

## Excitation Functions for Inelastic Scattering Reactions of Cadmium-111 and Indium-115\*

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(Received August 4, 1960; revised manuscript received September 15, 1960)

Excitation functions have been measured for the formation of  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$  in the bombardment of  $\text{Cd}^{111}$  and  $\text{In}^{115}$ , respectively, with 6–10 Mev protons, 10–20 Mev deuterons, and 15–40 Mev alpha particles. The excitation function for the  $\text{In}^{115}(\alpha, \alpha n)\text{In}^{114m}$  reaction has also been obtained. All the excitation functions increase monotonically with energy. The cross sections for the formation of  $\text{In}^{115m}$  at the highest bombarding energies are 1.5 mb, 42 mb, and 5.2 mb, for the proton-, deuteron-, and alpha-induced reactions, respectively. The corresponding cross sections for the formation of  $\text{Cd}^{111m}$  are 2–3 times larger. The results have been compared with the predictions of the statistical theory. The only definite conclusion can be drawn in the case of the  $(\alpha, \alpha \gamma)$  reaction where it is shown that an evaporation mechanism cannot account for the measured cross sections at bombarding energies above 25 Mev. The contribution of Coulomb excitation to the observed yields has been calculated. It is found that this process contributes substantially to the  $(\alpha, \alpha \gamma)$  and  $(p, p \gamma)$  reactions, particularly at low bombarding energies.

### I. INTRODUCTION

A NUMBER of studies of nuclear reactions involving inelastic scattering of medium energy particles by medium weight elements have been performed in recent years.<sup>1–4</sup> In these studies the angular distribution and energy spectra of the scattered particles were measured. The following are the main results of these experiments: The scattered particles are predominantly emitted in a forward direction; the emission of energetic particles is far more probable than would be expected on the basis of an evaporation mechanism; certain groups of levels of the residual nucleus appear to be particularly favored. These results have been widely interpreted as indicating that these reactions primarily involve a direct-interaction mechanism.

Reactions of this type have also been investigated by the measurement of excitation functions for  $(\alpha, \alpha n)$ ,  $(\alpha, \alpha p n)$ , and  $(p, p x n)$  reactions.<sup>5–11</sup> A detailed comparison of the results of these studies with the predictions of the statistical theory has been performed recently by application of the Monte Carlo technique to low-energy evaporation processes.<sup>12</sup> It was shown in this study that fairly good agreement between experiment and theory could be obtained for a large

number of reactions up to an excitation energy of approximately 40 Mev, provided that proper consideration was given to such factors as pairing energies, closed shells, etc.

It is at first sight somewhat disturbing that experiments on the particles emitted in inelastic scattering reactions should favor a direct-interaction mechanism, while the corresponding measurements on the residual nuclei are more consistent with an evaporation mechanism. A closer examination of the situation indicates that the results on the scattered particles are to a large extent based on measurements of the most energetic particles emitted in these reactions. This contrasts with the situation of the excitation function measurements for the above reactions because the energy of the inelastically scattered particles in  $(\alpha, \alpha n)$  or  $(p, p x n)$  reactions is limited by the fact that additional particles are emitted. In order to be more comparable with the experiments on inelastically scattered particles, an excitation function measurement would thus have to be performed for a reaction in which the incident particle is the only emitted particle. An excitation function measurement for such a reaction, based on the observation of the radioactive decay of the residual nucleus, is only possible for those cases where the target nucleus has a reasonably long-lived isomeric level. The measured cross section for the formation of this isomeric level constitutes only a partial determination of the cross section for the reaction of interest since only a fraction of the excited levels of the residual nucleus preferentially deexcite to the isomeric level. Fortunately, however, the shape of the excitation function for a particular reaction appears to be rather insensitive to the spin of the observed isomeric level,<sup>13</sup> so that a comparison with the statistical theory is possible.

The present work reports the measurement of excitation functions for the formation of  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$ ,

\* Research performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup> H. W. Fulbright, N. O. Lassen, and N. O. Poulsen, *Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd.* **31**, 10 (1959).

<sup>2</sup> J. L. Yntema and B. Zeidman, *Phys. Rev.* **114**, 815 (1959).

<sup>3</sup> B. L. Cohen and A. G. Rubin, *Phys. Rev.* **111**, 1568 (1959).

<sup>4</sup> G. Igo, *Phys. Rev.* **106**, 256 (1957).

<sup>5</sup> N. T. Porile and D. L. Morrison, *Phys. Rev.* **116**, 1193 (1959).

<sup>6</sup> N. T. Porile, *Phys. Rev.* **115**, 939 (1959).

<sup>7</sup> S. Tanaka, M. Furukawa, T. Mikumo, S. Iwata, M. Yagi, and H. Amano, *J. Phys. Soc. (Japan)* **15**, 545 (1960).

<sup>8</sup> F. S. Houck and J. M. Miller (private communication).

<sup>9</sup> M. Blann, T. D. Thomas, and G. Seaborg, *Abstracts of Papers 133rd Meeting American Chemical Society, San Francisco, 1958*, p. 35Q.

<sup>10</sup> R. A. Sharp, R. M. Diamond, and G. Wilkinson, *Phys. Rev.* **101**, 1493 (1956).

<sup>11</sup> J. W. Meadows, *Phys. Rev.* **91**, 885 (1953).

<sup>12</sup> I. Dostrovsky, Z. Fraenkel, and G. Friedlander, *Phys. Rev.* **116**, 683 (1959).

<sup>13</sup> J. W. Meadows, R. M. Diamond, and R. A. Sharp, *Phys. Rev.* **102**, 190 (1956).

e.g., the  $(\alpha, \alpha\gamma)$ ,  $(d, d\gamma)$ , or  $(d, pn)$ , and  $(p, p\gamma)$  reaction on  $\text{Cd}^{111}$  and  $\text{In}^{115}$  for incident particles of, respectively, 15–40 Mev, 10–20 Mev, and 6–10 Mev. In addition, results were also obtained for the  $\text{In}^{115}(\alpha, \alpha n)\text{In}^{114m}$  reaction. The choice of targets was partly motivated by a desire to investigate any possible effects of the spin of the isomeric level on the shape of the excitation functions and the magnitude of the cross section. The spins of the various states under investigation thus are as follows:  $\text{Cd}^{111}$  ( $1/2+$ ),  $\text{Cd}^{111m}$  ( $11/2-$ ),  $\text{In}^{115}$  ( $9/2+$ ), and  $\text{In}^{115m}$  ( $1/2-$ ). The experimental procedure and results are presented in Secs. II and III, respectively. The results are compared with values calculated on the basis of the statistical theory in Sec. IV. This section also includes an estimate of the contribution of Coulomb excitation to the measured cross sections.

