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PHYSICAL REVIEW C

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## Fission of $\text{Th}^{232}$ Induced by Intermediate-Energy $\text{He}^4$ Ions\*

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The cross sections for 34 nuclides covering the mass range 24 to 199 formed from the  $\text{He}^4$  ion-induced fission of  $\text{Th}^{232}$  at 20–41 MeV have been determined radiochemically. The resulting mass-yield curve clearly indicates contributions from at least three different types of fission: the symmetric binary, asymmetric binary, and asymmetric ternary modes. The identification of very light fragments and the low limits set on  $\text{Au}^{199}$  confirm the existence of ternary fission seen previously in this laboratory in plutonium compound nuclei. Certain similarities and differences in this fissioning system to other heavier elements are discussed.

### INTRODUCTION

Previous radiochemical studies from this laboratory on the fission yields of very low and very high masses have provided evidence for ternary fission in uranium irradiated with intermediate-energy helium ions.<sup>1-3</sup> Among the questions to study concerning the ternary-fission process is the generality with which it occurs in other compound nuclei. The present research on thorium is a part of such a study.

Preliminary experiments were carried out irradiating  $\text{Th}^{232}$  with protons. However, it became apparent that spallation products formed from the trace impurities in the best thorium foils which were available at the time made the interpretation of the low-yield ternary data very uncertain. A change to  $\text{He}^4$  ions on  $\text{Th}^{232}$  greatly improved the situation, and this communication is a report of that work.

### EXPERIMENTAL PROCEDURES

Irradiations using  $\text{He}^4$  ions were carried out at the 60-in. cyclotron at Argonne National Laboratory. The integrated currents ranged from 0.04 to 25  $\mu\text{A h}$  depending upon the estimated fission-product yield. Various  $\text{He}^4$  ion energies (22.9–41.3 MeV) were obtained by suitable aluminum degrading foils; the energy was known to  $\pm 0.5$  MeV. The 36-MeV proton bombardments were made at the Michigan State University cyclotron.

In most experiments 1.4–1.5-mil thorium metal foils were used. However, at the lowest energy a low-density thin thorium oxide target was prepared by electrodeposition using a procedure described elsewhere.<sup>4</sup> The target assembly consisted of the target foils and highly purified aluminum or silver cover foils,<sup>5</sup> some of which also served to collect fission products recoiling out of the thorium.

The metallic thorium foils were analyzed spec-

TABLE I. Cross sections for the fission of  $\text{Th}^{232}$  induced by intermediate-energy  $\text{He}^4$  ions. Values in square brackets were extrapolated from lower energies.

