were fed through an amplifying and discriminating system to a pair of independent scalers. Counting efficiency was checked regularly throughout the work by measuring the count rate from a 10 mc Ra-Be neutron source which could be inserted in the paraffin behind the target. Variations in counting efficiency were not greater than ± 1 per cent. from the mean value over a day's running.

The dead-time inherent in the system was the source of a counting loss which depended on the average input pulse rate. To take this into consideration reliably, a correction factor was determined experimentally as a function of average counting rate, and was applied to every point of the yield curve.

The radiation dose for each exposure was measured with a Victoreen $0\cdot 25$ r integrating monitor, used as a transmission chamber and placed behind the lead collimator so as to intercept the whole of the collimated beam. This ionization chamber was tested periodically for leakage, and was calibrated in terms of

" Lucite roentgens " by a comparison of its response with that of a 25 r thimble irradiated at the centre of an 8 cm " Perspex " cube.

Synchrotron energy was varied in steps of 0·2 MeV over the range 8–18 MeV to obtain a yield curve in terms of neutron counts per unit dose versus synchrotron energy. Except close to the Ta(γ,n) threshold, between 4000 and 10,000 counts were taken per run. Each run was repeated independently not less than four times, and average yields were plotted. Thus the statistical

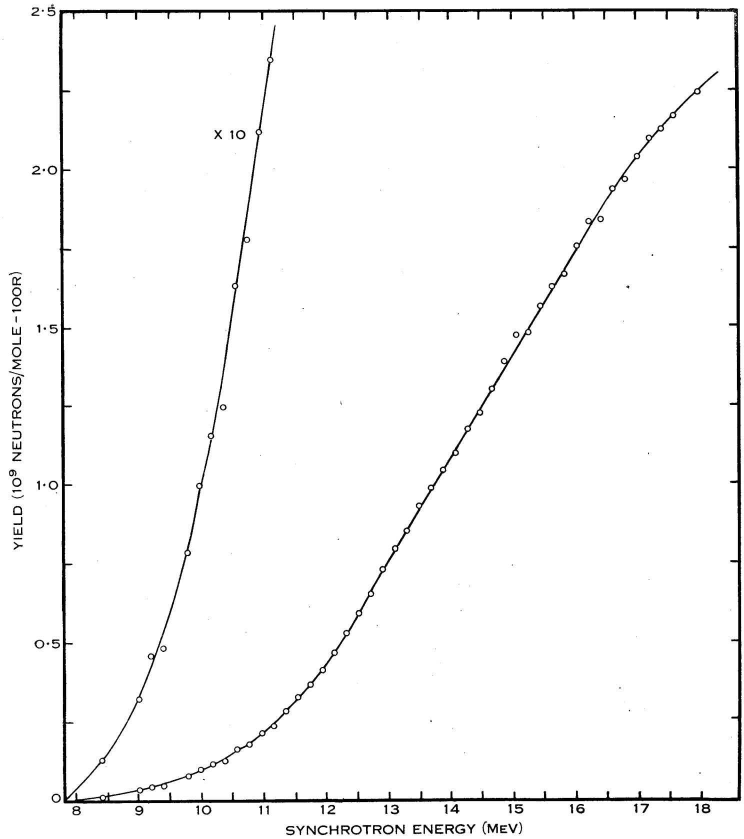

Fig. 1.—Measured yield curve of photoneutrons from tantalum as a function of synchroton energy.

counting error was between 0·5 and 1 per cent., except near the threshold. From this graph was subtracted a similarly obtained background curve, measured with all apparatus in place except the tantalum itself. At 17 MeV, it represented 4 per cent. of the total neutron yield.

A plot of the low energy yield curve points has indicated a value for the Ta(γ,n) threshold of $7 \cdot 7 \pm 0 \cdot 2$ MeV. This agrees with figures previously obtained by McElhinney *et al.* (1949) ($7 \cdot 7 \pm 0 \cdot 2$ MeV), by Sher, Halpern, and Mann (1951) ($7 \cdot 55 \pm 0 \cdot 20$ MeV), and by Cameron (1957), whose mass formula calculation gave 7·77 MeV.

