Photoexcitation of the Isomeric State of Indium-115*

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The cross section for photoexcitation of In^{115m} has been measured between 1 and 14 Mev. The cross section is found to rise to a narrow peak at 8 Mev, 2 Mev wide, of maximum value 1.2 mb. This peak is interpreted as the result of competition from neutron emission by the excited nucleus. Clear evidence for sharp levels in the excitation cross section is found below 2 Mev and the integrated cross sections for two levels are determined approximately.

THE excitation of nuclei to isomeric states by photon irradiation is interesting because it gives several types of information about the electromagnetic interactions of nuclei. If a nucleus has an isomeric state of conveniently measurable half-life, it is possible to study the inelastic photon scattering process by measuring the production of the isomer. Below particle emission thresholds, where elastic and inelastic photon scattering are the only possible photonuclear reactions, one can sometimes estimate crudely the expected branching ratio for isomer production and thus obtain an indication of the size and shape of the photon capture cross section. The behavior of the isomer excitation cross section near photoneutron threshold should show whether or not competition plays a role in nuclear photon scattering. And finally, at low energies the cross section might be expected to give information about the level structure of the nucleus being studied. All of the above points have been investigated by other workers in the past.[1–5]

The matrix element for direct excitation of an isomer is of course exceedingly small, this being the criterion for the existence of the isomeric state. Instead, isomer production must proceed by way of an excited intermediate state (only isomeric states of the target nucleus will be considered), although a compound nucleus in the sense of the statistical theory may not necessarily be formed. The excited intermediate state itself can decay in several ways: by particle emission with or without accompanying photon emission, if the energy is high enough, or by photon emission alone, directly or by cascade to the ground or isomeric state of the target nucleus.

In the experiments to be described below, the isomer chosen for study was In^{115m}, previously investigated in similar experiments at Notre Dame[2] and Saskatchewan.[4] The relatively long half-life (4.5 hours) and strongly converted gamma ray of this isomer make possible the use of standard Geiger-counting techniques, whose absolute efficiency can be determined with fair reliability. Also the large natural abundance (96%) of In^{115} is important because of the small cross section for the isomer-excitation reaction, while the absence of stable neighboring indium isotopes eliminates confusion from other possible reactions leading to In^{115m}. The spin of the ground state is 9/2, positive parity, and that of the isomeric state is $\frac{1}{2}$, negative parity; the 0.335 Mev isomeric transition is thus magnetic 2^4-pole. The conversion coefficient is known experimentally and predicted theoretically to be very nearly unity.[6]

I. EXPERIMENTAL PROCEDURE

A. High Energy Experiment

Two essentially separate experiments have been performed. In the first, two 2-mil indium foils were activated in each run by placing them on either side of a 5-mil tantalum radiator in the external analyzed electron beam of the Massachusetts Institute of Technology linear accelerator. This stacked-foil technique was used to obtain the highest possible yields from thin-target bremsstrahlung and also to facilitate background subtraction. Since the electron beam was to a first approximation unchanged in passing through the three foils, the electrodisintegration plus background activities in the two indium foils were approximately identical and the difference in activity was caused by forward-directed bremsstrahlung made in the tantalum foil. Runs were made at a series of electron energies from 2 Mev to 14 Mev, the high current output of the accelerator (approximately 0.2 μa analyzed beam) and the favorable exposure geometry making possible the low energy runs where activities were small. The lowest point (1.88Mev) was taken using only the Van de Graaff injector at a current of 1.25 μa. Exposures ranged from 15 to 90 minutes. To decrease radiation contamination, the electron beam collimators fitted to the linear accelerator were made of carbon. The beam cross section was 0.5 in. by 1 in., and the energy resolution of the analyzer was 3%. Beam current was measured by stopping the elec-

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trons in a carbon Faraday cage, shielded against secondary emission and grounded through a low impedance dc amplifier. To suppress ionization currents the Faraday cage was placed inside an evacuated chamber, with a thin aluminum window to admit the electron beam, the foils under exposure being placed between the vacuum chamber and the linear accelerator exit port. An excellent account of an almost identical experiment on copper has been given by Berman and Brown.[7]

As a check on the reliability of the exposure technique, several runs were made using a more conventional geometry. A thin foil of thorium was placed in front of enough graphite to absorb the electron beam, and this assembly was placed inside the analyzer vacuum chamber at the end of the collimator, with provision made for measuring beam current. Since the low atomic number of carbon makes it a very poor radiator, the radiation produced by this target was essentially thin-target bremsstrahlung from the thorium. Several runs were then made with indium foils placed outside the vacuum chamber in this bremsstrahlung beam, and the results agreed with those obtained with stacked foils. Because the broad angular distribution of the radiation at low energies made it difficult to intercept all the bremsstrahlung with the indium foil (separated from the radiator by approximately one inch of carbon), this technique was less satisfactory than the stacked foil method and was therefore abandoned.

