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Charge and Mass Identification
of Fission Fragments

by
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Thesis Director's signature:

[Signature]

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TABLE OF CONTENTS

I. Introduction 1

II. Background Information
   A. Spectrometer Requirements 4
   B. X-ray Detection as a Z Identification Method 8
   C. Inner-shell Ionization of Heavy Ions 17

III. Experiments
   A. X-ray Production by Accelerated Heavy Ions
      Stopping in a Silicon Surface-barrier Detector 35
   B. X-ray Production by Fission Fragments
      Passing Through Foils of CH₂, Ni, and Au 58
   C. X-ray Production by Fission Fragments Stopping
      in a Silicon Surface-barrier Detector 84

IV. Evaluation
   A. Further Development 99
   B. Time-of-Flight Mass Spectroscopy for Heavy Ions
      in the Fission Fragment Mass and Energy Range 103
   C. Conclusions 115

V. Appendices
   A. Study of Heavy Ion Beam Composition from a
      Tandem Van de Graaff Accelerator Utilizing
      a Gas Stripper A-1
   B. Gain Stabilization of Silicon Surface-barrier
      Detectors A-29
C. Elimination of Californium-252 Migration A-36
D. Pulse Height Resolution of Solid-state Particle Detectors for Heavy Ions A-44

Bibliography
I. Introduction

A major obstacle in understanding fission is that no one has ever been able to measure the charge \( Z \) and mass \( M \) of each pair of fission fragments. The purpose of this thesis is to evaluate the use of characteristic x-ray detection combined with energy and time-of-flight measurements for the simultaneous determination of \( Z \) and \( M \).

First, the efficiency and resolution requirements for a general heavy ion spectrometer system will be discussed. It will be seen that: 1) Any type of particle identification technique must be of high efficiency to be of practical use; 2) A good criterion for acceptable resolution (80\% probability for correct identification) is when the full width at half maximum (FWHM) of the identified line is equal to the separation from its adjacent lines.

Next, use of characteristic x-rays for \( Z \) identification is discussed. The important ideas covered are: 1) The need for high system efficiency requires that a Si(Li) or Ge(Li) x-ray detector be used; 2) When such a detector is used to identify one characteristic L x-ray, the probability for correct \( Z \) identification is poor due to line multiplicity and overlap; 3) When such a detector is used to identify one characteristic K x-ray, the
probability for correct Z identification is generally much improved but widely varying with x-ray energy since line overlap is still present; 4) If sources of x-ray background are significant, they may reduce the probability for correct Z identification. This is particularly true in fission where huge gamma-ray cascades are emitted in coincidence with the nuclear event; 5) There is a high probability that heavy nuclei (especially fission fragments) deexcite via K shell internal conversion. Use of the subsequent K x-rays for Z identification will result in an efficiency that is dependent on the nuclear process, and hence ambiguous; 6) X-ray production by impact ionization will have an efficiency dependent only on particle energy if the nuclear excitation is first allowed to relax over a long flight path (typically 1 meter for fission fragments).

The experimental section proceeds to investigate x-ray production when fission fragments and similar heavy ions slow down in a silicon surface-barrier detector and in different materials. Indications are that: 1) The nuclear deexcitation K x-ray yield for fission fragments might be reduced to negligible levels for flight paths greater than 1 meter; 2) The K x-ray yield from the silicon detector is much too low (0.018 K x-rays per fragment) to be of practical use; 3) The behavior of the L x-ray yield in different materials indicates that a
higher K x-ray yield might be obtained if a stopping material of higher Z, such as Ge, is used.

The conclusion is that it may be possible to fabricate a Z spectrometer that utilizes a Ge particle detector for x-ray production and a cooled Ge crystal for x-ray detection with an efficiency on the order of 18% for the K x-rays of fission fragments. Since the x-ray background depends on the nuclear process and detector arrangement, no conclusion is made about the probability for correct Z identification.

Finally, it was determined that, due to nuclear straggling, a solid-state particle detector cannot be used as the energy measurement in a fission fragment time-of-flight spectrometer of 1 amu (FWHM) resolution. Therefore the proposed system must either have poor mass resolution or low efficiency. For this reason it is believed that a greater potential for development lies in a dE-E-TOF measurement utilizing a multiwire light gas ion chamber to obtain a dE-E energy loss curve.
II. Background Information

A. Spectrometer Requirements

Experimental studies of nuclear reactions involving heavy ions are hampered by some intrinsic limitations that are not always encountered in light ion reactions. The requirements for spectrometer efficiency and resolution are consequently more stringent.

