Photoalpha Reaction in Sb^{121}†

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The $Sb^{121}(\gamma,\alpha)In^{117-117m}$ reaction has been studied by determining the radioactivities of the product nuclei in samples that were irradiated with bremsstrahlung of maximum energy varying from 15.5 to 24 Mev. Excitation functions for the total cross section and the cross sections to each isomer were obtained. The total cross section rises steadily over the energy range studied, reaching a value of 360 μb at 24 Mev. The ratio of the cross section for the direct production of the ground state to the cross section for the production of the isomer is constant over the energy range studied at a value of 2.60 ± 0.40. The total excitation function up to 18 Mev agrees well with cross sections calculated on the basis of a statistical theory for compound-nucleus decay. The cross-section predictions could not be made for higher energies. The observed ground-state-to-isomer cross-section ratio is consistent with that expected from a compound-nucleus mechanism and probably inconsistent with that expected from a direct mechanism. Thus, the reaction appears to involve compound-nucleus processes for the most part.

I. INTRODUCTION

PAST work on photonuclear reactions in which alpha particles are emitted seems to indicate that they are mainly compound-nucleus processes.[1–3] However, since the amount of the available data is quite limited, it is not known yet to what extent this is true. Also, very little of this experimental information is detailed in nature. Most of it has involved yield measurements made with bremsstrahlung of a single maximum energy, with detailed excitation functions having been determined for only seven cases outside of the light element region.[3–9] Thus, since it appears to be highly desirable to obtain additional information of a detailed nature about photoalpha reactions, we have made a study of the (γ,α) reaction in Sb^{121}.

The main reason for choosing this target nucleus was that In^{117} (the product nucleus) has an isomeric state. The determination of the relative yields of the isomeric and ground states (the so-called isomer ratio) gives an additional piece of information besides the excitation function that can be used in determining the mechanism of the reaction. Erdös *et al.*[8] have previously determined an excitation function for the production of the isomeric state, but they were not able to measure the direct yield to the ground state. They determined the yield by counting the β emission from indium samples that were chemically separated from the target material. We have chosen to use gamma-ray spectroscopy techniques in the present study to facilitate the observation of both the isomeric and ground state yields.

II. EXPERIMENTAL

Seventy-gram targets of saturated solutions of antimony trichloride were irradiated for 90 min in the external bremsstrahlung beam of the University of Illinois 22-Mev betatron. The only reactions that are important at the energies used in this study that produce indium isotopes are the $Sb^{121}(\gamma,\alpha)In^{117-117m}$ and the $Sb^{123}(\gamma,\alpha)In^{119-119m}$ reactions. Since the half-lives and decay properties of the In^{119} isomers are very different from those of the In^{117} isomers,[10] the $In^{119-119m}$ does not interfere with the determination of the $Sb^{121}(\gamma,\alpha)$ yield. Because of the large yields of (γ,n) reactions in this target material, it was necessary to chemically separate an indium fraction in order to observe the products of the (γ,α) reaction. This was done by precipitating indium with 8-hydroxyquinoline.[11] The chemical separation normally took about 1 hr. Corrections were made for the chemical yields in determining the absolute (γ,α) reaction yields.

The bremsstrahlung beam was monitored with an aluminum-walled air-filled ionization chamber placed behind a 2-in. thick lead converter in a geometrical position such that the chamber subtended the same solid angle as the target sample. The ionization current collected in the chamber was measured with a vibrating reed electrometer circuit. For the absolute yield measurement, the monitoring system was calibrated with a Victoreen "r" thimble to give the dose in roentgens. In the calibration, corrections were made for the beam distribution across the face of the sample and for absorption in the sample.

The radioactivity of the indium samples was determined with a gamma ray spectrometer consisting of a