Isotope/Mass <sup>a</sup>	22.9 MeV (mb)	33.2 MeV (mb)	39.1 MeV (mb)	41.3 MeV (mb)
Na <sup>24</sup>	...	...	...	$(730 \pm 200) \times 10^{-6}$
24	...	...	...	$730 \times 10^{-6}$
Mg <sup>28</sup>	...	...	...	$(125 \pm 20) \times 10^{-6}$
28	...	...	...	$125 \times 10^{-6}$
Si <sup>31</sup>	...	...	...	$(24 \pm 5) \times 10^{-6}$
31	...	...	...	$24 \times 10^{-6}$
S <sup>38</sup>	...	...	...	$(10 \pm 3) \times 10^{-6}$
38	...	...	...	$10 \times 10^{-6}$
Ca <sup>47</sup>	...	...	...	$\leq 1.2 \times 10^{-6}$
47	...	...	...	$\leq 1.2 \times 10^{-6}$
Mn <sup>56</sup>	...	...	...	$(565 \pm 50) \times 10^{-6}$
56	...	...	...	$565 \times 10^{-6}$
Ni <sup>66</sup>	...	...	...	$(29 \pm 4) \times 10^{-3}$
66	...	...	...	$30 \times 10^{-3}$
Zn <sup>72</sup>	...	...	...	$(170 \pm 10) \times 10^{-3}$
72	...	...	...	$172 \times 10^{-3}$
Br <sup>83</sup>	$0.72 \pm 0.04$	$8.98 \pm 0.43$	$13.5 \pm 0.20$	[14.5]
83	0.65	8.08	12.2	[14]
Sr <sup>89</sup>	$1.76 \pm 0.15$	$23.2 \pm 2.4$	$30.1 \pm 2.3$	[34]
89	1.76	23.2	30.1	[34]
Sr <sup>91</sup>	$2.52 \pm 0.25$	$29.4 \pm 2.6$	$38.7 \pm 2.6$	[40]
91	2.52	29.7	39.1	[40]
Y <sup>93</sup>	...	$34.8 \pm 5.0$	$50.5 \pm 5.0$	...
93	...	34.8	50.5	...
Zr <sup>95</sup>	$4.29 \pm 0.09$	$46.5 \pm 4.0$	$58.0 \pm 7.4$	[63]
95	4.29	46.5	58.0	[63]
Ir <sup>97</sup>	$3.34 \pm 0.35$	$37.4 \pm 2.6$	$49.7 \pm 6.7$	[56]
97	3.37	37.8	50.2	[57]
Ru <sup>103</sup>	$2.26 \pm 0.20$	$27.4 \pm 2.8$	$42.4 \pm 3.7$	[46]
103	2.26	27.4	42.4	[46]
Ru <sup>105</sup>	$1.41 \pm 0.22$	$19.6 \pm 1.0$	$31.9 \pm 4.5$	[34]
105	1.41	19.6	31.9	[34]
Ru <sup>106</sup>	$1.01 \pm 0.11$	$14.4 \pm 4.0$	$23.6 \pm 1.0$	[25]
106	1.01	14.4	23.8	[25]
Pd <sup>112</sup>	$0.57 \pm 0.06$	$15.0 \pm 1.8$	$24.8 \pm 0.7$	[28]
112	0.57	15.2	25.1	[28]
Ag <sup>113</sup>	$0.79 \pm 0.77$	$21.2 \pm 1.3$	$31.9 \pm 1.9$	[34]
113	0.79	21.2	31.9	[34]
Cd <sup>115</sup>	$0.58 \pm 0.67$	$14.8 \pm 0.8$	$27.8 \pm 1.6$	...
Cd <sup>115m</sup>	[0.05] <sup>b</sup>	$1.08 \pm 0.4$	$2.08 \pm 0.29$	...
Cd <sup>115 total</sup>	$0.63 \pm 0.07$	$16.1 \pm 0.8$	$30.6 \pm 0.9$	[37]
115	0.63	16.1	30.6	[37]
I <sup>131</sup>	$1.96 \pm 0.10$	$21.8 \pm 0.8$	$29.9 \pm 2.2$	[32]
131	3.02	34.6	47.9	[50]
I <sup>133</sup>	$2.86 \pm 0.13$	$32.7 \pm 1.1$	$40.9 \pm 1.2$	[45]
133	3.26	39.4	55.3	[60]

TABLE I (Continued)

Isotope/Mass <sup>a</sup>	22.9 MeV (mb)	33.2 MeV (mb)	39.1 MeV (mb)	41.3 MeV (mb)
Ce <sup>141</sup>	2.96 ± 0.49	28.8 ± 2.9	36.0 ± 1.5	[38]
141	2.96	28.8	36.4	[38]
Ce <sup>145</sup>	2.50 ± 0.45	24.3 ± 1.2	28.4 ± 0.10	[31]
145	2.53	24.8	29.3	[32]
Pr <sup>145</sup>	1.64 ± 0.03	15.4 ± 2.0	...	...
145	1.66	15.6	...	...
Nb <sup>147</sup>	1.08 ± 0.08	10.1 ± 0.80	12.7 ± 0.70	[13]
147	1.08	10.2	12.8	[13]
Sm <sup>153</sup>	0.119 ± 0.006	1.54 ± 0.33	2.44 ± 0.52	[2.8]
153	0.120	1.56	2.51	[2.9]
Eu <sup>156</sup>	(32.8 ± 4.0) × 10 <sup>-3</sup>	548 ± 33 × 10 <sup>-3</sup>	990 ± 270 × 10 <sup>-3</sup>	[1000] × 10 <sup>-3</sup>
156	33.0 × 10 <sup>-3</sup>	565 × 10 <sup>-3</sup>	1004 × 10 <sup>-3</sup>	[1100] × 10 <sup>-3</sup>
Eu <sup>157</sup>	(25.0 ± 3.0) × 10 <sup>-3</sup>	(305 ± 50) × 10 <sup>-3</sup>	(780 ± 22) × 10 <sup>-3</sup>	[900] × 10 <sup>-3</sup>
157	26.0 × 10 <sup>-3</sup>	328 × 10 <sup>-3</sup>	886 × 10 <sup>-3</sup>	[990] × 10 <sup>-3</sup>
Gd <sup>159</sup>	(10.3 ± 1.5) × 10 <sup>-3</sup>	(183 ± 7) × 10 <sup>-3</sup>	(383 ± 7) × 10 <sup>-3</sup>	[450] × 10 <sup>-3</sup>
159	11.0 × 10 <sup>-3</sup>	193 × 10 <sup>-3</sup>	416 × 10 <sup>-3</sup>	[500] × 10 <sup>-3</sup>
Tb <sup>161</sup>	(5.49 ± 6.0) × 10 <sup>-3</sup>	(95.0 ± 24.0) × 10 <sup>-3</sup>	(100 ± 0.25) × 10 <sup>-3</sup>	[105] × 10 <sup>-3</sup>
161	6.0 × 10 <sup>-3</sup>	98.0 × 10 <sup>-3</sup>	105 × 10 <sup>-3</sup>	[110] × 10 <sup>-3</sup>
Au <sup>199</sup>	...	...	...	≤ 1.3 × 10 <sup>-6</sup>
199	...	...	...	≤ 1.3 × 10 <sup>-6</sup>