## II. EXPERIMENTAL

The irradiations were performed with the deflected beam of the Brookhaven 60-inch cyclotron. Detailed descriptions of the target assembly and of the techniques used to determine the beam intensity and beam energy have been given elsewhere<sup>5,14</sup> for bombardment with alpha particles. In the experiments with incident protons and deuterons, the beam energy was not monitored directly but was assumed to be equal to, respectively, 0.25 and 0.5 of the energy of the incident alpha particles. The intensity of the proton and deuteron beams were determined in the same fashion as for the alpha-particle beam,<sup>14</sup> keeping in mind the fact that the number of net charges registered for each incident deuteron and proton are 1 and 0.5, respectively. The proton value follows from the fact that the cyclotron actually accelerates  $\text{H}_2^+$  ions which are stripped in a foil that is in electrical contact with the beam stopping plate.<sup>14</sup> The irradiations made use of the stacked-foil technique and aluminum foils were used as energy degraders. A range-energy relation based on the range-energy relation for protons of Bichsel *et al.*<sup>15</sup> was used to determine the energy of the degraded beam. The number of target foils irradiated in any one bombardment ranged from one to eight. Irradiation times varied between a few seconds and 3 hours, and the beam current varied between 0.2 and 1.5 microamperes. In the course of this work a total of 21 irradiations was performed.

The experiments with  $\text{In}^{115}$  were performed with high-purity foils of natural indium, 0.00075 inch in thickness, and uniform to within 5%. These foils were thick enough to make the loss of recoils from the target negligible. The formation of  $\text{In}^{115m}$  by the  $(\alpha, 2p)$  reaction on  $\text{In}^{113}$  was investigated by the use of enriched  $\text{In}^{113}$  (65.4 atom percent  $\text{In}^{113}$ )<sup>16</sup> electroplated on gold. One irradiation was performed at 40 Mev, and it was

found that the observed yield of  $\text{In}^{115m}$  could be entirely accounted for by the  $\text{In}^{115}(\alpha, \alpha\gamma)$  reaction. On this basis an upper limit of 2% may be set for the contribution of the  $\text{In}^{113}(\alpha, 2p)$  reaction. Since it is not expected that the excitation function for this reaction goes through a maximum below 40 Mev, the above upper limit is probably applicable to the entire energy range. An upper limit of 5% over the entire energy range was similarly established for the contribution of the  $\text{In}^{113}(\alpha, 2pn)$  reaction to the observed yield of  $\text{In}^{114m}$ .

The experiments with  $\text{Cd}^{111}$  were performed in all cases with enriched  $\text{Cd}^{111}$  (86.1 atom percent  $\text{Cd}^{111}$ )<sup>17</sup> electroplated on gold, and with 0.0003-inch thick cadmium foil having a normal isotopic composition. The electroplated targets had a thickness of approximately 1 mg/cm<sup>2</sup>. The uniformity was occasionally checked by chemical analysis of different portions of the deposit and the variation in thickness was found to be less than 10%. The electroplated targets were always positioned so that the forward recoils were caught in the gold backing. No correction was made for the negligible loss of recoils in the backward direction. The loss of recoils was likewise neglected for the thicker target foils of ordinary cadmium. The  $\text{Cd}^{111m}$  activity induced in the enriched isotope targets was partially due to reactions with  $\text{Cd}^{110}$  (2.3 atom percent) and  $\text{Cd}^{112}$  (7.5 atom percent). The interfering reactions were  $\text{Cd}^{112}(p, pn)$ ,  $\text{Cd}^{112}(d, p2n)$ ,  $\text{Cd}^{110}(d, p)$ ,  $\text{Cd}^{112}(\alpha, \alpha n)$ , and  $\text{Cd}^{110}(\alpha, 2pn)$ . The contribution of these reactions was determined in the bombardments of normal cadmium foil. The total contribution of interfering reactions was found to range from 0 to 1.1% for proton bombardment, 1.2 to 2.5% for deuteron bombardment, and  $\leq 0.5$  to 20.6% for alpha-particle bombardment. In cases where interfering reactions could occur with two isotopes the correction was determined from estimates of the relative magnitude of the cross sections for the interfering reactions. The largest error due to this procedure would occur for the  $(\alpha, \alpha\gamma)$  reaction at 40 Mev. An increase by a factor of 4 in the cross section of the  $\text{Cd}^{110}(\alpha, 2pn)$  reaction relative to that of the  $\text{Cd}^{112}(\alpha, \alpha n)$  reaction would thus lead to a lowering of the cross section of the  $\text{Cd}^{111}(\alpha, \alpha\gamma)$  reaction by 6%.

After bombardment the foils were dissolved and cadmium and indium were separated and decontaminated by conventional radiochemical procedures. The activity of the samples was determined with a  $3 \times 3$  inch sodium iodide scintillation counter and the decay of a particular photopeak was followed with the aid of a 100-channel pulse-height analyzer. The cross sections for the formation of  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$  were, respectively, determined on the basis of measurements on the 246-keV and 335-keV transitions. The  $\text{Cd}^{111m}$  results were corrected for photopeak losses arising from the coincidence between the 150-keV and 246-keV transitions. The decay of 50-day  $\text{In}^{114m}$  was followed by observation of

<sup>14</sup> S. Amiel and N. T. Porile, Rev. Sci. Instr. **29**, 1112 (1958).

<sup>15</sup> H. Bichsel, R. F. Mozley, and W. A. Aron, Phys. Rev. **105**, 1788 (1957).

<sup>16</sup> Obtained from Oak Ridge National Laboratory.

<sup>17</sup> Obtained from the Atomic Energy Research Establishment, Harwell, England.

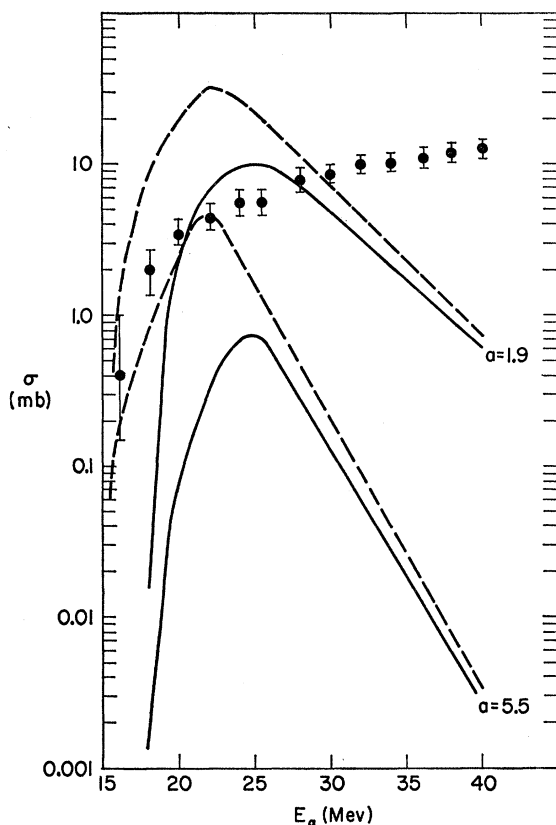


FIG. 1. Excitation function for the  $\text{Cd}^{111}(\alpha, \alpha\gamma)\text{Cd}^{111m}$  reaction. The top 2 curves were calculated for  $a = 1.9 \text{ Mev}^{-1}$  and the bottom 2 curves for  $a = 5.5 \text{ Mev}^{-1}$ . The dashed curves refer to  $r_0 = 1.7 \times 10^{-13} \text{ cm}$  and the solid curves to  $r_0 = 1.5 \times 10^{-13} \text{ cm}$ .

