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Production of ^{38}Ar and ^{39}Ar in the interaction of gold and thorium with 1, 2.5, and 24 GeV protons

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Cross sections and thick-target recoil properties of ^{38}Ar and ^{39}Ar, formed in the interaction of Au and Th with 1, 2.5, and 24 GeV protons, have been determined in order to study their variations with incident energy. The measurement of the experimental range, $2W(F+B)$, and of the forward-to-backward ratio F/B permits (with the help of a mathematical formalism based on the two-step model) the determination of some characteristics of nuclear reactions; i.e., the mean kinetic energy of the observed products $\langle T \rangle$, the mean forward component of the velocity imparted to the struck nucleus in the first step, $\langle v_{\parallel} \rangle$, and the excitation energy E^* of the residual nucleus after cascade. The results are discussed in terms of very asymmetric fission (at 1 GeV) and deep spallation (at 2.5 and 24 GeV) mechanisms. These results are very similar to those obtained for neutron-deficient krypton and xenon isotopes formed in thorium and presented in a previous publication from this laboratory.

[NUCLEAR REACTIONS Measured σ, $2W(F+B)$, and F/B of ^{38}Ar and ^{39}Ar isotopes formed in interaction of Au and Th with 1, 2.5, and 24 GeV protons.]

I. INTRODUCTION

The study of light or medium fragments ($A \leq 50$) formed in heavy targets by high-energy protons has retained the attention of many authors for more than twenty years. One of the first assumptions concerning this process, often designated by the term of "fragmentation," has been to consider that it was rapid. Forward-peaked angular distributions of ^{24}Na produced by bombardment of Bi by 2.9 GeV protons had been interpreted by Cumming *et al.*,[1] in 1964, as the signature of the emission of this fragment in a time comparable to that of the intranuclear cascade. Other experiments[2–5] concerning fragments produced with $2-5$ GeV protons in heavy targets have confirmed that a large part of the light nuclides were formed by a rapid one-step mechanism.

For a number of years, several authors[6–8] have felt there was a link between the productions of light fragments and of neutron-deficient isotopes. In that connection, various assumptions had been made without the correlation between fragmentation and deep spallation appearing very clearly. But in 1975, a very interesting result was obtained by Remsberg and Perry[9] who found a sideways peaking for the angular distributions of light fragments ($6 < Z < 12$) produced from 28 GeV proton irradiation of U and Au. This reduction of forward peaking (between 3 and 28 GeV) for light fragments was already known to be one of the characteristics of neutron-deficient isotopes production.

Another important study has emphasized this idea. In 1976, Scheidemann and Porile[10] measured cross sections and recoil properties of scandium isotopes formed in the interaction of ^{238}U with $1-300$ GeV protons. These authors have shown that cross sections, F/B ratios, and experimental ranges $2W(F+B)$ of such fragments exhibited the same behavior as heavier neutron-deficient isotopes. This experiment has greatly clarified the situation by proving that deep spallation could also explain the formation of medium fragments. It should be noted that this behavior of scandium differs from that of lighter nuclides, such as ^{24}Na or ^{28}Mg,[11,12] since Kaufman *et al.*[12] find no important variation of range values between 1 and 300 GeV for ^{24}Na produced in gold by high-energy protons.

In order to study the production characteristics of the intermediate mass region between magnesium and scandium, we have measured cross sections and recoil properties of ^{38}Ar and ^{39}Ar, formed in gold and thorium by protons of 1, 2.5, and 24 GeV.

II. EXPERIMENTAL PROCEDURE

We have used the well-known thick-target, thick-catchers technique in a manner already described in a recent publication.[13] The target stacks consisted of 50 μm thorium foil, sandwiched between two pairs of 50 μm aluminium foils. The first pair served as recoil catchers and the second pair as guard foils. Several aluminium foils were added at different locations in the stack to serve as beam monitors.

The irradiations were performed at Saturne I and II (1.05 and 2.5 GeV) and at CERN proton synchrotron (24 GeV). At 1 GeV, the irradiations were carried out in the internal beam, while at 2.5 and 24 GeV, external beams were used. The flux was monitored by the reaction $^{27}Al(p,3p3n)^{22}Na$, for which the cross sections have been compiled by Tobailem *et al.*[14] The total number of protons varied from 3×10^{16} to 3×10^{17}, depending on the irradiation. An interval of at least several months separated the irradiations from the analyses.