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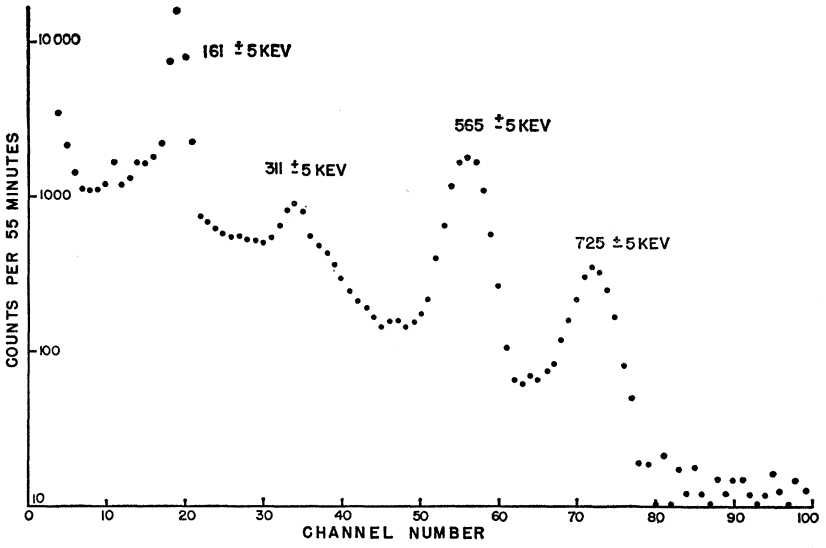

FIG. 1. Typical gamma-ray spectrum of an $In^{117-117m}$ sample.

$1\frac{1}{2}$-in.-diameter by $1\frac{1}{2}$-in.-long NaI(Tl) crystal coupled with a 100-channel pulse-height analyzer. Three 55-min counts were made on each sample, starting 1 hr after the end of the irradiation. A typical gamma-ray spectrum is shown in Fig. 1. Reference to the decay scheme shown in Fig. 2 shows the origin of the various photopeaks seen in the spectrum. (This decay scheme is a modification of the decay scheme reported by McGinnis.[12] The modifications were determined during the preliminary stages of this study, and they will be reported in detail elsewhere.) The 161-kev peak arises from the decay of both In^{117m} and In^{117}. The 311-kev peak arises only in the decay of the isomer. The 565-kev peak is associated only with the ground-state decay, and the 725-kev photopeak is a coincidence sum peak (161 kev plus 565 kev) associated with the ground-state decay. (The peaks associated with the ground-state decay will have a decay curve that involves both growth and decay because of the 28% isomeric transition in the decay of the isomer.)

The numbers of ground-state and isomer nuclei present at the end of each irradiation can be determined from the counting data in several different ways. Because of the growth of ground-state nuclei as a result of the decay of the isomer, the number of 161-kev events in two of the counts on a sample can be used along with decay and growth equations to determine the numbers of ground-state and isomer nuclei present at the end of the irradiation. The 565-kev events can also be used in the same way; or the 311-kev events and the 161- or 565-kev events in the same count can be used. Because the counting statistics associated with the 161-kev photopeak were much better than for the other photopeaks, the 161-kev events in two of the counts on each sample were used in making the analysis. The alternative methods were applied in several cases and gave results consistent with those gotten from only the 161-kev data. In this analysis, corrections were made for the geometry, crystal efficiency,[13,14] absorption in the crystal housing,[14] the abundances of the gamma rays in the decay schemes, and for the fact that the primary gamma-ray spectrum was modified by the coincidence summing of 161- and 565-kev photons in the ground-state decay.[15]

The calculation of the direct nuclear reaction yields of the isomer and ground state follows from the number of isomer and ground-state nuclei present at the end of the irradiation. In this calculation, the straightforward correction for the production of ground-state nuclei by decay of the isomer during the bombardment period must be made in order to get the direct nuclear reaction yield to the ground state. This correction involves the use of standard decay and growth relationships.

III. RESULTS

The ratio of the direct yields to the ground state and to the isomer was determined for each run. This ratio was constant to 7% at a value of 2.60 over the range of energies studied (15.5 to 24 Mev). If one includes a contribution arising from the uncertainties in the decay scheme, the yield ratio with its uncertainty becomes 2.60 ± 0.40. The constancy of the yield ratio as a function of betatron energy means that the ratio of cross sections as a function of gamma-ray energy is also constant.

The activation curve for the production of the ground state plus the isomer (total yield) is shown in Fig. 3, where the number of reactions per mole of Sb^{121} per roentgen is plotted against the bremsstrahlung maxi-

