Production of Gallium Isotopes in Copper by High-Energy Protons*

JOHN WARREN
Enrico Fermi Institute for Nuclear Studies, University of Chicago, Chicago, Illinois, and Chemistry Department, University of California, Davis, California

AND

ANTHONY TURKEVICH
Enrico Fermi Institute for Nuclear Studies, University of Chicago, Chicago, Illinois
(Received 30 November 1964; revised manuscript received 8 February 1965)

The production of Ga^{66}, Ga^{67}, and Ga^{68} in copper foils by high-energy protons has been determined by radiochemical techniques. Cross-section measurements were made for a series of copper foils ranging in thickness from 8 to 555 mg/cm² at each of the four proton energies 123, 254, 351, and 440 MeV. The thickness variations of the cross sections for all three isotopes were found to be consistent with the concept that these isotopes are formed by the capture in copper nuclei of alpha particles produced in primary interactions. Spectra of alpha particles with energies greater than 14 MeV which satisfactorily reproduced the experimental thickness variations were determined at each proton energy. These spectra were found to consist of a large number of evaporation alpha particles with a small forward-directed alpha-particle component. No evidence was found for the primary reaction $Cu^{65}(p,\pi^-)Ga^{66}$; the contribution of such a reaction to the total Ga^{66} production cross section is less than one-third of a microbarn in natural copper at a proton energy of 440 MeV.

I. INTRODUCTION

In reactions involving complex nuclei and high-energy protons, alpha particles are often emitted with high energies. These particles can in turn react with other nuclei of the target material to create products with an atomic number greater by two units than that of the target. Measurements of the production of isotopes formed by secondary reactions can be used to estimate the mean energy and energy spectrum of these secondary particles. This information is valuable for the complete understanding of the mechanism for formation of alpha particles and other fragments in the primary reactions.

The production of such "trans-target" nuclei has been studied in many radiochemical investigations.[1–11] The characteristics of the alpha spectra responsible have been deduced from the probabilities of formation of different isotopes by using theoretical estimates of the cross sections of (α,xn) reactions.[12] Another method has involved the measurement of the production of such trans-target nuclides in stacked foils.[8,10] The conclusion has been reached that some of the alpha particles responsible can be attributed to conventional nuclear evaporation processes. However, a high-energy, forward-directed component, usually ascribed to "direct interactions," is also needed to explain the results. The spectra deduced in this way have been compared with direct observations of the alpha particles by photographic techniques.[13]

In heavy elements like bismuth, direct-interaction alpha particles seem to predominate[8,10,14] at the lower bombarding energies, but the importance of the evaporation process increases with increasing energy. In a lighter element such as copper the evaporation alpha particles appear to be the more numerous even at incident proton energies below 100 MeV.[14,15] For example, at 56 MeV a total cross section for alpha-particle production of 159±5 mb has been estimated[14] with 134±5 mb attributable to evaporation and 25±7 mb to direct interaction.

Radiochemical studies of the formation of trans-target nuclei in light elements[2,3,5] have been less extensive than in the case of heavy elements. Copper is a satisfactory target for such investigations because, although it has two stable isotopes, the $Cu(\alpha,xn)$ reactions lead to three radioactive gallium isotopes of convenient half-life, Ga^{66}, Ga^{67}, and Ga^{68}. The cross sections for forming these nuclides in copper targets under low-

* This work was supported in part by the U. S. Atomic Energy Commission.
[1] A. E. Metzger and J. M. Miller, Phys. Rev. **113**, 1125 (1959).
[2] R. E. Batzel, D. R. Miller, and Glenn T. Seaborg, Phys. Rev. **84**, 671 (1951).
[3] A. Turkevich and N. Sugarman, Phys. Rev. **94**, 728 (1954).
[4] B. V. Kurchatov, V. N. Mekhedov, L. V. Chistiakov, M. I. Kuźnetsova, N. I. Borisova, and V. G. Solov'ev, Zh. Eksperim. i Teor. Fiz. **35**, 56 (1958), [English transl.: Soviet Phys.—JETP **8**, 40 (1959)].
[5] A. K. Lavrukhina, I. M. Grechishcheva, B. A. Kholin, At. Energ. (USSR) **6**, 1945 (1959); J. Nucl. Energy: Pt A: Reactor Science **11**, 200 (1960).
[6] M. Lefort, G. Simonoff and X. Tarrago, Nucl. Phys. **19**, 173 (1960).
[7] Wang Yung-Yü, V. V. Kuznetsov, M. Ya Kuznetsova, and V. A. Khalkin, Zh. Eksperim. i Teor. Fiz. **39**, 230 (1960) [English transl.: Soviet Phys.—JETP **12**, 166 (1961)].
[8] H. Gauvin, M. Lefort, and X. Tarrago, Nucl. Phys. **39**, 447 (1962).
[9] B. N. Belyaev, N. S. Mal'tseva, V. N. Mekhedov, Ming Nam Buk, and R. A. Szymczak, Zh. Eksperim. i Teor. Fiz. **43**, 1129 (1962) [English transl.: Soviet Phys.—JETP **16**, 757 (1963)].
[10] M. Lefort and X. Tarrago, Nucl. Phys. **46**, 161 (1963).
[11] N. S. Mal'tseva, V. N. Mekhedov, and V. N. Rybakov, Zh. Eksperim. i Teor. Fiz. **45**, 852 (1963) [English transl.: Soviet Phys.—JETP **18**, 585 (1964)].
[12] J. D. Jackson, Can. J. Phys. **34**, 767 (1956).
[13] L. E. Bailey, University of California Radiation Laboratory Report No. UCRL-3334, 1956 (unpublished).
[14] J. Muto, H. Itoh, K. Okano, N. Shiomi, K. Fukuda, Y. Omori, and M. Kihara, Nucl. Phys. **47**, 19 (1963).
[15] C. B. Fulmer and C. D. Goodman, Phys. Rev. **117**, 1139 (1960).

energy alpha-particle bombardments have recently been determined.[16] Turkevich and Sugarman[3] measured cross sections for production of Ga^{66}, Ga^{67}, and Ga^{68} with 2.2-GeV protons. An analysis of the results indicated that a mean alpha particle energy of 35 MeV satisfied the experimentally observed variation of the apparent production cross sections of gallium with foil thickness. They estimated that the cross section for production of alpha particles produced by 2.2 -GeV protons amounted to approximately 300 mb.

