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Yields of Polonium and Bismuth Nuclides from Bi^{209} and Recoil Studies of $Bi^{209}(p,p2n)Bi^{207}$ Reaction at 450 MeV*

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The radioactivity of Bi^{207} produced in the 450-MeV proton bombardment of Bi^{209} has been measured. The yield ratios Bi^{205}/Bi^{207}, Bi^{206}/Bi^{207}, and Po^{207}/Bi^{207} were determined to be 0.77 ± 0.16, 0.83 ± 0.19, and 0.224 ± 0.013, respectively. These ratios, and Po/Bi ratios for other isobaric pairs of nuclides, are lower than values given by the Metropolis *et al.* Monte Carlo intranuclear cascade calculations. Examination of available data on absolute yields of Po and Bi nuclides shows that there is an underestimation of $Bi^{209}(p,pxn)$ yields in the calculation, a larger discrepancy occurring at Bi^{207} than for lighter Bi nuclides (larger x values). New Monte Carlo calculations by Bertini and by Chen *et al.* give better agreement with experiments. Recoil properties of Bi^{207} from 450-MeV proton bombardment of Bi^{209} have been measured and compared with results derived from the Metropolis *et al.* calculations. Over-all agreement is satisfactory, but the transverse momentum component seems overestimated by the calculations, a result found earlier in work with lighter Bi nuclides.

I. INTRODUCTION

The object of this study is to provide experimental data on yields and recoil properties of nuclides and to compare these data with Monte Carlo intranuclear cascade calculations of high-energy nuclear reactions. The calculations used are for the most part those of Metropolis *et al.*[1]

In an earlier paper,[2] relative yields of Bi^{205} and Bi^{206} and their Po isobars from 450-MeV proton bombardment of Bi^{209} were presented, along with the recoil properties of Bi^{203}, Bi^{204}, Bi^{205}, and Bi^{206}. Studies of a similar nature for mass number 207 are presented[3] in this paper.

A summary of (p,pxn) yields in the energy range 100–900 MeV and a comparison with calculated results reveals discrepancies not noted earlier.

II. RECOIL AND YIELD MEASUREMENTS

A. Experimental

The method employed was essentially that described previously.[2] The radioactivity of Bi^{207} was measured with a 3-mm-thick NaI(Tl) crystal[4] by scintillation counting of the associated Pb K x rays, in the $BiPO_4$ samples isolated 8–12 months earlier for determination of Bi^{205} and Bi^{206}. By this time the contribution of species other than 28-yr Bi^{207} was negligible. As a check on radiochemical purity, target samples from some of the thick-target experiments were counted with a 3-in. ×3-in. NaI(Tl) crystal and a 400-channel analyzer. After background subtraction, the samples all displayed the Bi^{207} gamma-ray spectrum[5] with no evidence of any other component. It was not possible to check the catcher-foil samples in this way because of the low counting rates (a few tenths of a count/min), but the ratio of counting rates of catcher and target in the channel was always about equal to the ratio of integral counting rates, indicating good radiochemical purity. Background in the channel was 0.989 ± 0.008 counts/min.

B. Results

Yield ratios were calculated by the method employed previously.[2] The recoil quantities measured for Bi^{207} are, in the terminology of the earlier paper, F_F for thin targets, and F_FW, F_PW, and F_BW for thick targets.

Half-periods assumed in the yield ratio measurements were 6.0 h for Po^{207} and 28 yr for Bi^{207} (both from the *Nuclear Data Sheets*[6]) and values given earlier[2] for the other species. There is considerable variance in reported[7–10] Bi^{207} half-periods; however, the earlier accepted[7] value of 8.0 yr does not seem likely to be correct in view of measurements by other authors[8–10] of 28, 30.2, and 38 yr. Because of the method employed

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[1] N. Metropolis, R. Bivins, M. Storm, A. Turkevich, J. M. Miller, and G. Friedlander, Phys. Rev. **110**, 185 (1958); N. Metropolis, R. Bivins, M. Storm, J. M. Miller, G. Friedlander, and A. Turkevich, *ibid.* **110**, 204 (1958).

[2] W. R. Pierson and N. Sugarman, Phys. Rev. **130**, 2417 (1963).

[3] A preliminary report of these results was given earlier; see W. R. Pierson and N. Sugarman, Bull. Am. Phys. Soc. **8**, 389 (1963).

