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# Interaction of 0.6-300 GeV protons with U, Bi, Au, and Ag; mica track detector study*

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(Abstract)

Mica track detectors were utilized to investigate interactions of 0.6-300 GeV protons with U, Bi, Au, and Ag. Fission cross sections of heavy elements are shown to remain nearly constant as the incident proton energy is increased from 13 to 300 GeV. The values of $\sigma_f$ for U, Bi, Au, and Ag at 300 GeV are 890 ± 130, 181 ± 35, 118 ± 24, and ~1.3 mb, respectively. There is also no substantial change in the frequency of ternary relative to binary fission over this energy range; at 300 GeV it is about 0.4% for U, 1.1% for Bi, and 1.3% for Au. From observation of the forward to backward ratios of single (unpaired) tracks it was concluded that forward momentum transfer to heavy element targets peaks at ~2 GeV and then decreases at higher beam energies.

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[NUCLEAR REACTIONS U(p,f), Bi(p,f), Au(p,f), Ag(p,f); binary and ternary fission; E = 0.6-300 GeV; measured σ(E). Mica track detectors.]

## Introduction

The interaction between high-energy protons and complex nuclei has been studied intensively[1-6] at incident energies up to 29 GeV. It was shown[7] that product yield distributions are essentially energy independent between 10 and 29 GeV. When 300-GeV proton beams became available at the Fermi National Accelerator Laboratory (FNAL) several investigations with various target nuclei were initiated to test if this energy independence of product cross sections continues into this new energy range.[8-12] In all of these experiments formation cross sections of individual radioactive nuclides were measured. For light and medium mass target elements, where the spallation mechanism predominates, product cross sections were found[8,9] not to vary appreciably with proton energy between 11.5 GeV and 300 GeV. However, with U and Au targets small changes were observed[10-12] in the product cross sections, in the recoil ranges, and in the forward to backward ratios.

From heavy element targets, fission as well as spallation contributes to the product distribution, and yield measurements of individual radioactive product nuclides cannot always identify the specific formation mechanisms which lead to their formation. In the work reported here $4\pi$ mica track detectors were used to record all events in which energetic fragments ($Z \geq 15$ and K.E. $\gtrsim$ 10 MeV) are produced.[2] Thin targets of U, Bi, Au, and Ag sandwiched between sheets of mica were irradiated with high-energy protons up to 300 GeV and fission into two or more energetic heavy fragments was observed directly. Cross sections were determined for binary and ternary fission and also for production of single energetic recoil fragments. Some information was obtained on momentum transfer by study of forward to backward

ratios. Scanning of mica detectors from previous work[2] with protons of 0.6-29 GeV was repeated to ensure that the criteria for identifying ternary, binary, and unpaired events were identical over the whole energy range up to 300 GeV.

## Experimental Procedure and Results

Three stacks of mica sheets containing thin evaporated layers of target elements were irradiated at normal incidence with one or two pulses of 300-GeV protons at FNAL. One stack consisted of 5 pieces of annealed and pre-etched mica, 50 x 20 x 0.1 mm, vacuum sealed between two sheets of 0.1 mm Mylar.[13] The second and third stacks were similar except that they contained, in addition, 5 pieces of 0.08 mm Lexan plastic.[14] The thicknesses of the targets, prepared by evaporation, were chosen to obtain convenient track densities from all of the targets when irradiated with the same beam: U, 8.7 $\mu g/cm^2$; Bi, 52 $\mu g/cm^2$; Au, 55 $\mu g/cm^2$; Ag, 147 $\mu g/cm^2$. An aluminum foil, 21.4 $mg/cm^2$ thick, was attached to each stack to serve as a monitor for the proton beam by means of the $^{27}Al(p,3pn)^{24}Na$ reaction. The total thickness in each irradiation was about 200-250 $mg/cm^2$ (mica, plastic, targets, and Al) and therefore the effect of secondary particles was small.[2,10] The integrated beam in each irradiation was $\sim 3 \times 10^{10}$ protons and it was confined to an area of about 6 x 2 mm. Determinations of the beam intensities were made by measuring the $^{24}Na$ produced in the aluminum foils with a calibrated end-window proportional counter. The cross section values used for the $^{27}Al(p,3pn)^{24}Na$ reaction at 13, 29, and 300 GeV were 8.3, 8.0, and 8.0 mb, respectively, and they are based on recent measurements.[15] For lower energies the cross sections were taken from the review by Cumming.[16]

After the irradiations 1-mm diameter alignment holes were drilled through the corners of the target stacks. The target layer was carefully dissolved off each mica and its thickness determined by chemical analysis. The micas were etched in 48% HF for 30 min to 1.5 hr at 23°C. Pairs of adjacent pieces were realigned, mounted on lucite microscope slides, and scanned at a magnification of 150X for binary and ternary events and for single tracks with projected length $\gtrsim$ 4 μm. This minimum length was chosen because it serves as a convenient threshold for scanning; possible singles with projected length < 4 μ are much less clearly defined. Detection efficiencies, determined by duplicate scanning, were 95-99% for fission events and 80-90% for single unpaired tracks.