The technique employed for the measurement of argon produced by nuclear reactions has been described elsewhere.[15–17] Argon is extracted by melting the target (or catcher) under vacuum in a molybdenum crucible, heated by electronic bombardment (600–1800 °C, depending on the metal). After purification of the extracted gases on titanium and copper oxide-palladium getters, argon is analyzed in a 60° sector, 12 cm radius, mass spectrometer (a modified MICROMASS 12). The mass spectrometer is calibrated by means of a pipette which contains ^{36}Ar and ^{38}Ar and which allows a precisely known quantity of gas to be introduced. The isotope ratios are corrected for memory or pumping effects and isotopic discrimination in the mass spectrometer. Another correction is sometimes necessary because of the presence of hydrocarbon at mass 39. As few as 10^8 to 10^{12} atoms of argon can so be measured.

III. DETERMINATION OF RECOIL PROPERTIES

The very simple thick-target, thick-catcher technique provides the kinetic energy $\langle T\rangle$ of the reactions products and the excitation energy E^* of the cascade residual nucleus. The mathematical formalism used here has already been presented in a recent publication from this laboratory.[13]

The analysis of the data uses the two-step vector model of high-energy reactions first developed by Sugarman *et al.*[18–20] The nuclear interaction is described by the ejection of prompt nucleons, followed by deexcitation of the residual cascade nucleus by neutron or charged-particle evaporation. These two steps are characterized by the velocity vectors $\vec{v}$ and $\vec{V}$, respectively. Their resultant can lead to the emission of the product nuclei from the target material, to be caught in the aluminium catchers. The fractions F and B of the activity collected in the forward and backward directions may thus be determined.

The mathematical development of the thick-target, thick-catcher theory expresses the forward and backward activities F and B as follows:

$$F=\frac{R}{16\eta^4 W}\left\{(1+\eta)^{N+1}\left[\frac{4\eta^2}{N+3}(1+\eta)^2+\frac{4\eta^2}{N+1}(\eta^2-1)\right]\right.$$
$$\left.-(1-\eta^2)\frac{N+1}{2}\left[\frac{4\eta^2}{N+3}(1-\eta^2)+\frac{4\eta^2}{N+1}(\eta^2-1)\right]\right\}. \quad (1)$$

If η is replaced by $-\eta$, relation (1) gives the expression for B [designated (1a)].

In these equations, R is the mean range in the target material corresponding to the velocity V, η is the ratio of the cascade velocity v to the second step velocity V. (The perpendicular component of the cascade velocity, $v_\perp$, is assumed to be zero.) N is a constant characteristic of the nuclear reaction; $N\simeq1$ for fission and $N\simeq2$ for the spallation.

As explained elsewhere,[13,21,22] the range R is determined from the confrontation of the experimental values of $2W(F+B)$ and $W(F-B)$ with the calculated values considered as functions of R, η (varying from 0 to 1), and N (varying from 1 to 2). The range is then corrected for scattering. The mean kinetic energy $\langle T\rangle$ is calculated by means of the Northcliffe-Schilling range-energy relations.[23]

The velocity v imparted to the target nucleus by cascade can be determined from $T=\frac{1}{2}AV^2$ and η. Intranuclear cascade calculations have shown a correlation between momentum component P_t transferred to the target nucleus and parallel to the beam, and the average excitation energy E^* of cascade residues. Using Metropolis calculations,[24] Porile[25] found that the relation

$$\frac{E^*}{E_{\rm CN}}=0.8\frac{P_t}{P_{\rm CN}} \qquad (2)$$

was satisfied for many targets and bombarding energies up to 1.8 GeV. In Eq. (2), $P_{\rm CN}$ and $E_{\rm CN}$ are, respectively, the momentum and excitation energy of a hypothetical-compound nucleus formed by the fusion of proton and target nucleus. Kaufman *et al.*[12] have tested this correlation at 3 GeV and have found it valid for this case also.