1. Efficiency of Detection

In order to resolve closely lying states, line broadening due to kinematics and target thickness must be considered. In heavy ion reactions, the variation of energy of the reaction product with scattering angle is very large, requiring a very small solid angle (typically 0.1 millisteradians) subtended by the detector. The energy lost by the beam and reaction product in the finite thickness of the target increases rapidly with the atomic number of the ion involved. The use of very thin targets is required to minimize energy resolution deterioration due to energy loss straggling. The high energy loss in the target also limits the maximum useable heavy ion beam intensity to values that will not destroy the target.

Furthermore, the cross section for a particular heavy ion reaction is typically 10 to 100 times smaller
than those for typical light ion reactions due to the
increased coulomb barrier and the fact that the total
cross section must be distributed over a larger number
of outgoing channels.

As a consequence of the above, even a well-designed
heavy ion experiment must run for a very long time to
accumulate sufficient statistics. Hence a heavy ion
identification technique must generally be of high
efficiency to be of practical use.

2. Particle Identification Resolution

The large number of outgoing states typical of the
heavy ion reaction requires that efficient particle
identification be done. This means that the identification
technique must be of high resolution, as well as of high
efficiency.

Suppose the spectrum of measurements of discrete
quantities M (such as masses) obtained from a spectrometer
system is made up of a series of adjacent gaussian
distributions of roughly equal size. Window limits can
be placed at the minima between the peaks (Figure 1).

The probability for correctly identifying an event
falling in window M is just the area under the gaussian
inside the window for M. Since the distance between
adjacent peaks is 1 unit, the windows are 1 unit wide.
Resolution (FWHM) vs. probability for correct identification
Figure 1

Spectrometer window limits for adjacent gaussian distributions of roughly equal size
Figure 2

Probability of correct identification for an event falling in the spectrometer window of Figure 1 as a function of spectrometer resolution
Probability for Correct Identification (%) vs. Resolution (FWHM) in units of adjacent peak separation.
can be calculated using tables [BE 69]. This curve is shown in Figure 2. A good criterion for acceptable resolution is that 80% probability of correct identification is achieved at FWHM = 1.0 unit. Note that little is to be gained below FWHM = 0.5 units.

B. X-ray Detection as a Z Identification Method

The development of high efficiency Si and Ge detectors for photon spectroscopy capable of resolving Kα x-ray lines down to Z = 11 has led to many applications of such detectors as atomic number spectrometers. The principle involved is that the x-ray energy is characteristic of the atom it originates from.

In reality, an x-ray spectrum contains x-rays originating from many different sources. For spectra containing high statistics the contribution from each source may be separated by fitting with elemental x-ray signatures. However when using characteristic x-ray detection for identifying the Z of a nuclear reaction product, one generally has to deal with a relatively small number of events due to the low nuclear cross sections and the fact that the x-ray production efficiency, at least for fission fragments, is considerably less than one per particle. Due to the possibility of background contributions and characteristic x-ray line multiplicity and overlap, it is difficult to precisely
define Z resolution. It is more significant to think in terms of the probability of correct identification of one x-ray.

Consider the situation of observing one particular x-ray. Since it may have originated from different sources, the most information that can be inferred is that the event has a probability \( P_x \) for having originated at source \( x \).

1. Contributing Processes

One can list the major sources that contribute to x-ray production in the x-ray energy region of interest. In the system described one expects to see (Figure 3) fragments coincident with:

1) Characteristic fragment lines from:
   a) Interaction with the stopping medium;
   b) In-flight internal conversion deexcitation;
   c) In-flight nuclear (gamma-ray) deexcitation;

2) Background from:
   a) Compton scattering of fission gammas;
   b) Fission neutron decay and activation;
   c) Fragment bremsstrahlung in stopping medium;
   d) X- and gamma-rays Compton scattered from stopping medium.

Unless all such contributions are negligible compared to 1a), the probability for correct Z identification
Some photon-particle coincidence processes

(1a) Characteristic x-ray of particle produced when particle interacts with the stopping medium

(1b) Characteristic x-ray of particle due to in-flight internal conversion of nuclear excitation

(1c) Characteristic gamma-ray of particle due to in-flight nuclear deexcitation

(2a) Background due to Compton scattering of x- and gamma-rays from support material

(2b) Background from neutron decay and activation of support material, and (n, α) reactions in the x-ray detector

(2c) Background due to beta particle and delta-ray bremsstrahlung in stopping medium

(2d) Background from x- and gamma-rays Compton scattered from stopping medium
will be too low, even for K lines, for the spectrometer to be practical. Sources 2c) and 2d) represent inseparable sources of background but were observed not to dominate the spectra in the experiments of section III. Sources 2a) and 2b) can be reduced by improved shielding and moving the spectrometer farther from the fission event. The contributions from sources 1b) and 1c) can be reduced by increasing fragment flight times. The experiments of section III will show that for fission fragments a flight path of one meter is sufficient to accomplish this.