[4] Intercalibration of the counters at Ford where the Bi^{207} measurements were made, with those at Chicago where the lighter nuclei were measured, was necessary for determining the Bi^{205}/Bi^{207} and Bi^{206}/Bi^{207} ratios; this was effected by means of a Tl^{204} source (Hg K x ray).

[5] R. L. Heath, Atomic Energy Commission Report IDO-16408, 1957 (unpublished).

[6] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington 25, D. C.)

[7] L. S. Cheng, V. C. Ridolfo, M. L. Pool, and D. N. Kundu, Phys. Rev. **98**, 231A (1955).

[8] J. Sosniak and R. E. Bell, Can. J. Phys. **37**, 1 (1959).

[9] G. Harbottle, J. Inorg. Nucl. Chem. **12**, 6 (1959).

[10] E. H. Appelman, Phys. Rev. **121**, 253 (1961).

TABLE I. Yields of Bi^{205}, Bi^{206}, and Po^{207} relative to Bi^{207}.

	Experimental		Calculated		
	Hunter and Miller[a] 380-MeV protons	Pierson and Sugarman 450-MeV protons	Hunter and Miller[b] 450-MeV protons	Chen *et al.*[c] 370-MeV protons	Bertini[d] 400-MeV protons
$\mathrm{Bi}^{205}/\mathrm{Bi}^{207}$	0.91±0.24	0.77 ±0.16	1.41±0.64	1.08±0.37	1.27±0.31
$\mathrm{Bi}^{206}/\mathrm{Bi}^{207}$	0.90±0.23	0.83 ±0.19	2.22±0.93	1.08±0.37	1.47±0.35
$\mathrm{Po}^{207}/\mathrm{Bi}^{207}$	0	0.224±0.013	0.80±0.42	0.87±0.30	0.47±0.15
$\mathrm{Po}^{206}/\mathrm{Bi}^{206}$	0.16±0.12	0.28 ±0.15[e]	0.21±0.12	0.23±0.12	0.23±0.08
$\mathrm{Po}^{205}/\mathrm{Bi}^{205}$	0.26±0.19	0.39 ±0.06[e]	0.78±0.34	0.73±0.27	0.50±0.14

[a] From Ref. 14, but changing the assumed Bi^{207} half-period from 8.0 to 28 yr.

[b] From Ref. 14, based on the Metropolis cascade calculations (Ref. 1) and evaporation calculations of I. Dostrovsky, P. Rabinowitz, and R. Bivins, Phys. Rev. **111**, 1659 (1958).

[c] From calculations performed by K. Chen, Z. Fraenkel, G. Friedlander, J. R. Grover, J. M. Miller, and Y. Shimamoto (communicated by J. M. Miller, 1963), using a seven-step "nonuniform" density distribution (see text) and the evaporation calculations of I. Dostrovsky, Z. Fraenkel, and G. Friedlander [Phys. Rev. **116**, 683 (1960)].

[d] From H. W. Bertini [Phys. Rev. **131**, 1801 (1963); Oak Ridge National Laboratory Report ORNL-TM-480, 1963 (unpublished); Bull. Am. Phys. Soc. **8**, 314 (1963); and private communication] using the three-step "medium nonuniform" density distribution (see text) and the evaporation calculations of L. Dresner [Oak Ridge National Laboratory Report CF-61-12-30, 1961 (unpublished)] incorporating the work of Dostrovsky *et al.* (Ref. c above).

[e] From Ref. 2.

in these experiments (namely, the comparison of the radioactivity of Bi^{207} in samples separated at different times after the bombardment), the $\mathrm{Po}^{207}/\mathrm{Bi}^{207}$ yield ratio reported (Table I) does not depend on the Bi^{207} half-period. The $\mathrm{Bi}^{205}/\mathrm{Bi}^{207}$ and $\mathrm{Bi}^{206}/\mathrm{Bi}^{207}$ yield ratios do, however, depend upon the Bi^{207} half-period chosen. Although no effort was made to determine the Bi^{207} half-period in this work, a half-period greater than 20 yr is indicated by our observations.

The Bi^{205}, Bi^{206}, and Bi^{207} counting efficiencies were assumed to be equal within 20% for calculations of the $\mathrm{Bi}^{205}/\mathrm{Bi}^{207}$ and $\mathrm{Bi}^{206}/\mathrm{Bi}^{207}$ ratios.