IV. RESULTS

A. Cross sections

Table I gives the cross sections of ^{38}Ar and ^{39}Ar formed in gold and thorium at 0.15, 1, 2.5, and 24 GeV. All the cross sections are cumulative. Several values, marked with an asterisk, are due to Regnier.[15] In general, each value is the average of three independent measurements. The uncertainties have been determined as explained elsewhere.[13,16] The total uncertainty of a cross section measurement is generally from 10 to 20%. The mean of the independent measurements, x_i, is then

TABLE I. Cumulative ^{38}Ar and ^{39}Ar cross sections, in mb, measured in gold (above) and thorium (below) with 1, 2.5, and 24 GeV protons. Results marked with an asterisk are due to Regnier (Ref. 15).

	Proton energy (GeV)		
Ar in Au	1	2.5	24
38	*0.43±0.03	4.3±1.1	*6.5±0.8
39	*0.43±0.07	4.0±0.9	*5.3±0.7
Ar in Th	1	2.5	24
38	*0.56±0.06	9.3±1.8	13.0±2.6
39	*0.51±0.06	8.6±1.5	11.5±1.9

calculated, weighting each of these by the inverse of the square of its uncertainty, Δx_i. Thus σ is given by

$$\sum_i (x/\Delta x^2)/\sum_i (1/\Delta x)^2 .$$

The uncertainty given in Table I is the standard deviation from the mean of the independent measurements, or the quantity

$$\Delta\sigma = | \sum (1/\Delta x)^2 |^{1/2} ,$$

whichever is larger.

B. Recoil properties

Table II shows the results obtained. The uncertainties cited are the rms deviations observed for repeated experiments. The forward-to-backward ratio F/B is a measure of the forward peaking of the nuclide in the beam direction. The experimental range in the target material, $2W(F+B)$ (in mg/cm^2), is corrected for scattering at the target-catcher interface and for edge effects.

The mean kinetic energy $\langle T \rangle$ (in MeV) is calculated from the ranges using the Northcliffe-Schilling relations, as mentioned above. These experimental values are compared with those calculated theoretically, $\bar{E}$, by Nix and Swiatecki,[26] according to the liquid-drop model. The ratio $\langle T \rangle/\bar{E}$ may be considered to be a good test for fission.

The mean forward component of the cascade velocity $\langle v_{\parallel} \rangle$ has not been corrected for the possible overlap between the cascade and the deexcitation velocities (respectively, $\vec{v}$ and $\vec{V}$), since this effect does not qualitatively change the result.[27] The excitation energies E^* of the residual cascade nuclei leading to ^{38}Ar and ^{39}Ar have been calculated with formula (2) given above.

V. DISCUSSION

A. Cross sections, F/B ratios, experimental ranges, and kinetic energies

Figure 1 shows, on the same graph, the variations of cross sections, F/B ratios, and experimental ranges $2W(F+B)$ with incident energy. Biswas and Porile[28] used this representation in their work concerning the formation of cerium, lanthanum, and barium isotopes in uranium by GeV protons. Note, in Fig. 1, the increasing excitation functions,

TABLE II. Recoil data for ^{38}Ar and ^{39}Ar formed in gold and thorium with 1, 2.5, and 24 GeV protons. $\langle T \rangle / \bar{E}$ represents the ratio of experimental kinetic energy $\langle T \rangle$ to calculated kinetic energy $\bar{E}$ (see text).

E_p (GeV)	F/B	$2W(F+B)$ (mg/cm^2)	$\langle T \rangle$ (MeV)	$\langle T \rangle / \bar{E}$	$\langle v_{\parallel} \rangle$ (MeV/amu)$^{1/2}$	E^* (MeV)
			^{38}Ar in gold			
1	1.41±0.07	11.23±0.82	64.2±7.8	0.89±0.11	0.149±0.023	423± 66
2.5	1.73±0.10	8.49±0.69	40.2±6.0	0.56±0.08	0.193±0.034	699±122
24	1.06±0.04	7.58±0.72	34.1±5.8	0.47±0.08	0.027±0.005	116± 21
			^{39}Ar in gold			
1	1.37±0.08	10.82±0.73	60.1±7.0	0.84±0.10	0.133±0.021	379± 61
2.5	1.62±0.08	8.76±0.76	41.8±6.5	0.58±0.09	0.173±0.030	624±114
24	1.09±0.05	7.13±0.60	30.0±4.5	0.42±0.06	0.026±0.005	113± 21
			^{38}Ar in thorium			
1	1.28±0.09	15.84±1.35	95.1±12.4	1.26±0.16	0.134±0.025	448± 83
2.5	1.52±0.10	11.20±1.04	61.2± 8.9	0.81±0.12	0.179±0.038	765±163
24	1.09±0.05	9.13±0.82	44.3± 7.4	0.59±0.06	0.034±0.006	173± 32
			^{39}Ar in thorium			
1	1.34±0.11	14.67±1.24	87.8±12.1	1.14±0.16	0.149±0.028	497± 93
2.5	1.59±0.13	10.82±0.93	57.3± 7.9	0.75±0.10	0.189±0.045	803±132
24	1.07±0.03	8.41±0.94	37.4± 6.6	0.49±0.08	0.028±0.005	143± 24

