Differential Range Study of Na^{24} Formed by 2.9-GeV Proton Irradiation of Light Elements*

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Thin targets (≈ 50 $\mu g/cm^2$) of copper, vanadium, and chlorine (Saran) were irradiated with 2.9-GeV protons. Products recoiling at 90° to the beam were collected in a stack of thin plastic films, at a geometry of 2%. The range distributions of Na^{24} were determined by radiochemical separation, and energy spectra were deduced from the measurements. The mean energies of Na^{24} from copper, vanadium, and chlorine are 8.2, 6.1, and 4.3 MeV, respectively. The observed energy spectra were compared with the results of Monte Carlo cascade-evaporation calculations, and very good agreement was obtained for copper and vanadium. The calculation predicts significantly larger recoil energies than observed in the case of the chlorine target. It is concluded that this is due to an overestimate of the transverse component of momentum imparted to the residual nuclei ln the cascade process.

I. INTRODUCTION

SPALLATION reactions induced by high-energy protons are thought to proceed by a two-step cascade-evaporation process. Calculations based on the application of the Monte Carlo technique to both phases of the reaction have yielded spallation cross sections that are in fair agreement with experiment.[1–9] A further test of the calculations and of the model on which they are based can be obtained from a comparison of experimental and calculated recoil properties of the spallation products. Several comparisons with calculation of the average effective ranges of spallation products obtained in thick-target experiments have been presented.[9–12] While fairly good agreement has been obtained below 1 GeV, discrepancies between experiment and calculation have been noted for studies involving GeV protons.[10,12]

The measurement of differential ranges of spallation products provides more detailed information about the recoil behavior of the products than can be obtained from average range determinations. A comparison of these measurements with Monte Carlo cascade-evaporation calculations is consequently of interest. The differential ranges of a number of products formed in the interaction of 2.9-GeV protons with silver have recently been measured.[13] Comparison of the differential energy spectra based on these results with calculation has given good agreement for several products with $A = 43-83$. On the other hand, the results for Na^{24} did not agree with calculations, and it was concluded that a two-body breakup process was responsible for the formation of this product. In view of this fact it was of interest to extend the measurements on Na^{24} to lighter target elements. Chlorine and vanadium were chosen as typical targets from which Na^{24} could be produced by spallation reactions involving excitation energies of about 100 and 300 MeV, respectively. A comparison of these results with the Monte Carlo calculation provides a further test of the cascade-evaporation model in the light element region.

The formation of Na^{24} from copper could conceivably result from a spallation process involving very high excitation energies or from a two-body breakup process. The first mechanism has been advocated by Caretto *et al.*[14] on the basis of their measurements of the variation of the Na^{24} cross section with target mass number. On the other hand, Crespo *et al.*[15] have suggested that this reaction involves a fragmentation process on the basis of their investigation of thick-target recoil properties of Na^{24} produced from a number of elements. The present results on the differential range of Na^{24} produced in the interaction of 2.9-GeV protons with copper provide further evidence on this point.

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

[2] See J. M. Miller and J. Hudis, Ann. Rev. Nucl. Sci. **9**, 159 (1959) for references up to 1959.

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

[4] M. L. Linder and A. Turkevich, Phys. Rev. **119**, 1632 (1960).

[5] B. D. Pate and A. M. Poskanzer, Phys. Rev. **123**, 647 (1961).

[6] N. T. Porile, Phys. Rev. **125**, 1379 (1962).

[7] N. T. Porile and S. Tanaka, Phys. Rev. **132**, 397 (1963).

[8] N. T. Porile and L. B. Church, Phys. Rev. **133**, B310 (1964).

[9] W. R. Pierson and N. Sugarman, Phys. Rev. **133**, B384 (1964).

[10] A. M. Poskanzer, J. B. Cumming, and R. Wolfgang, Phys. Rev. **129**, 374 (1963).

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

[12] N. T. Porile and S. Tanaka, Phys. Rev. **135**, B122 (1964).

[13] J. B. Cumming, S. Katcoff, N. T. Porile, S. Tanaka, and A. Wyttenbach, Phys. Rev. **134**, B1262 (1964).