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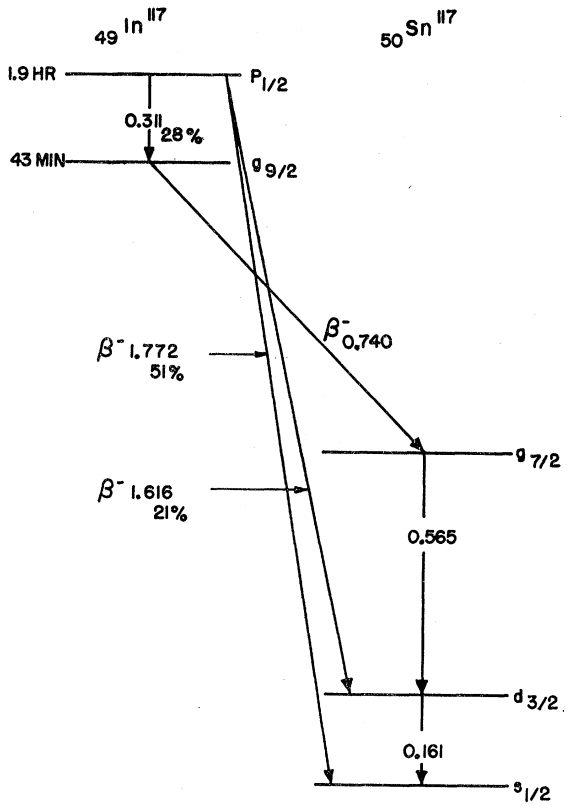

FIG. 2. $In^{117-117m}$ decay scheme.

mum energy (betatron energy). The photon difference method[16] was applied to extract the cross sections from the smoothed yield curve. The resulting total cross section as a function of gamma-ray energy is shown in Fig. 4. Also shown in Fig. 4 are the cross sections for the production of the isomer and for the ground state gotten by using the value of 2.60 for the ratios of the two cross sections. The excitation function measured by Erdös[8] for the production of the isomer is also shown. One notes poor agreement between the two excitation functions for the isomer production. However, a large part of the disagreement can be eliminated by shifting one or both of the curves along the energy axis.

IV. DISCUSSION

A. Total Cross Section

The circles in Fig. 4 represent cross sections that were calculated on the basis of compound nucleus formation followed by the evaporation of alpha particles. These predicted cross sections were obtained by calculating the ratio of the (γ,α) cross section to the (γ,n) cross section from a statistical theory for compound nucleus decay[17] and multiplying this ratio by the experimental $Sb^{121}(\gamma,n)$ cross section reported by Katz and Cameron.[16] Since the details of a similar calculation for the $V^{51}(\gamma,\alpha)$ reaction have been recently described,[3] they will not be reproduced here. One notes good agreement between the calculated cross sections and those obtained from the experiment. The extent of the agreement is probably accidental, since many other choices of certain parameters (nuclear-radius and level-density parameters, for instance) could have been used in the calculation. A change in the calculated values by a factor of two could be obtained without using unreasonable choices for these parameters. One very serious limitation in these calculations is the lack of (γ,n) cross-section data above 18 Mev. The (γ,n) cross section reaches its maximum value at 14.5 Mev. Above 18 Mev the cross section is quite small and very difficult to measure accurately with bremsstrahlung techniques. Thus, it appears that the compound-nucleus calculations can only serve as a detailed test of the reaction mechanism over a relatively small energy range. Over this small range, we do observe satisfactory agreement.

B. Ground-State to Isomer Cross-Section Ratio

Next, it is of interest to see what the observed ground-state to isomer cross-section ratio indicates about the

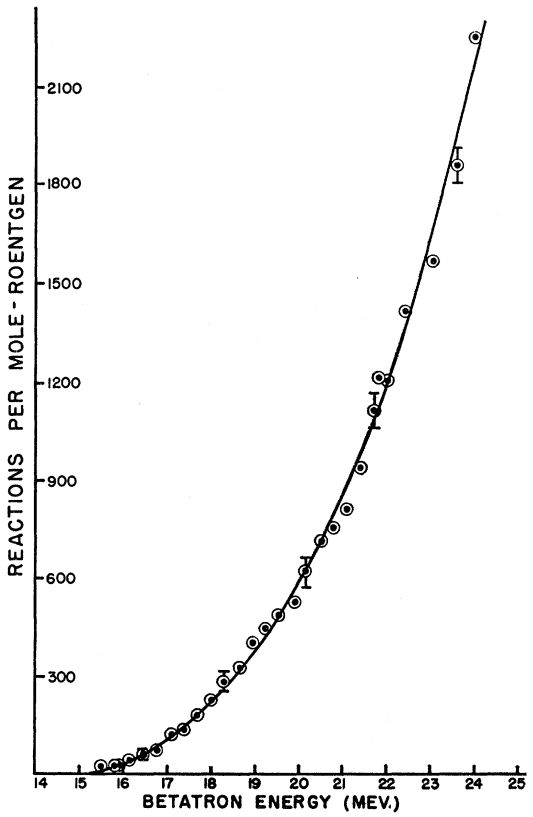

FIG. 3. Activation curve for the total yield of the $Sb^{121}(\gamma,\alpha)$ reaction

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reaction mechanism. Huizenga and Vandenbosch[18,19] have recently discussed the theoretical calculation of isomeric cross-section ratios for compound-nucleus reactions, and they have applied these calculations to several different situations. Their calculation of several isomer ratios for (γ,n) reactions is of direct interest here, and we will follow their methods very closely.