Cross sections for the production of Ga^{66} in natural copper have been determined by Lavrukhina *et al.*[5] at proton energies from 130 to 480 MeV. Because of the large increase in cross section observed as the proton energy was increased from 130–350 MeV, the authors argued that the yield of Ga^{66} could not result wholly from secondary reactions involving the production and reabsorption of alpha particles. They suggested that at 130 MeV the yield resulted from the reaction $Cu^{65}(\alpha,3n)Ga^{66}$, and above this proton energy the primary reaction $Cu^{65}(p,\pi^-)Ga^{66}$ became increasingly important. They observed that the yield of Ga^{66} decreased slightly at 480 MeV and postulated that reactions of the type $Cu^{65}(p,\pi^- xn)$ began to compete with the simpler reaction.

The interest in a more quantitative study of the formation of gallium isotopes in high-energy proton bombardments of copper is thus twofold: It would give more information on the numbers and characteristics of alpha particles emitted during high-energy reactions, and it would check the proposal of Lavrukhina *et al.* that the (p,π^-) reaction in a complex nucleus has a measurable cross section. For these reasons it was decided to study the production of gallium isotopes in copper targets of various thicknesses at a number of energies from 123 to 440 MeV.

II. EXPERIMENTAL PROCEDURES

Irradiations took place in the internal beam of the University of Chicago 170-in. Synchrocyclotron. The energy of the proton beam was determined by the radius at which the probe target was placed. Irradiations generally lasted for 1 h; in some cases when beam intensities were high, half-hour periods were used.

In all cases high-purity copper was used for target material. Spectrographic analysis revealed that the quantities of impurities with atomic weights greater than copper were too low to contribute to the production of the gallium isotopes. In most instances single copper foils were irradiated, but in the cases where thick targets were desired several of the thinner foils were fastened together in a foil stack. In these cases the leading edges of the stacks were carefully aligned and then machined so that all foils making up a stack were exposed to the same beam intensity.

In the study of primary nuclear reactions, target foils are generally wrapped with aluminum so that the reaction $Al^{27}(p,3pn)Na^{24}$ may be used as beam monitor. In order to avoid possible secondary reactions in the copper resulting from alpha particles born in the aluminum, no wrapping was used. The cross sections for the gallium isotopes were related to Ni^{57} produced by the primary reactions $Cu^{63}(p,2p5n)Ni^{57}$ and $Cu^{65}(p,2p7n)Ni^{57}$. The cross section for production of this nuclide relative to that of Na^{24} produced in aluminum, and the absolute cross section for Na^{24} over the proton energy range used in these experiments, were taken from the work of Fung *et al.*[17]

After irradiation, the first 4 mm from the leading edge was cut from the target foil and dissolved in nitric acid. Gallium and nickel carriers were then added as well as the appropriate holdback carriers. After removal of the sulphide group by precipitation from an acid solution, gallium and iron were extracted into isopropyl ether and back-extracted into a water solution. Iron was precipitated from a basic solution and then the solution was acidified. The appropriate holdback carriers were added and the cycle repeated. Gallium was finally precipitated from a neutral solution by the addition of 8-hydroxyquinoline. Nickel was purified by repeated precipitations of nickel dimethyl glyoxime with and then without holdback carriers.

All samples were counted with end-window methane-flow beta proportional counters. The gross decay curves, which showed three components, were resolved graphically and the saturation activity for each component was determined. The half-lives used in the analyses were 9.45 h for Ga^{66}, 77.9 h for Ga^{67}, 68 min for Ga^{68}, and 36 h for Ni^{57} as given by Strominger *et al.*[18] Corrections were applied to the saturation activities for chemical yield, self-scattering and self-absorption in the samples, absorption in the air and counter window, counter geometry, branching ratios, and undetected electron capture.

Ga^{67} decays wholly by electron capture but conversion electrons result from the decay of the 93-keV level. The beta proportional counters were used to count these soft electrons. The internal-conversion coefficient for the 93-keV transition was taken as 0.63 and the branching ratio to the level was taken as 48%.[18] All saturation activities of Ga^{67} were corrected for these effects as well as for scattering and absorption in the sample itself, in the air and the counter window.

III. RESULTS

The results are listed in Table I for the various isotopes studied, for the foil thicknesses and energies of protons used. Each cross section reported is based on natural copper and is the mean value determined from the results of two or more bombardments except for certain values at the lower proton energies where only

[16] N. T. Porile and D. L. Morrison, Phys. Rev. **116**, 1193 (1959).

[17] Si-Chang Fung and A. Turkevich, Phys. Rev. **95**, 176 (1954).

[18] D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Mod. Phys. **30**, 585 (1958).

TABLE I. Cross sections for production of Ga^{66}, Ga^{67}, and Ga^{68} by protons on natural copper in foils of various thicknesses.

Proton energy (MeV)	Foil thickness (mg/cm^2)	Production cross sections (microbarns) Ga^{66}	Ga^{67}	Ga^{68}
123	555	6.75[a]	1.93[a]	4.17[a]
	278	...	...	...
	92.5	3.90[a]	1.05[a]	3.36[a]
	30.1	2.76[a]	0.64[a]	2.48[a]
	15.6	2.09	0.43	1.80
	8.06	1.40	0.43	1.47
254	555	10.1[a]	3.81[a]	8.45[a]
	278	8.99	3.54	8.32
	92.5	6.42	1.96	5.81
	30.1	4.45	1.13	4.00
	15.6	3.28	0.83	2.73
	8.06	...	...	...
351	555	14.0	5.16	10.3
	278	13.0	3.74	9.51
	92.5	9.35	2.90	11.2
	30.1	5.59	1.43	5.82
	15.6	4.20	1.20	3.61
	8.06	...	...	...
440	555	14.5	5.04	10.5
	278	16.1	6.97	11.2
	92.5	12.6	3.73	9.04
	30.1	7.85	2.51	7.00
	15.6	5.08	1.33	4.37
	8.06	3.54	1.02	3.05

[a] One bombardment only.

one bombardment was obtained. The spread between results from duplicate irradiations was found to be about 5% for Ga^{66} and Ga^{67} and about 8% for Ga^{68}. The larger error for Ga^{68} arises from the difficulty in resolving the third component of a complex decay curve.

The production cross sections for Ga^{66}, Ga^{67}, and Ga^{68} at 123 MeV for the thick foils are 6.75, 1.93, and 4.71 μb, respectively. It is apparent from the table that the cross section at a given energy and foil thickness for Ga^{66} is almost always slightly larger than that of Ga^{68}, while the cross section of Ga^{67} is about a factor of 3 lower. As the proton energy is increased the cross sections of all three isotopes increase by a factor of 2 to 3 to 350 MeV, and then there is an apparent leveling off of values. Table I also shows that at a given proton energy, as the foil thickness is increased from 8.06 mg/cm^2 there is a sharp rise in the production probability for the three isotopes. After a thickness of 92.5 mg/cm^2 the rate of increase becomes considerably less. This dependence on target thickness is consistent with the idea that the gallium isotopes are produced by secondary reactions.

