tron density within ~0.2A of the nucleus contributes to the transition probability for internal conversion. The greater part of the effect is attributed to the $4p$ electrons, which account for possibly one-tenth of the total transition probability. In $KTcO_4$ an increase of 3 percent in the $4p$ electron density in the critical region, compared to the density in Tc metal, could produce an effect of the magnitude and direction observed.

The greater $4p$ electron density of Tc^{99m} in $KTcO_4$ compared to that in Tc metal is attributed to the fact that the Tc atom is constrained to a smaller volume in $KTcO_4$. The Tc—O separation in the TcO_4 tetrahedral ion[26] is ~1.75A compared to ~2.73A for the separation of the nearest neighbors in the metal.[27]

[26] W. H. Zachariasen, Am. Crystallographic Assoc. Abstracts, F-4, February, 1951 meeting, National Bureau of Standards, Washington, D. C.
[27] C. L. Mooney, Phys. Rev. **72**, 1269 (1947); Acta Cryst. **1**, 161 (1948).

The observed effects demonstrate that γ-ray emission and internal conversion take place competitively rather successively, in agreement with the accepted theory of internal conversion. The determination of the changes in the decay constant of isomers in different chemical combinations may help in understanding the electronic structure of these compounds.

An attempt is being made to attain increased accuracy with improved equipment and stronger sources so that the work need not be confined solely to very low energy isomeric transitions.

We are indebted to Mr. M. McKeown for checking the sources with a NaI scintillation spectrometer, to Dr. G. Friedlander for advice on some chemical aspects of this work, to Miss Jean Snover for much valuable help, particularly in the computations, and Dr. V. W. Cohen and Dr. Raymond Davis for their hospitality in providing air-conditioned space for setting up the equipment in their laboratories.

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Inelastic Nuclear Scattering of Photons by Rh^{103}†

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A lower limit to the inelastic nuclear photon scattering cross section of Rh^{103} was determined as function of energy. Rhodium foils were exposed to a bremsstrahlung beam and the yield of $Rh^{103}(\gamma,\gamma')Rh^{103m}$ was measured as a function of the upper limit of the spectrum. Activities were counted with a windowless 4π proportional counter and $Ta^{181}(\gamma,n)Ta^{180}$ was used as a monitor above 8 Mev, adopting the improved results of the Sasketchewan group. It was ascertained that the fast neutron contamination of the beam contributed but a small fraction of Rh^{103m} activity. The cross section, obtained from the yield curve by customary methods, is found to go through a peak of the order of 10 mb at about 13 Mev and to be appreciable below the $Rh^{103}(\gamma,n)Rh^{102}$ threshold (9.35 Mev). The observed location of the peak is considerably lower than would be expected for $Rh^{103}(\gamma,n)$ from the empirical A-dependence of (γ,n) cross sections, and is also at variance with the observations of Katz and Cameron on $Au^{197}(\gamma,\gamma')Au^{197}$, but appears plausible on theoretical grounds as does the magnitude of the cross section.

I. INTRODUCTION

THE importance of nuclear photon scattering, both elastic and inelastic, has long been emphasized in connection with the mechanism[1–3] of electromagnetic absorption in nuclei. Various experiments designed to detect the scattered photons directly have been reported. They differed mainly in the detectors used [cloud chamber,[4] activation of Pr^{141} by (γ,n) reaction,[5] NaI(Tl) crystal with pulse-height selection[6]] and suffered from the common shortcoming of low counting rates and no or insufficient ability to discriminate between elastic and inelastic scattering. Another method, first used at low energies, by the Notre Dame group,[7,8] consists in irradiating a stable isotope possessing an isomeric state with photons and determining the isomeric activity which is induced in a fraction of the inelastic scattering processes. This fraction being unknown experimentally, and but unreliably predictable theoretically, only lower limits to cross sections can be obtained in this way. Furthermore, in the "resonance" region, i.e., at energies above the threshold for neutron emission, this method is unfortunately confined to those isotopes whose isomeric states cannot

† Work assisted by joint program of the U. S. Atomic Energy Commission and U. S. Office of Naval Research.
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[1] M. Goldhaber and E. Teller, Phys. Rev. **74**, 1046 (1948).
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[5] Dressel, Goldhaber, and Hanson, Phys. Rev. **77**, 754 (1950).
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be excited by a (γ,n) reaction on a heavier isotope of the same element, which are very few. On the other hand, this method is far superior in sensitivity to those mentioned above and provides for an easy determination of the energy-dependence of the partial cross section from bremsstrahlung activation curves by customary methods.[9,10] It was therefore decided to apply it to a number of elements; in this paper we report results on Rh.