the 191-keV transition. The observed half-lives of the nuclides in question were in all cases consistent with reported values,<sup>18</sup> indicating that the contribution of other nuclides with transitions of about the same energy was negligible at the time of counting. The over-all photopeak efficiency was determined from measurements with calibrated sources having transitions in the energy region of interest. The calibrations were checked in some instances by measurement in a known geometry of the x-ray peak corresponding to the internal conversion of the  $\gamma$  ray of interest. These measurements were performed with essentially 100% efficiency by use of thin NaI counters. The photopeak efficiencies were obtained from the known<sup>18</sup> conversion coefficients and agreed with the measured values to within 10%. The disintegration rates of the samples were then obtained with the aid of the latest decay scheme data.<sup>18</sup>

The products isolated in the present study may also be formed by  $(n, n')$  reactions induced by secondary or stray neutrons. The contribution of these interfering reactions may be significant in view of the small primary cross sections for some of the reactions of

interest and in view of the much larger cross sections expected for  $(n, n')$  reactions. The cross section for the  $\text{In}^{115}(n, n\gamma)\text{In}^{115m}$  reaction has, in fact, been measured and is 370 mb for 2.5-MeV neutrons.<sup>19</sup> In addition to the above difficulty contamination of the proton and alpha beams with deuterons can also be a source of error, in view of the much larger cross sections for the deuteron induced reactions.

It was ascertained that interfering reactions did indeed contribute to the observed activities since  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$  were detected in cadmium and indium foils located in a foil stack beyond the range of the bombarding particles. The following additional experiments were performed in order to ascertain the nature of the interfering reactions. First, the beam collimator was replaced with an aluminum plate thick enough to stop all the incident particles and the yield of  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$  produced by the general background of stray neutrons was determined. Second, the proton and alpha beams were stopped in aluminum degrading foils and the activity of  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$  induced in foils located

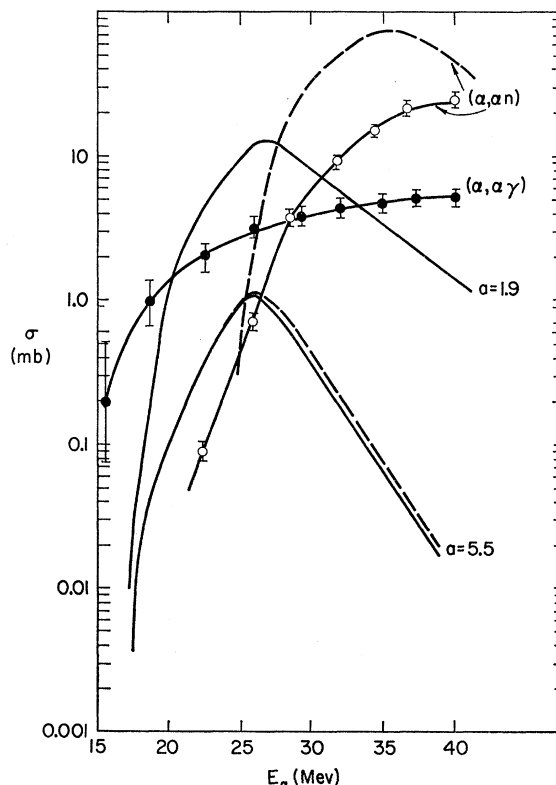


FIG. 2. Excitation functions for the  $\text{In}^{115}(\alpha, \alpha\gamma)\text{In}^{115m}$  and  $\text{In}^{115}(\alpha, an)\text{In}^{114m}$  reactions.  $\bullet$ ,  $(\alpha, \alpha\gamma)$  reaction.  $\circ$ ,  $(\alpha, an)$  reaction. The two solid curves without points represent calculated values for the  $(\alpha, \alpha\gamma)$  reaction for  $a = 1.9 \text{ Mev}^{-1}$  and  $a = 5.5 \text{ Mev}^{-1}$ . The dashed curve for  $a = 5.5$  indicates the effect of competition between gamma-ray de-excitation and neutron emission. The dashed curve for the  $(\alpha, an)$  reaction is the result of a Monte Carlo evaporation calculation.

<sup>18</sup> Nuclear Data Group, *Nuclear Data Sheets* (National Academy of Sciences, National Research Council, 1959).

<sup>19</sup> H. C. Martin, B. C. Divek, and R. F. Taschek, *Phys. Rev.* **93**, 199 (1954).

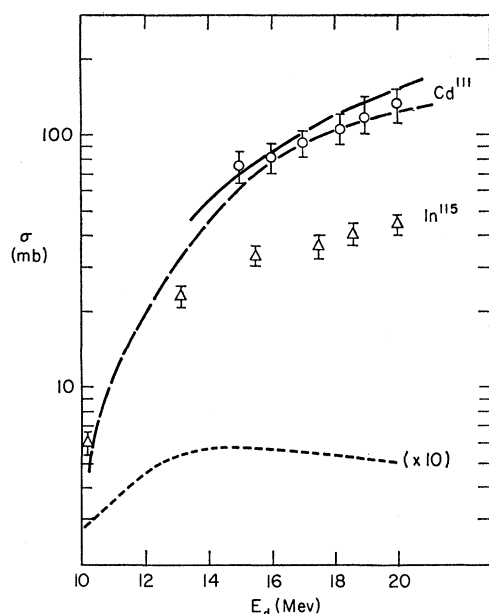


FIG. 3. Excitation functions for the  $\text{Cd}^{111}(d,d\gamma)\text{Cd}^{111m}$  and  $\text{In}^{115}(d,d\gamma)\text{In}^{115m}$  reactions.  $\Delta$ ,  $\text{In}^{115}$ ;  $\circ$ ,  $\text{Cd}^{111}$ ; — evaporation calculation for  $\text{In}^{115}$ ; — evaporation calculation for  $\text{Cd}^{111}$ ; - - - Coulomb excitation contribution to the  $\text{In}^{115}(d,d\gamma)\text{In}^{115m}$  reaction. The cross sections for this reaction are a factor of 10 lower than is indicated by the ordinate scale.

just beyond the range of a 10-Mev proton and a 20-Mev deuteron was determined. It was found for both the proton and the alpha-particle bombardments that about 95% of the observed activity originated from secondary particles produced in the target stack and only about 5% of the reactions were induced by stray neutrons. In addition it was also determined that the bulk of the secondary particles producing the activities in question consisted of neutrons. The contribution of deuterons in the alpha or proton beams appeared to be small.