The cross sections reported here for thick targets at 350 MeV are in good agreement with the approximate values determined by Batzel *et al.*[2] The values of the production cross sections for Ga^{66}, Ga^{67}, and Ga^{68} given by Batzel *et al.* for thick targets are 10, 6, and 10 μb, respectively. The thick target cross sections reported by Turkevich and Sugarman[3] for 2.2-GeV protons are a factor of 3 to 4 times larger than the values at 440 MeV in the case of Ga^{66} and Ga^{68}. The cross section for Ga^{67} is about a factor of 12 greater than the 440-MeV value, but it should be noted that the counting efficiency used by Turkevich and Sugarman for this isotope is particularly uncertain.[19]

Table II compares the cross sections for Ga^{66} production in copper for thick targets determined in the current experiments and the cross sections reported by Lavrukhina *et al.*[5] In both experiments the same monitor reaction was used with only slightly different values of the Na^{24} cross section. It is not apparent from their paper whether the Ga^{66} cross sections were calculated on the basis of natural copper or on Cu^{65} alone. If natural copper was used their cross sections are greater than those reported here by a factor of slightly greater than 3. On the assumption that they reported the Ga^{66} cross sections on the basis of Cu^{65}, their values have been converted to natural copper and are given in column 3 of Table II.

TABLE II. Comparison of cross sections for production of Ga^{66} in natural copper.

Proton energy (MeV)	Cross sections for Ga^{66} (microbarns) Lavrukhina *et al.*[a]	Lavrukhina *et al.* corrected[b]	This work[c]
123	...	...	6.75
130	13±1.5	4.0±0.5	...
190	20±2.0	6.2±0.6	...
250	31±2.0	9.6±0.6	10.1
350	44±2.5	13.6±0.8	14.0
440	...	...	14.5
480	35±2.0	10.8±0.6	...

[a] Reference 5.
[b] Cross sections corrected assuming those given in column 2 are based on Cu^{65} rather than natural copper.
[c] The values in this column are results for thick foils (555 mg/cm^2).

An examination of columns 3 and 4 of Table II reveals excellent agreement at 250 and 350 MeV. At the low- and high-energy ends the results of the current experiments are about 40% higher, and no evidence is found that the cross section may be decreasing at the high-energy end. However, it was not possible in these experiments to reach a proton energy of 480 MeV as did Lavrukhina *et al.*

IV. ANALYSIS OF DATA

Table I indicates that the measured cross sections increase markedly at a given proton energy, as the foil thickness is increased. This behavior is characteristic of the formation of gallium by a secondary-reaction mechanism. Consequently, it seemed appropriate to explore whether the experimental data could be explained quantitatively in terms of the production and capture of alpha particles in the copper targets. It was first assumed that the alpha particles were emitted in the primary

[19] A. Turkevich (private communication).

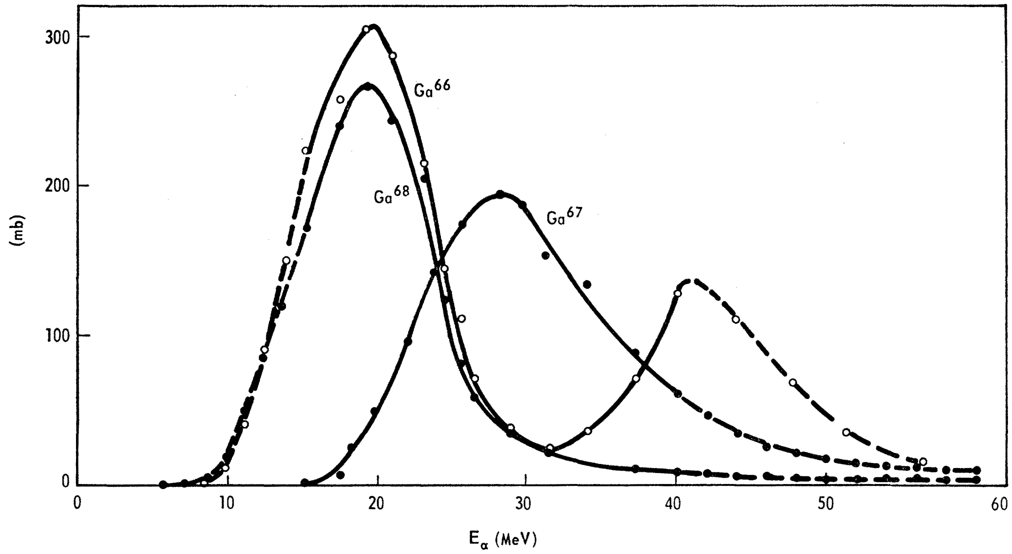

FIG. 1. Excitation functions determined by Porile and Morrison (Ref. 16) for the production of Ga^{66}, Ga^{67}, and Ga^{68} in natural copper under alpha-particle bombardment. The dashed portions represent reasonable extrapolations for energy regions not investigated by these authors.

interaction with a unique energy in the direction of the proton beam. Attempts were then made to find a value of the alpha energy which best reproduced the experimental cross sections for each isotope independently. The results were unsatisfactory both in the quality of the fit to the experimental data and the physical reasonableness of the resultant energies. In order to improve the fit for each isotope the assumption was then made that the unique-energy alpha particles were emitted isotropically in the laboratory system. This likewise led to unsatisfactory results. Lastly, attempts were made to find an alpha-particle spectrum which best reproduced the cross-section dependence on thickness and energy for all three isotopes.

In these analyses the excitation functions of the reactions Cu(α,xn) for incident alpha particles of 15 to 41 MeV were taken from the data of Porile and Morrison[16] and are reproduced in Fig. 1. Their excitation functions were extrapolated by reasonable methods[20] from the thresholds of the various reactions to 15 MeV and from 41 MeV to about 60 MeV. The ranges and energy losses of alpha particles in copper were calculated from Sternheimer's values[21] for protons in the same element.

(a) Monoenergetic, Unique-Angle Alpha-Particle Emission

In view of the fact that there is evidence for a direct interaction mechanism for the production of alpha particles in heavy nuclei, an attempt was made to fit the experimental data first on the assumption that the alpha particles are monoenergetic and emitted wholly in the forward direction. This case also covers the situation where the monoenergetic alpha particles are given off at a unique angle other than at zero degrees relative to the proton beam.