the maximum of F/B about 2.5−3 GeV, followed by a continuous decrease at higher energies, and the sharp decrease of experimental ranges beyond 1 GeV. This behavior of ^{38}Ar and ^{39}Ar produced in

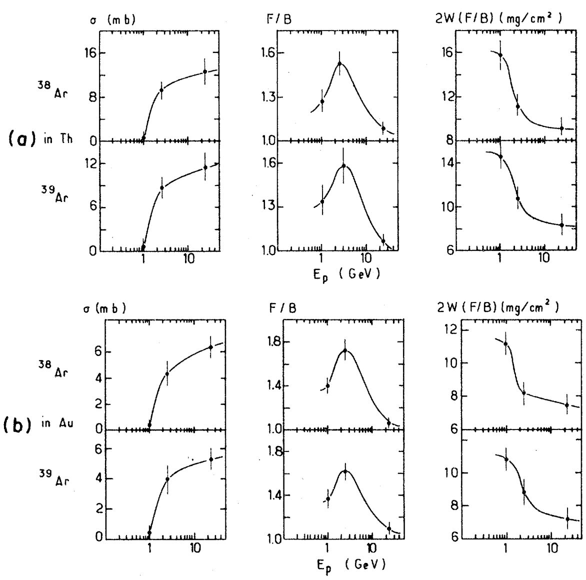

FIG. 1. Incident energy dependence of σ (left), F/B (middle), and range (right) of ^{38}Ar and ^{39}Ar formed by interaction of (a) ^{232}Th and (b) ^{197}Au with high energy protons.

gold and uranium is very similar to that of neutron-deficient isotopes in the mass region $A \sim 80-140$, formed in thorium[13] or uranium.[28] Scandium isotopes produced in uranium by 1−300 GeV protons also exhibit these characteristics. It thus seems justified to explain the formation of fragments such as argon or scandium in heavy targets by a mechanism similar to deep spallation. The fall of the ranges beyond 1 GeV and the peak of F/B in the neighborhood of 3 GeV are generally interpreted as a transition between fission (up to 1 GeV) and deep spallation (beyond 5−6 GeV).

The comparison between experimental kinetic energies $\langle T \rangle$ and those calculated theoretically, $\bar{E}$, by the simplified liquid-drop model[26] provides a possible measurement of the fission contribution. As has already been mentioned,[10,13] agreement between experimental and calculated values does not mean that the production mechanism is necessarily binary fission, but simply that the results are consistent with such a process. Moreover, the model of Nix and Swiatecki[26] is not perfectly adapted to binary fissions as asymmetric as those leading to the formation of fragments such as argon. It may be noted, for instance, that the values of $\langle T \rangle / \bar{E}$ for ^{38}Ar and ^{39}Ar produced in thorium (the most heavy target) are greater than unity. The value of 1.26 for ^{38}Ar is too high, since the target nucleus is assumed to be the fissioning nucleus. In this case, the mass difference due to nucleons lost during the cascade step and to prefission evaporation is not taken into account. In addition, the Northcliffe-Schilling relations have been used in the present work, while other range-energy relations, such as those of Winsberg,[29] would lead to kinetic energies which would be smaller by about 10 to 12%.