[14] A. A. Caretto, Jr., J. Hudis, and G. Friedlander, Phys. Rev. **110**, 1130 (1958).

[15] V. P. Crespo, J. M. Alexander, and E. K. Hyde, Phys. Rev. **131**, 1765 (1963).

TABLE I. Irradiation data.

Irradiation	Target element	Target thickness ($\mu g/cm^2$)	Backing thickness ($\mu g/cm^2$)	Integrated circulating beam intensity
1	Cu	100	199	3.0×10^{14}
2	Cu	40	78	4.4×10^{14}
3	V	125	135	4.0×10^{14}
4	V	40	50	2.5×10^{14}
5	Cl	24[a]	73	2.5×10^{14}

[a] The actual Saran thickness was 32 $\mu g/cm^2$. The chlorine thickness was calculated from the target composition.

II. EXPERIMENTAL

The experimental procedures used in this study were similar to those employed in the previous investigation of the differential ranges of products from silver.[13] Only a brief summary of the details will be presented here.

The differential ranges of Na^{24} were measured with the target assembly shown in Fig. 1. The copper and vanadium targets were vacuum deposited on a thin Formvar (polyvinyl formal) backing and covered an area of 1 cm^2. The target for the chlorine experiment consisted of a thin Saran (polydichloroethylene) film, 1 cm^2 in area, laminated onto the Formvar backing. Several target and backing thicknesses, summarized in Table I, were used to investigate the effects of scattering on the observed range distributions. The target foils were inclined at an angle of 30° to the beam direction so that the recoil path lengths in the target would be less than 15% longer than the normal target thickness.

The catcher foils consisted of Formvar films with a thickness of 50–250 $\mu g/cm^2$. The thickness and uniformity of the foils were measured with an alpha thickness gauge as described elsewhere.[13] Only foils which were uniform to better than 10% were used. The catcher foils were mounted on aluminum frames having 2-in.-sq holes. To insure that all foils subtended the same solid angle, an aluminum baffle with a $1\frac{1}{2}$-in.-sq hole was placed over the catcher foil stack. Both baffle and foil frames were coated with plastic to prevent contamination of the foils by sodium recoils from the aluminum. The target-to-baffle distance was 3 in., the solid angle subtended was ~2% of 4π sr, and the angular acceptance was 74–106°.

Five irradiations were performed with 2.9-GeV protons, as indicated in Table I. The catchers were shielded from the proton beam during irradiation by means of a copper "shadow" block, while a retractable shutter prevented low-energy protons from striking the target foil. For the irradiations involving the thinner targets, the cosmotron magnet was operated in the "flat-topped" mode.[13] The number of proton traversals through the target was thereby substantially increased over the corresponding number for the irradiations of the thicker targets.

After irradiation, the target assembly was taken apart and the individual foils were carefully separated from their frames and transferred to centrifuge cones containing 10 mg of sodium carrier. The foils were dissolved in aqua regia and sodium was separated by the previously described procedure.[13]

The radioactivity of the sodium samples was assayed with calibrated end-window beta-proportional counters. Anticoincidence shielding was used to reduce the counter backgrounds to ~0.3 counts per minute for the assay of samples having low activities. It was assumed that the variation of counting efficiency with sample thickness could be ignored for the narrow range in thickness encountered in this study. The activity in the samples decayed with the 15.0-h half-life of Na^{24} and a small amount (~5%) of a much longer lived component.

In most cases, samples were separated from foils beyond the range of Na^{24}. These gave a measure of the activation blank and were usually very small.

III. RESULTS

The differential range data are summarized in Tables II–IV and in Fig. 2. The results of the duplicate irradiations are in very good agreement with each other except for the first one or two foils, where the experiments with the thicker targets gave somewhat higher counting rates. This difference can be attributed to the backscattering of recoils in the target. Since the difference between the relative activities of the first catcher foils is less than 20% for a factor-of-3 variation in target thickness, it is felt that the residual effect for the thinner targets is negligible.