[20] M. M. Shapiro, Phys. Rev. **90**, 171 (1953).
[21] R. M. Sternheimer, Phys. Rev. **115**, 137 (1959).

Using the cross-section data of Porile and Morrison[16] and the stopping-power functions of Sternheimer,[21] the production of each gallium isotope by secondary reactions at a given proton energy in foils of different thickness was calculated from geometrical considerations. Optimizing the fit to the observed production cross sections as a function of foil thickness gave values for the energies and numbers of the alpha particles responsible for the gallium production in copper.

The results of this analysis were completely unsatisfactory in providing an explanation for the results obtained experimentally. Alpha particle energies in the region from 16–19 MeV were needed to fit the Ga^{66} and Ga^{68} data, but energies of the order of 27.5 MeV were required to explain the production of Ga^{67}. Moreover, even with different alpha particle energies for each isotope and each proton bombarding energy, the fit to the thickness curve was only to a standard deviation of about 20% (as compared to the experimentally expected 5–10%) and, in addition, showed systematic deviations.

In an effort to improve the fit of calculated to observed values, mixtures of two discrete alpha-particle groups were investigated sufficiently to establish that it was impossible to get a satisfactory representation of the production of all three gallium isotopes as a function of target thickness at a given proton bombarding energy using such a composite alpha-particle spectrum.

(b) Monoenergetic, Isotropic Alpha-Particle Emission

It is known that, at the proton energies used in these experiments, the evaporation process for the emission

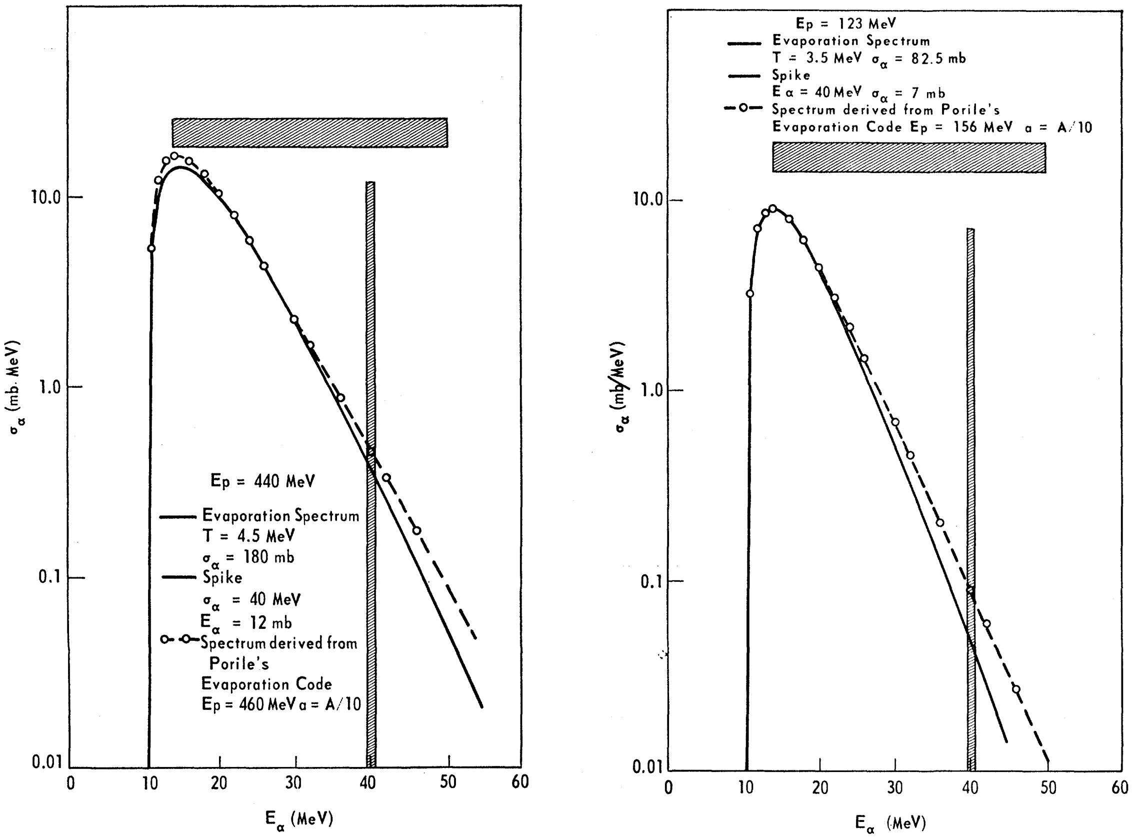

FIG. 2. (a) Comparison of the alpha-particle spectrum produced in copper which best satisfies the observed thickness variations for 440-MeV protons with the spectrum derived from Porile's evaporation code (Ref. 27) applied to the residual nuclei from the intranuclear-cascade calculations of Metropolis *et al.* (Refs. 25, 26) for 460-MeV protons. The spectra have been normalized at 20 MeV. The cross-hatched region along the top indicates the energy region to which the present experiments are sensitive. (b) Comparison of the alpha-particle spectrum produced in copper which best satisfies the observed thickness variations for 123-MeV protons with the spectrum derived from Porile's evaporation code (Ref. 27) applied to the residual nuclei from the intranuclear-cascade calculations of Metropolis *et al.* (Refs. 25, 26) for 156-MeV protons. The spectra have been normalized at 20 MeV. The cross-hatched region along the top indicates the energy region to which the present experiments are sensitive.

of neutrons and charged particles plays an important part in nuclear reactions. As a consequence it was decided to see if the fit to the experimental data could be improved by assuming that the alpha particles were emitted monoenergetically and isotropically in the laboratory system. As in the previous case, the secondary production of each gallium isotope at a given proton energy in foils of different thickness was calculated from geometric considerations incorporating the data of Porile and Morrison and of Sternheimer and assuming various alpha particle energies. The energies and numbers of alpha particles which best reproduced the gallium production in copper were then deduced.

The results of such calculations indicated that considerably better fits to the experimental data are obtained in this way than in the case of unique angle emission. The fits to the Ga^{66} and Ga^{68} data approach those expected from the experimental errors. However, in the case of Ga^{67} while the fits are improved, the fractional standard deviations remain large and once more different alpha-particle energies are needed to form the different gallium isotopes. The production of Ga^{66} and Ga^{68} requires alpha particles of 24 MeV while that of Ga^{67} requires 35-MeV alpha particles in about one-third the amount needed for the other gallium isotopes.