2. X-ray Line Overlap

Characteristic x-ray line overlap resulting from the multiplicity of x-ray lines and detector energy resolution will affect the ability of the spectrometer to correctly identify one x-ray. Line overlap and detector energy resolution will define the best Z identification performance of any particular x-ray detector. X-ray energies and transition probabilities (line intensities) have been observed to vary strongly with particle energy and Z in heavy ion-atom collisions but very few actual values have been measured. Reality may be approximated by employing the fluorescent x-ray energies and radiative transition probabilities used in a similar study [FI 71].
Assume that the number of particles entering the spectrometer and their x-ray production probabilities are such that an equal number of characteristic x-rays are produced for each atomic number in a restricted $Z$ range (e.g., for fission fragments). Assume also the contribution from all background sources is negligible.

The probability $P_j$ that one x-ray originates from element $Z_j$ is just $1/N_Z$, where $N_Z$ is the number of elements considered, since each is assumed to have equal probability for producing a characteristic x-ray. For each element $Z_j$ this probability is spread over x-ray energy ($E_x$) according to the relative transition probabilities for the characteristic lines involved.

The characteristic line $L_{ij}$ from element $Z_j$ can be represented by a gaussian distribution in energy:

$$A_{ij} e^{-\frac{1}{2} \left( \frac{E_x - E_{ij}}{\sigma_{ij}} \right)^2}$$

where $A_{ij}$ is an amplitude proportional to the relative intensity of line $L_{ij}$ of element $Z_j$, $E_{ij}$ is the energy of the line, and $\sigma_{ij}$ is the detector resolution (FWHM/2.355) at $E_{ij}$. The probability distribution in $E_x$ is a sum over all lines of $Z_j$ with the requirement that the total probability be $1/N_Z$:

$$P_j(E_x) = \sum_i A_{ij} e^{-\frac{1}{2} \left( \frac{E_x - E_{ij}}{\sigma_{ij}} \right)^2}$$
and
\[
\frac{1}{N_x} = \int p_j(E_x) dE_x = \sum \frac{A_{i,j} \sigma_{i,j}}{\sqrt{2\pi}}.
\]

The energy dependence of the Si(Li) x-ray detector resolution is determined from $FWHM^2 = STAT^2 + NOISE^2$, where $STAT^2 = (2.355)^2 \varepsilon FE$, $\varepsilon$ is the energy required to produce an electron-hole pair in silicon (3.8 eV), $F$ is the Fano factor (0.13 in silicon), and $E$ is the x-ray energy.

Using these relations and an optimistic estimate of $FWHM = 170$ eV @ 6.4 keV (NOISE = 107 eV), a map of the probability that a detected x-ray of energy is a characteristic line from element $Z$ was computed. One finds that at very few x-ray energies is there greater than 90% certainty of positive $Z$ identification. The $K\alpha$-$K\beta$ overlap is large for $Z+2$ in the range $Z = 33$ to 41 and $Z+3$ for $Z = 48$ to 56, which unfortunately just happen to be the regions of the two fission peaks.

The ability of the spectrometer to identify a single x-ray can be estimated by calculating the spread of $Z$ in the above map as a function of $E_x$ (Figure 4). It must be pointed out here that most of the x-rays produced in the stopping medium are L x-rays, while the K x-ray yield is prohibitively lower. This is unfortunate since, as can be seen from Figure 4, the $Z$ spread for L x-rays
Figure 4

Width (FWHM) of x-ray occurrence probability map (described in text) in the Z parameter as a function of Ex.

The sharp variations reflect the overlap of x-ray lines. Weighting by the total occurrence probability for a given Ex only serves to accentuate these variations.

(a)  K lines

(b)  L lines
is large. The Z spread for K x-rays is on the average less, but widely varying. This is because at many points the Kα line from one Z is large but overlaps strongly with a Kγ line from another Z.

Recapitulating, the above evaluation of the line overlap situation is contingent on the following assumptions: 1) Equal probability of a characteristic x-ray being detected from each Z in a restricted range of Z; 2) Line ratios same for all Z and independent of particle energy; 3) Line energies independent of Z and particle energy; 4) All background sources can be reduced to negligible importance; 5) No interference from any characteristic lines in the stopping medium. In reality the situation is much more complex, since none of assumptions 1)-3) hold for heavy ion x-ray production.