The separation times were soon enough after bombardment that the effect on the recoil results, due to Bi^{207} derived from Po^{207} decay, should be small (e.g., $\leq 6\%$ even if the F_FW value of Po^{207} is zero). Other sources of error have already been discussed.[2] Worthy of attention for the present work, however, is the error introduced in correcting the thin-target results for the fact that the target thickness is not really zero. The recoil ranges of Bi^{207} are shorter than those of the lighter species studied previously[2]; about 22% of the total Bi^{207} activity remains in the target layer. It is not clear how this activity should be apportioned between the backward and forward catcher foils; in these calculations the target activity was discarded. The error in F_F occasioned by discarding such a large fraction of the total activity is probably not more than 10%.

The yields of Bi^{205}, Bi^{206}, and Po^{207} relative to Bi^{207} are given in Table I. These ratios are averages of two experiments. The ratios have been computed on the basis of a 28-yr half-period for Bi^{207}. Included also in Table I are pertinent data from earlier[2] work. The recoil results for Bi^{207} are given in Table II.

The possibility was considered that an appreciable contribution to the polonium and bismuth yields might arise from reactions initiated by secondary protons and neutrons. The reactions most likely to be of importance are $\mathrm{Bi}^{209}(p,3n)\mathrm{Po}^{207}$ and $\mathrm{Bi}^{209}(n,3n)\mathrm{Bi}^{207}$. Using the $\mathrm{Bi}^{209}(p,3n)$ excitation function of Bell and Skarsgard,[11] the Monte Carlo data[1] on the number and energy distribution of emitted cascade protons (i.e., secondary protons), the range-energy curves for protons,[12] and the Bi target thickness (~40 mg/cm²), and assuming isotropic emission of the secondary protons, it is calculated that secondary $(p,3n)$ reactions might account for about 1% of the total Po^{207} yield, assuming that the cross section for production of Po^{207} by primary 450-MeV protons is about 15 mb (see Table III). Since the secondary protons are emitted preferentially in the forward direction, this 1% should be regarded as an upper limit. Similar considerations indicate that the amount of secondary $(n,3n)$ reactions should be about 3 times as great, i.e., up to about 0.5% of the Bi^{207} yield, using 65 mb for the production cross section of Bi^{207} by 450-MeV protons. Secondary protons and neutrons from the upstream catcher and wrapper foils (total material about 20 mg/cm²) also give rise to reactions in the target, to about the same extent as secondaries produced in the target. Unpublished analyses[13] of Bi^{206} production in that portion of the

TABLE II. Recoil properties of Bi^{207}.

	Observed	Calculated[a]
Thick target F_FW, mg/cm² Bi[b]	0.0137±0.0008[c]	0.032 ±0.013[d]
F_BW, mg/cm² Bi[b]	0.0058±0.0004	0.0125±0.0048[d]
F_PW, mg/cm² Bi[b,e]	0.0115±0.0004	0.032 ±0.015[f]
Thin target F_F[g]	0.73 ±0.06	0.71 ±0.16[h]

[a] Not the same as given in Ref. 2. See text for explanation.

[b] Average of gold and aluminum catchers.

[c] Errors quoted are random errors. For a discussion of systematic errors, see Ref. 2.

[d] Calculated according to the assumption (see Ref. 2) of a Gaussian scatter in range, with straggling parameter 0.41, ignoring evaporation.

[e] F_PW is taken as the average of the results of both the catcher foils. These values were 0.0126 and 0.0104 mg/cm² Bi and differ because of the 10° inclination of the plane of the target with respect to the beam in order to reduce multiple scattering (see Ref. 2).

[f] Calculated according to the assumption (see Ref. 2) of a "ball model" scattering, ignoring evaporation. Use of Gaussian scatter assumption as in calculation of F_FW and F_BW would raise the value.

[g] Aluminum catchers.

[h] Calculated on the assumption of no scattering of Bi recoils in Al, and each neutron evaporation imparting a recoil momentum of 80 MeV/c or the maximum possible, whichever is less (see Ref. 2).

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[11] R. E. Bell and H. M. Skarsgard, Can. J. Phys. **34**, 745 (1956).

[12] M. Rich and R. Madey, University of California Radiation Laboratory Report UCRL-2301, 1954 (unpublished).