Since inelastic neutron scattering can also produce In^{115m},[8,9] an experiment was performed to check the remote possibility that the observed activity differences between front and rear foils might be caused by neutrons made in the radiator or elsewhere in the target room. Obviously this effect could only occur at high energies, above neutron threshold. A single indium foil was placed behind a four-inch lead target in the undeflected beam of the accelerator and exposed for 15 minutes at 2 μa and 13.5 Mev. The copious neutron flux thus produced gave rise to some neutron capture activity (54 minute In^{116m}) but no observable inelastic scattering. The upper limit thus placed on the ratio of (n,n') to (n,γ) assured that the neutron-produced activity in the separate foils was negligible, and hence that the neutron contribution to the rear-front difference was negligible to a higher order.

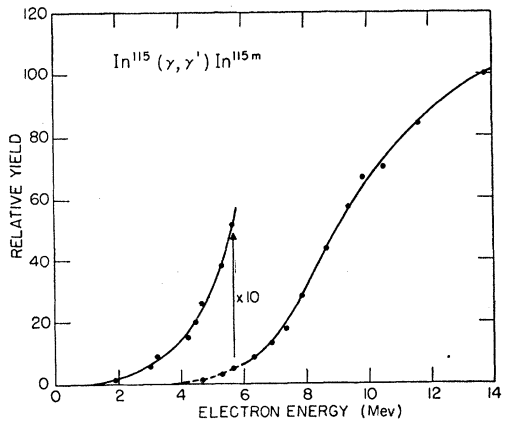

FIG. 1. Activation curve for the photoexcitation of In^{115m}, using thin-target bremsstrahlung.

The activated foils were counted on standard end-window Geiger counters. Activity was plentiful at the higher energies, but below 4.5 Mev the net counting rates were generally below background, so that statistical errors on individual points became large. Above 5 Mev, these errors are estimated to be less than $\pm 5\%$; at lower energies they may become as high as $\pm 20\%$, but in this region other corrections probably mask the statistical errors anyway. To determine the absolute efficiency of the Geiger counting procedure an activated indium foil was counted first in the standard manner and then in a 4π methane flow counter. Knowing this efficiency and the decay scheme of the isomer one can determine the true number of disintegrations occurring in the foil and hence, with the use of the bremsstrahlung spectrum, the magnitude of the activation cross section.

B. Low-Energy Experiment

The second experiment was performed with the Electrical Engineering Department Van de Graaff generator in Building 28 at Massachusetts Institute of Technology. Thick target bremsstrahlung was produced by stopping the electron beam in a $\frac{1}{4}$-inch thick water-cooled gold target at the base of the accelerating tube. Single 12-mil indium foils were exposed immediately below the aluminum window at the end of the tube, approximately one inch from the target. The target and tube extension were electrically connected and the current to them measured by a microammeter, all runs being made with 200 microamperes beam current. Machine energy was measured with a generating voltmeter whose linear voltage scale was calibrated against the beryllium and deuterium photoneutron thresholds and is estimated to be accurate to within ± 50 kv. A very satisfactory check on its accuracy was obtained by comparison with the energies of Na^{24}, Co^{60}, and Cs^{134} gamma rays, using scintillation counters.

The activation curve for In^{115m} produced by thick-target bremsstrahlung was obtained with the above apparatus between 1 and 2 Mev, using the same counting technique as in the high-energy experiment. Counting errors were less serious for the present experiment because of the high electron beam current employed. The bremsstrahlung spectrum of the machine was measured in a separate experiment reported elsewhere.[10]

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II. RESULTS AND DISCUSSION

A. High-Energy Experiment

In the linear accelerator experiment several corrections to the raw activity data were needed, besides the trivial normalizations to standard beam current, exposure time, and foil mass. The electrodisintegration contributions to the activities in the foils are different in that the electron beam energy is lower in the rear foil than in the front, and the self-induced photodisintegration activities (produced in each foil by bremsstrahlung from the same foil) differ for the same reasons. Corrections for these effects were made using the energy-loss data of Paul and Reich[11] and Goldwasser *et al.*,[12] in conjunction with the electrodisintegration theory of Blair[13] and experiments of Scott *et al.*[14] and Brown and Wilson.[15] These energy corrections were relatively unimportant at high energies, but below about 4 Mev their magnitude increased considerably, amounting to roughly a factor of two on the individual foil activities at 2 Mev. The large correction arises from the fact that the total foil thickness for the three foils (0.186 g/cm²) corresponds to an average energy loss of about 0.25 Mev for 2 Mev incident electrons. No correction was needed for background activities produced by photon contamination of the electron beam because these activities were equal in the two foils.