The absolute yield curve, obtained by taking account of counter efficiency and absorption of X-rays in the target, is shown in Figure 1. Since the test neutron source calibration was quoted to ± 10 per cent., the absolute values may be in error by this amount. However, the relative values will be very much better than this, because of the good counting statistics.

IV. Analysis of Results

Derivation of the cross section from the yield curve was carried out by the Leiss-Penfold method (Penfold and Leiss 1954), using effective energy intervals of 0·5 MeV (Fig. 2). First differences of the yield curve were smoothed, to minimize errors due to the drawing of a curve through the experimental points.

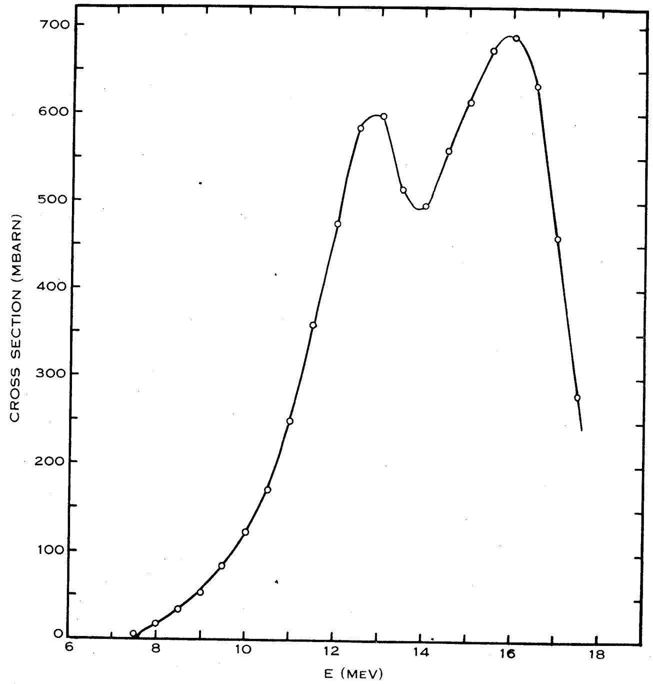

Fig. 2.—Cross section calculated by the Leiss-Penfold method from the neutron yield curve. This cross section plot represents $\sigma(\gamma,n)+2\sigma(\gamma,2n)$ as a function of energy.

Above the threshold of the ^{181}Ta$(\gamma,2n)$ reaction, account has to be taken of multiple neutron production, since the cross section derived from the neutron yield curve is the sum of $\sigma(\gamma,n)+2\sigma(\gamma,2n)+3\sigma(\gamma,3n)$ and so on. To obtain the absorption cross section, the extra weighting of multiple neutron reactions must be allowed for. For our purposes, the absorption cross section is $\sigma(\gamma,n)+\sigma(\gamma,2n)$, since the threshold for the ^{181}Ta$(\gamma,3n)$ reaction is greater than 18 MeV.

There being very little experimental evidence available on the subject, statistical theory of nuclear reactions as described by Blatt and Weisskopf (1952) was used. The result which they derive is

$$\sigma(\gamma,2n)=\sigma_{\mathrm{abs}}\{1-(1+\varepsilon_{\mathrm{sec}}/\Theta)\exp(-\varepsilon_{\mathrm{sec}}/\Theta)\},$$

where σ_{abs} is the total cross section for nuclear absorption of a γ-ray. ε_{sec} is the maximum energy which could be given to a second neutron emitted from the nucleus, that is, it is equal to the difference between the γ-ray energy and the energy of the $(\gamma,2n)$ threshold. Θ is the maximum possible nuclear temperature of the ^{180}Ta nucleus, which is formed after the emission of the first neutron, and before the second neutron is emitted. Θ is defined by the equation

$$\Theta=\frac{1}{a}\{E_\gamma-E_{th}\}^{\frac{1}{2}},$$

where a is a constant, estimated by Blatt and Weisskopf (1952) to have the value 10 in our case. E_{th} is the (γ,n) threshold energy.

