

# Isobaric Yields in the Reactions of 2.9-Bev Protons with Arsenic-75†

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(Received December 28, 1961)

Cross sections for the formation of four isobars of  $A = 72$  from bombardment of  $\text{As}^{75}$  with 2.9-Bev protons are reported. The cross sections as a function of  $Z$  fit a Gaussian curve with a peak at  $Z = 32.3$  and a width at half-maximum of 1.4 charge units. Cross sections for six other nuclides in the mass range  $66 \leq A \leq 74$  were also measured, and their cross sections lie close to the Gaussian when plotted against  $Z - Z_A$ , with the exception of  $\text{As}^{74}$ . An analysis of the yields of  $\text{Ga}^{72}$  and  $\text{Zn}^{72}$  indicates that most of the  $\text{Zn}^{72}$  is not formed in a  $(p, 4p)$  reaction but in a  $(p, 3pn\pi^+)$  reaction, or similar process in which one or more  $\pi^+$  mesons are emitted, with a cross section of  $> 0.018$  mb.

## INTRODUCTION

THE importance of relative isobaric yields in interpreting high-energy nuclear reactions has been emphasized in a recent review article.<sup>1</sup> There is often a large yield ratio for neighboring isobars, and these relative yields are nearly independent of both target and bombarding energy, as long as the isobars are at least a few mass and atomic numbers away from the target. These observations are well accounted for by the evaporation step of the cascade-evaporation model. Comparison of the yield ratios with those observed in low-energy compound nucleus reactions confirms this view.

The evaporation steps of a nuclear reaction can be calculated, provided the level density parameters of the nuclei involved are known. Dostrovsky, Fraenkel, and Friedlander<sup>2</sup> have made such calculations with a Monte Carlo technique, and adjusted the parameters to obtain the best fit to experimental excitation functions. One can then use these parameters to calculate other evaporation processes involving the same nuclei. Applying this to isobars produced in high-energy reactions, it may be possible to learn something about the preceding cascade step. Isobars are especially useful for this, since the evaporation calculations are more reliable when only an isobaric ratio is desired. In fact, the isobaric ratio is sensitive mainly to the odd-even parameter  $\delta$ , and not to the parameter  $a$  in the Fermi-gas level density expression.<sup>2</sup>

The isobars whose cross sections were measured in this work are those of  $A = 72$ , and the target was  $\text{As}^{75}$ . They are especially suitable because there are four radioactive isobars with well-known decay schemes and convenient half-lives,  $\text{Zn}^{72}$ ,  $\text{Ga}^{72}$ ,  $\text{As}^{72}$ , and  $\text{Se}^{72}$ , and only one stable isobar,  $\text{Ge}^{72}$ . The reactions producing them are of the type<sup>3</sup>  $(p, 4 \text{ nucleon})$ , and include the extreme

$(p, 4n)$  and  $(p, 4p)$  reaction types. Excitation functions for production of all four isobars by alpha-particle bombardment have been measured,<sup>4</sup> and the evaporation calculations<sup>2</sup> provide values of  $\delta$  for them. Cross sections of six other isotopes of Ga and As were also measured.

## EXPERIMENTAL DETAILS

Bombardments were done in the circulating proton beam of the Cosmotron, at an energy of 2.9 Bev. Arsenic was bombarded as  $\text{As}_2\text{O}_3$  powder wrapped in aluminum foil, and as metallic As evaporated onto 1-mil aluminum, with a thickness of 0.64 mg/cm<sup>2</sup>. The powder targets provided considerably more activity than the evaporated targets, but gave only relative cross sections. The evaporated targets were used with aluminum foil monitors to determine absolute cross sections by comparison with the  $\text{Al}^{27}(p, 3pn)\text{Na}^{24}$  cross section. Thin aluminum recoil catcher foils surrounded the target foil, and were dissolved with it. The monitor cross section used was 9.4 mb at 2.9 Bev.<sup>5,6</sup>

Chemical separations of Zn, Ga, As, and Se were performed using the procedures outlined in the Appendix. The Ga and As samples were counted directly, but the Zn and Se samples were "milked" for  $\text{Ga}^{72}$  and  $\text{As}^{72}$ , respectively, at a suitable time after purification.

Counting was done with beta-proportional counters and a 3×3-in. NaI crystal attached to a 100-channel pulse-height analyzer. The beta counters were used to count the  $\text{Ga}^{72}$  and  $\text{As}^{72}$  milked from Zn and Se, and they were calibrated for these two activities by sources of different thickness, whose absolute disintegration rates were measured as described below.