Much uncertainty and many calculational difficulties in data analysis can be bypassed by calibrating the system efficiency with heavy ion accelerator beams. One would have to measure the x-ray yield per particle

\[ Y_x \left( E_x, Z_j, E_{Z_j} \right) \]

which is the x-ray energy spectrum produced by atom \( Z_j \) of energy \( E_{Z_j} \) entering the spectrometer. Knowledge of these yields would then determine the probability that an arbitrary heavy ion producing a spectrometer
response be of atomic number $Z_j$. However, the large exposures necessary to calibrate the efficiency would destroy the particle detector. Since most of the x-ray production occurs near the surface, the efficiency might be strongly dependent on surface effects, particularly if the particles channel in the detector crystal. So the x-ray production efficiency measured for one particle detector is not necessarily applicable to another detector. The sensitivity of the x-ray production efficiency (reproducibility) to surface (orientation) effects and charge state of the incident ion are unknown and from here on will be ignored.

C. Inner-shell Ionization of Heavy Ions

The inner-shell ionization resulting from photon or light particle (e, p, α) excitation is simply proportional to the number of atoms present, for a particular excitation method. X-ray production following nuclear processes is not so easily described. Quite often a product is produced in a nuclear excited state. Nuclear deexcitation may often proceed in such a way as to produce an inner-shell electronic vacancy, with subsequent x-ray production. This is particularly true for heavier ions that have many closely lying nuclear levels. The importance of such processes may vary strongly depending on nuclear parameters that are
particular to the reaction being studied. Thus the probability for characteristic x-ray emission by a nuclear reaction product has some quantitative uncertainty.

When identifying particles it is desirable for the spectrometer efficiency to be unambiguous. One possibility is to remove the uncertainty by producing inner-shell vacancies after the nuclear and atomic structure has decayed to a stable configuration. For fission fragment energies, the probability of inner-shell vacancy production by impact ionization is dependent only on the identity of the collision partners and their relative velocity. By using this method, the process being studied and the detection method are "decoupled". One may thus avoid the complex problem of having to know the particulars of the process before studying it. The following three subsections digress into details of the inner-shell ionization processes and may be bypassed without losing continuity.

1. Internal Processes of X-ray Production in Nuclear Decay

A radioactive nuclear species can be thought of as a complex system that is in a long-lived excited state. The system will eventually spontaneously pass to a more stable configuration by one of many allowed channels. Some of these channels involve interactions with the atomic electron cloud due to finite overlap of the
electron wavefunction with the nucleus. As a consequence, inner-shell vacancies can be produced. Once an inner-shell vacancy is produced, the atom decays via the x-ray/Auger electron competition. The probability of x-ray emission from a particular inner-shell vacancy is the fluorescent yield, shown in Figure 5.

Common channels of nuclear deexcitation are: 1) Gamma-ray emission/internal conversion; 2) Positron emission/electron capture; 3) Negatron emission; 4) Alpha particle emission; 5) Neutron emission; and 6) Fission. These may occur in combinations. The first two of these are commonly the most important for producing inner-shell vacancies.

Gamma-ray emission competes with internal conversion, the ejection of a bound electron from the electron cloud. Conversion in the K shell is in general more likely than in other shells because of the greater probability of K electrons being found in the nucleus. Conversion is more probable for higher Z because the K shell radius varies as 1/Z so the "effective volume" occupied by the K shell electrons varies as 1/Z^3 and nuclear overlap increases. Furthermore, low transition energies favor internal conversion. Heavier nuclides tend to have many closely lying states, so internal conversion is an important deexcitation process in these atoms. High
Figure 5

K and L shell fluorescent yields [BE 70]
Figure 6

K shell internal conversion coefficients as a function of gamma-ray energy [DY 73]

(a) $Z = 30$

(b) $Z = 78$
Figure 7

Ratio of electron capture to positron emission as a function of the maximum energy of the positron spectrum
multipolarity transitions have low probability of radiating electromagnetic energy, and in such cases the relative probability of internal conversion is high. The relative probability of internal conversion for different atomic shells also depends strongly on the character (electric or magnetic) and multipolarity of the transition, a fact that is exploited in nuclear spectroscopy for determining the type of transition. Figure 6 illustrates the behavior of some K shell internal conversion coefficients. The internal conversion coefficient is defined as the ratio of the probability of ejecting an electron from a particular inner shell to the probability for emission of a gamma-ray for the same nuclear transition.

Nuclei on the neutron deficient side of the line of beta stability tend to decay by the competing processes of positron emission and electron capture. Electron capture is strongly favored at high Z because: 1) The effective volume occupied by the K shell electrons decreases and nuclear overlap increases; 2) The nuclear coulomb barrier becomes increasingly difficult for a positron to penetrate. Electron capture is also favored for low transition energies for the same reason [DY 73]. This behavior is shown by the ratio of electron capture to positron emission, illustrated in Figure 7.