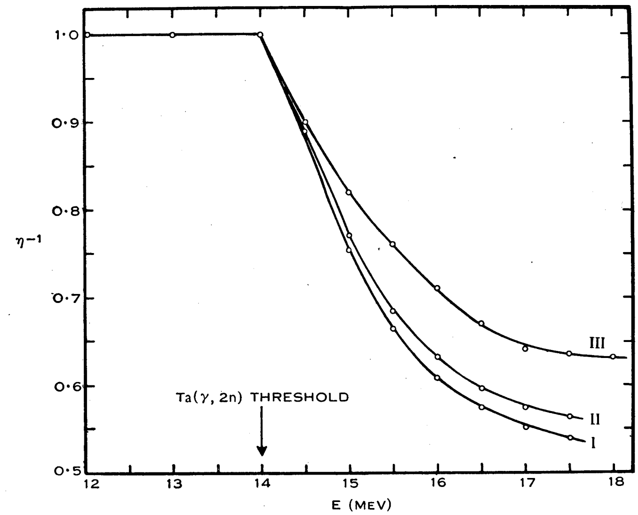

Fig. 3.—The reciprocal of the neutron multiplicity plotted as a function of γ-ray energy for tantalum. Curve I shows $(\eta_1)^{-1}$ calculated on the basis of the statistical theory of nuclear reactions. Curve II shows $(\eta_2)^{-1}$ calculated on statistical theory plus an assumed 10 per cent. of the absorption giving rise to direct photoeffect. Curve III shows $(\eta_3)^{-1}$ as found in the experiment of Whalin and Hanson (1953).

This formula for $\sigma(\gamma,2n)$ is based on the assumptions that $E_\gamma\gg\Theta$, and that neutron emission from the intermediate nucleus predominates immediately it becomes energetically possible. Both of these assumptions are good for our case.

In our calculation, a was put equal to 10, and the values of the (γ,n) and $(\gamma,2n)$ thresholds used were 7·7 and 14·0 MeV respectively. The result of the calculation was expressed in terms of a factor $\eta(E)$, known as the neutron multiplicity. This is defined as the average number of neutrons emitted for each photon absorbed. The measured neutron yield is then given by

$$Y(E_0)=0\cdot6023\int_0^{E_0}\sigma(E)\eta(E)P(E,E_0)\mathrm{d}E,$$

where $P(E, E_0)\mathrm{d}E$ is the number of photons of energy E to $E+\mathrm{d}E$ per cm^2 per 100 r in a spectrum of maximum energy E_0. The cross section is in barns in this formula. The neutron multiplicity can be less than unity owing to competition from proton emission, but, since the proton yield from tantalum is so small relative to the neutron emission, the minimum value of $\eta(E)$ in our case is unity. A graph of $1/\eta(E)$ for tantalum as a function of γ-ray energy is shown in Figure 3. The calculation using statistical theory leads to the curve $(\eta_1)^{-1}$ in that figure. Thus the absorption cross section is η^{-1} times the cross section derived from the neutron yield curve.

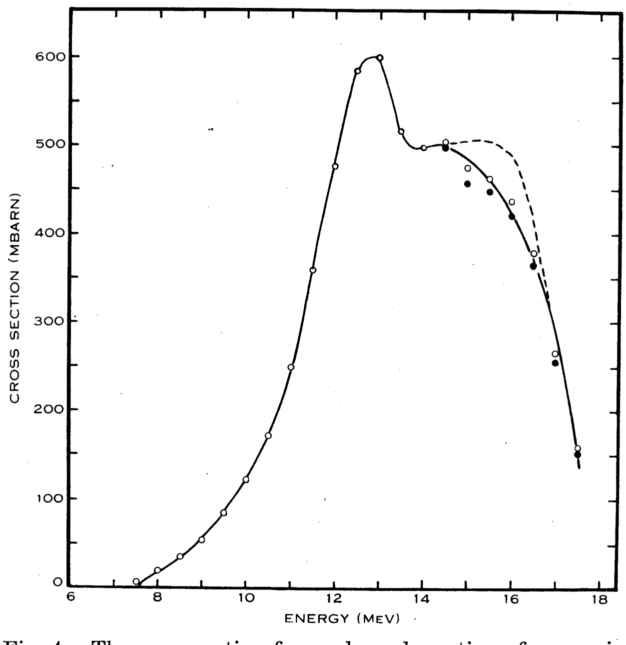

Fig. 4.—The cross section for nuclear absorption of γ-rays in tantalum. The full circles were calculated using curve I of Figure 3, while the open circles were the result of using curve II. Use of curve III led to the dotted cross section.