The NaI crystal was calibrated for photopeak efficiency by means of NBS standard sources of  $\text{Hg}^{203}$ ,  $\text{Sr}^{85}$ ,  $\text{Nb}^{95}$ ,  $\text{Mn}^{54}$ , and  $\text{Zn}^{65}$ . A  $\text{Co}^{57}$  source whose disintegration rate was measured by  $4\pi$   $\gamma$  counting was also

type  $(p, 4p)$  includes  $(p, 3pn\pi^+)$ . Cascade will be used to denote the actual cascade process.

<sup>4</sup> S. Amiel, Phys. Rev. **116**, 415 (1959).

<sup>5</sup> J. B. Cumming, G. Friedlander, and C. Swartz, Phys. Rev. **111**, 1386 (1958).

<sup>6</sup> J. B. Cumming, A. M. Poskanzer, and J. Hudis, Phys. Rev. Letters **6**, 484 (1961); J. B. Cumming (private communication).

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

<sup>1</sup> J. M. Miller and J. Hudis, Ann. Rev. Nuclear Sci. **9**, 159 (1959).

<sup>2</sup> I. Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev. **116**, 683 (1959).

<sup>3</sup> Reaction type will be used here to denote the change in mass and charge by writing the reaction as if only neutrons and protons were emitted. This does not imply a mechanism; thus reaction

TABLE I. Counting characteristics of nuclides studied.<sup>a</sup>

Nuclide	Half-life	$\gamma$ -ray energy (MeV)	Abundance
Zn <sup>72</sup>	49 hr	<sup>b</sup>	
Ga <sup>66</sup>	9.3 hr	1.037	37%
Ga <sup>67</sup>	78 hr	0.180+0.206	30%
Ga <sup>72</sup>	14.0 hr	0.835	96.7%
Ga <sup>73</sup>	5.0 hr	0.30	97%
As <sup>71</sup>	62 hr	0.175	100%; $e/\gamma=0.10$
As <sup>72</sup>	25.3 hr	0.835	76.6%
As <sup>73</sup>	76 days	0.054	100%; $e/\gamma=4.7$
As <sup>74</sup>	17 days	0.596+0.635	76.0%
Se <sup>72</sup>	8.5 days	<sup>c</sup>	

<sup>a</sup> Taken from reference 8.<sup>b</sup> Ga<sup>72</sup> daughter counted.<sup>c</sup> As<sup>72</sup> daughter counted.

used. The photopeak efficiency plotted against  $\gamma$ -ray energy gave a straight line on log-log paper,<sup>7</sup> for  $0.15 \leq E \leq 1.12$ . Since each of the nuclides studied had a prominent  $\gamma$  ray, their absolute disintegration rates could be determined from the photopeak counting rate and the abundance of the  $\gamma$  ray. Table I lists the nuclides whose cross sections were measured, their half-lives, the  $\gamma$  ray used for counting, and its abundance.<sup>8</sup> In most cases the subtraction of the Compton background due to higher energy  $\gamma$  rays could be done graphically. The  $\gamma$  spectra of Ga<sup>66</sup> and Ga<sup>72</sup> prepared separately, however, showed that each one had a peak in the same position as the main peak of the other one. Therefore, the individual spectra were used to calculate the fraction of counts in each peak due to each activity. The 0.30-MeV  $\gamma$  of Ga<sup>67</sup> interfered with the 0.30-MeV  $\gamma$  of Ga<sup>73</sup>, but the widely different half-lives allowed them to be resolved by the decay curve.

As<sup>73</sup> was counted with a 2-mm thick NaI crystal, whose geometry was measured with the 60-keV  $\gamma$  of Am<sup>241</sup>, using a source calibrated by alpha counting.<sup>9</sup> The difference in  $\gamma$ -ray energies of As<sup>73</sup> and Am<sup>241</sup> makes necessary a small correction for the amount of iodine x-rays escaping from the crystal.<sup>10</sup>

The nuclides Ga<sup>66</sup> and As<sup>72</sup> were also standardized by measuring their total  $\beta^+$  emission rate by 0.51-0.51  $\gamma$ -coincidence counting, using a standardized Na<sup>22</sup> source for comparison. For Ga<sup>66</sup>, the ratio of total positrons to 1.037 $\gamma$  was  $1.39 \pm 0.04$ ; for As<sup>72</sup>, the ratio of total positrons to 0.835 $\gamma$  was  $1.04 \pm 0.05$ ; these numbers are in agreement with published decay schemes.<sup>8</sup>

The aluminum monitors were counted in the beta-proportional counters, which were calibrated for Na<sup>24</sup> in different thicknesses of aluminum foil by  $\beta$ - $\gamma$  coincidence counting. Many of the decay curves obtained with the beta counters were resolved by a computer

<sup>7</sup> N. H. Lazar, R. C. Davis, and P. R. Bell, *Nucleonics* 14, (No. 4), 52 (1956).