In order to convert the range distributions to energy spectra, a range-energy relation for Na^{24} in Formvar is needed. The relation used in the present work, depicted in Fig. 3, is identical to that used in previous work.[13] It is based on the calculation of Lindhard, Scharff, and Schiott,[16] on the experimental data of McCargo *et al.*[17]

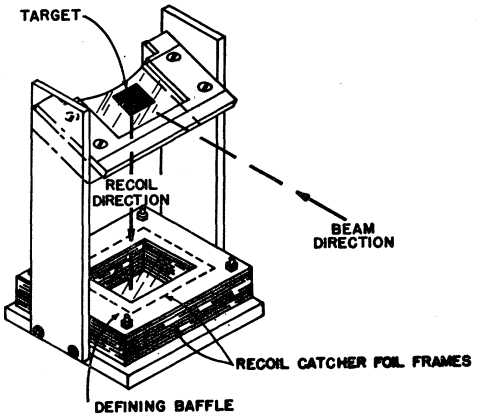

FIG. 1. Apparatus for measuring differential ranges at 90° to beam direction.

[16] J. Lindhard, M. Scharff, and H. E. Schiott, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **33**, No. 14 (1963).

[17] M. McCargo, F. Brown, and J. A. Davies, Can. J. Chem. **41**, 2309 (1963).

TABLE II. Data for calculation of range and energy distributions at 90° of Na^{24} from copper.

Catcher foil	Irradiation No. 1 Foil thickness ($\mu g/cm^2$)	Mean energy (MeV)	Net activity (counts/min)	Irradiation No. 2 Foil thickness ($\mu g/cm^2$)	Mean energy (MeV)	Net activity (counts/min)
1	134	0.3	222	145	0.3	53.4
2	136	1.0	208	143	1.0	57.6
3	150	1.9	373	137	1.8	121
4	139	3.2	494	152	3.1	196
5	139	4.6	596	147	4.8	260
6	140	6.3	572	149	6.5	231
7	141	8.2	431	146	8.5	200
8	143	10.4	464	155	10.8	143
9	140	12.3	273	159	13.1	111
10	144	14.3	240	147	15.4	77.1
11	144	16.2	168	148	17.1	57.9
12	148	18.0	137	159	19.0	12.0
13–14	292	20.8	190	489	22.3	75.0
15–16	298	23.8	106	475	27.3	27.6
17–18	295	26.5	74.8	479	32.5	0.9
19–20	305	29.5	49.6			

on the range of Na^{24} in Al, of Poskanzer[18] on the range of Ne^{22} in Al, and of Northcliffe[19] on the range of Ne^{20} in Al. It is estimated that the recoil energies based on the use of Fig. 3 may be in error by as much as 15%.

Differential energy spectra have been obtained from the data in Tables II–IV by plotting the activity of each foil per unit energy interval. A small correction was made to each set of data for energy loss in the target. The results have also been corrected for range straggling on the basis of Poskanzer's[18] measurement of a half-width of 0.2 mg/cm² for the range of 1–3-MeV Ne^{22} ions in aluminum. On the assumption that this value applies to Na^{24} over the entire energy range under consideration, the correction is found to be negligible for all but the first one or two foils. The corrected results are shown in Fig. 4 as the smooth curves drawn through the experimental points. The spectra show a steady shift towards higher energies as the mass difference between target and product increases. The spectrum for the chlorine target peaks at zero energy, while the other spectra show most probable energies well above zero. The mean energies are summarized in Table V.

The results of the present study, coupled with the data for Na^{24} from silver[13] and similar measurements for Na^{24} from bismuth,[20] provide an over-all picture of the variation with target mass number of the energy spectrum of Na^{24} recoils emitted at 90° to an incident beam of 2.9-GeV protons. A plot of the variation of the mean recoil energy with target A is shown in Fig. 5. The general trend of the data is expected on rather general grounds regardless of whether spallation or two-body breakup processes predominate. Another quantity of interest is the coefficient of skewness χ which is a measure of the asymmetry of the energy spectra. This

TABLE III. Data for calculation of range and energy distributions at 90° of Na^{24} from vanadium.