Nevertheless, one of the most important points to be noted is the variation of $\langle T \rangle / \bar{E}$ with E_p, as shown in Fig. 2. This ratio falls by a factor of about 2 between 1 and 24 GeV. This behavior is very similar to that of heavier neutron-deficient nuclides, for example, $^{78-81}$Kr or $^{124-129}$Xe formed in thorium.[13] Scandium isotopes produced in uranium also exhibit this decrease of kinetic energy. In all cases, it is naturally a consequence of the drop-off of the ranges. This behavior has probably a single interpretation. It is generally accepted that the decrease of kinetic energy (or range) beyond 1 GeV represents the transition from fission to deep spallation. It is perhaps difficult to consider that fission may be responsible for argon production at 1 GeV. It should be noted, however, that the weak cross sections (0.4—0.5 mb) are compatible with such a process. Another argument for fragment production by asymetric binary fission around 1 GeV comes from the study of differential ranges of scandium nuclides formed in the interaction of ^{238}U by 0.8 GeV protons.[30] Fortney and Porile consider that their results are consistent with highly asymetric binary fission at 0.8 GeV, but suggest an increasing contribution of deep spallation at the higher energies.

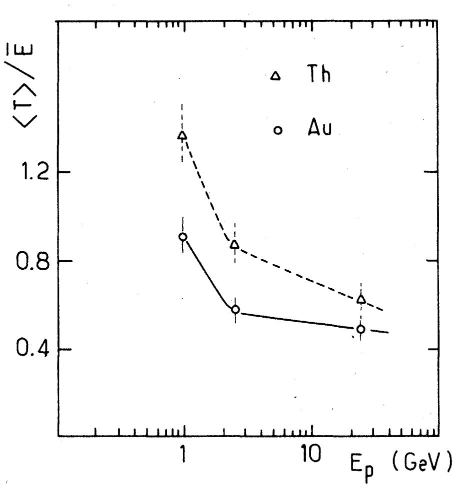

FIG. 2. Incident energy dependence of experimental to calculated kinetic energy $\langle T \rangle / \bar{E}$ ratio of ^{38}Ar formed in thorium and gold.

B. Velocity of the struck nucleus and excitation energies of the residual cascade nucleus

A noticeable characteristic of argon production in heavy targets above 1 GeV is the very important decrease of $\langle v_{\parallel} \rangle$ (the mean forward component of the velocity of the struck nucleus) beyond 3 GeV (Fig. 3). This type of variation much resembles that observed for scandium isotopes and for the most neutron-deficient isotopes (e.g., ^{131}Ce) produced in uranium.[10,28] If the two-step representation remains valid at these high energies, this fall of $\langle v_{\parallel} \rangle$ means that the new mechanism replacing fission is characterized by weak impulsion transfer to the struck nucleus as the incident energy increases. This is another characteristic of deep spallation. The immediate consequence of the decrease of $\langle v_{\parallel} \rangle$ is a decrease of the excitation energies E^* (Fig. 4) because of the linear dependence $v_{\parallel}$-E^*. In the framework of the conventional two-step model, Porile and Sugarman[31] have shown that the shape of the excitation functions of high-energy reaction products (such as argon formed in heavy targets) is not compatible with such a decrease of E^* beyond 3 GeV. The confrontation of the shape of the excitation functions with the variation of excitation energy versus incident energy may be seen in Fig. 4. Here again is found the same contradiction between increasing excitation functions and highly decreasing excitation energies beyond 3 GeV that has been observed for neutron-deficient krypton and xenon isotopes produced in thorium.[13] We have already mentioned that the relation between $v_{\parallel}$ and E^* arise from Monte Carlo cascade calculations.[24,25] The breakdown of the E^* values at 24 GeV may consequently mean that the classical

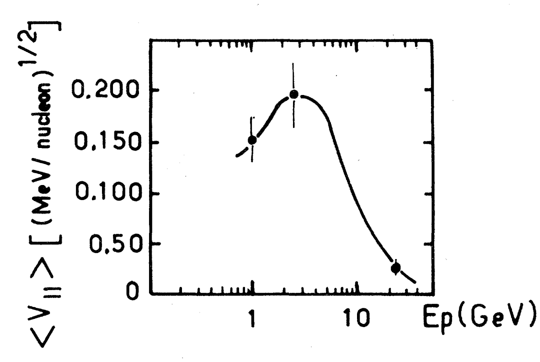

FIG. 3. Incident energy dependence of the cascade velocity $\langle v_{\parallel} \rangle$ of nuclei leading to the formation of ^{38}Ar in gold.