<sup>a</sup>Corrected for independent yields (see text).<sup>b</sup>Estimated as 9% of Cd<sup>115</sup> total.

troscopically for contaminants which form interfering products.<sup>6,7</sup> The spallation contributions to the total observed Mg<sup>28</sup> in the target foil were small (≤2%), but were significant for Na<sup>24</sup>, Si<sup>31</sup>, Mn<sup>56</sup>, Ni<sup>66</sup>, and Zn<sup>72</sup>. Consequently, only the fractions recoiling into the backward (i.e., 180° from the direction of the beam) catcher foils were determined for these nuclides, and their ranges were assumed to be the same as observed for Mg<sup>28</sup> and previously<sup>1</sup> for Sr<sup>89</sup>. Appropriate factors were then applied to obtain the total formation cross section. Additional targets were run at 11.6 MeV to determine the contribution to binary fission induced by secondary neutrons at the target, and at 36 MeV to evaluate spallation contributions to the ternary yields.

Chemical separations of the binary products have been described elsewhere.<sup>4</sup> The procedures used for ternary products and Au<sup>199</sup> required extensive and lengthy separations sometimes requiring 24 h.<sup>2,8</sup>

Previously standardized β<sup>4</sup> and γ<sup>9</sup> counting systems were used for the higher-yield species and anticoincidence low-background (≤0.24-counts/min) Geiger counters shielded with several tons of lead were used for the lower-yield species to determine absolute disintegration rates.

Isobaric cross sections obtained from cumula-

tive isotopic yields were based on independent-yield corrections using the constant charge ratio (CCR)<sup>10</sup> rule. Such corrections amounted to a few percent except for iodine isotopes (35–40%) and some heavier rare-earth isotopes (≤12%). The CCR rule has been shown to give sufficiently accurate corrections for the purposes of the present research in the fission of heavy elements at intermediate energies.<sup>10–12</sup> The binary-fission cross sections are believed accurate to about ±10% and the lower-yield ternary products to ±20%.

#### EXPERIMENTAL RESULTS

The observed and corrected (to total isobaric yield) cross sections are summarized in Table I. The cross sections for Pd<sup>112</sup> were obtained by “milking” and counting the Ag<sup>112</sup> daughter. At two energies, 22.9 and 33.2 MeV, the cross section for mass 113 obtained by counting Ag<sup>113</sup> appeared to be higher than the mass-115 yield. It is now believed that this is due to the inherent difficulties in resolving the decay curves for silver. Cross sections for mass 106 were lower than those observed for masses 112, 113, and 115 at the higher energies, but not at 22.9 MeV.

The average number of neutrons emitted during fission increased from ~3 to ~5 between 22.9 and

39.1 MeV, as determined by reflection of the data for certain masses between 75–90 and the reflected heavy-fragment peak at masses 140–155.

These neutron numbers were, in turn, used to help fix the total isobaric yields for those isotopes where independent-yield corrections were significant.