The yields of  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$  due to these interfering reactions had the following values compared with the corresponding yields at the highest bombarding energies: 0.10 for incident alpha particles, 0.03 for incident deuterons, and 0.02 for incident protons. The results for  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$  were substantially the same in this respect. At the lowest bombarding energies the secondary reactions actually account for most of the observed alpha induced yields of  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$ . Although at the highest bombarding energies the actual correction is probably even smaller than indicated because of the preponderant forward emission of the secondary neutrons a constant correction has been applied at all energies. The over-all uncertainties in the measured cross sections are 10–20% at the higher energies and increase to a factor of two or more at the very lowest bombarding energies. The contribution of secondaries to the  $\text{In}^{115}(\alpha,an)$  reaction is entirely negligible and the cross sections for this reaction have an uncertainty of about 10%.

### III. RESULTS

The results are presented in the form of excitation functions in Figs. 1–4. The cross sections for the  $(p,p\gamma)$  reaction are seen to increase monotonically with bombarding energy. This increase is due in approximately equal measure to the increase in the total reaction cross section and to the greater probability for proton emission with increasing excitation energy. The cross sections for 10-Mev incident protons are 1.5 mb for  $\text{In}^{115}$  and 3.6 mb for  $\text{Cd}^{111}$ . These values constitute only a small fraction of the total inelastic cross section, indicating that neutron emission is the most probable mode of de-excitation. The cross section for the  $\text{Cd}^{111}(p,n)$  reaction at 7 Mev thus is about 300 mb<sup>20</sup> while the corresponding value of the  $(p,p\gamma)$  partial cross section is 0.4 mb. The cross section for the  $\text{In}^{115}(p,p\gamma)\text{In}^{115m}$  reaction has previously been measured for a proton energy of 5.8 Mev. A value of 0.01 mb was obtained in this early experiment.<sup>21</sup>

The cross sections for the  $(d,d\gamma)$  reaction at 20 Mev

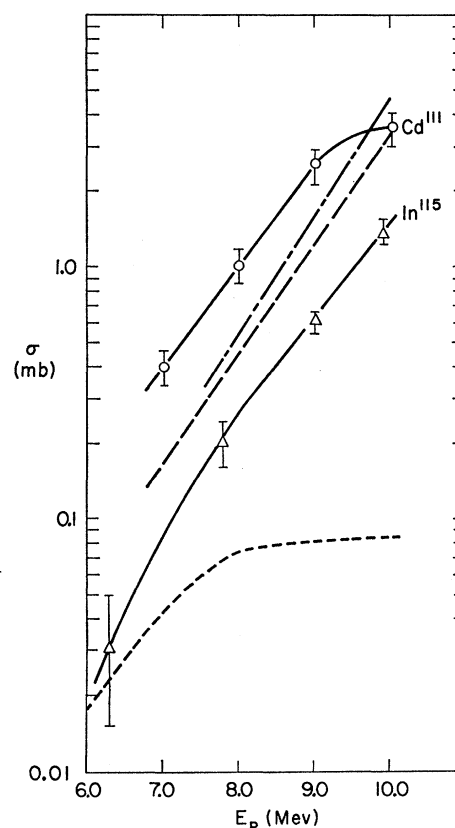


FIG. 4. Excitation functions for the  $\text{Cd}^{111}(p,p\gamma)\text{Cd}^{111m}$  and  $\text{In}^{115}(p,p\gamma)\text{In}^{115m}$  reactions.  $\Delta$ ,  $\text{In}^{115}$ ;  $\circ$ ,  $\text{Cd}^{111}$ ; — evaporation calculation for  $\text{In}^{115}$ ; — evaporation calculation for  $\text{Cd}^{111}$ ; - - - Coulomb excitation contribution to the  $\text{In}^{115}(p,p\gamma)\text{In}^{115m}$  reaction.

<sup>20</sup> J. P. Blaser, F. Boehm, P. Marmier, and D. C. Peaslee, *Helv. Phys. Acta* **24**, 3 (1951).

<sup>21</sup> S. W. Barnes and P. W. Aradine, *Phys. Rev.* **55**, 50 (1939).

are 42 mb for  $\text{In}^{115}$  and 132 mb for  $\text{Cd}^{111}$ , and are smaller at lower bombarding energies. The observed products may, of course, be the result of the  $(d, pn)$  reaction as well. The evaporation calculations discussed in the following section indicate, in fact, that the latter reaction is more probable than the  $(d, d\gamma)$  reaction and is able to account for both the shape of the excitation function and the magnitude of the cross sections. The inelastic scattering of deuterons has been studied by Yntema and Zeidman.<sup>2</sup> Integration of the measured differential cross sections and energy spectra gives a value of approximately 85 mb for the cross section for deuteron emission associated with the  $(d, d\gamma)$  reaction. This value applies to 21.6-Mev deuterons incident on rhodium and silver targets. If it is assumed that this value also applies to  $\text{Cd}^{111}$  and  $\text{In}^{115}$ , then deuteron emission would have to lead primarily to the respective ground states. This follows from the fact that the observed cross sections for the formation of the metastable state may be adequately accounted for by the evaporation of a proton and a neutron. This conclusion is reasonable, since it is not expected that direct inelastic scattering would involve substantial transfers of angular momentum with large probability.

The cross sections for the  $(\alpha, \alpha\gamma)$  reaction increase with increasing bombarding energy in the energy region covered in this experiment. The values obtained at 40 Mev are 5.2 mb and 12.2 mb for  $\text{In}^{115}$ , and  $\text{Cd}^{111}$ , respectively. The  $(\alpha, 2p2n)$  reaction becomes energetically possible above approximately 30 Mev. It is very unlikely, however, that this reaction contributes appreciably to the observed activity even at 40 Mev since the evaporated protons must be emitted with substantial kinetic energies because of the Coulomb barrier. The  $(\alpha, \text{He}^3n)$  and  $(\alpha, Tp)$  reactions become energetically possible above approximately 20 Mev and the measured cross sections may include a small contribution from these reactions. The cross section for the  $\text{In}^{115}(\alpha, \alpha\gamma)\text{In}^{115m}$  reaction has previously been measured with 16-Mev alpha particles and was found to be 0.3 mb.<sup>22</sup> This value is in good agreement with the result of the present study. The cross section for the  $\text{In}^{115}(\alpha, \alpha n)\text{In}^{114m}$  reaction at 40 Mev is 24 mb and the excitation function does not appear to peak in the energy region covered in this study. The peak thus appears to be shifted to somewhat higher excitation energies than the peak for the corresponding reaction with targets in the  $A = 50-70$  region.<sup>5, 6, 8, 9</sup> This behavior is reasonable in view of the fact that the higher Coulomb barrier for alpha particles at  $Z \sim 50$  tends to shift the whole excitation function for the  $(\alpha, \alpha n)$  reaction to higher excitation energies.