[13] This result was obtained by Sugarman *et al.* while engaged in the work reported in Ref. 32, but is not reported therein.

TABLE III. Experimental and calculated absolute yields (mb) of Po and Bi nuclides.

	Experimental yield						Calculated yield					
									Bertini,[h] 400 MeV			
									Small		Medium	
Nuclide	Hunter and Miller,[a] 380 MeV	Pierson and Sugarman,[b] 450 MeV	Murin *et al.*,[c] 480 MeV	Murin *et al.*,[c] 660 MeV	Vinogradov *et al.*,[d] 480 MeV	Malysheva and Alimarin,[e] 660 MeV	Hunter and Miller,[f] 450 MeV	Chen *et al.*,[g] 370 MeV	Uniform	Nonuniform	Uniform	Nonuniform
Po^{207}	0	14.6± 3.7					12.3± 4.8	32± 8	3.2± 1.4	8.4± 2.3	13.2± 3.3	11.5± 3.1
Po^{206}	7.7± 5.6	15.1± 8.9	2.6	3.0			7.0± 3.6	9.3± 4	6.5± 2.0	12.2± 2.8	23.0± 4.4	8.2± 2.6
Po^{205}	12.9± 9.4	19.5± 4.1					16.8± 5.6	29± 8	12.3± 2.8	9.7± 2.5	18.1± 3.9	15.6± 3.6
Po^{204}	8.9± 6.5						19.9± 6.1	29± 8	19.4± 3.5	9.0± 2.4	21.4± 4.2	6.6± 2.3
Po^{203}	12.5± 9.1						13.3± 5.0	29± 8	6.5± 2.0	6.4± 2.0	16.5± 3.7	6.6± 2.3
Po^{202}	5.2± 3.8						21.1± 6.3	12± 5	12.9± 2.9	10.9± 2.7	14.8± 3.5	14.8± 3.6
Po^{201}	13.3± 9.7						16.0± 5.5	22± 7	5.8± 1.9	2.6± 1.3	11.5± 3.1	9.9± 2.9
Po^{200}	10.0± 7.3						21.8± 6.4	17± 7	11.0± 2.7	7.7± 2.2	14.0± 3.4	4.9± 2.0
Σ Po (201–206)	61 ±45[i]						94 ±13	130±17	63.3± 6.4	50.8± 5.7	105.3± 9.3	61.7± 7.1
Bi^{209}							1.9± 1.9[j]		1.3± 0.9	1.3± 0.9	1.6± 1.2	7.4± 2.5
Bi^{208}							13.0± 4.9[j]	64±12	11.6± 2.7	16.7± 3.3	23.0± 4.4	48.5± 6.3
Bi^{207}	55.0±13[k]	65.1±15.8					15.3± 5.3	37± 9	11.0± 2.7	23.8± 3.9	28.0± 4.8	24.7± 4.5
Bi^{206}	49.3± 5.9	53.8±13.1	71	75	60	25	33.9± 7.9	40±10	27.2± 4.2	26.4± 4.1	51.8± 6.5	36.2± 5.5
Bi^{205}	50.0± 7.0	50.0± 7.0[l]	77	78		22	21.6± 6.3	40±10	23.3± 3.9	30.2± 4.4	41.1± 5.8	31.3± 5.1
Bi^{204}	37.1± 3.2				11[m]	21	16.9± 5.6	34± 9	28.2± 4.3	21.9± 3.8	45.2± 6.1	31.3± 5.1
Bi^{203}	47.6± 7.6					9	25.0± 6.8	49±11	36.2± 4.8	28.3± 4.3	49.3± 6.4	24.7± 4.5
Bi^{202}	55.8± 9.4				10	8	36.6± 8.2	54±12	37.5± 4.9	23.2± 3.9	58.4± 6.9	35.4± 5.4
Bi^{201}	49.6± 4.4					4	27.5± 7.1	44±10	28.4± 4.3	24.4± 4.0	45.2± 6.1	36.2± 5.5
Bi^{200}	64.4						30.2± 7.5	30± 8	38.1± 5.0	28.9± 4.3	54.3± 6.7	20.6± 4.1
Bi^{199}	68.6[n]						38.9±11.8	30± 8	39.4± 5.1	27.0± 4.2	56.8± 6.8	27.1± 4.7
Bi^{198}	60.1[n]						34.2±11.0	47±11	44.6± 5.4	20.6± 3.6	51.8± 6.5	26.3± 4.7
Σ Bi (201–206)	289 ±16						161.5±17	261±25	181 ±11	154 ±10	291 ±15.5	195 ±13