In one experiment information on the range of  $Mg^{28}$  in thorium was obtained. In the irradiation of  $Th^{232}$  with 41-MeV  $He^4$  ions,  $3.1 \times 10^6$   $Mg^{28}$  nuclei were observed in a 1.38-mil target foil, and  $3.08 \times 10^5$  were found in the "backward" catcher foil. For thick targets the relationship<sup>13</sup>  $R = 2W(F+B)$  can be used, where  $W$  is the target-foil thickness in  $mg/cm^2$ , and  $F$  and  $B$  are the fraction of the total  $Mg^{28}$  nuclei which recoil into the forward and backward catcher foils, respectively. Since the presence of spallation precluded measuring  $F$ , the approximate relationship  $R \cong 4WB$  was used to estimate the range as  $12.5 \pm 1.3$   $mg/cm^2$ . This range can be converted to the corresponding value in aluminum from the range ratios<sup>14</sup>:

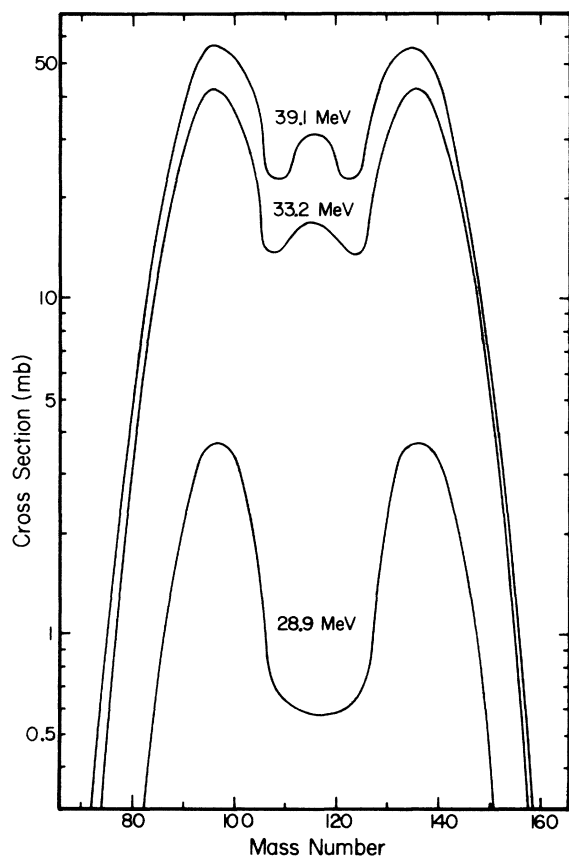


FIG. 1. Binary-fission mass-yield distributions for  $Th^{232}$  induced by 22.9-, 33.2-, and 39.1-MeV  $He^4$  ions (data from Table I).

$$R_{Al}/R_{Th} = (Z_{Th}^{1/3} A_{Al} / Z_{Al}^{1/3} A_{Th})^{0.687}.$$

The range of  $Mg^{28}$  in aluminum so calculated is  $4.4$   $mg/cm^2$ . From the heavy-ion data of Northcliffe, the kinetic energy is then estimated to be  $48 \pm 5$  MeV,<sup>15</sup> similar to that observed previously.<sup>1,2</sup>

The ternary-fission cross section for  $Mg^{28}$  was also measured in the irradiation of  $Th^{232}$  with 36-MeV protons and was found to be  $90 \pm 10$  nb, corrected to  $68 \pm 15$  nb for spallation impurities.

## DISCUSSION

The fission-mass-yield curves constructed from the data are given in Figs. 1 and 2. These illustrate a number of interesting features, not the least of which is resolution of three different types