<sup>8</sup> *Nuclear Data Sheets* (National Research Council and National Academy of Science, Washington, D. C., 1961).

<sup>9</sup> The alpha counting was done by Dr. A. M. Poskanzer.

<sup>10</sup> P. Axel, *Rev. Sci. Instr.* 25, 391 (1954).

least-squares program.<sup>11</sup> Several half-lives were determined using this program, and they are: Ga<sup>66</sup>,  $9.30 \pm 0.03$  hr; Ga<sup>72</sup>,  $13.98 \pm 0.05$  hr; As<sup>72</sup>,  $25.3 \pm 0.1$  hr.

The results are summarized in Table II. The error assigned to each cross section was obtained from the spread of individual determinations. In addition, there are systematic errors of the following kind: (a) the subtraction of the Compton background under the photopeak; (b) the  $\gamma$ -ray abundances; (c) the monitor cross section. It is difficult to estimate a highly subjective error such as (a); one can say that all peaks were analyzed in as nearly the same way as possible. In the case of As<sup>74</sup>, whose absolute cross section is of interest, a standard spectrum of annihilation radiation was used to subtract out the 0.51-MeV peak from the 0.60-MeV +0.64-MeV peak. In the case of As<sup>71</sup>, the 0.51-MeV spectrum was used to subtract out the back-scatter peak in the region of the 0.175-MeV peak.

Error (b) can be very small, as in the cases of Ga<sup>72</sup>, Ga<sup>73</sup>, and As<sup>71</sup>, when the abundance is close to 100% and the decay scheme has been confirmed. The relative isobaric cross sections, Zn<sup>72</sup>:Ga<sup>72</sup> and Se<sup>72</sup>:As<sup>72</sup>, are independent of this, since the daughter nuclide was used to determine the parent. The conversion coefficient of the 54-keV  $\gamma$  of As<sup>73</sup> is  $4.7 \pm 0.6$ ; this results in a 10% uncertainty in the As<sup>73</sup> cross section. Error (c) will not affect the relative cross sections, but will be important in comparing the results with other data in the literature, since the recent revision<sup>6</sup> of the monitor cross section.

The Ga<sup>72</sup>, As<sup>72</sup>, and As<sup>74</sup> yields are independent, while the other yields are cumulative, except for Ga<sup>66</sup> which is partly independent. A consideration of the isobaric-yield curve indicates that all these nuclides have parents with cross sections less than one-tenth of theirs, with the exception of As<sup>73</sup>. The experimental cross sections are thus approximately independent ones.

## DISCUSSION

The cross sections for the isobars of  $A=72$  are plotted as open circles in Fig. 1, and those of the other nuclides

TABLE II. Experimental cross sections.

Nuclide	Cross section mb	Number of determinations
Zn <sup>72</sup>	$0.025 \pm 0.003$	6
Ga <sup>66</sup>	$5.1 \pm 0.5$	11
Ga <sup>67</sup>	$8.0 \pm 0.8$	8
Ga <sup>72</sup>	$2.28 \pm 0.19$	6 <sup>a</sup>
Ga <sup>73</sup>	$0.62 \pm 0.07$	6
As <sup>71</sup>	$7.0 \pm 0.6$	7
As <sup>72</sup>	$14.3 \pm 0.9$	7
As <sup>73</sup>	$17 \pm 2$	2
As <sup>74</sup>	$47 \pm 3$	5
Se <sup>72</sup>	$0.45 \pm 0.08$	6

<sup>a</sup> Ga<sup>72</sup> used to normalize other cross sections in As<sub>2</sub>O<sub>3</sub> bombardments.

<sup>11</sup> Programmed by J. B. Cumming, Brookhaven National Laboratory.

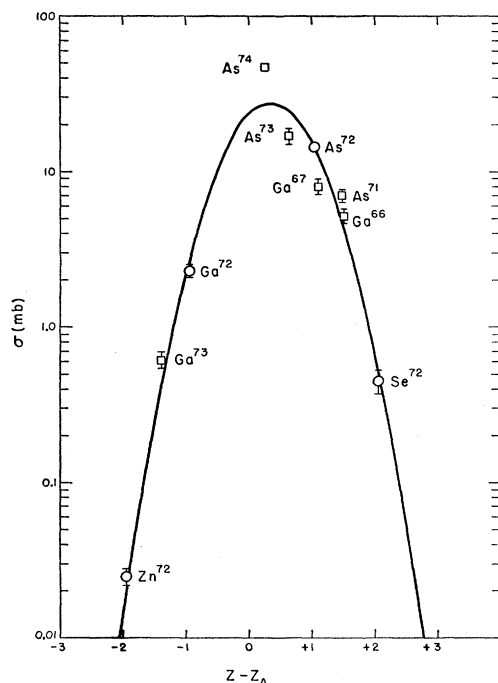


FIG. 1. Experimental cross sections vs  $Z - Z_A$ . Curve is Gaussian which best fits  $A = 72$  cross sections (circles).