[a] Ref. 14.
[b] This paper and Ref. 2.
[c] Ref. 16.
[d] Ref. 17.
[e] Ref. 18.
[f] Calculated in Ref. 14 from the calculations of Metropolis *et al.* (Ref. 1).
[g] Calculations performed by K. Chen, Z. Fraenkel, G. Friedlander, J. R. Grover, J. M. Miller, and Y. Shimamoto (communicated by J. M. Miller, 1963).
[h] H. W. Bertini, Phys. Rev. **131**, 1801 (1963); Oak Ridge National Laboratory Report ORNL-TM-480, 1963 (unpublished); Bull. Am. Phys. Soc. **8**, 314 (1963); and private communication. For explanation of column headings, see text.
[i] Error is taken to be 73% since the errors quoted on the individual Po yields were all about 73%.
[j] Not given by Hunter and Miller; estimates made by us, with Metropolis calculations and our own evaporation treatment.
[k] Cross section assuming a Bi^{207} half-period of 28 years.
[l] Accepted cross section; other values in this column are normalized to this value.
[m] Average of Bi^{203} and Bi^{204}.
[n] Cumulative cross section.

target not directly bombarded by the primary proton beam indicate that production of Bi^{207} by stray neutrons should also be small. The effect of secondaries on the results is therefore believed to be minor.

III. DISCUSSION

A. Yields of Po and Bi Nuclides

The yield ratios Bi^{205}/Bi^{207} and Bi^{206}/Bi^{207} from the present and earlier[2] work at 450 MeV (Table I) are in good agreement with those derived from the results of Hunter and Miller[14] at 380 MeV, after correction for the Bi^{207} half-period of 28 yr. The yield ratio Po^{207}/Bi^{207} from the present work, however, is sizeably different from that of Hunter and Miller who apparently observed no radioactivity attributable to Po^{207} in their experiments.

The yield ratios predicted by current Monte Carlo calculations are shown in Table I. The first set of calculated yield ratios is obtained from Hunter and Miller and represents the calculations of Metropolis *et al.*, employing a constant-density nucleus of radius 7.71×10^{-13} cm. The second set is obtained from a similar but later calculation by Chen *et al.*, in which refraction effects were included, pion production was neglected, and the uniform density distribution was replaced with a step-function density distribution approximating, in seven steps, the charge distribution obtained by Hofstadter.[15] The radius at which the density is one-half its central value was chosen to be 1.07×10^{-13} $A^{1/3}$ cm, and the outermost step ($0.04\times$central density) extends to 8.85×10^{-13} cm.

The third set comes from calculations by Bertini, similar to those of Chen *et al.*, except that refraction is neglected, and the Hofstadter density distribution is approximated in three steps rather than seven. The last step extends to 8.85×10^{-13} cm.

A comparison of the observed and calculated Po/Bi yield ratios shows that the calculated ratios are, in general, higher than the observed ones. Over the mass interval 205–207, our experimental ratio of total Po yield to total Bi yield is 0.291 ± 0.064, while the ratios for the three calculations shown are 0.51 ± 0.14, 0.60 ± 0.13, and 0.38 ± 0.07, respectively, giving a level of significance of 48%, 96%, and 66% for the respective discrepancies. Thus the calculations all tend to overestimate the likelihood for zero protons to be ejected, relative to that for one proton.

Comparison of the experimental and calculated *absolute* yields of Po and Bi nuclides can be made from Table III. The experimental absolute yields are from Hunter and Miller at 380 MeV, from the present work at 450 MeV (calculated from the ratios reported earlier,[2] and from Table I normalizing to a yield of 50 mb[14] for Bi^{205}), and from Russian investigators[16–18] at 480 and 660 MeV. The calculated yields of Table III are from Hunter and Miller calculated from the Metropolis *et al.* cascade data at 450 MeV, from Chen *et al.* at 370 MeV, and from Bertini at 400 MeV.