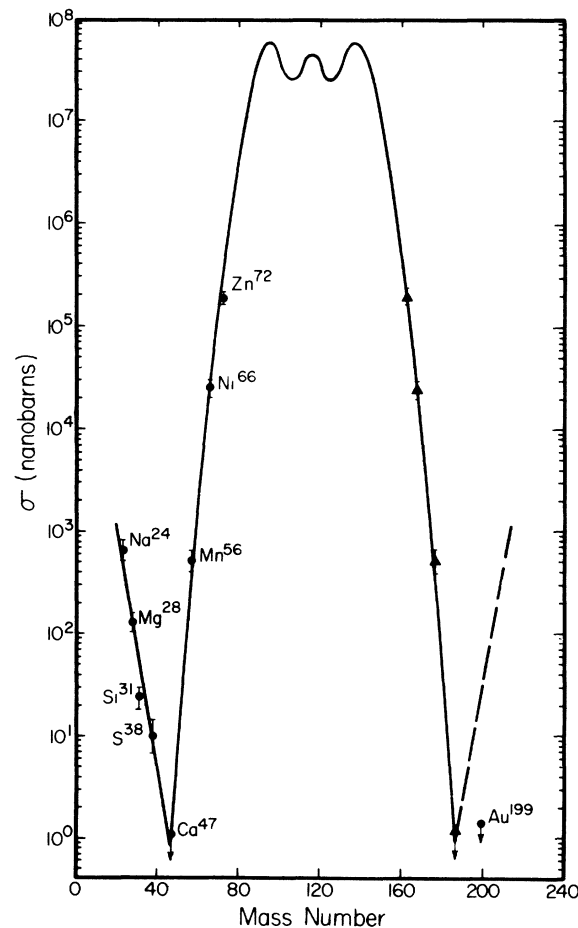


FIG. 2. Mass-yield distribution for the fission of  $Th^{232}$  induced by 41.3-MeV  $He^4$  ions. The dashed line is the expected heavy-mass distribution which would be produced if the light masses resulted from binary fission. The upper part of the mass-yield curve was extrapolated from lower-energy data (Table I). The unlabeled symbols are the reflected points.

of mass distributions resulting from the binary symmetric, binary asymmetric, and ternary asymmetric modes. The resolution of the symmetric binary mode (Fig. 1) was not unexpected, since it has been seen in a few other similar fissioning systems.<sup>16,17</sup> The growing in of the symmetric peak with increasing energy appears reasonable for this fission system. The total binary-fission cross section determined by integration of the mass-yield curves were 65, 810, and 1180 mb at 22.9, 33.2, and 39.1 MeV, respectively.

The light-mass region (Fig. 2) is separated enough from the asymmetric-mass binary region to indicate that ternary fission has been induced in the  $\text{U}^{236*}$  compound nucleus. This is the third ternary-fission system observed in this laboratory and tends to confirm that the phenomenon is a general one.<sup>1-3</sup> The low limit set on  $\text{Au}^{199}$  also confirms the ternary nature of the low-mass region, since the yield of mass 199 resulting from the binary split 37-199 would be expected to be 1 order of magnitude greater (dashed line, Fig. 2).

In this respect, it is of interest to examine further whether  $\text{Au}^{199}$  accurately monitors the total mass-199 chain. Using the CCR procedure,<sup>10</sup> it can be calculated that  $\text{Au}^{199}$  represents >99% of the total mass-199 yield. Alternatively, a modification of the equal charge displacement (ECD) rule can be used to estimate the independent-yield correction.<sup>12</sup> For the compound nucleus  $\text{U}^{236*}$ ,  $Z_P = 46 + \frac{1}{2}(Z_A - Z_{(236-A)})$ , where  $Z_P$  is the most probable charge of a fission fragment,  $Z_A$  and  $Z_{(236-A)}$  are determined by differentiation of the empirical-mass equation to obtain the charge of the fragment of mass  $A$  and  $(236 - A)$  which have the maximum binding energy. It is assumed that there is no significant neutron loss from such a highly deformed compound nucleus, in agreement with previous calculations.<sup>3</sup> The maximum binding-energy assumption for  $Z_A$  is<sup>18</sup>

$$Z_A = \frac{0.585A^{2/3} + 73.18A}{2(0.585A^{2/3} + 72.4)}$$

Using this procedure, the independent-yield corrections for mass 199 not monitored by  $\text{Au}^{199}$  would, again, be <1%.

It is also worthwhile to examine more closely the assumptions that the independent-yield corrections to the ternary species  $\text{Na}^{24}$ ,  $\text{Mg}^{28}$ ,  $\text{Si}^{31}$ ,  $\text{S}^{38}$ , and  $\text{Ca}^{47}$  are small and can be neglected. Using the CCR procedure of Colby and Cobble<sup>10</sup> this as-

sumption is valid; however, there is no evidence that the CCR rule holds for ternary fission. Using the ECD rule, when a light ternary fragment of mass  $A_L$  is formed along with two heavier fragments of masses  $A_B$  and  $A_C$ , the above  $Z_P$  equation becomes  $Z_P = 46 + \frac{1}{2}(Z_L - Z_B - Z_C)$ . If  $A_B$  is varied from 80 to 110, the mass of  $A_C$  is correspondingly fixed for the various ternary fragments being considered. All fragments are assumed to be formed with the maximum binding energy and  $Z_A$  is as given above. Values were computed for  $Z_{PL}$  for ternary fragments of mass  $A_L$  for all allowed sets of  $A_B$  and  $A_C$ . In all cases the independent-yield corrections for the ternary isotopes under consideration were 1%. The simplifying assumption that  $A_B = A_C$  may be made to obtain a value of  $Z_{PL}$ . In this case,  $Z_{PL} = 46 - \frac{1}{2}(Z_A - 2Z_{(236-A)/2})$  and only  $Z_A$  and  $Z_{(236-A)/2}$  need to be calculated.