We shall show (Sec. IV) that the conventional statistical theory of nuclear reactions is well able to account satisfactorily for all the features of the observed $Rh^{103}(\gamma,\gamma')Rh^{103m}$ cross section, starting from plausible assumptions as to the magnitude and energy-dependence of the cross section, σ_{abs}, for the absorption of electromagnetic radiation. While one cannot exclude the possibility that this agreement with theory be fortuitous, or peculiar to the isotope Rh^{103} (see the discussion of $Au^{197}(\gamma,\gamma')Au^{197m}$ in Sec. IV), we are led to believe that one should conversely be able to estimate with some safety *total* (γ,γ') cross sections from partial (γ,γ') cross sections measured through isomeric excitation.

II. EXPERIMENTAL PROCEDURE

Rhodium metal of high purity in the form of thin (30 mg/cm^2) disks of 14-mm and 24-mm diameter was irradiated in the beam of the University of Chicago betatron. The dimensions of these samples were chosen to achieve a compromise between maximum useful photon flux and uniform sensitivity of our counter and to reduce the contribution of accompanying activities to a minimum; large disks were used at low, small ones at high betatron energies. Disks of pure tantalum of identical diameters and 1 g/cm^2 thickness were juxtaposed with the rhodium samples and irradiated simultaneously in each run above 8 Mev for monitoring purposes. The runs lasted usually two hours.

Activities were counted with a 4π internal proportional flow counter somewhat similar to that of Borkowski.[11] With pure methane as a flow gas and an operating voltage of 4200 v this counter proved to be extremely stable. Calibrated P^{32} sources[12] yielded counting rates in agreement with the absolute activities indicated by the supplier within statistical errors. The efficiency of the counter was hence assumed to be 100 percent for weightless samples of β^- (or e) emitters and its constancy checked throughout the work with secondary standards.

The thickness of the Rh samples used was large with respect to the ranges of the K and L conversion electrons by which the 56-minute isomeric state of Rh^{103} of 40-kev exitation decays.[13] The over-all efficiency of the 4π counter for the Rh^{103m} activity from these thick samples was determined in the following way: One of the Rh disks was bombarded with fast neutrons from the small Chicago cyclotron simultaneously with 250 mg of rhodium chloride juxtaposed to it between Cellophane foils and distributed substantially over the same area (the Rh^{103m} activity is very strongly induced by inelastic scattering of neutrons). The chloride was dissolved in a few ml of water and repeatedly evaporated almost to dryness with concentrated HCl in order to eliminate possible activities from chlorine. The resultant chloride solution was diluted to 500 ml and aliquots of 0.1 ml were evaporated on plastic films of about 25 μg/cm^2 thickness. The activities from these samples and the neutron bombarded Rh disk were then compared to determine the actual efficiency of the counting procedure.

To obtain the counting efficiency for the rather complex Ta^{180} activity a self-absorption curve was taken *inside* the 4π counter by sandwiching an active Ta disk between an increasing number of thin (60 mg/cm^2) Ta disks. The mass absorption coefficient μ was thus determined and the correction calculated according to the formula $[1-\exp(\mu x)]/\mu x$, a fair agreement with the observed activity ratio of Ta disks of different thicknesses was obtained. The corrected saturated activities of the Ta monitoring disks were converted into x-ray doses by adopting the most recent $Ta^{181}(\gamma,n)Ta^{180}$ activation curve of the Sasketchewan group.[14] The term "roentgen," when used in this paper (e.g., Fig. 1), is consequently to be understood according to the definition of Johns *et al.*[15]