Catcher foil	Irradiation No. 3 Foil thickness ($\mu g/cm^2$)	Mean energy (MeV)	Net activity (counts/min)	Irradiation No. 4 Foil thickness ($\mu g/cm^2$)	Mean energy (MeV)	Net activity (counts/min)
1	78	0.3	316	68	0.2	467
2	76	0.6	174	69	0.4	245
3	74	1.0	197	73	0.8	287
4	78	1.5	296	68	1.2	328
5	76	2.0	391	68	1.6	459
6	78	2.7	452	66	2.1	586
7	78	3.4	424	68	2.6	750
8	80	4.2	496	69	3.2	744
9	71	5.0	333	65	4.0	781
10	70	5.8	362	68	4.7	788
11	72	6.8	283	73	5.6	769
12	70	7.7	240	70	6.5	634
13–14	229	10.0	583	460	10.2	2110
15–16	310	13.8	293	445	16.3	666
17–18	294	17.3	190	463	21.5	236
19–20	316	21.1	85	445	26.4	90

[18] A. M. Poskanzer, Phys. Rev. **129**, 385 (1963).
[19] L. C. Northcliffe, Phys. Rev. **120**, 1744 (1960).
[20] J. B. Cumming, R. J. Cross, Jr., J. Hudis, and A. M. Poskanzer, Phys. Rev. **134**, B167 (1964).

coefficient is given by

$$\chi=(\sum_{i=1}^{n} N(E_i))^{1/2} \sum_{i=1}^{n}(E_i-\bar{E})^3 N(E_i)/ (\sum_{i=1}^{n}(E_i-\bar{E})^2 N(E_i))^{3/2},$$

where $N(E_i)$ is the relative intensity of the spectrum at energy E_i, $\bar{E}$ is the mean recoil energy, and n is the number of points obtained from the smoothed energy spectra at regular intervals. It is readily seen that a symmetric distribution has $\chi=0$. In Fig. 5 χ has small positive values in the region where a two-body breakup process is indicated, but becomes increasingly positive as the mass difference between target and product decreases in the spallation region. We do not know whether this difference is connected with the change in mechanism or whether it merely reflects the fact that the low-energy ends of the spallation spectra are absent because negative energies are not possible.

IV. DISCUSSION

The energy spectra may be compared with the results of Monte Carlo cascade-evaporation calculations. These

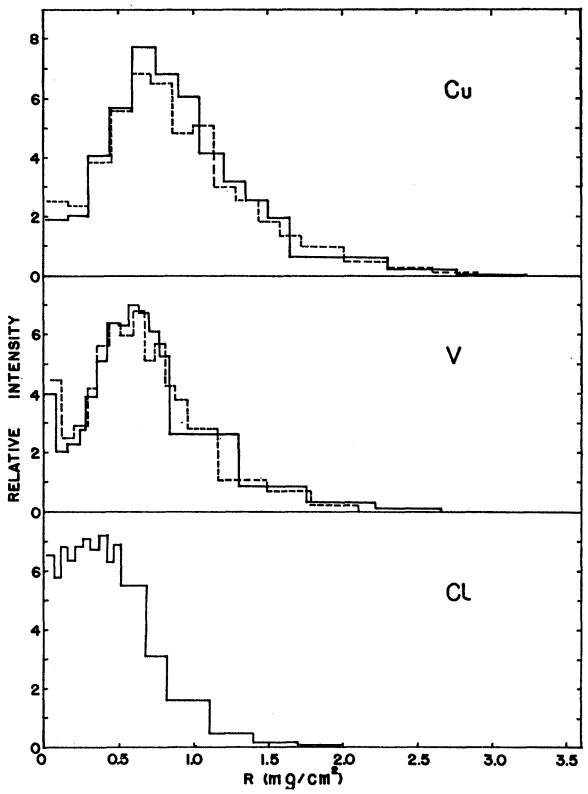

FIG. 2. Observed range distributions at 90° of Na^{24} from 2.9-GeV proton irradiation of the indicated targets. The dashed histograms are the results of duplicate experiments with thicker targets normalized to the same area as the corresponding thin target histograms.