It should be noted that the yield of  $\text{Na}^{24}$  is considerably larger than that for  $\text{Mg}^{28}$  for the  $\text{U}^{236*}$  compound nucleus. In this respect the present results differ from the other two compound nuclei,  $\text{Pa}^{241*}$  and  $\text{Pu}^{242*}$ , studied previously.<sup>1-3</sup> In these latter two cases,  $\text{Na}^{24}$  and  $\text{Mg}^{28}$  appeared to be near the peak on the ternary-mass-yield curve; there is no indication of such a peak from the present data. At present it is not known whether this is due to the shifting of the peak position or whether there is simply a continuous distribution of ternary fragments from mass 50 up to smaller particles.

Finally, the yield of  $\text{Mg}^{28}$  from 36-MeV protons on  $\text{Th}^{232}$  is lower by a factor of the 2 than the yield from 41-MeV  $\text{He}^4$  ions on  $\text{Th}^{232}$ . This factor is larger than expected, particularly when it is considered that the  $\text{Pa}^{233*}$  compound nucleus is formed with ~5 MeV more excitation energy than  $\text{U}^{236*}$ . The small differences in  $Z^2/A$  are not expected to cause an effect of this magnitude.<sup>19,20</sup> If this observation is correct, it suggests that the small but increased angular momentum imparted by the  $\text{He}^4$  ions *vis à vis* protons in the bombardments has some influence on the probability of inducing ternary fission.

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<sup>6</sup>Analysis carried out by National Spectrographic Laboratories, Inc., Cleveland, Ohio.

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## Study of Charge-Exchange Coupling in Proton-Induced Reactions on <sup>95,98,100</sup>Mo and <sup>92,94</sup>Zr<sup>†</sup>

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The presence of isospin coupling in the incident channel has been studied via proton-induced reactions on <sup>95,98,100</sup>Mo and <sup>92,94</sup>Zr. Anomalous behavior in the excitation functions at backward angles was observed in deuteron and proton outgoing channels. In the deuteron channels these anomalies, located near the (*p,n*) threshold to the ground-state analog, were, for almost all cases, similarly characterized by a double-dipped shape. Their strength is generally much weaker than the single minima observed in (*d,p*) reactions near mass 90 and can be categorized by the proton decay energy from the analog state formed in the charge-exchange process. The proton elastic excitation functions showed no structure near the (*p,n*) thresholds although the (*p,p'*) curves did exhibit fluctuations.

### I. INTRODUCTION

The observation of charge-exchange effects in single-nucleon-transfer reactions has been the subject of considerable study in recent years. Such effects were initially observed by Moore *et al.*<sup>1</sup> as an anomaly (dip or cusp) in the backward-angle <sup>90</sup>Zr(*d,p*)<sup>91</sup>Zr(g.s.) excitation function, centered about the threshold energy of the (*d,n*) channel to the isobaric analog state (IAS) in <sup>91</sup>Nb. Similar isospin coupling effects have been observed in many other nuclei, especially around *A* ≈ 90. The absence of such phenomena in other mass regions, or in transitions to particular states in the mass-90 region, has led to an understanding of the con-

ditions necessary for the observation of such anomalies<sup>2</sup> and the explanation of the effect as due to coupling of the exit channel to the charge-exchange channel.

While the majority of such observations has been restricted to (*d,p*) reactions, charge-exchange effects have also been observed<sup>3,4</sup> in the inverse (*p,d*) reaction. In this case, under the assumption of a charge-exchange process, there is coupling between the proton-plus-target channel and the virtual, neutron-plus-target analog channel. As illustrated in Fig. 1, the neutron in this latter channel can pick up a proton from the IAS and form a deuteron, leaving the residual nucleus in its ground state or in an excited state. The energy at which