Both of the above are single step processes and are correspondingly the most probable. "Secondary" processes
involving direct interactions (scattering) between a charged particle or photon escaping the nucleus and the atomic electrons occur much less frequently. This is surprising since every such particle must pass through the electron cloud in order to leave the atom. One would expect the probability of interaction to be higher than that for externally incident particles whose distance of closest approach to the nucleus is on the average much greater. In a recent review, Freedman [FR 74] asserts that "no current evidence firmly indicates direct interaction of real nuclear radiation with the atomic structure of the parent atom...". The reason for this is not clear, but it may be due to the way the radiation passes through the electron cloud. One expects the cross section for inner-shell ionization to be largest when the average distance of closest approach of the ionizing projectile ($\rho$) is near the K shell mean radius: the overlap of the projectile and electron wavefunctions should then be largest for the longest time (Figure 8). This at least is compatible with the observed cross sectional behavior. If on the other hand the inner-shell ionization is to be done by a projectile leaving the nucleus the average overlap is expected to be small.

Other atomic interactions in nuclear decay that have been observed but are generally much weaker than the single step processes are: 1) Internal bremsstrahlung
Figure 8

Illustrating average projectile-target electronic overlap as a function of average distance of closest approach $\rho$

(a) $E_p$ low, $\rho \gg$ shell radius, average overlap small, ionization cross section small

(b) $E_p$ larger, $\rho \approx$ shell radius, average overlap very large, ionization cross section maximum

(c) $E_p$ very high, $\rho \ll$ shell radius, average overlap less, ionization cross section decreasing
resulting from acceleration of a beta particle in the coulomb field of the nucleus or, in the case of electron capture, the sudden change in dipole moment of the electron/nucleus system; 2) "Shake-up" excitation and "shake-off" ionization are the reactions of the electron cloud to the relatively sudden change in its internal electrostatic environment as charged particles are emitted or relocated within the atom during the nuclear process. The nuclear process occurs much faster than the electron motion, so transfer of energy is possible via the potential energy change of the electron cloud. Shake-up involves excitation of bound electrons to unoccupied bound orbitals; or to the free electron state in shake-off. At all Z, 20 to 30% of the atoms subject to beta emission undergo shaking. Most of this is confined to the outer (valence) shells except in the lightest atoms. The probability for K electron shake-off per beta decay is [FR 74]

\[ P_K = \frac{a}{Z^2}, \quad 0.6 < a < 1.6 \]

The shake-off probability for the K and L shells for heavy ions such as fission fragments is small, on the order of a few percent.

Weaker processes which have been observed but are of negligible importance here are: 1) Internal pair production; and 2) Ionization by nuclear recoil. This
is important only for light alpha particle emitters (of which there are few) and for fission.

2. Internal X-ray Production in the Fission Process

The fission process [BO 39] may be represented as shown in Figure 9. Two neutron rich, excited fragments of the compound nucleus separate in a time short compared with the radiative lifetime of the compound state (less than $10^{-14}$ sec). At the instant of separation, "prompt" neutrons are emitted, mainly from the moving fragments. Further energy is released by the emission of "prompt" gamma-radiation from excited levels with lifetimes of the order $10^{-14}$ sec. The fragments are still neutron rich and continue to decay via beta, gamma, and delayed neutron emission.

Vacancies in the K shell due to disruption of the electron cloud during scission or coulomb ionization during the stopping of the fragments are not expected, since the fragment velocities are much less than the velocities of the K electrons [VA 73]. The enhanced ionization yield that is observed during fragment stopping is understood to be due to electron promotion; however this yield is still small relative to the amount of fragment deexcitation for times less than $10^{-8}$ sec after scission.

Almost all the x-rays emitted in fission resulting
Figure 9

The fission process [BO 39]

(a) Formation of compound nucleus \( t = 0 \)
(b) Scission: Prompt neutron emission
(c) Prompt gamma-ray emission \( t = 10^{-14} \) to \( 10^{-9} \) sec
(d) Beta, gamma, and delayed neutron emission \( t > 10^{-9} \) sec
(a) \( t = 0 \)

(b) \( t = 10^{-14} \)

(c) \( t = 10^{-14} \) to \( 10^{-9} \) (sec)