The shapes of the excitation functions for the reactions of  $\text{Cd}^{111}$  are very similar in all cases to those of the corresponding reactions of  $\text{In}^{115}$ . The reactions of  $\text{Cd}^{111}$  have the larger cross sections in all cases. The

value of  $\sigma(\text{Cd}^{111})/\sigma(\text{In}^{115})$  thus is 2.3-3.9 for the  $(p, p\gamma)$  reactions, 2.3-3.0 for the  $(d, d\gamma)$  reaction, and 2.3-2.5 for the  $(\alpha, \alpha\gamma)$  reaction. The formation of  $\text{Cd}^{111m}$  may be favored because of the larger statistical weight assigned to this nuclide.

#### IV. DISCUSSION

##### A. Comparison with the Statistical Theory

The experimental results may be compared with values calculated by use of the statistical theory in order to resolve the previously mentioned discrepancy. The cross section for the  $(\alpha, \alpha\gamma)$  reaction is given by the expression

$$\sigma(\alpha, \alpha\gamma) = \sigma_c(\alpha) F_{\alpha 1} / \sum_x F_{x \text{tot}},$$

where  $\sigma_c(\alpha)$  is the cross section for formation of the compound nucleus,  $F_{\alpha 1}$  is the emission function for one alpha particle and  $F_{x \text{tot}}$  is the total emission function for particle  $x$ . In the present calculation the emission of neutrons, protons, and alpha particles was considered. The emission function for particle  $x$  is given by<sup>6</sup>

$$F_x = M_x (2I_x + 1) \int_{\epsilon_{x \text{min}}}^{\epsilon_{x \text{max}}} \epsilon_x \sigma_c(\epsilon_x) \omega(\epsilon_x) d\epsilon_x,$$

where  $M_x$  and  $\epsilon_x$  are the reduced mass and kinetic energy of  $x$  in the center-of-mass system,  $I_x$  is the spin of particle  $x$ ,  $\sigma_c(\epsilon_x)$  is the inverse cross section for formation of the compound nucleus by  $x$ , and  $\omega(\epsilon_x)$  is the level density of the residual nucleus.

$$\omega(\epsilon_x) = C \exp\{[4a(E - \delta)]^{1/2}\},$$

where  $E$  is the excitation energy of the residual nucleus,  $\delta$  is the pairing energy,  $a$  is the level density parameter, and  $C$  is a constant. The pairing energy corresponds to the energy difference between the ground state and the characteristic level at which the exponential level density expression sets in. In the case of the total emission functions, the integration is performed over all possible values of  $\epsilon_x$ , while in the case of the single-particle emission function the integration is performed over all values of  $\epsilon_x$  for which further particle emission is energetically impossible. The analytic approximation for the inverse cross sections given by Dostrovsky *et al.*<sup>12</sup> and the pairing energies listed by Cameron<sup>23</sup> were used in the evaluation of the cross section. The calculation was performed for several values of the nuclear radius and level density parameters with the aid of an IBM 610 computer.<sup>24</sup>

In the case of the  $(d, d\gamma)$  or  $(d, pn)$  and  $(\alpha, \alpha n)$  reactions the expressions for multiple particle emissions have to be evaluated. This is a considerably more difficult task than the evaluation of the above-mentioned expressions for single-particle emission and it is convenient to use the Monte Carlo technique to obtain the desired cross

<sup>22</sup> J. R. Risser, K. Lark-Horovitz, and R. N. Smith, Phys. Rev. **57**, 355 (1940).

<sup>23</sup> A. G. W. Cameron, Can. J. Phys. **36**, 1040 (1958).

<sup>24</sup> The computer program is due to J. M. Miller.

sections.<sup>12</sup> The excitation functions for the  $(d,d\gamma)$  or  $(d,pn)$ , and  $(\alpha,\alpha n)$  reactions were calculated by application of this technique and the results are based on an over-all total of about 4000 interactions. The emission of deuterons, tritons, and  $\text{He}^3$  nuclei was considered in addition to that of protons, neutrons, and alpha particles. The calculation was done for  $r_0 = 1.7 \times 10^{-13}$  cm and the value of  $a$  was taken as 3.8 ( $a = A/30$ ). In cases where the residual nucleus had  $Z = 50$ , the pairing energy term was increased by 1 Mev in order to take account of the occurrence of a closed shell.

The above procedure was not followed for the  $(p,p\gamma)$  reaction since the above-mentioned analytic expression for the inverse cross section for charged particles is not well suited to excitation energies comparable to the Coulomb barrier for charged particle emission. The cross sections for the  $(p,p\gamma)$  reactions were calculated by numerical integration of the appropriate expressions using the continuum theory values<sup>25</sup> for  $r_0 = 1.5 \times 10^{-13}$  cm for the inverse cross sections for protons and neutrons. The contribution of the levels between the ground state and the characteristic level to the total level density was included in this calculation since it has been shown that this effect is important at excitation energies close to the effective threshold of a particular reaction.<sup>6</sup> The expression for the emission function for neutrons and protons then takes on the following form:

$$F_n = \int_0^{E^* - S_n - \delta} \epsilon_n \sigma_c(\epsilon_n) [\omega(\epsilon_n) + \omega'] d\epsilon + \int_{E^* - S_n - \delta}^{E^* - S_n} \epsilon_n \sigma_c(\epsilon_n) \omega' d\epsilon,$$

where  $E^*$  is the excitation energy of the compound nucleus,  $S_n$  is the binding energy of the emitted particle, and  $\omega'$  is the density of levels between the ground state and the characteristic level. The values of  $\omega'$  were evaluated on the basis of experimental data.<sup>18</sup> The constants in the level density expression were taken from the recent work of El-Nadi and Wafik<sup>26</sup> and for  $A = 115$  they are  $a = 3.45 \text{ Mev}^{-1}$  and  $C = 9.26 \text{ Mev}^{-1}$ . The contribution of the  $(p,\alpha)$  and  $(p,\gamma)$  reactions were neglected in this calculation so that only the  $(p,p\gamma)$  and  $(p,n)$  reactions were taken into account.