In the work performed previously[2] in this laboratory to study fission induced by 0.6-29-GeV protons a photographic process was used to facilitate scanning. Enlarged images of the original etched tracks were reproduced on high contrast film and scanning was performed on a light box with paired sheets of film. This method was tested[2] by measurements of the thermal neutron fission cross section of uranium. Excellent agreement was obtained with the accepted value, 4.18 ± 0.03 b, but (5±2)% of the fissions registered as single tracks rather than as pairs, probably because of decreased registration efficiency for fission fragments entering the mica at grazing incidence and possible self absorption in the target layer. In the present investigation scanning was performed directly under the microscope and this method was also shown to register nearly every thermal fission as a binary event. Test targets of U varying in thickness from 2.8 to 257 μg/cm$^2$ were sandwiched in mica, vacuum sealed in Mylar, and irradiated in the thermal column of the Brookhaven Medical Reactor. The

neutron fluxes were determined by activation of small gold foils ($^{197}Au(n,\gamma)^{198}Au$, $\sigma = 98.8 \times 10^{-24}$ cm$^2$) and the 412-keV $\gamma$ rays emitted by $^{198}Au$ (0.968 per disintegration) were measured with a calibrated Ge(Li) detector. After the irradiations the target stacks were processed and scanned as described above. About 1200 events were counted in each case. The results are given in Table I. The fission cross sections are based on the sum of the correlated pairs of tracks and the single tracks. Again, agreement with the accepted value for the natural U fission cross section is excellent. The unpaired tracks amount to (2.4 ± 1.3)% of the events, independent of target thickness (2.8-257 μg/cm$^2$); in (0.4-1.9)% of the cases both fission fragments record in a single mica because of scattering.

These test results clearly demonstrate that those fragments of mass and energy which result from thermal neutron fission are registered by both scanning techniques with efficiencies approaching 100%. From high-energy fission the distributions in fragment mass and kinetic energy are considerably broader and thus relatively more lightly-etched tracks, of low optical contrast, were produced in the mica detectors. It was found that some of these tracks were not reproduced on the photographic film. Such tracks must be from fragments whose mass and/or energy are outside the region of fragments produced in thermal fission. Thus the recording threshold was lowered when direct microscope observation was used instead of the photographic method. A substantial portion of the etched mica from each of the earlier runs (0.6-29 GeV) was re-scanned and thus the detection threshold is the same as for the 300-GeV run. Differences between results given below and those reported earlier[2]

are due mainly to the lower threshold. In general, the cross sections are higher, even for the unpaired single tracks.

Binary fission cross sections, $\sigma_f$, for U, Bi, and Au are shown in Table II for the energy range 0.6-300 GeV. About 500-2000 fission events were recorded for each case; the quoted uncertainties include statistical factors and possible errors of identification. For uranium, $\sigma_f$ decreases monotonically from $\sim$1700 mb at 0.6 GeV to 860 mb at 13 GeV and then it appears to remain constant up to 300 GeV. For bismuth, $\sigma_f$ decreases from 299 mb at 0.6 GeV to an asymptotic value of $\sim$ 180 mb at 13 GeV. In the case of gold the fission cross section reaches a constant value of 100-110 mb at 2.0 GeV; it appears to be increasing slightly between 0.6 and 2.0 GeV. Within the experimental errors, $\sigma_f$ for each of the three targets at 300 GeV is the same as at 13-29 GeV. The radio-activation work[10,12] on U has revealed decreases in cross sections of $\sim$20% for certain specific nuclides in the mass region 70-140 when the beam energy was changed from 11.5 to 300 GeV. However these decreases may be for the fractions of those products which are produced by non-fission mechanisms. There is evidence from the unpaired track data given below to support this hypothesis.

Comparison of the lower energy values in Table II with those published in Reference 2 shows that the fission cross sections are now generally larger by 1/3. This means that with the present experimental method more highly asymmetric splits are included among the fission events. The cross sections measured by Brandt et al.[17] in their mica track detector study of fission induced by 0.59-23-GeV protons are generally about 19% lower than those reported here. Rémy et al.[18] used polycarbonate plastic track detectors to study fission of U and Pb induced by 0.6-23-GeV protons.

Their values for binary fission of U are in good agreement with the results of the present study (except at 0.6 GeV where their number is 29% lower).