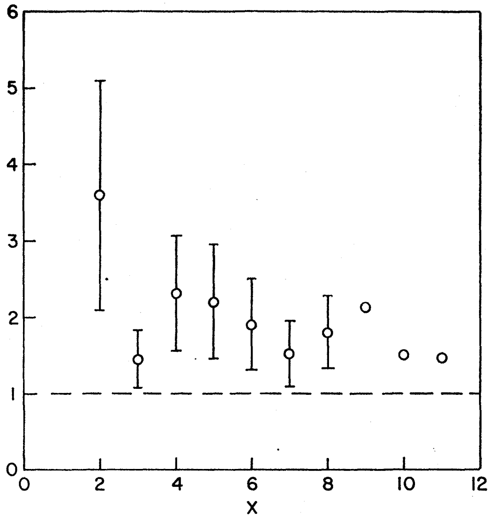

FIG. 1. Ratios of experimental $Bi^{209}(p,pxn)$ yields to calculated values (see Table III) of Hunter and Miller.

The Bertini calculations were carried out in the present case for four nuclear configurations as denoted by the column headings of Table III: "small uniform," "small nonuniform," "medium uniform," and "medium nonuniform." The "small uniform" configuration is that used by Metropolis *et al.*,[1] namely a uniform-density nucleus of radius 7.71×10^{-13} cm. The "small nonuniform" nucleus has the outermost step extending to the same radius as that of the "small uniform" nucleus; the density distribution otherwise is made to approximate the Hofstadter distribution obtained by using the value 1.07×10^{-13} $A^{1/3}$ for the radius at which the density is one-half its central value. The "medium uniform" nucleus is like the "small uniform" nucleus, but with radius equal to 8.85×10^{-13} cm. The "medium nonuniform" nucleus has already been described.

It is clear from Table III that there are discrepancies between the experimental Bi yields of Hunter and Miller and the yields calculated by Hunter and Miller from the Metropolis *et al.* cascade calculations. The average ratio of observed to calculated Bi yields for mass numbers 201–206, weighted by the individual errors, is 1.71 ± 0.13, and the ratio for Bi^{207} is 3.59 ± 1.51.

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[14] E. T. Hunter and J. M. Miller, Phys. Rev. **115**, 1053 (1959).

[15] R. Hofstadter, Rev. Mod. Phys. **28**, 214 (1956).

[16] A. N. Murin, B. K. Preobrazhensky, I. A. Yutlandov, and M. A. Yakimov, *Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, July 1955, Session of the Division of Chemical Science* (Akad. Nauk SSSR, 1955), p. 101 (translation: Consultants Bureau, AEC-tr-2435, part 2, available from the Superintendent of Documents, U. S. Government Printing Office, Washington 25, D. C.).

[17] A. P. Vinogradov, I. P. Alimarin, V. I. Baranov, A. K. Lavrukhina, T. V. Malysheva, and F. I. Pavlotskaya, see Ref. 16, p. 85.

[18] T. V. Malysheva and I. P. Alimarin, Zh. Eksperim. i Teor. Fiz. **35**, 1103 (1958) [translation: Soviet Phys.—JETP **8**, 772 (1959)].

It is thus seen that the observed Bi yields are larger than calculated, particularly in the case of Bi^{207}. This fact is demonstrated in Fig. 1 which shows the ratios of the Hunter and Miller experimental cross sections to the calculated cross sections for the $\mathrm{Bi}^{209}(p,pxn)$ products, using the data from Table III. The experimental data at $x=10$ and 11 were adjusted by reducing the cumulative yields by an arbitrary 10 mb as a correction for the presence of Po precursors. A better estimate of proton evaporation than that made by Hunter and Miller would result in even larger discrepancies between observed and calculated yields at low mass numbers.

It has commonly been assumed that those high-energy (p,pxn) reactions which show discrepantly high yields when compared to the Metropolis *et al.* calculations[1] fall into two principal categories, first, those with $x=1$ at all bombarding energies,[19] especially in the heavy elements, and second, those at BeV bombarding energies for larger x values,[20–23] again especially in the heavy elements. It is now noted (Table III, Fig. 1) that the absolute yields[14] of (p,pxn) products from Bi^{209} at 380 MeV are substantially higher than the yields from the Metropolis *et al.* calculations over the entire interval measured ($x=2$–11).