as open squares. The cross sections are plotted against  $Z - Z_A$ , the difference between the charge of a nuclide and the value of the most stable charge for that mass number. The latter were obtained from Coryell's compilation.<sup>12</sup> The smooth curve is the Gaussian which best fits the  $A = 72$  points. Most of the other points lie near this curve, the main exception being  $As^{74}$ , which is the  $(p, pn)$  product. Similar behavior of a number of spallation products has been observed before,<sup>13,14</sup> and the smooth curve is useful in estimating unmeasured cross sections. The implication here is that for  $66 \leq A \leq 73$  the mass yield is constant, and the peak of the isobaric yields is at the same value of  $Z - Z_A$ , which is  $+0.35$ . The width at half-maximum is 1.4 charge units.

The anomaly of  $As^{74}$  is understandable in view of the large magnitude of  $(p, pn)$  reactions at BeV energies.<sup>15-18</sup> These are due mainly to  $p$ - $n$  collisions in the periphery of the nucleus in which both collision partners escape, leaving the residual nucleus with insufficient excitation energy to evaporate additional particles.

The two extremes of the  $A = 72$  isobars,  $Zn^{72}$  and  $Se^{72}$ , are of particular interest, since they can be formed only

by a reaction in which four identical particles are emitted,  $(p, 4p)$  and  $(p, 4n)$ , respectively, or by reactions in which one or more charged mesons are emitted,  $(p, 3pn\pi^+)$  for  $Zn^{72}$ , for example. The  $Zn^{72}$  cross section is expected to be small, since only relatively improbable reactions will result in this nuclide. The  $(p, 4p)$  cascades are rare, and only a small fraction of these will leave the residual nucleus with insufficient excitation energy to evaporate any particles. While the  $(p, 3p)$  and  $(p, 2p)$  cascades are more probable, they would have to be followed by proton evaporation, which has a low probability. Finally, the reactions involving meson emission which could lead to the  $(p, 4p)$  product are also improbable. In the next section, the probabilities of these different paths will be calculated.

The  $(p, 4n)$  cascade should be even less probable than the  $(p, 4p)$  cascade, since in the former case the incident particle must be absorbed by the nucleus, or undergo a charge-exchange scattering. The alternative reactions, in which one or more  $\pi^-$  mesons are emitted, should have a low probability, as above. The  $Se^{72}$  is produced by  $(p, xn)$  cascades,  $x \leq 3$ , followed by  $4 - x$  neutron evaporations, which are much more probable than are proton evaporations following  $(p, xp)$  cascades. Thus  $Se^{72}$  has a much larger cross section than  $Zn^{72}$ .

The highest cross sections of these isobars are for the  $(p, p3n)$  and  $(p, 2p2n)$  type reactions, the latter being interpolated from the smooth curve. The total isobaric yield, including the interpolated value for  $Ge^{72}$ , is 41 mb.

The spallation of arsenic with protons has also been studied at 49 Mev,<sup>13</sup> 103 Mev,<sup>13</sup> 170 Mev,<sup>13</sup> and 380 Mev.<sup>19</sup> Cross sections for  $Zn^{72}$  and  $Ga^{72}$  from arsenic have been reported at energies of 100–400 Mev,<sup>20</sup> and for  $As^{74}$  at energies of 250–400 Mev.<sup>21</sup>

The cross sections of neutron-excess nuclides,  $Zn^{72}$ ,  $Ga^{72}$ , and  $Ga^{73}$ , increase with energy, the last two leveling off above 0.4 BeV. The cross sections of neutron-deficient nuclides decrease with energy, the sharpest decrease being for  $Se^{72}$ . The excitation functions for  $Ga^{66}$  and  $Ga^{67}$  go through a maximum near 0.2 BeV, after which they decrease with energy. The  $As^{74}$  excitation function is flat above 0.25 BeV, after a sharp decrease. The isobaric yield curves at the lower energies have a Gaussian shape, with the peak more neutron-deficient than at 2.9 BeV.