A point of the activation curve for $Rh^{103}(\gamma,\gamma')Rh^{103m}$ was taken at 7 Mev, i.e., below the $Ta^{181}(\gamma,n)$ threshold.[16] The dose was at this energy monitored directly with a Victoreen thimble imbedded in a Lucite block, again following the procedure of Johns *et al.*[15]

The contribution of the fast neutron contamination of the betatron beam to the observed Rh^{103m} activity, which could in principle be very important owing to the large inelastic neutron scattering cross section, was estimated by exposing during the same run pairs of Rh and Ta disks on and about 2 in. off the axis of the self-collimated betatron beam. Neglecting variations of the x-ray spectrum with angle and assuming equal neutron intensities at both locations, the activity attributable to neutrons was found to be of the order of 20 percent at 25 Mev and of 6 percent at 18 Mev. As a determination of $\sigma(\gamma,\gamma')$ above 18 Mev from the

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[10] L. V. Spencer, National Bureau of Standards Circular No. 1531, March, 1952.
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[13] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).
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[16] We are indebted for these irradiations, which were carried out with the Illinois Medical School betatron, to Dr. Harvey, and wish also to thank Dr. J. Ovadia and Mr. W. Beattie of his staff for their cooperation.

activation curve was not attempted, no correction for this contamination was applied.

III. RESULTS

Figure 1 shows the activation curve (A) obtained for $Rh^{103}(\gamma,\gamma')Rh^{103m}$. The errors indicated on the graph represent standard statistical deviations only. The cross section curve (σ) obtained from the activation curve by the "photon difference" method[9] of numerical differentiation is also reproduced in the same figure. It is seen to exhibit a resonance-like behavior, with a peak of 12.5 mb height and 2.9 Mev width at 12.8 Mev. Notwithstanding our efforts to count the activity in an absolute way, we estimate the uncertainty in the magnitude of the cross section to be ± 30 percent; similarly, owing to uncertainties in the measured activation curve and to those inherent to the differentiation process, the location of the maximum could possibly be displaced by ± 0.5 Mev. Furthermore, the reliability of this cross-section curve is also somewhat impaired by the fact that considerable weight is given to comparatively few experimental points in the steep part of the activation curve. It can be seen from Fig. 1 that the (γ,γ') process is of considerable magnitude below 9.35 Mev, the measured[17] (γ,n) theshold in Rh^{103}. This is in agreement with the work of the Notre Dame group and is to be expected theoretically.

The limited accuracy of our measurement would not have enabled us to detect any "fine structures" in the energy dependence of the cross section. One would, however, not expect the absorption into non-overlapping levels to be important for a medium heavy nucleus in the energy region investigated here.

IV. DISCUSSION

The salient features of the observed (γ,γ') cross section are its magnitude and the location of its peak. The $Rh^{103}(\gamma,n)Rh^{102}$ cross section is not known experimentally, and hence we cannot adopt it as an approximation to the total absorption cross section. However, from the empirical A dependence of absorption, or rather (γ,n), cross sections we would predict for Rh an absorption cross section of about 350 mb, peaked at approximately 16 Mev. Bearing in mind that the total (γ,γ') cross section is some factor α times ($\alpha \geq 2$) larger than the observed cross section, the probability for reemission of γ-rays from the highly excited target nucleus appears to be surprisingly large in the light of customary (but not necessarily correct) concepts. This finding is, however, in agreement with the results of Cameron and Katz[18] on $Au^{197}(\gamma,\gamma')Au^{197m}$. These authors have already pointed out that such a large width Γ_γ is compatible with electric dipole absorption only.

[17] Sher, Halpern, and Mann, Phys. Rev. 84, 387 (1951).
[18] A. G. W. Cameron and L. Katz, Phys. Rev. 84, 608 (1951).

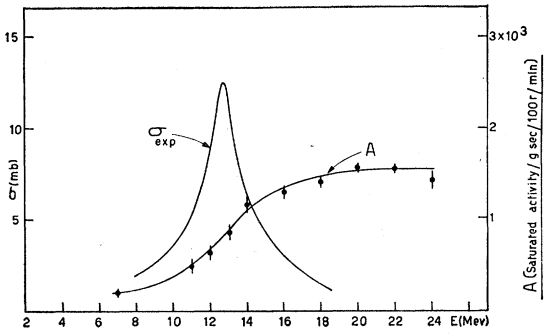

FIG. 1. Energy dependence of activation A and cross section σ_{exp} for the process $Rh^{103}(\gamma,\gamma')Rh^{103m}$.