The results on ternary fission are shown in Table III where data have been combined, in most cases, from runs of two adjacent energies in order to improve statistical accuracy. Each ratio of ternary-to-binary fission, $\sigma_t/\sigma_f$ in percent, is based on 1000-2000 binary events. The ratios for Au and Bi targets are about the same, and they increase from $\sim$0.3% at 0.6-1.0 GeV to $\sim$1.2% at 300 GeV; however, within the assigned errors the ratios are constant between 13 and 300 GeV. For U targets $\sigma_t/\sigma_f$ shows no clear trend with beam energy; above 2 GeV it varies between $\sim$0.2% and $\sim$0.7%. The fact that the present values are about 5-fold higher than those measured by the earlier method[2] strongly suggests that in most ternary fission events at least one of the fragments has a low mass or a low energy near the recording threshold, and thus break-up into three heavy fragments of approximately equal mass is very rare even in high-energy proton interactions. Brandt et al.[19] measured ratios of ternary to binary cross sections for U at 18 GeV and at 23 GeV. Their values of $\sim$0.2% and $\sim$0.5%, respectively, are in reasonable agreement with the results reported here.

Cross sections $\sigma_s$ for production of unpaired tracks, with no apparent partner, are given in Table IV. These single tracks are attributed mainly to very energetic spallation residues. Since the 4 μm projected length cut-off is not accurately determined and because there is some variation in the sensitivity of the mica, the results shown are approximate. The single track cross sections increase with energy and become almost as large as the binary fission cross sections

the F/B ratios near 3-GeV incident proton energy were first found[5] for certain neutron-deficient medium mass nuclides ($^{83}Sr$, $^{131}Ba$) produced from U targets. It was later shown[11,20] that lighter products such as Na, Mg, and Sc isotopes from U and Au also exhibit maximum F/B ratios near 2-3 GeV. The present nuclear track data show that this phenomenon is quite general and that forward momentum transfer from the incident proton decreases very substantially above 2-3 GeV.

All of the experimental results on the interaction of 0.6-300 GeV protons with Ag are summarized in Table VII. Identification of these events from Ag is considerably less certain than identification of the heavy element events, and the statistical uncertainties are greater. Therefore the apparent fluctuations in $\sigma_f$ and $\sigma_s$ shown in the Table may not be real. We can state, however, that with incident 1.0-300 GeV protons the binary fission cross sections are 3±2 mb and that the fission fragments are peaked strongly forward. Thus the fission cross section from Ag is about 30-fold smaller than it is from Au, and the momentum transfer in fission events is considerably greater. In the earlier work[2] upper limits had been set on $\sigma_f$: < 0.3 mb at 0.6 GeV; < 2 mb at 1.0 GeV; < 5 mb at 2, 3, and 13 GeV; < 6 mb at 29 GeV. No ternary events were seen from Ag. The F/B ratios of the unpaired tracks show very strong forward peaking at 0.6 GeV which decreases with increasing beam energy until 13 GeV when F/B becomes $\approx 1.4$.

## Conclusions

Within experimental error, the binary and ternary fission cross sections of U, Bi, and Au were shown to be constant in the energy range 13-300 GeV. Fractions of the total reaction cross sections[21,22] which

result in fission are: U, 0.45; Bi, 0.10; Au, 0.06; Ag, $\sim$ 0.003. No events were seen at any beam energy in which there was break-up into more than 3 heavy fragments. In non-fission events, the magnitude of forward momentum transfer to heavy energetic fragments from U, Bi, and Au targets is greatest at 2 GeV and then it decreases with increasing energy.

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Table I. $UF_4$ targets sandwiched between pieces of mica irradiated with thermal neutrons: percent of single uncorrelated tracks, percent of events in which both fission tracks record in the same mica, and fission cross section measured for natural uranium.

| U Target thickness ($\mu g/cm^2$) | Single Tracks | Paired tracks in same mica | Fission cross section (barns) |
|---|---|---|---|
| 2.80 | 2.1% | 0.6% | 4.05 ± 0.25 |
| 9.7 | 2.7 | 0.4 | 4.07 ± 0.25 |
| 27.9 | 1.3 | 0.6 | 4.26 ± 0.25 |
| 65.0 | 1.1 | 1.9 | 4.17 ± 0.25 |
| 257 | 4.6 | 1.5 | 4.10 ± 0.25 |

Table II. Binary fission cross sections $\sigma_f$ in mb

| Target | $E_p$ (GeV) | | | | | | |
|---|---|---|---|---|---|---|---|
| | 0.6 | 1.0 | 2.0 | 3.0 | 13 | 29 | 300 |
| U | 1700 ± 250 | 1530 ± 230 | 1520 ± 230 | 1210 ± 180 | 860 ± 130 | 830 ± 120 | 890 ± 130 |
| Bi | 299 ± 60 | 262 ± 50 | 255 ± 40 | 213 ± 40 | 188 ± 40 | 162 ± 30 | 181 ± 35 |
| Au | 76 ± 15 | 94 ± 19 | 112 ± 22 | 103 ± 21 | 117 ± 23 | 89 ± 18 | 118 ± 24 |