(c) Isotropic Alpha Emission with a Distribution of Energies

The idea of a monoenergetic emission of alpha fragments is clearly an over-simplification. In order to see if a more realistic physical model would yield an improved representation of the experimental gallium cross sections, the alpha particle emission was assumed to be due to an evaporation process. The energy distribution of the alpha particles was based on the well-known equation of Weisskopf[22]

$$W[E]dE=(E-V_{\mathrm{eff}}/T^2)\exp[-E(-V_{\mathrm{eff}})/T]dE. \quad \text{(I)}$$

[22] V. Weisskopf, Phys. Rev. **52**, 295 (1937).

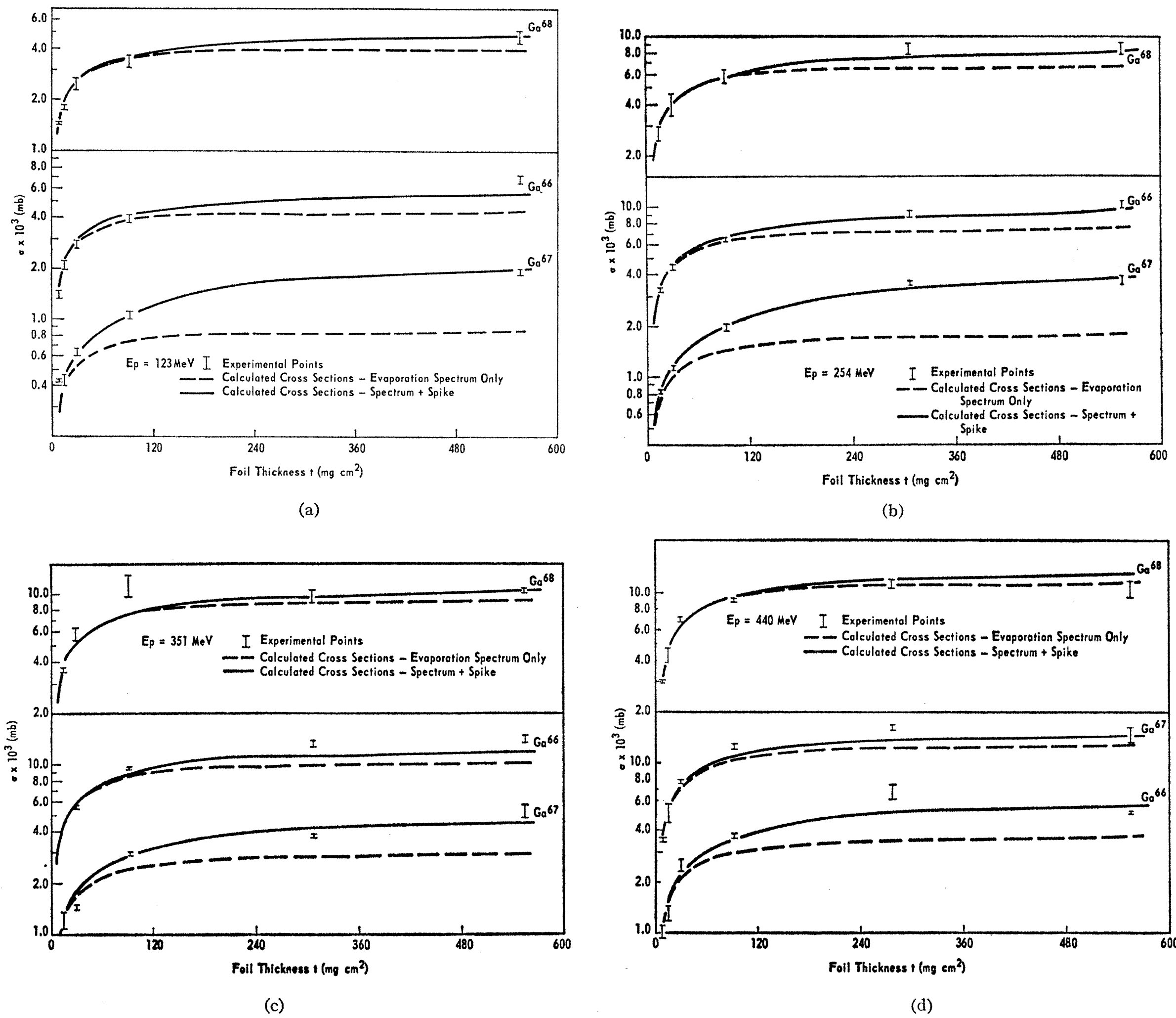

FIG. 3. Variations of the Ga^{66}, Ga^{67}, and Ga^{68} production cross sections in copper foils of various thicknesses bombarded with protons of energy 123 MeV (a); 254 Mev (b); 351 MeV (c); and 440 MeV (d).

In this expression $W(E)$ represents the probability of emitting an alpha particle with energy between E and $E+dE$, V_{eff} is the effective Coulomb barrier, and T the nuclear temperature. The effective Coulomb barrier of 10.5 MeV was calculated in the usual manner taking $r_0=1.5$ F and correcting for penetration by the factor given by Dostrovsky.[23] The use of this formula for the energy distribution in the laboratory system assumes a negligible contribution from possible motion by the emitting nucleus.

The number of secondary reactions occurring in the foil per alpha particle emitted at a given energy was calculated from formulas derived on the assumptions that the alpha particles were emitted monoenergetically and isotropically in the target foil. The total number of secondary reactions occurring in the foil per emitted alpha particle was then determined by simple integration over the energy spectrum of the emitted alpha particles as given by Eq. (I). The nuclear temperature associated with the distribution and the integrated alpha-particle-production cross section which gave the best fit to the thickness curves, were again calculated for each gallium isotope at each incident-proton energy. These best fit parameters as well as the fractional standard deviations of the experimental compared to the theoretical cross sections are presented in Table III. In the case of Ga^{67} the calculations indicated that the best fit corresponded to temperatures above 7.5 MeV, the largest value used in these analyses.

The use of the evaporation type spectrum reproduces the experimental thickness data very well for Ga^{66} and Ga^{68}. However, the nuclear temperatures needed are usually unrealistically high and do not appear to increase, as expected, with increasing incident proton energy. In the case of Ga^{67} the treatment breaks down completely.

[23] I. Dostrovksy, Z. Fraenkel, and G. Friedlander, Phys. Rev. **116**, 683 (1959).

TABLE III. Nuclear temperatures, alpha-particle production cross sections, and fractional standard deviations determined using the evaporation spectrum.