The calculated excitation functions for the  $\text{Cd}^{111}(\alpha,\alpha\gamma)$  reaction are given by the curves in Fig. 1. The solid curves are for  $r_0 = 1.5 \times 10^{-13}$  cm and  $a = 5.5$  and 1.9. The dashed curves are for  $r_0 = 1.7 \times 10^{-13}$  cm and the same values of  $a$ . It is seen that in all cases the calculated excitation functions peak for bombarding energies of 22–25 Mev, whereas the measured cross sections still increase with energy at 40 Mev. The peak in the calculated excitation functions is due, of course, to the

competition of the  $(\alpha,\alpha n)$  reaction. The subsequent decrease in cross section of the  $(\alpha,\alpha\gamma)$  reaction is larger for  $a = 5.5$  because of the increasing probability for further de-excitation with increasing level density. The peak in the excitation function occurs at lower energies for  $r_0 = 1.7 \times 10^{-13}$  cm because the lower Coulomb barrier for alpha particles in effect shifts the excitation functions for both the  $(\alpha,\alpha\gamma)$  and  $(\alpha,\alpha n)$  reactions to lower energies. The calculation indicates that the magnitude of the cross sections for the  $(\alpha,\alpha\gamma)$  reaction is strongly dependent on the values of  $r_0$  and  $a$ . The dependence on the nuclear radius parameter is once again related to the decrease in Coulomb barrier as  $r_0$  increases. This dependence is particularly striking at low bombarding energies where the  $(\alpha,\alpha\gamma)$  reaction competes primarily with the  $(\alpha,n)$  reaction. At higher energies the competition is mainly with the  $(\alpha,\alpha n)$  reaction and so the effect of the Coulomb barrier is much smaller. The dependence at low bombarding energies of the cross sections for the  $(\alpha,\alpha\gamma)$  reaction on the level density parameter is also related to the Coulomb barrier. As  $a$  decreases the nuclear temperature increases, and the alpha particles are on the average emitted with greater kinetic energies. The effect of the Coulomb barrier in depressing the emission of alpha particles is consequently reduced. The dependence on the level density parameter at higher energies is related to the competition between the  $(\alpha,\alpha\gamma)$  and  $(\alpha,\alpha n)$  reactions.

The calculated excitation functions for the  $\text{In}^{115}(\alpha,\alpha\gamma)$  reaction are given in Fig. 2. The solid curves are for  $r_0 = 1.5 \times 10^{-13}$  cm and  $a = 5.5$  and 1.9. It is seen that the curves are similar in all their features to the corresponding curves for  $\text{Cd}^{111}$ . The dashed curve was obtained by increasing the neutron separation energy for  $\text{In}^{115}$  by 0.2 Mev, in order to account for the competition between gamma-ray de-excitation and neutron emission from an excited  $\text{In}^{115}$  nucleus. It is seen that the resulting effect on the excitation function for the  $(\alpha,\alpha\gamma)$  reaction is very small.

It is concluded from these comparisons that the  $(\alpha,\alpha\gamma)$  reaction proceeds by a direct-interaction mechanism above 25 Mev, in agreement with the results of inelastic scattering experiments. The situation below 25 Mev is not clear in view of the wide range of possible values for the calculated cross sections. The experimental information in this mass region is too meager to permit a proper choice of parameters on the basis of which a distinction between mechanisms could be made. The results of Fulbright *et al.*<sup>1</sup> on the energy spectra of alpha particles indicate, however, that there is no discernible contribution from an evaporation mechanism even at these low energies.

The calculated excitation function for the  $\text{In}^{115}(\alpha,\alpha n)$  reaction is given by the dashed line in Fig. 2. The excitation function goes through a maximum at 35 Mev whereas the experimental curve still appears to be rising at 40 Mev. The calculated cross sections are larger than the experimental ones by as much as a

<sup>25</sup> J. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952).

<sup>26</sup> M. El-Nadi and M. Wafik, *Nuclear Phys.* **9**, 22 (1958)

factor of five. A different choice of  $r_0$  and  $a$  could change the cross sections by as much as an order of magnitude, however. The most likely mechanisms for the  $(\alpha, \alpha n)$  reaction appear to be either direct inelastic scattering followed by neutron evaporation or formation of a compound nucleus followed by the consecutive evaporation of an alpha particle and a neutron. For bombarding energies above 35 Mev the initial evaporation of a neutron followed by the evaporation of an alpha particle becomes important. The direct "knock-out" of a neutron by the inelastically scattered alpha particle does not appear to be of great importance in view of the very large  $(\alpha, \alpha n)$  to  $(\alpha, \alpha p)$  cross-section ratios obtained in the mass region.<sup>27</sup> The comparison of the calculated and experimental excitation functions indicates that the direct-interaction mechanism is of importance above 35 Mev. No definite conclusions about the reaction mechanism at the lower energies can be drawn from this comparison, however. The measurement of the ranges of the  $\text{In}^{114m}$  recoils might be of value in distinguishing between the above two mechanisms in this energy region. Considerably longer ranges are expected for those interactions in which evaporation of alpha particles takes place because the latter are emitted with symmetry about  $90^\circ$  in the center-of-mass system whereas the directly scattered alpha particles are emitted in a forward direction.

The calculated excitation functions for the  $(d, d\gamma)$  and  $(p, p\gamma)$  reactions are presented in Figs. 3 and 4, respectively. It is seen that for both reactions the calculated cross sections are larger for  $\text{Cd}^{111}$ , as is the case for the experimental results. The calculated cross sections are generally somewhat larger than the experimental values, and the shapes of the excitation functions are in reasonably good agreement. In view of the fact that the excitation functions do not peak within the energy range covered in this study, it is difficult to reach any definite conclusions from this comparison. The calculations for the  $(d, d\gamma)$  reaction indicate that the latter involves the emission of a proton and neutron rather than that of a deuteron in over 90% of the events so that this reaction is really in a different category than the other reactions considered in this study.

### B. Coulomb Excitation

One of the direct-interaction mechanisms that may contribute to the observed cross sections is Coulomb excitation and an estimate of the magnitude of this contribution may be made. For the purposes of this estimate only those classical paths that come no closer than a nuclear interaction radius will be considered. At smaller distances of approach nuclear interactions can take place and a more detailed analysis is necessary. The maximum deflection angle for which the above

condition holds is given by

$$\sin(\theta_{\max}/2) = \frac{1}{2(E/E_B) - 1},$$

where  $E$  is the energy of the incident particle and  $E_B$  is the Coulomb barrier. The value of  $\theta_{\max}$  for 40-Mev alpha particles on indium thus is  $28^\circ$ . The cross section for Coulomb excitation is then estimated by integrating the differential cross section<sup>28</sup> over the range  $0 \leq \theta \leq \theta_{\max}$ .