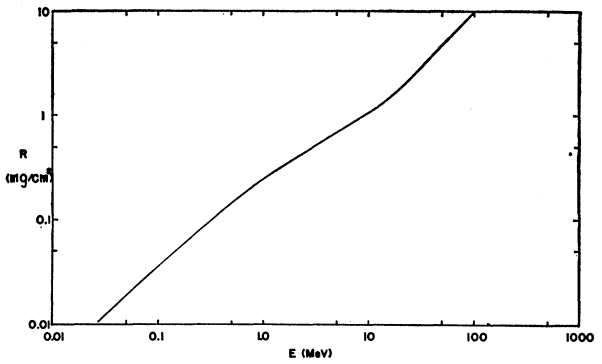

FIG. 3. Range-energy relation for Na^{24} in Formvar.

calculations have been performed in the manner described in detail in a previous publication.[13] The starting nuclei for the evaporation calculation were obtained from the cascade calculation of Metropolis *et al.*[1] for 1.8-GeV protons incident on copper and aluminum. Appropriate shifts in charge, mass number, and excitation energy were performed in order to apply the results to the target nuclei of interest. The validity of this transformation procedure is indicated by the fact that practically the same distribution of residual nuclei was obtained for Cl or V targets, whether the copper or the aluminum cascades were used. The momentum of the

TABLE IV. Data for calculation of range and energy distributions at 90° of Na^{24} from chlorine.

Catcher foil	Foil thickness ($\mu g/cm^2$)	Mean energy (MeV)	Net activity (counts/min)
1	47	0.1	445
2	50	0.5	417
3	49	0.7	483
4	51	1.0	468
5	49	1.3	485
6	49	1.6	503
7	47	1.9	457
8	50	2.4	520
9	50	2.8	456
10	52	3.4	518
11	158	5.6	1250
12	147	7.3	660
13–14	294	13.1	695
15–16	285	17.0	183
17–18	296	20.1	67.6
19–20	296	23.8	33.7

TABLE V. Mean ranges and energies of Na^{24}.

Target	Mean range in Formvar (mg/cm^2)	Mean energy Experimental (MeV)	Cascade-evaporation[a] (MeV)	Evaporation[b] (MeV)
Cl	0.34	4.3	6.2	1.9
V	0.59	6.1	6.6	3.7
Cu	0.84	8.2	9.7	6.4

[a] Cascade-evaporation calculation.
[b] Evaporation calculation assuming no cascade recoil.

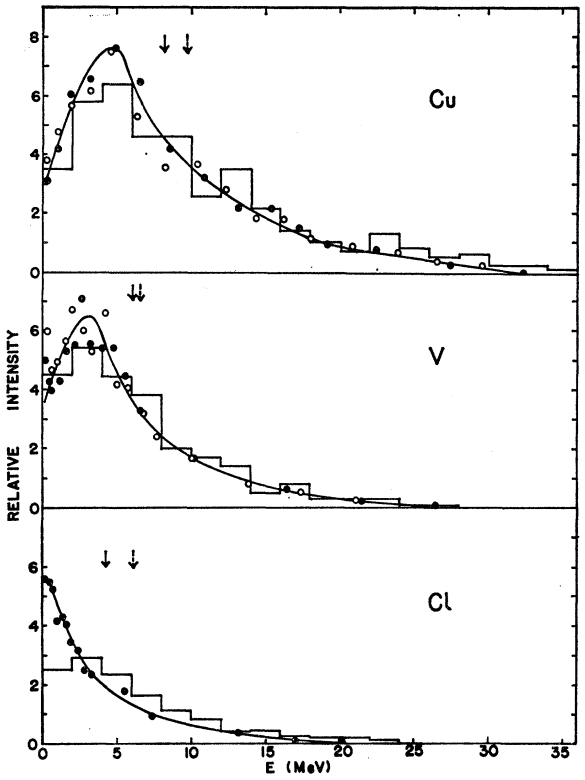

FIG. 4. Energy spectra at 90° of Na^{24} from 2.9-GeV proton irradiation of the indicated targets. Experimental data are shown by the points and the smooth curves through them. (Open points are the results of duplicate experiments with thicker targets.) The histograms show the results of the cascade-evaporation calculation. The solid and dashed arrows refer to mean experimental and calculated energies respectively.