The published literature was surveyed for other comparisons of calculated[1] and experimental (p,pxn) yields for $x>1$ in the proton energy range 100–900 MeV. A discrepancy is reported for U^{238} at 100 MeV ($x=9$ and 10),[24] and also for Au^{197} at 86 MeV and lower ($x=2$ and 3).[25] There is good agreement for Ga^{69} and Ga^{71} at 500 MeV ($x=2$–6),[23] I^{127} at 200 and 500 MeV ($x=2$–7),[20] Au^{197} at up to 150 MeV ($x=2$),[26] and Th^{232} ($x=4$–6) and U^{238} ($x=6$, 8, 9, and 10) at 340 MeV.[24] It should be noted that the Th^{232} data[24] constitute the only evidence for good agreement in the case of heavy (i.e., mass number 200 or more) nuclei at several hundred MeV, and even here there is involved an assumption on the fissionability of the cascade nuclei. (The U^{238} agreement is not overly significant on account of serious fission competition.) Moreover, there are other reported (p,pxn) yields in this energy region for which no comparison with the Metropolis calculations[1] has yet been made, but which are almost certainly much higher than the calculations would predict. These are the $\mathrm{Cs}^{133}(p,p2n)$ reaction[27] at 240 MeV (460 mb), and the $\mathrm{Ta}^{181}(p,pxn)$ reactions[28] at 340 MeV for x values of 1 (265 mb), 3 (152 mb), and 5 (also 265 mb).

Analysis of the Bertini data leads to the following conclusions: inclusion of the nonuniform density results in a substantial increase in the $\mathrm{Bi}^{209}(p,p2n)$ yield, in better (though still poor) agreement with experiment; however, there is a significant decrease in the yields of the lighter Bi nuclides, and therefore poorer agreement with experiment for those nuclides, unless the radial extension of the nucleus is increased as well. Even with an increase in nuclear size, however, agreement with experiment is not achieved, either for the $(p,p2n)$ or the rest of the (p,pxn) reactions. Moreover, the fission of the low mass Bi nuclides would increase the discrepancy since fission was not considered in the Bertini calculations. Thus the use of the nonuniform density results in better agreement between experimental and calculated (p,pxn) yields of Bi nuclides only if the size of the nucleus is increased, and agreement is not obtained even then.

Analysis of the calculations of Chen *et al.* shows that the average ratio of our experimental Bi yields (201–206) to those calculated weighted by the individual errors is 1.095 ± 0.040. The corresponding ratio for Bi^{207} is 1.49 ± 0.50. Evidently the inclusion of refraction and more steps in the density distribution, probably mainly the former, produces better agreement with experiment. However, no improvement is realized in the ability of the calculations to predict the relative probabilities of (p,pxn) and (p,xn) reactions.

The data on Po yields warrant only brief mention because of the discrepancies among the experimental results of the different investigators. The total yield over the mass number interval 205–207 as measured by Hunter and Miller (21 ± 15 mb) is significantly lower (88% confidence level) than that indicated by our experiments (49 ± 10 mb). The average ratio of our experimental Po yields (205–207) to those calculated weighted by the individual errors is 1.21 ± 0.14 with the Hunter and Miller calculations, 0.54 ± 0.11 with the calculations of Chen *et al.*, and as follows for the various Bertini calculations: small uniform 1.77 ± 0.42, small nonuniform 1.72 ± 0.26, medium uniform 0.97 ±0.14, and medium nonuniform 1.29 ± 0.10.

B. Recoil Behavior of Bi^{207}

The measured values of F_FW, F_BW, and F_PW for Bi^{207} for thick targets, and F_F for thin targets, are compared in Table II with those calculated[29] from the momentum

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[19] See for recent examples D. L. Morrison and A. A. Caretto, Jr., Phys. Rev. **127**, 1731 (1962); also Ref. 23.

[20] I. Ladenbauer and L. Winsberg, Phys. Rev. **119**, 1368 (1960).

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[24] M. Lindner and R. N. Osborne, Phys. Rev. **103**, 378 (1956); M. Lindner and A. Turkevich, *ibid.* **119**, 1632 (1960).

[25] T. M. Kavanagh and R. E. Bell, Can. J. Phys. **39**, 1172 (1961). Comparisons were made with Jackson's two-dimensional cascade calculations (Ref. 31), but the conclusions would be the same if the Metropolis *et al.* calculation, as given in Ref. 26, were used.