In the energy region considered here the classical description is still approximately valid; thus  $\eta$ , the parameter measuring the effective strength of the electromagnetic interaction,<sup>28</sup> is between 3 and 5. It is consequently possible to use the values given by Alder *et al.*<sup>28</sup> for the angular distribution of the Coulomb cross section.

The cross section for the formation of a given level by electric excitation of multipole order  $\lambda$  is given by the expression<sup>28</sup>:

$$\sigma_{E\lambda} = C_{E\lambda} E^{\lambda-2} (E - \Delta E)^{\lambda-1} B(E\lambda) \int_0^{\theta_{\max}} 2\pi \sin\theta f'_{E\lambda}(\theta) d\theta,$$

where  $C_{E\lambda}$  is a constant depending on the multipolarity of the transition and the charge and mass of the target and projectile,  $\Delta E$  is essentially the energy of the excited level,  $B(E\lambda)$  is the reduced transition probability, and  $f'$  is a tabulated<sup>28</sup> differential function of the deflection angle  $\theta$ , and in addition depends on the parameters of the interaction.

The electric multipoles  $E1$ ,  $E2$ ,  $E3$ , together with double  $E2$  excitations were considered. The contribution of magnetic excitations is negligibly small. The results depend, of course, on both the energies of the states populated by the excitation process and the strengths of the matrix elements. Two simplifying assumptions were made for the purposes of the present calculation in this connection. First, it was assumed that either single levels or simple distributions of levels were populated. Second, the values of the reduced transition probabilities were in general obtained by use of the sum rule. The spectrum of states populated by  $E1$  excitations was thus approximated by a continuous rectangular distribution centered at 20 Mev with a width of 10 Mev. In addition, the effect of the possibility that a small fraction of the  $E1$  transitions populate levels at lower excitation energies was investigated by assuming 1% branches to levels at 5 and 10 Mev. The  $E2$  excitation process in the mass region of interest has been interpreted as being vibrational in nature.<sup>28</sup> The value of  $B(E2)$  was therefore obtained on the assumption that the transition consisted of a single-phonon vibrational excitation. The value obtained in this fashion is some 30% lower than the sum rule value. The experimental values of  $B(E2)$  in even-even nuclei in the In,

<sup>27</sup> B. M. Foreman, Jr., Bull. Am. Phys. Soc. 5, 270 (1960).

<sup>28</sup> K. Alder, A. Bohr, T. Huus, B. Mottelson, and A. Winther, Revs. Modern Phys. 28, 432 (1956).

TABLE I. Cross sections for the  $\text{In}(\alpha, \alpha\gamma)$  and  $\text{In}(\alpha, \alpha n)$  reactions due to Coulomb excitation.

Energy	Process	$\sigma(\alpha, \alpha\gamma)$ mb	$\sigma(\alpha, \alpha n)$ mb
40 Mev	<i>E1</i>	0.3	$2 \times 10^{-3}$
	<i>E2</i>	15	$2 \times 10^{-3}$
	<i>E3</i>	2.0	$2 \times 10^{-3}$
	Double <i>E2</i>	1.6	...
	Total	19	$6 \times 10^{-3}$
20 Mev	<i>E1</i>	$2 \times 10^{-3}$	$5 \times 10^{-8}$
	<i>E2</i>	23	$10^{-10}$
	<i>E3</i>	2.0	$10^{-11}$
	Double <i>E2</i>	0.9	...
	Total	26	$5 \times 10^{-8}$

$\text{Cd}$  region indicate that the single-phonon transition practically accounts for the entire sum rule.<sup>28</sup> The cross section is insensitive to the value of the excitation energy for low excitations and the transition was assumed to populate a level at 1.2 Mev. In order to obtain an estimate about the *E2* contribution to the  $(\alpha, \alpha n)$  reaction, a 1% *E2* branch to a level at 10 Mev was assumed. The double (*E2*) excitation cross section was estimated by assuming that the second (*E2*) excitation populated levels at about twice the energy of the levels populated by the single (*E2*) excitation and that the second (*E2*) transition had the same cross section as the first transition. The cross section for the (*E3*) excitation was obtained on the assumption that the populated level was at 2 to 3 Mev and that essentially the whole *E3* sum rule is used up in the transition to this level. A 1% branch to a 10-Mev level was assumed in order to estimate the contribution to the  $(\alpha, \alpha n)$  reaction.

The results of calculations based on these assumptions for *Z* of indium are summarized in Table I for 20- and 40-Mev alpha particles. Results are given for the  $(\alpha, \alpha\gamma)$  and  $(\alpha, \alpha n)$  reactions. It is seen that the contribution of the Coulomb excitation process to the  $(\alpha, \alpha\gamma)$  reaction can be very substantial, particularly for *E2* excitations. The contribution to the  $(\alpha, \alpha n)$  reaction, on the other hand, is negligibly small. The calculated  $(\alpha, \alpha\gamma)$  excitation function is given by the solid curve in Fig. 5.

Having seen that Coulomb excitation can be important, the applicability of these results to the  $\text{In}^{115}(\alpha, \alpha\gamma)\text{In}^{115m}$  reaction must be scrutinized. Unfortunately, there is as yet not enough detailed information available about the level scheme and the electromagnetic transition probabilities of  $\text{In}^{115}$  to make conclusive statements possible. The following argument is therefore based partly on conjecture.

We are interested in the population of the  $(1/2-)$  isomeric level. This could occur through Coulomb excitation of a higher level followed by a cascade to the  $(1/2-)$  level. Since an *E2* single-phonon excitation appears to be most important, it is this process that will be considered. Suppose that there is a set of

excited levels based on the addition of one phonon to the  $(9/2+)$  ground state, i.e., a set of states of spin  $(5/2+)$ ,  $(7/2+)$ ,  $(9/2+)$ ,  $(11/2+)$ , and  $(13/2+)$ . The  $(7/2+)$ ,  $(9/2+)$ , and  $(11/2+)$  levels have, in fact, been identified in the level scheme of  $\text{In}^{115}$  and the de-excitations from these levels appears to feed the ground state primarily.<sup>18</sup> The  $(13/2+)$  level is expected to follow this pattern. The  $(5/2+)$  level, on the other hand, can readily de-excite to the known  $(3/2-)$  level via an *E1* transition and then cascade to the  $(1/2-)$  level by an *M1* transition. Since the relative population of these levels varies as  $(2J+1)$ , it is clear that the *E2* excitation of the  $(5/2+)$  level constitutes only a small fraction of the whole phonon excitation. A reasonable estimate of the branching ratio to the isomeric level might thus be  $\sim 6\%$ . On these further assumptions, taken together with the above computations, the estimate given by the dashed curve in Fig. 5 is made. It is seen that a substantial fraction of the observed yield of  $\text{In}^{115m}$  may be accounted for by Coulomb excitation, particularly at the lower bombarding energies.