Basically, the calculations involve following the distribution of nuclear spins through the various steps of the reaction and then deciding on the relative populations of the ground state and isomer in the final step. The steps to be considered are the absorption of the gamma ray by the Sb^{121} target to give the compound nucleus, the evaporation of the alpha particle to give an excited In^{117} nucleus, and the de-excitation by the gamma-ray cascade that eventually leads either to the isomer or to the ground state. We next discuss the various steps in detail.

Following Huizenga and Vandenbosch,[18] we assume that all of the gamma-ray absorption by Sb^{121} is electric dipole in nature. Since the spin of Sb^{121} is $\frac{5}{2}$,[10] dipole absorption would lead to compound states of spin J_c equal to $\frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$. It is further assumed that the relative abundance of each spin state is proportional to $(2J_c+1)$.

Next, this distribution is modified by the emission of alpha particles which carry away various amounts of angular momentum. For a given compound nucleus spin J_c the relative probability for the emission of alpha particles that lead to a final state with angular momentum of J_f is given by[19]

$$P(J_f) \propto \rho(J_f) \sum_{l=|J_c-J_f|}^{J_c+J_f} T_l(E), \qquad (1)$$

where $T_l(E)$ is the barrier transmission coefficient for an alpha particle of energy E and orbital angular momentum l and $\rho(J_f)$ is the nuclear level density for spin J_f. The spin dependence of the nuclear level density is given by [20,21]

$$\rho(J)=\rho(0)(2J+1)\exp[-(J+\tfrac{1}{2})^2/2\sigma^2], \qquad (2)$$

where $\rho(J)$ is the density of levels of spin J, $\rho(0)$ is the density of levels for spin zero, and σ is a parameter that characterizes the distribution. Although the calculations were made for several values of the parameter σ, it should be pointed out that Huizenga and Vandenbosch were successful in fitting the measured isomer ratios for several different reactions by using a value of about 4 for σ.

The calculation of the spin distribution after the emission of the alpha particles proceeds by applying Eq. (1) to each of the compound-nucleus spin states

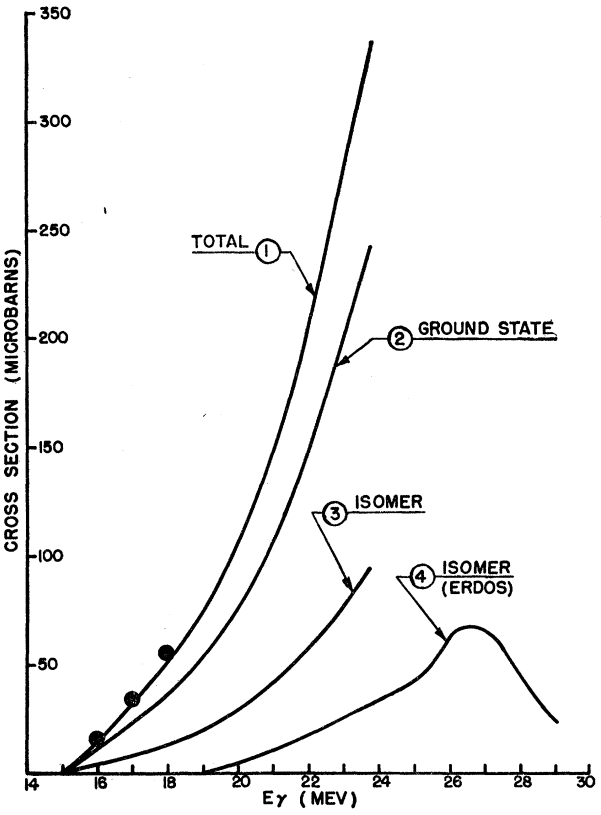

FIG. 4. $Sb^{121}(\gamma,\alpha)$ cross sections as a function of gamma-ray energy. Curves 1, 2, and 3 are the results of this study. Curve 1 is the total cross section. Curve 2 is the cross section for the production of the ground state. Curve 3 is the cross section for production of the isomeric state. Curve 4 is the cross section for the production of the isomeric state previously reported by Erdös *et al.*[8] The closed circles are total cross sections calculated on the basis of a compound-nucleus mechanism.