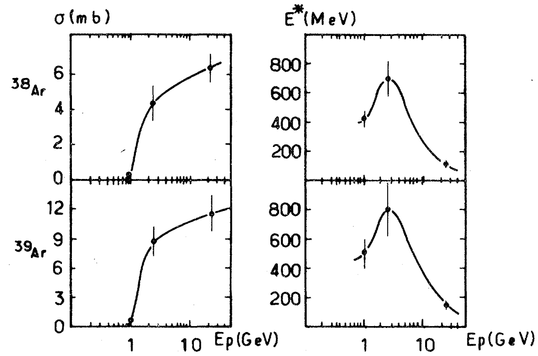

FIG. 4. Incident energy dependence of cross sections and excitation energies for ^{38}Ar formed in gold and for ^{39}Ar formed in thorium.

representation of the intranuclear cascade ceases to be valid at multi-GeV energies. In these conditions, instead of a cascade of individual and quasi-free nucleons, one must probably envisage a collective interaction between incident proton and target nucleus.[32,33] Several authors have used this new assumption to try to explain some deep spallation characteristics.[28,34–36] According to these workers, a nearly central collision of a highly-relativistic proton with a heavy nucleus may be represented as follows: Target nucleons in the path of the incident proton may act collectively, constituting an "effective target." These nucleons are rapidly ejected from the target nucleus in the forward direction, carrying off most of the impulsion of the incident proton. Other nucleons may be emitted from the surface of the resulting "hole" punched out in the target along the beam direction. Consequently, the target residue is almost stationary in the laboratory system and the resulting fragments are preferentially ejected transversely to the beam direction because of Coulombic repulsion. The observed products resulting from the deexcitation of these fragments will thus have low values of F/B and $\langle v_{\parallel} \rangle$. Argon produced in gold and thorium at 24 GeV exhibits precisely these characteristics.

Such a collective model seems able to account qualitatively for some experimental features of deep spallation reactions. But one of the principal difficulties is to explain the extensive mass loss occurring prior to breakup, as attested by the decrease of the ranges. On the other hand, except for a calculation of Cumming,[34] very little in the way of quantitative tests presently exists for this collective model.

However, the idea of fragment production by a collective interaction between incident proton and target nucleus has already been advanced before the application of the "collective tube model" to deep spallation reactions. Thus, Remsberg and Perry[9] have assumed the generation of nuclear shock waves to explain the sideways peaking in angular distributions of light fragments formed in gold and uranium by 28 GeV protons. Scheidemann and Porile[10] have also considered the propagation of a shock wave inside the nucleus as one of the possible explanations of the low F/B values for scandium isotopes produced by GeV protons in uranium. It may be noted that the shock wave assumption[37] and other hydrodynamical descriptions[38] have been proposed by heavy ions physicists.

Thus, various data seem to indicate that the assumption of a collective interaction between incident proton and target nucleus is slowly gaining ground in attempts to account for the experimental characteristics of deep spallation or fragmentation reactions. Although these explanations are as yet qualitative, some quantitative results, such as those indicating that fast secondary particles created in high-energy proton-nucleus interaction do not cascade[32]—also call into question the classical representation of intranuclear cascade. More experiments and calculations in this area are clearly needed.

VI. CONCLUSIONS

The thick-target, thick-catchers technique and mass spectrometry measurements have been used to study the production of ^{38}Ar and ^{39}Ar in gold and thorium by high-energy protons. The incident energy dependence of cross sections and recoil properties are compatible with the following two mechanisms: very asymmetric fission is responsible for argon production at 1 GeV; the contribution of deep spallation is already important at 2.5 GeV and total at 24 GeV. No appreciable difference is observed between the results for gold and for thorium. The transition between fission and deep spallation is interpreted as a change of the first step of the reaction from a classical intranuclear cascade to a collective interaction.

Thus the principal result of this work is to show that fragments as light as argon are produced by a mechanism very similar to deep spallation responsible for the production of neutron deficient isotopes ($80 < A < 140$) in heavy targets (Refs. 6, 7, 8, 12, 13, 17, 22, and 28).

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