An attempt to allow for the effect of the direct photoeffect was made. Once again, experimental evidence was lacking, and to estimate the significance of the effect the cross section for direct interaction between photons and nucleons of the tantalum nucleus was assumed to be a constant 10 per cent. of the total cross section for absorption of photons by that nucleus. Including this consideration, an alternative factor was found, and this is plotted as $(\eta_2)^{-1}$ in Figure 3.

Experimental values for the ratio of $\sigma(\gamma,n)$ to $\sigma(\gamma,2n)$ have been obtained by Whalin and Hanson (1953) and by Carver, Edge, and Lokan (1957). This was done by measuring a total neutron yield curve, and subtracting from it the yield curve for the (γ,n) reaction only. The (γ,n) yield was obtained by counting the β^+-activity of ^{180}Ta. This subtraction gives the yield curve for the $(\gamma,2n)$ reaction. These results disagree with the statistical theory very considerably.

The values of η^{-1} obtained in the experiment of Whalin and Hanson are plotted in Figure 3 as $(\eta_3)^{-1}$.

The effects of η_1 and η_2 respectively are shown in the two sets of plotted points in Figure 4, which shows the cross section for photon absorption in the tantalum nucleus. Precisely where the actual curve should lie cannot be said. However, the essential features of the final cross section are not at all dependent on these doubtful multiplying factors. If the experimental values of $\eta(E)$ are

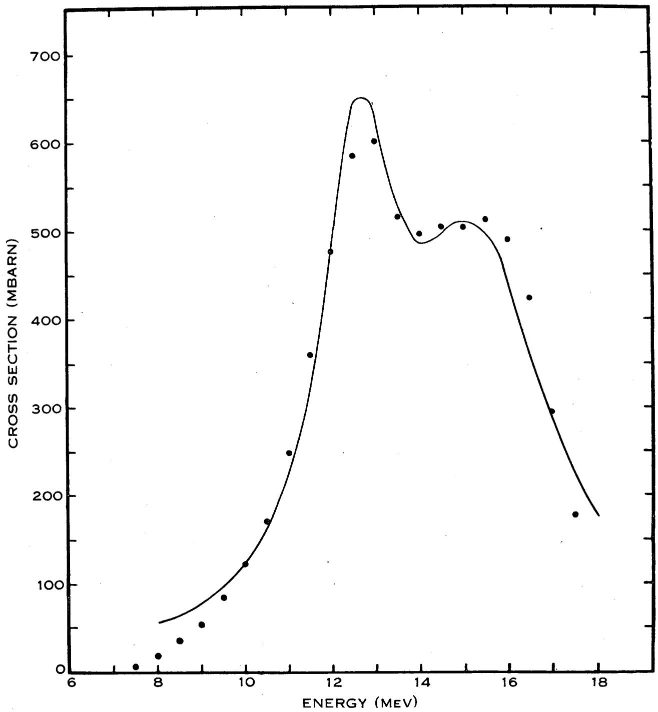

Fig. 5.—The fit of two Breit-Wigner shape resonance curves to the cross section calculated using curve III of Figure 3. Parameters of Breit-Wigner fit to observed σ: $E_1=12\cdot6$ MeV, $\sigma_{1m}=500$ mbarn, $\Gamma_1=2$ MeV; $E_2=15\cdot3$ MeV, $\sigma_{2m}=450$ mbarn, $\Gamma_2=4$ MeV.

used, the dotted curve of Figure 4 is obtained. In this case, the evidence for a second peak in the cross section for the absorption of γ-rays is even more pronounced.

In view of the limits of error shown and the constancy of the characteristic general shape of the cross section under different methods of allowing for the $(\gamma,2n)$ reaction, it is considered that the evidence for two peaks in the absorption cross section is quite definite. The width of the resonance curve of Figure 4 (full width at half maximum) is $5\cdot4$ MeV, the maximum cross section is 615 mbarn, and the integrated cross section up to 18 MeV is $3\cdot2$ MeV-barn.