Table III. Ratio of ternary to binary fission, $\sigma_t/\sigma_f$, in percent. The data were combined from two adjacent beam energies so that each ratio is based on 1000-2000 binary events.

| Target | $E_p$ (GeV) | | | |
|---|---|---|---|---|
| | 0.6 and 1.0 | 2.0 and 3.0 | 13 and 29 | 300 |
| U | <0.10 ± 0.10 | 0.66 ± 0.25 | 0.22 ± 0.11 | 0.40 ± 0.16 |
| Bi | 0.33 ± 0.17 | 0.62 ± 0.22 | 0.82 ± 0.26 | 1.15 ± 0.24 |
| Au | 0.32 ± 0.18 | 0.61 ± 0.23 | 1.02 ± 0.29 | 1.31 ± 0.35 |

Table IV. Approximate cross sections, $\sigma_s$, for production of single (unpaired) tracks; ratios, $\sigma_s/\sigma_f$; and total observed cross sections, $\sigma_s + \sigma_f$.

| Target | | $E_p$ (GeV) 0.6 | 1.0 | 2.0 | 3.0 | 13 | 29 | 300 |
|---|---|---|---|---|---|---|---|---|
| U | $\sigma_s/\sigma_f$ | 0.09 | 0.19 | 0.35 | 0.62 | 0.80 | 0.75 | 0.28 |
| | $\sigma_s$ (mb) | 150 | 290 | 530 | 750 | 690 | 620 | 250 |
| | $\sigma_f + \sigma_s$ (mb) | 1850 | 1820 | 2050 | 1960 | 1550 | 1450 | 1140 |
| Bi | $\sigma_s/\sigma_f$ | 0.27 | | 0.27 | 0.80 | 1.6 | 1.5 | 0.88 |
| | $\sigma_s$ (mb) | 80 | | 70 | 170 | 295 | 250 | 160 |
| | $\sigma_f + \sigma_s$ | 379 | | 325 | 383 | 483 | 412 | 341 |
| Au | $\sigma_s/\sigma_f$ | 0.53 | 0.48 | 0.63 | 0.91 | 2.9 | 1.8 | 2.0 |
| | $\sigma_s$ (mb) | 40 | 45 | 70 | 94 | 340 | 160 | 240 |
| | $\sigma_f + \sigma_s$ (mb) | 116 | 139 | 182 | 197 | 457 | 249 | 358 |

Table V. Fractional excess of fissions with both fragments in forward hemisphere. Experimental uncertainties are ±30% (see text). Data at 300 GeV not directly comparable with those at 0.6–29 GeV.

| Target | $E_p$ (GeV) | | | | | | |
|---|---|---|---|---|---|---|---|
| | 0.6 | 1.0 | 2.0 | 3.0 | 13 | 29 | 300 |
| U | 0.048 | 0.045 | 0.058 | 0.069 | 0.038 | 0.054 | (0.025) |
| Bi | 0.059 | 0.070 | 0.100 | 0.109 | 0.084 | 0.119 | (0.028) |
| Au | 0.103 | 0.074 | 0.096 | 0.114 | 0.090 | 0.102 | (0.079) |

Table VI. Ratios of unpaired tracks forward to unpaired tracks backward (F/B ratios). Experimental uncertainties are 15-20%.

| Target | $E_p$ (GeV) | | | | | | |
|---|---|---|---|---|---|---|---|
| | 0.6 | 1.0 | 2.0 | 3.0 | 13 | 29 | 300 |
| U | 0.96 | 0.90 | 1.35 | 0.81 | 0.95 | 1.20 | 0.86 |
| Bi | 0.90 | 1.88 | 1.67 | 1.25 | 1.16 | 1.16 | 1.05 |
| Au | 1.29 | 2.37 | 3.04 | 0.9 | 1.42 | 1.07 | 0.93 |

Table VII. Interaction of 0.6-300 GeV protons with Ag; recorded in mica track detectors.

| Proton Energy (GeV) | Number of Binary Events | Binary Fission, $\sigma_f$ (mb) | Binaries Forward (net fraction) | Single Tracks, $\sigma_s$ (mb) | Single Tracks (F/B) |
|---|---|---|---|---|---|
| 0.6 | 11 | 0.6 | 0.36 | 5 | 20 |
| 1.0 | 11 | 1.0 | 0.36 | 13 | 3.6 |
| 2.0 | 10 | 4.8 | 0.50 | 58 | 5.1 |
| 3.0 | 8 | 3.5 | 0.25 | 53 | 3.2 |
| 13 | 14 | 4.9 | 0.36 | 41 | 1.4 |
| 29 | 20 | 3.2 | 0.39 | 120 | 1.3 |
| 300 | 27 | 1.3 | 0.26 | 21 | 1.4 |