residual nuclei was obtained from the calculation of Porile.[21] The same corrections were applied to these results as discussed elsewhere.[12,13]

The evaporation calculation was based on the treatment of Dostrovsky *et al.*[22] and included the modifications for the calculation of the recoil energy due to evaporation.[12,13] In addition to the evaporation of the six lightest particles through He^4, heavy particle evaporation was considered by replacement of all heavy particles with $A \leq 10$ by Li^7 in its ground state. This procedure has been described in detail in previous publications.[12,13] The calculation was performed with the same evaporation parameters used in earlier work.[12,13] In order to improve the statistical accuracy of the calculation, ten evaporation calculations were performed for each of approximately 400 starting nuclides. Also, the results for all products with $A = 23$–25 were combined for comparison with the results for Na^{24}. The output of the calculation gave the kinetic energy resulting from the cascade-evaporation process of all $A = 23$–25 products recoiling in an angular range of 74–106° with respect to the beam.

The calculated energy spectra are plotted as histograms in Fig. 4. In order to facilitate comparison, they have been normalized to the same area as the experimental spectra. It is seen that calculation and experiment are in very good agreement for the copper and vanadium targets. The mean recoil energies, compared in Table V, agree to within about 15% in these two cases. By contrast, there is a significant difference between the calculated and experimental spectra for the chlorine target. While the experimental spectrum has a pronounced peak for zero recoil energy followed by a sharp drop-off in intensity, the calculated spectrum remains essentially constant out to 5 MeV. The calculated mean energy for chlorine is some 40% larger than the experimental value.

To evaluate the significance of this discrepancy it is worthwhile to recall that the energy spectrum is determined by the recoil momentum imparted during both the cascade and evaporation phases of the reaction. We have computed the energy spectrum expected for the evaporation phase by starting with the same distribution of residual nuclei as before and setting the momentum imparted in the cascade equal to zero. The resulting spectra are compared with the cascade-evaporation results in Fig. 6. It is seen that the evaporation spectra tend to peak close to the origin in all cases. Evidently the displacement of the most probable energy from zero obtained in the cascade-evaporation calculations can be attributed to the recoil resulting from the cascade. In spite of this difference between the two sets of spectra, it is clear that the evaporation process makes the major contribution to the cascade-evaporation spectrum in the case of copper. On the other hand, the evaporation calculation for chlorine predicts much lower recoil energies than the cascade-evaporation calculation, indicating that the contribution from the cascade predominates. The mean energies predicted by the two calculations are summarized in Table V,

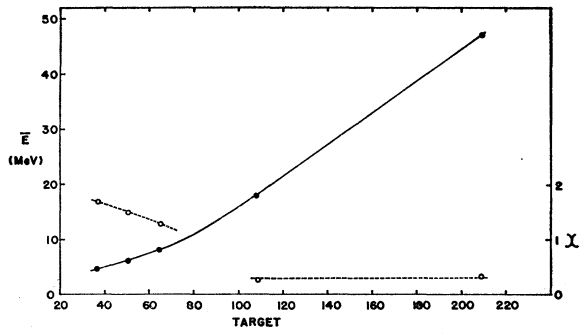

FIG. 5. Solid curve-variation with target A of the mean energy of Na^{24} emitted at 90° from 2.9-GeV proton irradiation of various targets. The ordinate scale at the left applies. Dashed curve variation with target A of the skewness coefficient χ for the energy spectra of Na^{24}. The ordinate scale at the right applies. Data in this figure are from the present study and from Refs. 13 and 20.

[21] N. T. Porile, Phys. Rev. **120**, 572 (1960).
[22] I. Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev. **116**, 683 (1959).