With the above corrections the activation curve plotted in Fig. 1 was obtained. It shows the relative yield of In^{115m} produced by the bremsstrahlung radiation from one indium and one tantalum foil bombarded with electrons of various energies. This activation curve was differentiated numerically using the Bethe-Heitler extreme relativistic bremsstrahlung spectrum, including screening,[16] at 0.5-Mev intervals, and the resulting cross section is shown in Fig. 2. The uncertainty of the cross section in the peak region is ~30%, assuming the Bethe-Heitler spectrum is correct. The position of the peak is sensitive to the exact shape of the activation curve and is thus uncertain by about ±1 Mev. The full width at half-maximum, likewise sensitive to the activation curve shape, is 2 Mev, with an estimated uncertainty of ±1 Mev. The low-energy end of the cross-section curve may be overestimated somewhat by the use of the extreme relativistic bremsstrahlung formula.

The above cross section compares reasonably well in the location of the peak, but not in width or magnitude, with that of the Saskatchewan group[4] which finds a peak value of 2.2×10^{-27} cm² (corrected to an internal conversion coefficient of unity) at 9 Mev with a width of 9 Mev. That the narrower width is more plausible can be seen from the following considerations. First, it is apparent from both experiments that the peaked shape does not coincide with the dipole resonance maximum, but rather that the isomer excitation seems to peak near the (γ,n) threshold around 9 Mev. Neutron emission should be much more probable than photon emission when both are energetically possible, so that one would expect the isomer excitation cross section to fall off rapidly above photoneutron threshold, forming a sharp peak as observed in this experiment. Further evidence in support of this view is contributed by the Rh^{103m} excitation cross section measured at Chicago,[5] which has a width of 3 Mev, and the photon scattering cross sections investigated at the National Bureau of Standards,[17] which show clearly the effects of neutron emission competition.

With this explanation of the peak in terms of competition, it would seem that the photon capture cross section should be smooth in the neighborhood of the neutron threshold, because capture and emission are then not directly related. Since the photoneutron cross section is a good representation of the capture cross section just above threshold, one would therefore expect the total photon capture cross section below neutron threshold to join smoothly on to the extrapolated photoneutron curve. Using the Saskatchewan In^{115} (γ,n) cross section,[4] it appears that the peak value of the photon capture cross section to be expected just below neutron threshold should be roughly 50×10^{-27} cm². To compare this value with the measured cross section, one must estimate the fraction of excited nuclei which decay into the isomeric state. Since low multipole photon transitions are strongly favored when suitable spin states are available, a nucleus excited out of a 9/2+ state is much more likely to return to that state than to a nearby ½− state. By analogy with some measured isomer/ground state ratios in (γ,n) reactions, Goldemberg and Katz estimate as ⅕ the fraction going to the isomer by photon de-excitation in In^{115}. This estimate seems unconvincing, however, since selection rules and statistical factors for neu-

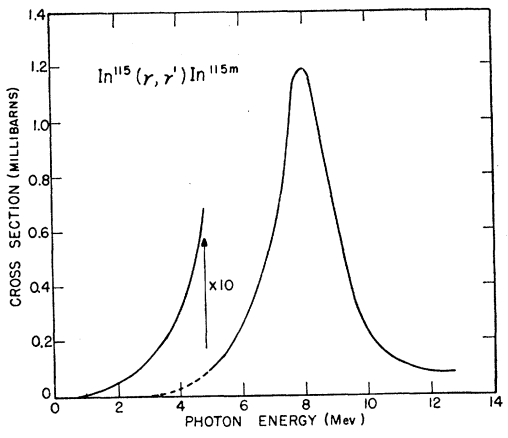

FIG. 2. Cross section for the photoexcitation of In^{115m}.