(d) \( t > 10^{-9} \) (sec)
from nuclear deexcitation are produced by internal conversion, with times of emission on the order of $10^{-10}$ to $10^{-9}$ sec. The fragments, especially the heavy ones around mass numbers $A = 140$ to $150$, are characterized by many low energy rotational levels. Heavy primary fragments emit a cascade of several low energy, high multiplicity gamma-rays. The probability for deexcitation of these levels by internal conversion is high. The yield of x-rays from the even $Z$ elements is low, because of proton pairing in the nucleus which produce first excited states far above the ground state. High energy gammas from such transitions have a low internal conversion rate and hence a low x-ray yield. On the other hand odd $Z$ elements have many low energy levels and a correspondingly high x-ray yield. This even-odd effect disappears around $Z = 58$, near the onset of stable nuclear deformation as the internal conversion x-ray yield increases for both kinds of nuclei, reflecting more and more the effects of nuclear shape rather than pairing energy. In general, prompt x-rays (faster than 1 nsec) show more of these effects than do the delayed (1 µsec) ones [RE 71].

The K x-ray yields in fission are therefore strongly dependent on the nuclear structure, and this feature has been exploited to extract information from the fission products soon after production of the fragments. The energies of the K x-rays identify the $Z$ of the fragments,
and pre-neutron emission fragment masses can be determined through conservation of mass and linear momentum if the fragment energies are known. The strong dependency of the internal conversion probability on type of gamma-ray has yielded general information on fragment structure and excitation even for fragments far from the line of beta stability which could not be measured using normal chemical means. However, the very fact that there is a strong (and not very well known) dependence on fragment charge and mass has been a limitation of identifying fragment Z with K x-rays since it can bias the results. Also, the observed yields are really integrated quantities for gamma-ray cascades averaged over several isotopes. For this reason it is difficult to make unambiguous conclusions about most of the nuclear species involved except the ones which are well studied.

Some difficulties that are inherent in fission-photon coincidence experiments are: 1) A high x-ray background in true coincidence, e.g., from (n, γ) and (γ, γ') reactions in the support structure; and 2) The extremely large number of outgoing channels for fission, many of which are very weak. To be statistically conclusive, studies involving the weaker channels require that a very large total number of events be collected. This is seldom achieved. Some fission fragment x-ray yields measured in other studies are shown in Figure 10.
Figure 10

Fission x-ray yields

(a) K x-rays emitted within 1 nsec after fission as a function of fragment post-neutron emission mass, for $^{233}\text{U}(n,f)$, $^{235}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$ and $^{252}\text{Cf}(sf)$ [RE 71]

(b) K x-rays per fragment for $^{252}\text{Cf}(sf)$ as a function of fragment atomic number:
   1) Within 1 nsec after fission
   2) Within 1 µsec after fission [RE 71]

(c) K x-rays per fragment for $^{236}\text{U}(n,f)$ within 1 µsec after fission as a function of fragment atomic number [KA 71A]
3. X-ray Production in Heavy Ion-Atom Collisions

X-ray production by light ions (p, d, α) is well understood and is successfully described theoretically by the perturbation caused by an incident point charge (coulomb ionization). However a direct interaction model is increasingly less satisfactory for lighter targets and at low ion energies. For heavier ion-atom collisions, the x-ray production cross sections observed are orders of magnitude higher than those predicted by coulomb ionization. The mechanism causing inner-shell ionization for collision velocities smaller than the orbital velocities of the electrons concerned is mainly electron promotion in the unbound (quasi-molecular) atom-atom system formed during the collision. The term "quasi-molecule" indicates that the internuclear separation is changing slowly enough for the electrons to adjust to the new levels in the atom-atom system.

The cross-sections at low energies are nonmonotonic with target atomic number but approach monotonicity with increasing energy. This has been interpreted in terms of increased probability of electron promotion resulting when the binding energy of the electron to be excited matches the binding energy of an electron of the collision partner [FA 65, BA 72, GA 73]. Figure 11 [KU 73] shows the bromine x-ray production cross section for bromine incident on various targets as a function of atomic
Figure 11

Br K\textsubscript{α} excitation cross section for Br beam as a function of target atomic number for beam energies 45, 60, 85, and 110 MeV [KU 73]
number for energies in the fission fragment region. The first peak occurs when K shells are matched in target and projectile, and the second peak occurs when the bromine K shell matches the target L shell. It is not clear where the molecular orbital picture of the interaction breaks down and direct coulomb excitation takes over, but electron promotion is expected to be the predominant mechanism for inner shell ionization as long as the projectile velocity is less than the velocity of the bound electron to be ejected.