## CALCULATIONS

The cascade-evaporation model has been used successfully to describe the general features of high-energy reactions. The difficulty of an exact calculation of the nuclear cascade has prevented any but a Monte Carlo

<sup>12</sup> C. D. Coryell, Ann. Rev. Nuclear Sci. 2, 305 (1953).

<sup>13</sup> G. Rudstam, thesis, University of Uppsala, Uppsala, Sweden, 1956 (unpublished).

<sup>14</sup> E. Belmont and J. M. Miller, Phys. Rev. 95, 1554 (1954).

<sup>15</sup> S. M. Markowitz, F. S. Rowland, and G. Friedlander, Phys. Rev. 112, 1295 (1958).

<sup>16</sup> I. M. Landenbauer and L. Winsberg, Phys. Rev. 119, 1368 (1960).

<sup>17</sup> D. R. Nethaway and L. Winsberg, Phys. Rev. 119, 1375 (1960).

<sup>18</sup> B. D. Pate and A. M. Poskanzer, Phys. Rev. 123, 647 (1961).

<sup>19</sup> J. B. Cumming, Atomic Energy Commission Document NYO-6141, 1954 (unpublished); J. B. Cumming (private communication).

<sup>20</sup> D. L. Morrison, Atomic Energy Commission Document NYO-8921, 1961 (unpublished).

<sup>21</sup> P. P. Strohal and A. A. Caretto, Phys. Rev. 121, 1815 (1961).

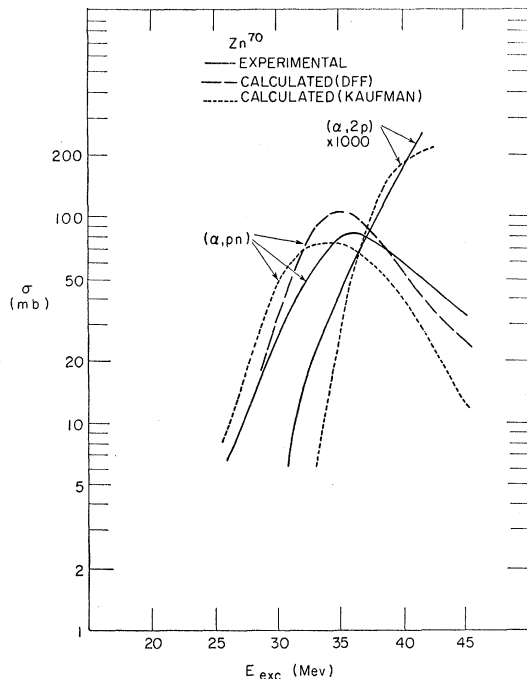


FIG. 2. Experimental and calculated excitation functions for  $Zn^{70}(\alpha, pn)$  and  $(\alpha, 2p)$  reactions. The curve labeled (DFF) was taken from reference 2, the smooth experimental curves from reference 4.

calculation from being made.<sup>22</sup> Monte Carlo calculations of the evaporation behavior of the highly excited nuclei formed in the cascade have also been made.<sup>23</sup> However, it would not be informative to use the cascade calculations to obtain cross sections for comparison with these experimental results, because the limited number of cascades calculated results in poor statistics for cross sections less than a few millibarns.

A more satisfactory approach is to calculate *ratios* of isobaric cross sections to compare with experiment. These ratios usually are less dependent on the fast cascade than on the evaporation stage, and therefore are less uncertain than the absolute cross sections.

As pointed out by Hudis and Miller,<sup>1</sup> many isobaric yield ratios are similar when measured in high-energy reactions and in low-energy evaporation reactions. The ratio of  $Ga^{72}$  to  $Zn^{72}$  cross sections measured in low-energy alpha-particle bombardments is shown in Fig. 3. The same ratio measured in this work is  $90 \pm 11$ , which fits the low-energy data at 40-Mev excitation energy. Since  $Zn^{72}$  can be formed in three different processes, as discussed above, we will estimate the contribution of the evaporation process to the  $Zn^{72}$  cross section.