populated in the original gamma-ray absorption step. In this calculation, the transmission coefficients for the alpha particles were taken as those for an alpha particle whose energy corresponded to the mean kinetic energy given by the evaporation calculations referred to in part A. This approximation was shown to be quite accurate for the case of neutron emission by Huizenga and Vandenbosch. Two different sets of alpha-particle transmission coefficients were used. Those derived from a square-well potential were taken from Feshbach *et al.*[22]; coefficients calculated on the basis of the optical model were taken from the work of Huizenga and Igo.[23]

Next, the resulting distribution is further modified by the gamma-ray cascade that follows the emission of the alpha particle. We have assumed that only dipole radiation is emitted in the cascade except for the last gamma ray. For each step, the spin distribution is calculated assuming that the relative population of the

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TABLE I. Calculated ground-state to isomer cross-section ratios for different sources of the alpha-particle transmission coefficients (square well and optical model), various values of the parameter σ, and various numbers (N_γ) of gamma rays in the de-excitation cascade for a compound nucleus excitation energy of 22 Mev.

	$N_\gamma=2$	$N_\gamma=3$	$N_\gamma=4$
$\sigma=4$ (square-well)	2.18	2.27	2.38
(optical model)	2.53	2.59	2.67
$\sigma=6$ (square-well)	2.98	3.27	3.43
(optical model)	3.60	3.88	4.05
$\sigma=\infty$ (square-well)	4.11	4.56	5.00
(optical model)	5.14	5.60	6.00

various possible spin states is given by the $\rho(J)$ in Eq. (2). The last gamma ray in the cascade is assumed to populate the final state (the ground or isomeric state) that is closest in spin to the emitting state. For emitting states having a J of $\frac{5}{2}$, equal populations to the ground and isomeric states are assumed. The calculations were made for several different numbers of gamma rays in the cascade.

The results of the calculations for a compound nucleus excitation energy of 22 Mev are shown in Table I, where the calculated ground to isomer cross-section ratio is given for various combinations of the two sources of the transmission coefficients, three values of the level density parameter σ, and three different gamma-ray cascades. In this case, the evaporation calculations gave a mean alpha-particle kinetic energy of 13.5 Mev. After taking account of the 3.4-Mev threshold for the reaction,[24] the average energy associated with the gamma-ray cascade becomes 5.1 Mev. We would expect an average of about three gamma rays per cascade for this energy. One notes good agreement between the experimental and calculated cross-section ratios for this number of gamma rays in the cascade and a σ value of 4, using the optical-model transmission coefficients, or a σ value of about 5, using the square-well coefficients. Thus, the compound-nucleus calculations based on a reasonable value for the parameter σ give good agreement with the experimentally measured ratio. Furthermore, the energy dependence of this ratio is expected to be very slow in this energy region mainly because the average energies of the evaporation spectra do not change very rapidly with changes in the compound nucleus excitation energy. For example, calculations made for a σ of 4 and a compound-nucleus excitation energy of 15 Mev gave results that are only 10% smaller than those appearing in Table I.

No detailed theory for direct photoalpha reactions has as yet been proposed. However, qualitatively, we might expect the direct reaction to give us resulting nuclei with their spins closer to the target nucleus than the evaporation process does. This would lead to comparable numbers of ground-state and isomer nuclei. A quantitative estimate can be made on the following basis. If we assume that the vast majority of the direct alpha-particle emission processes involve neutrons and protons that were originally paired in the nuclei in much the same way that favored alpha decay in odd mass nuclei seems to involve this,[25] we would expect the excited In^{117} nuclei to be born with a spin of $\frac{5}{2}$ (due to the same odd proton that is responsible for the $\frac{5}{2}$ spin of Sb^{121}). Since these nuclei would probably have relatively low excitation energies, they would be expected to de-excite by only one, or, at most, two gamma-ray transitions. For de-excitation involving only one gamma ray, we would expect a ground to isomer ratio of unity. For two gamma rays in a cascade and a σ of 4, we would expect a value of 1.27 for the ratio. Thus, we would predict much smaller values for the ground to isomer cross section ratio than are obtained experimentally.

We thus conclude that the $Sb^{121}(\gamma,\alpha)$ reaction is a compound nucleus reaction on the basis of both the magnitude of the observed cross section and the observed ground-state to isomer cross-section ratio.

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