There remains some doubt about our assessment of the absolute yield, mainly owing to the uncertainty in calibration of the neutron source used to test the counter efficiency and to uncertainty in the calibration of the Victoreen thimble as used in the 10–20 MeV region. If instead we standardize our neutron yield from tantalum against that from copper, and use the value quoted by Montalbetti, Katz, and Goldemberg (1953) for the neutron yield from this element, the peak cross section becomes 540 mbarn.

V. Conclusions

The γ-ray absorption resonance observed for tantalum exhibits distinct features of shape and width not normally found in curves of this type. This result can be satisfactorily explained in terms of a sum of two components, and a "resonance" showing just these features might well be expected on the basis of the theory of Danos (1958). To demonstrate the feasibility of such a resolution

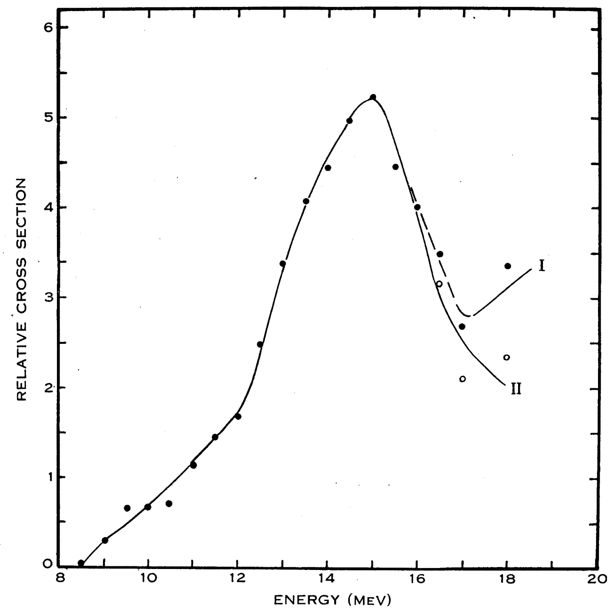

Fig. 6.—Cross section for nuclear absorption of γ-rays in lanthanum. Curve I is the cross section calculated from the neutron yield curve. Curve II is corrected for the effects of neutron multiplicity.

into two peaks as required by the Danos theory, the cross section obtained using the experimental values of $\eta(E)$ has been decomposed into two components, σ_1 and σ_2 (see Fig. 5). Both these component cross sections were assumed to have Breit-Wigner shape, with parameters as stated in Figure 5. This decomposition is not unique, of course, but serves to show the compatibility of this result with the predictions of Danos (1958). The energy separation of the two peaks in this decomposition leads to a value of the intrinsic electric quadrupole moment for tantalum of $6 \cdot 1$ barns (compared with the previously quoted values of $5 \cdot 8$ and $6 \cdot 8$ barns). This value was obtained assuming $R = R_0 A^{\frac{1}{3}}$, $R_0 = 1 \cdot 2 \times 10^{-13}$ cm. Further, the ratio of area under the lower energy peak to that under the higher energy peak is 0·55, in approximate agreement with the predicted value of 0·5.

Whilst the above results thus lend clear support to the validity of Danos's improvement to the hydrodynamic model, it is not claimed that this theory is the correct or the only interpretation of them. Before any adequate verification of the theory is obtained, the γ-ray absorption cross sections must be measured for a number of nuclei of varying deformations. In particular, it must be demonstrated that this effect does not occur for undeformed nuclei. To test this, the above technique has been applied to the measurement of the γ-ray absorption cross section in lanthanum. This is a nucleus with a closed shell of neutrons ($N=82$) and thus has very small deformation. The absorption cross section obtained for this nucleus is shown in Figure 6, and shows no evidence whatever for two components. This is also in agreement with the hydrodynamic model theory.

We are grateful to have been informed of a series of experiments, with an object identical to the present one, recently performed by Weiss and Fuller (Fuller, personal communication 1957). The cross section for absorption of γ-rays by the tantalum nucleus has been resolved into two peaks at 12·8 and 15·7 MeV respectively. The integrated cross section to 20 MeV was found to be 3·27 MeV-barn and the width of the resonance about 6 MeV. This is in good agreement with the present work.

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