If we represent the evaporation contribution to a

cross section as  $\sigma_E$ , then

$$\frac{\sigma_E(Zn^{72})}{\sigma_E(Ga^{72})} = \frac{\int_{S_p}^{E_m} N(U)G_p(U)dU}{\int_{S_n}^{E_m} N(U)G_n(U)dU}. \quad (1)$$

In this equation,  $G_p(U)$  and  $G_n(U)$  are the probabilities that a  $Ga^{73}$  nucleus with excitation energy  $U$  will evaporate a proton and neutron, respectively, without any additional particles being evaporated,  $N(U)$  is the distribution of excitation energy in  $Ga^{73}$  nuclei as formed in the cascade,  $S_p$  and  $S_n$  are the separation energies of a proton and a neutron, respectively, from  $Ga^{73}$ , and  $E_m$  is the maximum excitation energy in  $Ga^{73}$ . The observed cross section of  $Ga^{72}$  is obviously an upper limit to the evaporation cross section,  $\sigma_E(Ga^{72})$ , so

$$\sigma_E(Zn^{72}) \leq (2.28 \text{ mb}) \times \frac{\int_{S_p}^{E_m} N(U)G_p(U)dU}{\int_{S_n}^{E_m} N(U)G_n(U)dU}. \quad (2)$$

$G_p(U)$  and  $G_n(U)$  are evaluated by the evaporation formalism. The computer program described previously<sup>24</sup> was used, with some modifications:

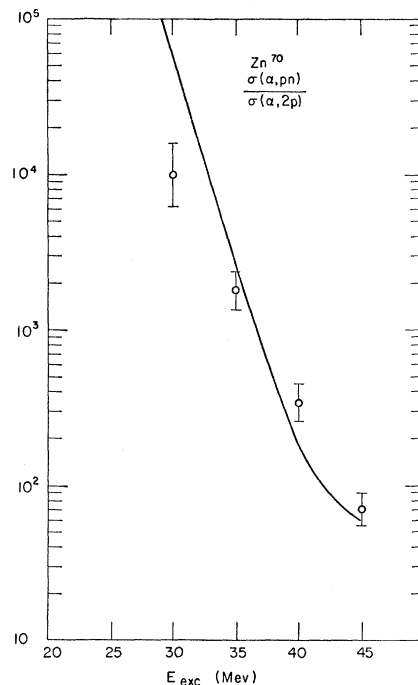


FIG. 3. Experimental points and calculated curve for the cross section ratio  $Zn^{70}(\alpha, pn):Zn^{70}(\alpha, 2p)$ . The points were taken from the smooth curves of reference 4, and the errors from typical experimental points.

<sup>22</sup> N. Metropolis, R. Bivins, M. Storm, J. M. Miller, G. Friedlander, and A. Turkevich, Phys. Rev. **110**, 204 (1958).

<sup>23</sup> I. Dostrovsky, P. Rabinowitz, and R. Bivins, Phys. Rev. **111**, 1659 (1958).

<sup>24</sup> S. Kaufman, Phys. Rev. **117**, 1532 (1960).

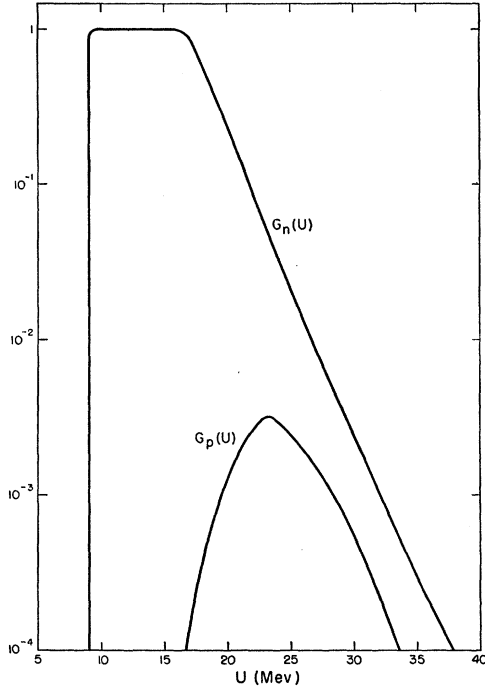


FIG. 4. Calculated values of  $G_n(U)$  and  $G_p(U)$ , probabilities of evaporating one and only one neutron and proton, respectively, from  $Ga^{73}$ , at excitation energy  $U$ .

(a) The level-density function used was

$$\begin{aligned} \omega(E) &= C, \quad 0 < E < \delta; \\ \omega(E) &= C \exp\{2[a(E-\delta)]^{1/2}\}, \quad E > \delta. \end{aligned} \quad (3)$$

This function still retains the possibility of transitions to states below the characteristic level, and makes the computations simpler. The alteration from the function used previously,<sup>24</sup> which did not have a break at  $E=\delta$ , made little difference in the results.

(b) The expression for the inverse cross section for neutrons given by Dostrovsky, Fraenkel, and Friedlander<sup>2</sup> was used:

$$\sigma(n) = \pi R^2(1.29 + 0.072/\epsilon), \quad \text{for } A=72. \quad (4)$$

(c) Competition from a possible third evaporating particle was added to the program; this was not necessary at the low energy used previously.