A similar analysis of  $\text{Cd}^{111}$  is not possible at this time

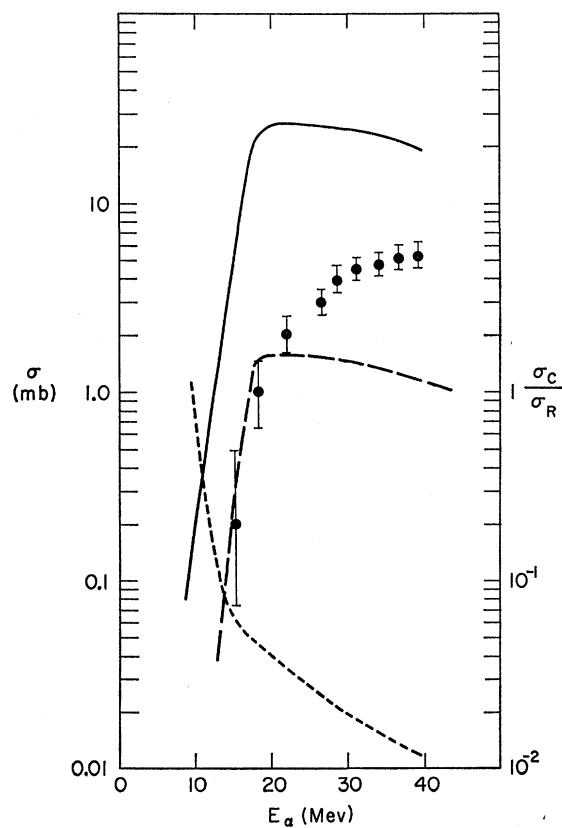


FIG. 5. Contribution of Coulomb excitation to the  $\text{In}(\alpha, \alpha\gamma)$  reaction (solid curve) and to the  $\text{In}^{115}(\alpha, \alpha\gamma)\text{In}^{115m}$  reaction (heavy dashed curve);  $\bullet$ , experimental points for  $\text{In}^{115}(\alpha, \alpha\gamma)\text{In}^{115m}$  reaction; - · - · - ratio of total Coulomb excitation and reaction cross sections for  $\text{In}^{115}$ . The ordinate scale at the right applies to the last curve.



since the level scheme is not well known. It seems quite likely though that the In results might apply to Cd<sup>111</sup> as well. To summarize the situation, it appears that the observed yield of the  $(\alpha, \alpha\gamma)$  reaction below about 25 Mev can be accounted for by contributions from both evaporation and Coulomb excitation mechanisms. Above 25 Mev the main contribution to the observed yield appears to come from a nuclear direct-interaction mechanism.

The ratio of the total Coulomb excitation and reaction cross sections for In<sup>115</sup> is given by the dashed line in Fig. 5. The total reaction cross section was obtained by use of the continuum theory<sup>25</sup> with a value of  $1.5 \times 10^{-13}$  cm for the nuclear radius parameter. The two cross sections are about equal at 10 Mev and the ratio decreases sharply with increasing energy so that at the energy corresponding to the Coulomb barrier (15.5 Mev) the ratio is only 0.06. This sharp decrease is due to the much steeper increase with energy of the total reaction cross section for energies below the Coulomb barrier. At higher energies the ratio decreases less sharply as the reaction cross section approaches its asymptotic value. While the contribution of the Coulomb excitation process thus is fairly substantial, it should be kept in mind that this process only affects the  $(\alpha, \alpha\gamma)$  reaction to any appreciable extent.

The calculation of the Coulomb excitation cross

section has also been carried out for proton and deuteron bombardment of In<sup>115</sup>. The calculated excitation function for the In<sup>115</sup>( $d, d\gamma$ )In<sup>115m</sup> reaction is given by the dashed line in Fig. 3. It is seen that the calculated values are lower than the experimental points by at least a factor of 30. This is not surprising in view of the fact that most of the observed yield is probably due to the  $(d, pn)$  reaction. The calculated excitation function for the In<sup>115</sup>( $p, p\gamma$ )In<sup>115m</sup> reaction is given by the dashed line in Fig. 4. It is seen that the contribution of the Coulomb excitation process is substantial at the lowest bombarding energy. The calculated cross section at 10 Mev is, on the other hand, much smaller than the experimental value.

#### ACKNOWLEDGMENTS

The continued interest in this work of Dr. G. Friedlander is appreciated. The author wishes to thank Professor J. M. Miller and Dr. J. Weneser for valuable discussions. Dr. I. Dostrovsky and Dr. Z. Fraenkel kindly performed the Monte Carlo evaporation calculations. The cooperation of Dr. C. P. Baker and the crew of the 60-inch cyclotron is gratefully acknowledged. The chemical yield analyses were performed by Dr. R. W. Stoenner, Dr. J. K. Rowley, and members of the analytical chemistry group.

## Nucleon Transfer Reactions in Grazing Collisions of Heavy Ions\*

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 (Received August 30, 1960)

Reactions in which several nucleons are transferred between complex nuclei have been studied by measurement of angular distributions and excitation functions of the recoiling projectile residues. The results show that multinucleon transfer does not proceed either through a compound nucleus or through a mechanism in which the Coulomb barrier is not penetrated (such as the tunneling mechanism for single-nucleon transfer). Instead, the data indicate the existence of a "grazing contact" mechanism. In such a grazing reaction, it appears that a high-energy projectile, though deflected by the Coulomb barrier, still penetrates the region of nuclear binding of the target. It moves along the surface of the target, with the zone of contact between the nuclei being frictionally excited and thus preventing formation of a compound nucleus. The system separates after half a rotation, or less, because the repulsive Coulombic and centrifugal forces exceed the nuclear binding force. Depending on the mode of separation, such grazing contact may result in nucleon transfer, inelastic scattering, or breakup of the projectile. At energies well above the Coulomb barrier, such grazing processes appear to represent an important fraction of the geometric cross section.

### I. INTRODUCTION

TWO large classes of heavy-ion nuclear reactions have been identified and studied: compound nucleus interactions and various Coulomb scattering processes occurring outside the normal range of nuclear binding forces. This paper explores a third and intermediate

class of heavy-ion reactions which may be termed grazing processes. At bombarding energies only slightly above the Coulomb barrier, the reactions of heavy ions can be separated into two classes. For relatively large impact parameters, the Coulomb barrier is not penetrated and Rutherford scattering, Coulomb excitation,<sup>1</sup>

\* Contribution No. 1637 from the Sterling Chemistry Laboratory, Yale University.

<sup>1</sup> D. G. Alkhazov, D. S. Andreyev, A. P. Greenberg, and I. N. Lemberg, *Physica* 22, 1129 (1956).