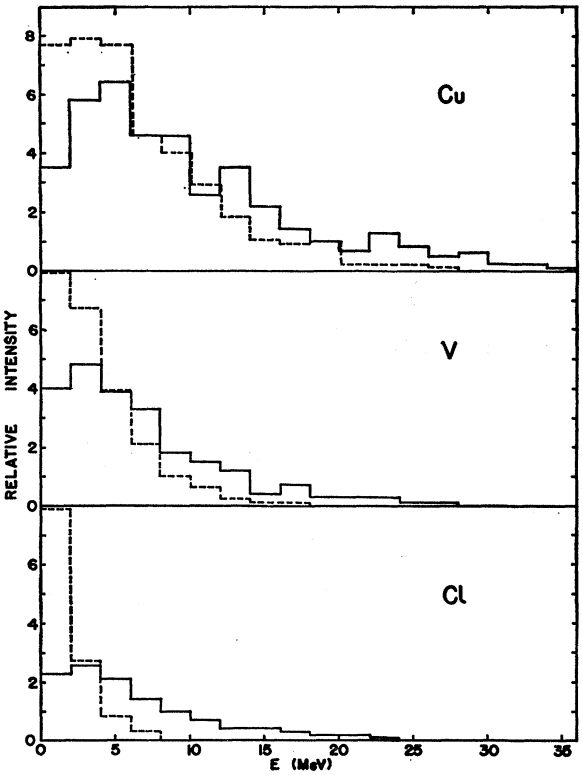

FIG. 6. Comparison of energy spectra for Na^{24} obtained from cascade-evaporation calculations (solid histograms) with those obtained from evaporation calculations (dashed histograms).

showing that the cascade-evaporation mean exceeds the evaporation mean by factors of 1.5, 1.8, and 3.3 in order of decreasing target A.

The discrepancy between experiment and calculation observed for chlorine can therefore be attributed to an overestimate of the momentum imparted to the residual nuclei in the cascade process. Since the spectra were obtained at 90° to the beam, this overestimate refers primarily to the transverse component of momentum. The calculation is not particularly sensitive to the forward component of momentum imparted in the cascade, and no conclusions can be drawn about this quantity. It has been previously concluded, however, on the basis of measurements of forward and backward effective ranges,[12] that the calculation also overestimates the value of the forward component of momentum.

In spite of the discrepancy observed for chlorine, it may be concluded that the energy spectra are fully consistent with a spallation process. This agreement does not necessarily rule out a contribution from two-body breakup processes for Na^{24} from copper since there may not be a very large difference between Na^{24} spectra for these two mechanisms. A crude estimate of the mean recoil energy for a two-body breakup process may be obtained from the results for Na^{24} from silver[13] and bismuth.[20] In both of these cases a twobody breakup process is indicated and the ratio of the mean energy to the Coulomb energy, calculated for a division of the target nucleus into two spheres with $r_0=1.44$ F, is about 0.5. A similar ratio for copper would result in Na^{24} fragments with a mean energy of only 10 MeV, in fair agreement with the experimental value.

On the other hand, a reasonable case can be made against a large contribution from a two-body breakup process to the formation of Na^{24} from copper on the basis of the relatively small difference in mass number between target and product. The estimate by Crespo *et al.*[15] of the average excitation energy required for the production of Na^{24} from Cu, as determined by the shape of the excitation function for this reaction, indicates a value of about 500 MeV. Nearly all of this energy will be available for particle emission, even in the case of two-body breakup, because the energy required to separate a nucleus in the copper region into two fragments is only about 20 MeV. Since an excitation energy of 500 MeV can lead to the formation of Na^{24} by spallation, it would appear that a two-body breakup process would have to lead to fragments with a lower mass number than Na^{24}. This conclusion is consistent with the evidence cited by Cumming *et al.*[13] to the effect that the most probable two-body breakup products from a target as heavy as silver lie in the $A=20$–30 region. The argument is also consistent with the results of the Monte Carlo cascade-evaporation calculation of the present authors,[12] which predicts a value for the cross section for Na^{24} formation from copper that is in good agreement with the experimental value at GeV energies.

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