Other processes in heavy ion-atom collisions affecting x-ray production are: 1) Multiple inner shell ionization; 2) Charge state of the incident ion; 3) X-ray bands characteristic of molecular orbital x-rays due to the possible relaxations of preexisting vacancies during a close encounter; 4) Electron capture at high ion velocities with x-ray emission dependent on velocity, and initial and final electron bound states; and 5) High energy bremsstrahlung from initially bound target electrons in the coulomb field of the projectile.
III. Experiments

A. X-ray Production by Accelerated Heavy Ions

Stopping in a Silicon Surface-barrier Detector

1. Introduction

Heavy ion beams from the Rice Tandem Van de Graaff accelerator were used to produce x-rays when the heavy ions were stopped in a silicon surface-barrier detector. X-rays from the particle detector were observed with a Si(Li) detector at right angles to the incident particle direction. X-ray-particle coincidence yields were obtained as a function of particle type and energy. Cl K x-ray yields and Br and I L x-ray yields were measured. Upper limits were determined for Br and I K x-ray yields. The Si K x-ray was observed for all beams.

2. Experimental

a. Chamber, Detectors

Since placing the surface-barrier detector directly in the beam line was not advisable, it was decided to scatter the beam at forward angles from a thin foil. The amount of the heavier beams reaching the target gave prohibitively small count rates at the minimum angle allowed by the ports of the available scattering chamber. The gas stripper of the accelerator produced large numbers of the lower energy, lower charge states only. Since
the x-ray cross section was expected to drop off rapidly below 1 MeV/amu, it was important to lose as little of the beam energy as possible in the scattering process in order to keep the x-ray yield up to measureable levels. The elastic scattering cross section becomes large at small forward angles, and the energy of the scattered particle at forward angles is a large fraction of the incident energy. Both the elastic cross section and the energy of the scattered ion increase with target mass, so it was decided to scatter the heavy ions at small forward angles from a thin gold foil.

The arrangement used to accomplish this is shown in Figure 12. The body of the target holder was positioned so that the beam spot on the thin gold scattering foil was near the scattering arm's rotational center. The direct beam was stopped on the side of the collimator holder when the axis of the scattering arm was moved away from 0°. The angle of the scattering arm was kept in the range 3° to 14° so that the direct beam would strike on the side of the collimator holder. This reduced background from the beam hitting the scattering arm wall upstream of these collimators. The pump holes in the collimator holder were positioned on the side of the scattering arm furthest from the primary beam axis for the same reason.

The scattered beam then passed into the detector
arrangement through a 6.35 mm diameter carbon collimator, Figure 13. This chamber was constructed to provide as large an x-ray solid angle as possible. The particle detector (Ortec 5-836D, 400 mm² x 40 μm, 40 μg/cm² gold surface electrode, 300 ohm-cm resistivity) could be rotated about a vertical axis from 0° to 90° to the incident particle direction while maintaining the front surface of the detector centered in the chamber. In the experiment an angle of 45° ± 5° was used. The x-rays produced passed through an uncollimated 25.4 μm mylar vacuum window to the Si(Li) detector, external to the vacuum. The Ortec 7116 Si(Li) x-ray detector had a 25.4 μm Be entrance window and a 30 mm² x 3.49 mm crystal; resolution was 190 eV FWHM at the 5.895 keV line of $^{55}$Fe.

The vacuum in the system was maintained by a 5 cm diameter oil diffusion pump in front of the detection chamber, aided by a 7.5 cm diameter pump about 2 meters upstream. Pressure at the 7.5 cm pump was about 2 x 10⁻⁵ Torr.

b. Electronics

The electronics used to accumulate the data are shown schematically in Figure 14. The large delay required for the linear signals was necessary to compensate for the delay resulting from crossover timing on the slow bipolar output of the x-ray spectroscopy amplifier.
Figure 12

Small angle scattering apparatus (not to scale)

C1 = 3.18 mm diameter tantalum collimator
C2 = 3.97 mm diameter tantalum collimator
F = 145 µg/cm² gold scattering foil
A = moveable scattering arm
D = detector assembly
HI = heavy ion beam from accelerator
Figure 13

Detector assembly closeup

C3  =  6.35 mm diameter x 2 mm graphite collimator
Ep  =  particle detector
M   =  25.4 µm mylar vacuum window
A   =  2.4 mm air gap
Be  =  25.4 µm Be detector window
Ex  =  Si(Li) x-ray detector crystal
C   =  cryostat
X   =  x-ray
HI  =  heavy ion
Br  =  brass
Al  =  aluminum
Lu  =  lucite
Figure 14