The level density parameter,  $\delta$ , is best evaluated from low-energy excitation functions. Amiel<sup>4</sup> has measured excitation functions for alpha-particle-induced reactions leading to the four  $A=72$  isobars studied here, and Dostrovsky, Fraenkel, and Friedlander<sup>2</sup> have obtained values of  $\delta$  which fit three of these quite well. The cross section for the  $Zn^{70}(\alpha, 2p)Zn^{72}$  reaction is so small that no Monte Carlo cascades were observed for that reaction. These  $\delta$  values were used in the present calculation. Additional parameters used were: the level-density parameter  $a = A/20$ ; the nuclear radius  $R = 1.5 \times 10^{-13} A^{1/3}$

cm; barrier penetrabilities for a square well potential<sup>25</sup>; separation energies from a recent compilation.<sup>26</sup>

The experimental and calculated excitation functions are shown in Fig. 2. The ratio of  $Ga^{72}$  to  $Zn^{72}$ , which is the important quantity, is shown in Fig. 3. The agreement between the calculated curve and the experimental points over a wide range in this ratio is satisfactory, and one therefore can assume that the evaporation behavior of  $Ga^{73}$ , which is the critical factor in this ratio, has been calculated satisfactorily. The values of  $G_p(U)$  and  $G_n(U)$  for  $Ga^{73}$  are shown in Fig. 4, which shows the rapid decrease in these quantities due to competition from a second evaporated particle.

The excitation energy distribution,  $N(U)$ , in  $Ga^{73}$  was obtained from the results of the cascade calculations.<sup>22</sup> The calculations for 1840-Mev protons incident on  $Cu^{64}$  nuclei were used, and all  $(p, 3n)$  cascades were combined to get  $N(U)$  after a  $(p, 3p)$  cascade. It was felt that the improvement in statistics justified this procedure. A small fraction of  $Ge^{74}$  nuclei produced in  $(p, 2p)$  cascades evaporates a proton to give  $Ga^{73}$ , and  $N(U)$  was corrected for this contribution.

An experimental check on the resulting spectrum can be obtained by considering the cross sections of  $Ga^{73}$  and  $Ga^{72}$ . Since  $Ga^{73}$  nuclei with  $U < 9$  Mev will de-excite by gamma emission, giving  $Ga^{73}$ , while those with  $9 < U < 19$  Mev will evaporate one neutron,

$$\frac{\sigma(Ga^{73})}{\sigma_E(Ga^{72})} = \int_0^9 N(U) dU / \int_9^{19} N(U) dU. \quad (5)$$

Since  $\sigma_E(Ga^{72}) \leq 2.28$  mb, and  $\sigma(Ga^{73}) = 0.62$  mb,

$$\int_0^9 N(U) dU / \int_9^{19} N(U) dU \geq 0.27. \quad (6)$$

The  $N(U)$  spectrum obtained from the cascade calculations was a histogram, but a smooth curve could be drawn which obeyed the lower limit set by Eq. (6). Evaluation of the integrals in Eq. (2) gave

$$\begin{aligned} \sigma_E(Zn^{72}) &\leq (2.28 \text{ mb})(3.3 \times 10^{-3}) \\ &\leq 7.5 \times 10^{-3} \text{ mb}. \end{aligned} \quad (7)$$

Since the peak of  $G_p(U)$  is at 23 Mev, while  $G_n(U)$  decreases above 16 Mev, any  $N(U)$  which increased less steeply, still obeying Eq. (6), would decrease the calculated fraction of proton evaporation, giving a ratio of  $\sigma_E(Zn^{72})/\sigma_E(Ga^{72})$  less than  $3.3 \times 10^{-3}$ . Therefore,  $7.5 \times 10^{-3}$  mb is a definite upper limit to  $\sigma_E(Zn^{72})$ . Since this is considerably less than the observed value of  $25 \times 10^{-3}$  mb, the difference must be accounted for. From the discussion above, the two possibilities are direct formation of  $Zn^{72}$  by a  $(p, 4p)$  cascade with less than 8.3 Mev of residual excitation energy, or by

<sup>25</sup> M. M. Shapiro, Phys. Rev. **90**, 171 (1953).

<sup>26</sup> F. Everling, L. A. König, and J. H. E. Mattauch, Nuclear Phys. **18**, 529 (1960).

formation of  $\text{Zn}^{73}$  or  $\text{Zn}^{74}$  excited nuclei, which evaporate one or two neutrons, respectively. The latter nuclei can only be formed in cascades in which mesons are emitted,  $(p, 3p\pi^+)$  or  $(p, 2p2\pi^+)$ .