On the other hand, the (γ,γ') cross section given by Cameron and Katz has its peak at much higher energy (15 Mev) than the Rh cross section reported here; this is particularly striking inasmuch as the peak energies for (γ,n) cross sections are known to decrease with A. The results of the experiment of Cameron and Katz were, however, rather inaccurate, as was pointed out by these authors themselves, and the discrepancy noted here might well be due to experimental reasons.

It is perhaps of interest to make some crude estimates of the magnitude and the energy dependence of the cross section for $Rh^{103}(\gamma,\gamma')Rh^{103m}$ on the basis of the statistical theory of nuclear reactions and to compare them with our results.

For an excitation of E_a Mev, we divide the energy range available for the inelastically scattered γ-rays into two regions: (1) $0 \leq \hbar\omega \leq E_a - \epsilon$, (2) $E_a - \epsilon \leq \hbar\omega \leq E_a$. In region (1), above the neutron binding energy ϵ, γ-emission can be followed by neutron emission; in region (2), not. We can split the total width Γ_γ (at E_a) into two parts $\Gamma_\gamma = \Gamma_< + \Gamma_>$, $\Gamma_<$ and $\Gamma_>$ being the widths for emission into regions (1) and (2), respectively.

Introducing the neutron width, Γ_n, the total absorption cross section $\sigma_{abs}(E_a)$ and a factor α to account for the loss of cascades not leading to the isomeric state, one derives for the *observed* cross section $\sigma(\gamma,\gamma')_{obs}$ the approximate expression

$$\sigma(\gamma,\gamma')_{obs} = \alpha^{-1}\sigma_{abs}(E_a)\left(\frac{\Gamma_\gamma}{\Gamma_\gamma+\Gamma_n}\right)_{E_a}\left\{\left(\frac{\Gamma_\gamma-\Gamma_<}{\Gamma_\gamma}\right)_{E_a} + \left(\frac{\Gamma_<}{\Gamma_\gamma}\right)_{E_a}\left(\frac{\Gamma_\gamma}{\Gamma_\gamma+\Gamma_n}\right)_{E'}\right\}. \quad (1)$$

The subscripts E_a, E' are energies at which the branching ratios are to be evaluated. E' is some mean energy in region (1), and can for instance be taken as the most probable excitation after emission of a photon from E_a; E' thus depends on E_a.

To evaluate $\sigma(\gamma,\gamma')_{obs}$ as a function of E_a, we have

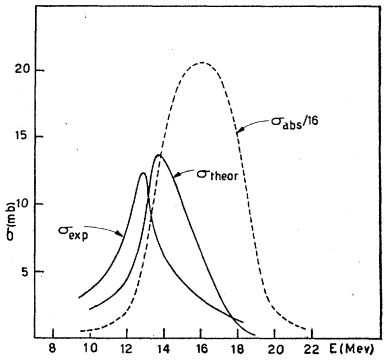

FIG. 2. Comparison of experimental cross section σ_{exp} for $Rh^{103}(\gamma,\gamma')Rh^{103m}$ with the cross section σ_{theor} predicted on the basis of an *assumed* absorption cross section σ_{abs}. Note: σ_{theor} is σ_{obs} of Eq. (1).

determined the quantities which enter (1) as follows:

$$\Gamma_\gamma(E)=\frac{1}{4}\cdot 3\cdot\left(\frac{1}{137}\right)\left(\frac{R}{\hbar c}\right)^2\left(\frac{1}{D_0}\right)\exp[-2(aE)^{\frac{1}{2}}]$$
$$\times\int_0^E(\hbar\omega)^3\exp[2a^{\frac{1}{2}}(E-\hbar\omega)^{\frac{1}{2}}]d(\hbar\omega), \quad (2)$$
$$\Gamma_<(E_a)=\Gamma_\gamma(E_a-\epsilon).$$

This is the electric dipole width formula proposed by Blatt and Weisskopf.[19] It was evaluated assuming $D_0=1$ Mev and using $a=8$ Mev^{-1} given by the same authors. For the nuclear radius R the value 6.6×10^{-13} cm was adopted.