Isotope	Proton energy (MeV)	Nuclear temperature (MeV)	Alpha production cross section (mb)	Fractional standard deviation (percent)
Ga^{66}	123	6.1	65.1	11.9
	254	6.2	99.9	7.4
	351	7.1	128.0	4.8
	440	5.8	174.3	6.0
Ga^{67}	123	≥ 7.5	≤ 32.2	~ 19.9
	254	>7.5	<58.0	<16.2
	351	>7.5	<76.7	<15.6
	440	>7.5	<103.3	<15.3
Ga^{68}	123	4.0	79.5	9.6
	254	6.3	101.1	5.2
	351	5.8	146.0	14.7
	440	4.4	184.5	6.6

(d) Combination of Evaporation Spectrum and Unique-Angle Monoenergetic Emission

The treatments summarized above were all unsatisfactory in one respect or another in explaining the thickness variations of the production cross sections of the gallium isotopes and indicated that a two component spectrum was needed. It was noticed that at each proton energy the cross sections for the production of all three gallium isotopes in the thin foils could be fitted by the same evaporation type spectrum of reasonable temperature; it was the thicker foils that involved extra production in varying amounts of the different isotopes. This extra production could be explained by a high-energy, forward-directed component of the alpha-particle spectrum. This component had to be chosen to be high enough in energy and directed enough forward not to contribute significantly to the thinnest foils or to the production of Ga^{68} in the thick foils.

Specific spectra at the different proton bombarding energies were derived from the experimental data in the following manner. At each proton energy the experimental production cross sections for Ga^{66}, Ga^{67}, and Ga^{68} for the two or three thinnest foils (<50 mg/cm^2) were used to determine the temperature and the alpha-particle-production cross section of the isotropic evaporation spectrum that best fitted the data. This fit for all isotopes at the same proton energy for the thin foils was to a standard deviation of 11% or better. The resulting temperatures and alpha-particle-production cross sections are given in columns 2 and 3 of Table IV.

The contribution of these spectra to the cross sections for the three thickest foils showed the greatest (fractional) deficiency for Ga^{67}. A high-energy, forward-directed component was then introduced in an amount such that the total calculated cross section represented a best fit to all the data at a given proton bombarding energy. The high-energy, forward-directed component

TABLE IV. Parameters for evaporation spectra and unique-angle emission which "best fit" the experimental thickness variations.

Proton energy (MeV)	Nuclear temperature (MeV)	Alpha-production cross section due to spectrum (mb)	Unique-angle energy (MeV)	Alpha-production cross section due to spike (mb)
123	3.5	85.2	40	7
254	4.0	120	40	13
351	4.5	145	40	10
440	4.5	180	40	12

was chosen to be monoenergetic at 40 MeV in all cases, although calculations have shown that a spread of about 5 MeV above and below this value does not significantly change the agreement with the experimental data. The amounts needed corresponded to the production of such forward moving alpha particles with a cross section increasing from 7 mb at 123 MeV to 12 mb at 440 MeV and were found to be insensitive functions of the alpha-particle energy chosen.

The parameters of these spectra, involving an isotropic evaporation spectrum and a high-energy forward component, are summarized in Table IV. Two of the spectra are illustrated in Figs. 2(a) and 2(b). The extent to which these spectra reproduced the experimental thickness variations is illustrated in Fig. 3. In these graphs the contributions of the isotropic evaporation spectra are shown separately. Table V gives the values of the calculated gallium cross sections as determined from

TABLE V. Calculated gallium-production cross sections based on the "best fit" parameters.

Proton energy (MeV)	Foil thickness (mg/cm^2)	Calculated cross sections (microbarns) Ga^{66}	Ga^{67}	Ga^{68}	Fractional standard deviations (percent) Ga^{66}	Ga^{67}	Ga^{68}
123	555	5.62	2.00	4.88			
	278	5.13	1.78	4.49			
	92.5	4.07	1.04	3.54	9.3	15.8	7.0
	30.1	2.91	0.610	2.56			
	15.6	2.15	0.432	1.89			
	8.06	1.47	0.296	1.31			
254	555	9.56	3.90	8.24			
	278	8.65	3.47	7.49			
	92.5	6.64	2.05	5.70	4.1	5.0	5.9
	30.1	4.57	1.19	3.97			
	15.6	3.31	0.829	2.87			
	8.06	2.23	0.558	1.96			
351	555	11.93	4.61	10.35			
	278	11.00	4.19	9.59			
	92.5	8.74	2.83	7.58	11.3	15.3	17.3
	30.1	5.86	1.80	5.12			
	15.6	4.19	1.23	3.64			
	8.06	2.78	0.838	2.45			
440	555	14.76	5.67	12.82			
	278	13.63	5.15	11.90			
	92.5	10.85	3.51	9.43	9.9	14.9	10.5
	30.1	7.28	2.23	6.37			
	15.6	5.20	1.52	4.53			
	8.06	3.46	1.03	3.05			

the parameters in Table IV as well as the fractional standard deviations of the calculated from the observed cross sections for all isotopes at each proton energy.

Table V shows that the fractional standard deviations varied from 5% at 254 MeV to 15% at 351 MeV. Although this is somewhat higher than the reproducibility errors would predict, it is not unreasonable in view of the possible contribution of systematic errors from the relative counting efficiencies in this work as compared to those in the fundamental Cu(α,xn) cross section data. On the positive side, the same spectrum, at a given proton energy, now explains the production of all three isotopes with no systematic deviations from the experimental data.

V. DISCUSSION

The interpretation of the experimental data attributing the production of the gallium isotopes at all proton energies to secondary reactions by alpha particles is in contradiction to the proposal of Lavrukhina *et al.*[5] that a large fraction of the observed Ga^{66} yield is produced in copper by a (p,π^-) reaction on Cu^{65} at a proton bombarding energy above 200 MeV. This proposal was based primarily on the course of the excitation function obtained in their work.

The thinnest foil for which we have data at all four energies (15.6 mg/cm^2) is thick enough to retain Ga^{66} if it were formed by the (p,π^-) reaction. The data at this thickness show a smooth increase in the production cross section of Ga^{66} as a function of energy as shown in Fig. 4. Thus, the present results on the excitation function for the production of Ga^{66} in copper show no features requiring a (p,π^-) mechanism.