The latter processes would be expected to have a sharply decreasing cross section as the bombarding energy is decreased, becoming zero below the meson production threshold. On the other hand, the Monte Carlo cascade calculations do not indicate any striking change in the probability of the  $(p, 4p)$  cascade with energy.

The  $\text{Ga}^{72}$  cross section is 2.5 mb at 380 Mev,<sup>19</sup> while the  $\text{Zn}^{72}$  cross section is about 0.008 mb<sup>19,20</sup>; from the preceding calculations it appears that the  $\text{Zn}^{72}$  cross section at 380 Mev is entirely due to evaporation of a proton from  $\text{Ga}^{73}$ . So the "excess"  $\text{Zn}^{72}$  rises sharply from a threshold of 400 Mev or greater to a value of more than 0.018 mb at 2.9 Bev. This behavior agrees with that expected from the meson mechanism.

It should be emphasized that a number of reactions, all involving emission of one or more  $\pi^+$  mesons, may contribute to the "excess"  $\text{Zn}^{72}$ , since only those reactions which proceed through  $\text{Ga}^{73}$  as intermediate nucleus and the  $(p, 4p)$  cascades have been eliminated. The only such reactions whose cross sections have been measured are  $(p, p\pi^+)$  in  $\text{Al}^{27}$ ,  $\text{Cu}^{65}$ , and  $\text{In}^{115}$ , with cross sections of 0.1–0.2 mb,<sup>17,27,28</sup> and  $(p, p2\pi^+)$  in  $\text{I}^{127}$ , with a cross section of <0.02 mb.<sup>16</sup>

#### ACKNOWLEDGMENTS

The author is indebted to Dr. R. W. Dodson and the staff of the Chemistry Department for their hospitality during his visit to Brookhaven National Laboratory, and to Dr. G. Friedlander, who suggested this research. He wishes to thank the many people there who assisted in this work, especially Dr. R. W. Stoenner and Dr. J. K. Rowley for performing chemical yield determinations. Discussions with Dr. J. M. Miller, Dr. A. M. Poskanzer, and Dr. J. B. Cumming were especially

helpful. Finally, he wishes to thank the Chemistry Department of Princeton University for the grant of a leave of absence.

#### APPENDIX. CHEMICAL PROCEDURES

Targets were dissolved with the aluminum wrapping or catcher foils in a mixture of HCl and  $\text{H}_2\text{O}_2$  containing carriers of Zn, Ga, Ge, and Se. The  $\text{H}_2\text{O}_2$  served to oxidize As and Se to their highest valence states and prevent volatilization as chlorides or hydrides. Ge was removed by two distillations in a stream of chlorine. As and Se were then distilled in a stream of HBr, which caused their reduction to the volatile bromides. As and Se were separated and purified from each other by a series of precipitations of Se from concentrated HCl with  $\text{SO}_2$ . The Se purification was done several days after the bombardment, to insure that all the  $\text{Se}^{73}$  had decayed to  $\text{As}^{73}$ , which otherwise would be present in the As milk. As carrier was added to the purified Se and an aliquot taken for determination of the Se chemical yield. After 2–4 days, a Se-As separation was done, and the As milk counted as  $\text{As}_2\text{S}_3$ .

After the Se scavenges, the As fraction was precipitated as  $\text{As}_2\text{S}_3$ , dissolved in  $\text{HCl} + \text{H}_2\text{O}_2$ , and a Ge distillation carried out, followed by distillation of As.  $\text{As}_2\text{S}_3$  was precipitated from the distillate and counted with the NaI crystal.

The residue after the first distillation was diluted with 6M HCl, and Ga was extracted into diethyl ether. After washing and back-extraction into water, two  $\text{Fe}(\text{OH})_3$  scavenges were done, the solution made 6M with HCl, KI added to reduce Fe, and the Ga extracted as before. Ga was precipitated as the 8-hydroxyquinolate.

The HCl solution after Ga extraction was boiled to dryness, the residue dissolved in 0.5M HCl, and passed through a Dowex-1 ion exchange column. The column was washed with 0.5M HCl until no more activity appeared in the washings, and Zn was eluted with 0.001M HCl. The eluate was made 6M in HCl and a Ga extraction into diethyl ether done. Ga carrier was added to the purified Zn, an aliquot taken for determination of the Zn chemical yield, and after 1–2 days Ga was milked and counted as the 8-hydroxyquinolate.

<sup>27</sup> P. A. Benioff, Phys. Rev. **119**, 316 (1960).

<sup>28</sup> D. W. Barr, Atomic Energy Commission Document, University of California Radiation Laboratory Report UCRL-3793, 1957 (unpublished).