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[29] The calculated values are based on a range-energy relation (given in Ref. 2) derived from work from various sources. Recent experiments [I. Bergström, J. A. Davies, B. Domeij, and J. Uhler, Arkiv Fysik **24**, 389 (1963); B. Domeij, I. Bergström, J. A. Davies, and J. Uhler, *ibid.* **24**, 399 (1963)] on Rn^{222} ions accelerated to hundreds of keV kinetic energies give mean ranges

predictions[30] of the Metropolis *et al.*[1] Monte Carlo calculations. (A similar detailed comparison with results of the other calculations cannot be made because the information is not available.) It may be noted that the calculated values in Table II are *not* the same as those given in the earlier[2] paper. The reason for this is that the evaporation treatment used here is different from that used formerly. Instead of assuming that one neutron is evaporated when the excitation of the residual nucleus is between the neutron binding energy and 22.8 MeV, and that more than one neutron evaporates when the initial excitation of the residual nucleus is greater, now 18.3 MeV is assumed to be the dividing line, in crude approximation to the evaporation treatment by Jackson[31] with a nuclear temperature of 1.9 MeV. This assumption changes somewhat the assignments of the individual residual nuclei to their final mass numbers, so that many recoil nuclei formerly ending up in the $A=207$ "bin" are now in the $A=206$ "bin."

The experimental values for Bi^{207} fit well on the curves drawn through the experimental data for the lighter Bi nuclides,[2,32] showing, in general, a linear decrease in F_F, F_FW, and F_PW with increasing mass number, and a fairly constant value of F_BW. The agreement between calculated and experimental thin-target F_F is excellent. The discrepancies between the experimental thick-target values for Bi^{207} and the calculated values might be significantly related to the large yield discrepancy noted earlier, or they may be a consequence of the large scatter in the calculated thick-target values because of the relatively small number of cases of each type available, and of the crudeness of the selection process used for binning nuclei after evaporation.

The tendency of the calculations to overestimate the average transverse component of momentum for the lighter Bi nuclides[2] seems to extend to Bi^{207}. This statement is *not* made on the basis of the data of Table II, but rather on the basis of the fact that the observed F_PW/F_FW ratio (0.84 ± 0.08) falls close to the line running through the F_PW/F_FW values for the lighter nuclides in Ref. 2, and on the fact that the observed F_PW/F_FW ratios always lie below the calculated ones, as shown in Fig. 8 of Ref. 2, whereas the observed and calculated F_FW values, in general, agree fairly well as can be seen in Fig. 6 of Ref. 2. The calculated F_PW and F_FW values for Bi^{207} (Table II) are both well above the trend established by the Monte Carlo calculations and represent only 7 events, and for these reasons no attempt was made to base any conclusions on them. It is worth noting that the Bertini cascade calculations using the medium nonuniform nucleus predict average transverse momenta 20% smaller than those calculated by Porile[30] from the Metropolis *et al.* cascade data for those prompt cascades involving emission of 1 proton and 0–4 neutrons; the corresponding average forward component of momentum for these cases is about 10% larger.

It appears that the discrepancies between the experimental (p,pxn) yields for $x=2$–11 and those calculated from the Metropolis *et al.* data are not accompanied by any systematic discrepancies in recoil properties other than a slight overestimate of transverse momentum. The especially large discrepancy in the $(p,p2n)$ cross section, possibly related in origin to that observed in (p,pn) reactions,[19] is accompanied by a difference between experimental and calculated recoil behavior not outside the limits of expectation.

Note added in proof. Bertini has discovered an error in his calculations. When the corrected results are available, an erratum will be submitted.

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in tungsten much larger than consistent with this range-energy relation. This effect is attributed to channeling associated with a preferred crystal orientation induced in the thin rolled tungsten foils. [See also G. R. Piercy, F. Brown, J. A. Davies, and M. McCargo, Phys. Rev. Letters **10**, 399 (1963).] There is no reason to believe that significant orientation has occurred in the bismuth foils used in the present work, but should it have occurred then the comparison of experimental and calculated thick-target recoil properties would be seriously affected. The theory of Lindhard *et al.* [J. Lindhard and M. Scharff, Phys. Rev. **124**, 128 (1961); J. Lindhard, M. Scharff, and H. E. Schiøtt (private communication, 1962)], which generally agrees well with experimental results in this energy region, gives calculated F_FW, F_BW, and F_PW values larger than those given in Table II by 14, 7, and 7%, respectively.

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[31] J. D. Jackson, Can. J. Phys. **34**, 767 (1956).

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