A second argument against the proposal of Lavrukhina *et al.* can be made by comparing the Ga^{66}-production cross section to that of Ga^{68} for the 15.6 mg/cm^2 foils at the incident proton energies used in this experiment. The ratios of the two cross sections are 1.15, 1.20, 1.16, and 1.16 at the proton energies of 123, 254, 351, and 440 MeV, respectively The ratio is almost constant over the proton-energy range, the small differences being

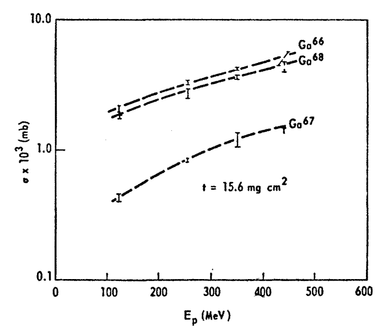

FIG. 4. Thin-target (15.6 mg/cm^2) excitation functions for the production of Ga^{66}, Ga^{67}, and Ga^{68} in natural copper induced by protons. The dashed curves represent the calculated excitation functions on the basis of secondary production while the points represent the experimentally determined cross sections.

apparently random in nature This constancy suggests that the same mechanism is responsible for the production of the two isotopes and that the contribution by the (p,π^-) reaction to the observed yield of Ga^{66} must be small since Ga^{68} can be made from copper by means of a secondary reaction only.

An even stronger argument against a significant contribution by the (p,π^-) reaction is the adequate explanation for the production of all three gallium isotopes in the thin foils by secondary alpha particles possessing a reasonable evaporation type spectrum. The adequacy of such an interpretation to account for the thickness dependence of the production of Ga^{66}, Ga^{67}, and Ga^{68} and the consistency of the excitation functions of all three isotopes argues very strongly that secondary alpha particles are responsible for the Ga^{66} seen in copper targets bombarded with 200–440-MeV protons.

Because of the significance of the (p,π^-) reaction an estimate of the maximum amount that might be present is desirable This estimate is most reliable at 440 MeV where a one-third microbarn cross section based on natural copper (one microbarn on Cu^{65}) would seem more than can be allowed on the basis of the fit to the thickness curves.

The technique employed here to investigate the spectra of alpha particles emitted in high-energy reactions has obvious limitations deriving primarily from its integral character. Thus, even with the detailed knowledge available for the excitation functions of the three gallium isotopes only a limited number of characteristics of the alpha spectra responsible for their production are determinable from a small number of experimental cross section values as a function of target-foil thickness. The spectra we have deduced cannot be considered as unique, but rather, as possible spectra that satisfy the observations. Some comments can be made, however, as to the general characteristics of spectra that can or cannot be determined by data of this kind.

Firstly, it is clear that the results are insensitive to the number of alpha particles in certain energy regions. In the low-energy region, this is a consequence of the low gallium cross sections and the large energy loss of the alpha particles per unit path length. Calculations indicate that the absolute number of alpha particles postulated as being below about 14 MeV could be changed by a factor of two without significantly affecting the agreement of calculated and experimental results. Similarly, at the high-energy end, because of the low cross sections, long ranges, and finite sizes of the targets used, the results are probably insensitive to alpha particles above about 50 MeV moving predominantly in the forward direction.

Within the energy range from 14–50 MeV, however, the results are quite sensitive to spectrum characteristics as indicated by the calculations summarized in the section on analysis of data. The thin-foil data require alpha particles of low energy (of around 25 MeV) with an appreciable angular spread. The thick-foil data require significant additional alpha particles particularly in the case of Ga^{67}. Since this nuclide is produced predominantly by alpha particles of greater than 30 MeV (when energy-loss considerations are taken together with the cross sections of Fig. 1) the high-energy component must have at least this energy. It must be directed primarily forward to avoid significant production of the other nuclides. Within these general features it is of course possible to vary somewhat the amounts and spectra of the two major components. Thus the forward-directed contribution illustrated in Fig. 2 at 40 MeV should be taken as indicating the presence of forward high-energy alpha particles with energies in the region of 35 to 45 MeV and total amounts equal to that illustrated. This situation is similar at the lower bombarding energies, except that at 123 MeV the presence of an anomalous experimental point for Ga^{67} for one of the two thinnest foils makes the division into the spectrum and forward component somewhat uncertain, e.g., an increase in the nuclear temperature of the spectrum from 3.5 MeV–3.75 MeV would result in a decrease in the forward component needed from 7 mb–5 mb with very little loss in over-all goodness of fit, although systematic deviations of the calculated and experimental data become prominent.

The nuclear temperatures and amounts of the alpha evaporation spectra deduced from this work are quite reasonable when compared to other experimental data and to theoretical expectations. Thus the temperature deduced from the present data varies from 3.5 MeV–4.5 MeV as the incident-proton energy is increased from 123 MeV–440 MeV. These results are in line with the results of Bailey[13] and Deutsch[24] who found that low-energy alpha-particle spectra, particularly backward-directed spectra, are consistent with an evaporation origin while the high-energy components in the forward direction require a nonevaporation mechanism.

The temperatures expected for the residual nuclei after the prompt cascade is over depend directly on the excitation energies of these nuclei. Average excitation energies can be estimated theoretically by interpolation of the Monte Carlo calculations of Metropolis *et al.*[25,26] In the case of Cu^{64} targets, average excitation energies at the proton energies used in this work are given in Table VI. Assuming a level density parameter of $A/10$, the corresponding nuclear temperatures increase from 2.9 MeV–3.8 MeV as the incident-proton energy is increased from 123 MeV–440 MeV. If the parameter is taken as $A/20$, the temperatures have a range from 4.0–4.9 MeV. These theoretical temperatures are close to those deduced from the experimental data. As a refinement it can be noted that the distribution in excitation energies of the residual nuclei from the cascade will tend to give the appearance to the over-all spectrum of a higher temperature in the region of interest for the work here. Thus a calculation of the energy spectrum of alpha particles emitted from the assemblage of excited nuclei left after a cascade produced by incident 460-MeV protons is compared with the $T=4.5$-MeV spectrum deduced from this work in Fig. 2(a). In this calculation the evaporation code of Porile[27] was used to calculate the probability of emitting the alpha particles. The Coulomb barrier used in this evaporation code was determined in the manner described by Dostrovsky *et al.*[23] A level density parameter of $a=A/10$ was used both for the evaporation probability and to obtain the energy distribution of the alpha particles. These two spectra were normalized at 20 MeV. Comparison of the two spectra indicates negligible difference in the energy range from 18 MeV–35 MeV. The deviations at low energies are not detectable by this work, and it is clear that this type of evaporation spectrum cannot introduce enough high-energy alpha particles (around 40 MeV) to produce the extra Ga^{67} seen in the thick foils. A similar comparison of the spectrum calculated for 156-MeV protons is made in Fig. 2(b) with the 3.5-MeV temperature spectrum deduced from this work for 123-MeV incident protons. Again, in the energy region of interest the shape is the same.

TABLE VI. Average excitation energies from Monte Carlo calculations and resulting nuclear temperatures.