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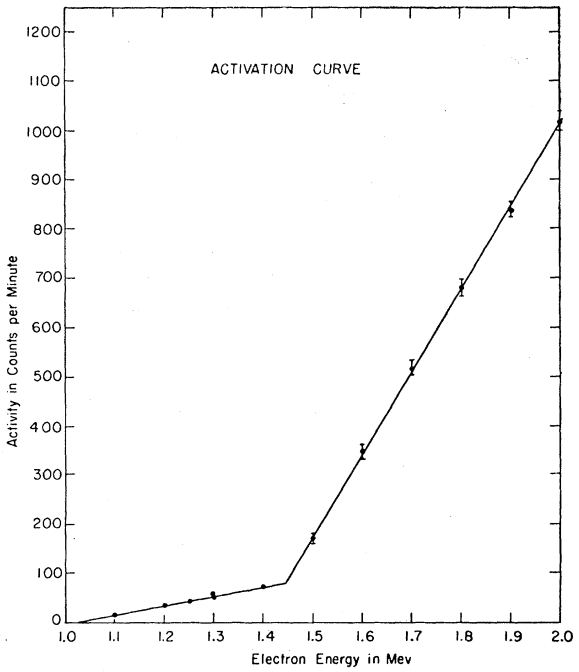

FIG. 3. Activation curve for the photoexcitation of In^{115m} at low energies, using thick-target bremsstrahlung.

tron and photon de-excitation are quite different. Moreover, Telegdi estimates the isomer/ground state ratio in rhodium to be $\frac{1}{4}$, and Rh^{103m} is only an $E3$ isomer, considerably less forbidden than the $M4$ In^{115m} case. His 12 millibarns peak measured cross section thus leads to about 50×10^{-27} cm^2 for the capture peak in rhodium, in good agreement with the above expectation. It would appear that a branching ratio of the order of 1/50 for In^{115} near the peak of the isomer cross section is by no means out of the question. Since the branching ratio is very likely energy-dependent, one cannot deduce a curve for the photon absorption cross section *versus* energy.

B. Low Energy Experiment

The activation curve obtained in the low energy experiment is shown in Fig. 3. The experimental errors are shown on the graph and are primarily statistical in origin. Where no vertical bar is indicated, the estimated uncertainty is smaller than the size of the corresponding point. This activation curve is excellent agreement with that obtained by the Notre Dame group,[2] both in shape and location of breaks.

In order to obtain a cross section from the activation curve of Fig. 3, it is necessary to know the thick-target bremsstrahlung spectrum from the particular target used in this experiment. Since this spectrum should be a relatively smooth function of energy, it is evident that the cross section between 1 and 2 Mev consists mainly of sharp lines at 1.02 ± 0.05 Mev and 1.45 ± 0.05 Mev, with the possibility of some smaller unresolved levels and/or a continuous contribution as well. The only reason for repeating these results, all of which were obtained by the Notre Dame group earlier, is that the shape and intensity of the bremsstrahlung spectrum used here has been measured,[10] and it is thus possible to obtain values for the integrated cross sections of the two levels detected. Assuming that only these levels are important, it is found that $\int\sigma dE$ for the 1.02-Mev level is 2×10^{-33} Mev-cm^2, and that for the 1.45-Mev level is 1×10^{-32} Mev-cm^2, where σ is the cross section for photoexcitation of the isomer, not the photon capture cross section. The estimated uncertainty of these values is of the order of a factor of four, resulting primarily from the difficulty of measuring the slope of the bremsstrahlung spectrum accurately.

For sharp lines of the sort excited in this experiment, one would expect an integrated photon-capture cross section of the order of $\lambda^2\Gamma$, where λ is the wavelength of the resonance x-rays and Γ the partial width of the level for decay directly to the ground state. If the fraction of the excited nuclei which decay into the isomeric state is called β, it is then expected that $\int\sigma dE$ as measured here should be of the order of magnitude of $\lambda^2\Gamma\beta$. Thus for the 1.45-Mev level, using the experimental integrated cross section, $\Gamma\beta$ should be about 10^{-12} Mev.

In the absence of detailed knowledge of the level scheme and transition matrix elements of In^{115}, one can only comment that this number appears reasonable, indicating that the observed transition is relatively improbable. (For example, Γ might be expected to be of the order of millivolts, making β about 10^{-3}.) It is interesting to note that Varma and Mandeville[18] found a level in In^{115} at 1.42 Mev, but they did not observe direct transitions between it and the ground state or any transitions connecting it with the isomeric state. In fact, none of the levels they observed connects directly or by cascade to both the ground and isomeric states with measurable intensity, which fact agrees with the small value of $\Gamma\beta$ quoted above and the absence of evidence in the isomer excitation cross section for the several other levels shown in their paper.

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