To calculate Γ_n, we start from the formula[20]

$$\Gamma_n=T(E_n)D/2\pi, \quad (3)$$

where D is the average level distance and $T(E_n)$ the transmission coefficient toward the outside for neutrons of energy E_n. We now integrate over all open channels, approximating the sticking probability $\sum_l(2l+1)T(E_n)$ by $(k_nR+1)^2$ (k_n=wave number of a neutron of energy E_n). The result—apart from factors which should appear also in (2) and are irrelevant for our calculations—is:

$$\Gamma_n(E)=(2D/2\pi)\int_0^{E-\epsilon}(k_nR+1)^2\rho_0(E-E_n)dE_n, \quad (4)$$

where $\rho_0(E-E_n)$ is the density of levels with $J=0$ at the final state of the transition. (We have assumed $\rho_J=(2J+1)\rho_0$.)

The quantity α must be at least 2, because after a number of cascades the ground state is reached at least with the same probability as the isomeric state just above. On the other hand, there is a large spin difference between these two states; the isomeric transition in Rh^{103} is classified[13] as $E3(g_{\frac{7}{2}}\rightarrow p_{\frac{1}{2}})$. Whereas a direct re-emission by $E1$ to the ground state is possible (elastic scattering), direct transition to the isomeric state from E_a requires at least $E2$. The quadrupole width $\Gamma_\gamma{}^{(2)}$ at E_a is however much smaller than the dipole width at the same energy. To estimate the factor α we have thus to compare the cascades starting with an $E1$ transition at E_a only. To do this, we have drawn all these cascades (differing in the J of the state at E_a, and the J' at E' to which γ-emission leads) and have evaluated the branching ratios from the statistical factors[21] $S(J,J')$ which govern the transition probabilities between two states of given energy difference for a given multipolarity. These branching ratios, and hence our result, are independent of the matrix elements between these states; they are, however, slightly dependent on the model (the single particle model in Moszkowski's case). We obtain $\alpha\simeq4$, and believe that this number would not change radically by assuming some other model.

Figure 2 shows the assumed absorption cross section σ_{abs} and the result of the calculation, σ_{theor}, together with the experimentally determined cross-section curve σ_{exp}.

The agreement is surprisingly good in view of the crudeness of our assumption and could perhaps be fortuitous. Only further experiments on other elements or a direct measurement of the scattered photons can bear our conclusion out that the commonly accepted notions of statistical theory can well account for the processes following the absorption of electromagnetic radiation by nuclei.‡

It should perhaps be mentioned that in $Au^{197}(\gamma,\gamma')$ one would also expect to find a displacement of the peak towards lower energies owing to the (γ,n) and $(\gamma,\gamma'n)$ competition. Adopting the same σ_{abs} for Au, we find a peak 3.3 Mev wide at 14.2 Mev, in contrast to available evidence. Besides the reasons mentioned earlier, this disagreement might perhaps be due to some neutron contribution to the activity measured by Cameron and Katz, and it would be of interest to reinvestigate the inelastic scattering of photons by Au.

We would like to thank Mr. B. C. Cook for his untiring assistance in operating the betatron, and Dr. M. Gell-Mann for his interesting discussions. One of the authors (C. S. R.) would also like to express his gratitude to Professor S. K. Allison, director of the Institute for Nuclear Studies, for the kind hospitality extended to him.

[19] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, New York, 1952), p. 649.
[20] J. M. Blatt and V. F. Weisskopf, reference 19, p. 389.
[21] S. Moszkowski, Ph.D. thesis, University of Chicago, 1952; Phys. Rev. **89**, 474 (1953).
‡ *Note added in proof:*—Recent measurements by J. Goldemberg and L. Katz [Phys. Rev. (to be published)] on indium, similar to those reported here, bear out our conclusions fully.