Target nucleus	Proton energy (MeV)	Average excitation energy of residual nucleus[a] (MeV)	Nuclear tempera- (Mev) $a=A/10$	Nuclear tempera- (MeV) $a=A/20$
Cu^{64}	123	52.3	2.9	4.0
Cu^{64}	254	61.7	3.1	4.3
Cu^{64}	351	66.1	3.2	4.4
Cu^{64}	440	77.3	3.8	4.9

[a] References 25 and 26.

Strictly speaking the theoretical spectrum should be corrected for center-of-mass motion during the emission process. Reasonable values for this velocity indicate less than a ten percent velocity contribution to the evaporation alpha particles in the energy region of interest. Since this will both add and subtract to an isotropic spectrum, the results in the laboratory system will be primarily a slight broadening of the energy distribution near the peak of the spectrum which is below the energy of interest for this work.

The total cross section for the production of alpha particles in the evaporation spectrum as deduced from this work varies from 85 mb at 123 MeV to 180 mb at 440 MeV (see Table IV). This increase is reasonable in magnitude. For example, the Porile[27] evaporation code using the numbers and excitation energies

[24] R. W. Deutsch, Phys. Rev. **97**, 1110 (1955).

[25] N. Metropolis, R. Bivins, M. Storm, A. Turkevich, J. M. Miller, and G. Friedlander, Phys. Rev. **110**, 185 (1958).

[26] N. Metropolis, R. Bivins, M. Storm, A. Turkevich, J. M. Miller, and G. Friedlander, Phys. Rev. **110**, 204 (1958).

[27] N. T. Porile (private communication).

of the residual nuclei obtained from the Monte Carlo calculations[25,26] predicts a total alpha-particle production cross section varying from 87 mb for 82-MeV incident protons to 188 mb for 460-MeV incident protons. These theoretical yields are expected to be very sensitive to the choice of level density parameters; the agreement in magnitude and rate of increase is, however, satisfying.

These production cross-section values for evaporation alpha particles appear to be low compared to values reported by Muto *et al.*[14] and by Bailey[13] for targets in the region of copper. The discrepancies are probably due to the fact that the experiments reported here are sensitive only to alpha particles with energies above 14 MeV and to the fact that the total production cross sections were determined in this work by extrapolation to lower energies involving an assumed, rather high value for the Coulomb barrier (10.5 MeV). Similarly, although it is difficult to compare data obtained from target nuclei with very different nuclear charge, the observations of Gauvin *et al.*[8] and Lefort *et al.*[10] that the number of evaporation alpha particles produced from bismuth increases as the proton energy is increased from 40 MeV–550 MeV are consistent with the results of the current experiments.

The need for a high-energy, forward-directed component to the alpha spectrum arises primarily from the thick-foil cross sections for the production of Ga^{67}. As seen in Fig. 3, only 50% of the yield in the thickest foils is accounted for by the evaporation spectrum that sucessfully explains the data for the thin foils for all the gallium isotopes. The energy of this component is required to be about 40 ± 5 MeV to avoid contributions to the thinner foils or to other isotopes. Likewise, the forward nature of this component is dictated by similar considerations, although probably it could be spread over angles comparable to 45° relative to the beam direction without changing the nature of its contribution.

The presence of such high-energy tails in the spectra of alpha particles has been noted numerous times in the past.[6,13,14,24,28] In all investigations, the vast majority of these relatively high-energy alpha particles were found to be emitted in the forward direction relative to the direction of the incident-proton beam. Ostroumov, Perfilov, and Filov,[29] in a study of alpha particles with energies greater than 30 MeV produced in the silver and bromine nuclei of nuclear emulsion by 360- and 660-MeV protons, found that the degree of anisotropy of these particles increased as their kinetic energy increased.

The origin of the high-energy alpha particles is not clear. They may be a result of the interaction of cascade nucleons with alpha-particle clusters in the nucleus. On this basis, it is reasonable to expect that they should have a preferential forward direction. However, the relatively small increase in the number of such alpha particles as the incident-proton energy is increased is surprising on this interpretation.

The cross sections of these high-energy alpha particles needed to explain the results of the present experiments appear to be smaller than might be deduced from other work. Thus, Muto *et al.*[14] derived a direct-interaction cross section for alpha particles of 25 ± 7 mb for copper at an incident-proton energy of 56 MeV. At a proton energy of 190 MeV, Bailey[13] finds a cross section of about 28 mb for the production of nonevaporation alpha particles in nickel. Ostroumov and Filov[30] have measured the cross sections of cascade alpha particles with energies greater than 30 MeV produced in the heavy nuclei of emulsions exposed to protons ranging in energy from 100 MeV–660 MeV. At an incident energy of 100 MeV, they find a production cross section of 22 mb rising to a value of 82 mb at an incident energy of 660 MeV. Lefort *et al.*[10] quote values of 84, 94, and 105 mb for the production cross sections of all cascade alpha particles with energies greater than 20 MeV produced in bismuth targets by 240-, 420-, and 550-MeV protons, respectively.

A possible explanation for the small values reported here is based on the fact that only quite high-energy alpha particles have been considered as cascade particles in this experiment. Muto *et al.*[14] have shown that for copper bombarded by 56-MeV protons, forward-directed alpha particles attributed to the cascade process are produced with energies as low as 14 MeV. A rough estimate based on their cascade spectrum indicates that about 70% of the reported direct-interaction cross section of 25 mb is due to alpha particles with energies between 14 and 30 MeV. Similarly, Bailey[13] attributed all alpha particles with energies greater than 20 MeV produced in nickel by 190-MeV incident protons to a direct-interaction process. In light of these facts the cross sections reported here for forward-directed alpha particles, the largest value found being 13 mb at 254 MeV, are not unreasonable.

ACKNOWLEDGMENTS

The authors are indebted to Professor Nathan Sugarman for very many helpful discussions and to Professor N. Porile for making available his evaporation code. The cooperation of the operating staff of the University of Chicago Synchrocyclotron is gratefully acknowledged.

[28] P. A. Vaganov and V. I. Ostroumov, Zh. Eksperim. i Teor. Fiz. **33**, 1131 (1957) [English transl.: Soviet Phys.—JETP **6**, 871 (1958)].

[29] V. I. Ostroumov, N. A. Perfilov, and R. A. Filov, Zh. Eksperim. i Teor. Fiz. **36**, 367 (1959) [English transl.: Soviet Phys.—JETP **9**, 254 (1959)].

[30] V. I. Ostroumov and R. A. Filov, Zh. Eksperim. i Teor. Fiz. **37**, 643 (1959) [English transl.: Soviet Phys.—JETP **10**, 459 (1960)].