Electronics schematic

Ex = Si(Li) x-ray detector
P = preamplifier
HV = detector bias supply
S = spectroscopy amplifier
TSCA = timing single channel analyzer
CTSCA = constant fraction TSCA
TAC = time to pulse height converter
DA = delay-line amplifier
Ep = silicon surface-barrier particle detector
PS = preamplifier power supply and amplifier
PL = pulser
Pa = pulser signal attenuated x10
Pd = direct pulser signal
GDG = gate and delay generator
C = overlap coincidence
D = linear delay
ADC = analog to digital converter
BI = bipolar
This delay was eliminated in later work by timing on the Si(Li) preamp output, which also improved time resolution by a factor of 10 without noticeably degrading the energy resolution. The best time resolution obtained with timing on the slow bipolar crossover was about 70 nsec FWHM for fixed amplitude pulser signals. The TAC produced an output for events falling within ± 500 nsec of the pulser coincidence peak. The random rate was practically zero, so the poor timing did not influence the results. The slow coincidence output was used to gate the ADC's for x-ray energy (Ex), particle energy (Ep), and TAC output (TAC). The 3-parameter events were streamed to magnetic tape for off-line analysis. Separate ADC's were used to accumulate free spectra for both energy parameters as monitors.

c. Beam Production

Negative ion beams of O, Cl, Br, and I were produced by direct extraction from the duoplasmatron ion source and injected into the tandem. There was some confusion in identifying the beams passing the accelerator's 90° analyzing magnet, since these were observed to have much lower energy than expected for the same charge states leaving the accelerator.

To resolve this uncertainty, the nature of the beam was studied by scattering from a foil before the 90°
magnet. Details of this study are contained in Appendix 1. It was discovered that it is the residual stripper gas in the high energy acceleration tube and between the tandem and the 90° magnet that strips lower charge states to higher states, enabling them to pass the 90° magnet. Only for Cl and O were the beams sufficiently well defined to allow the terminal voltage regulation system to operate. A very broad distribution of energies is produced when the heavier ions Br and I change charge during acceleration in the high energy tube. Under these conditions, roughly equal amounts of several charge states were able to pass the 90° magnet and no terminal voltage regulation was achieved. Though it was possible to selectively tune one or a few of these charge states through the system, this was not done. The 90° magnet apertures defined the energies of these beams regardless of the terminal voltage so several different energy data points were obtained in each run. The multienergetic nature of the beam is apparent in the typical particle spectra of Figure 15.

3. Procedure
a. Angular Scale

The angle of the scattering arm was determined by a scale on a horizontal support bar arm and a mark on the arm indicating its axis. The scale was calibrated
Figure 15

Typical particle monitor spectra

Vertical scale for each plot is logarithmic from $10^0$ to $10^5$ counts per channel. Horizontal scale is the channel number. The peak label in parentheses is the charge state of the particle as it passes the 90° magnet. The other label is the calculated particle energy in MeV.
\[ \text{OH}^- \text{ injected} \]
\[ \text{TV} = 3.4, 5 \text{ MV} \]
\[ \text{NMR } f = 29.4958 \text{ MHz} \]

\[ \text{Cl}^- \text{ injected} \]
\[ \text{TV} = 5 \text{ MV} \]
\[ \text{NMR } f = 32.9377 \text{ MHz} \]

\[ \text{Cl}^- \text{ injected} \]
\[ \text{TV} = 5 \text{ MV} \]
\[ \text{NMR } f = 37.9123 \text{ MHz} \]

\[ \text{Br}^- \text{ injected} \]
\[ \text{TV} = 3 \text{ MV} \]
\[ \text{NMR } f = 44.6844 \text{ MHz} \]
Br$^-$ injected
TV = 4 MV
NMR $f = 44.6844$ MHz

Br$^-$ injected
TV = 4.5 MV
NMR $f = 44.6844$ MHz

I$^-$ injected
TV = 3 MV
NMR $f = 45.9108$ MHz

I$^-$ injected
TV = unknown
NMR $f = 44.6844$ MHz

Log$_{10}$ (counts per channel) vs. Channel Number
geometrically with knowledge of the approximate position of the beam axis. Angular resolution on this scale was 0.6°/cm. The angle used for most of the data was 6.6°. The angular aperture of the detection system was 0.27° (17 microsteradians).

b. Target Thickness

The Au scattering foil thickness was measured to be 145 ± 10 μg/cm² by absorption of the 5.895 keV x-ray line of $^{55}$Fe. This value was used despite the fact that thickness measurements by x-ray absorption are notorious for giving low average thickness values for non-uniform foils.

c. Energy Calibration

Calibration of the x-ray monitor energy axis was done with $^{57}$Co and $^{55}$Fe radioactive sources. The x-ray calibration remained stable throughout the run except when the gain of the x-ray amplifier was deliberately changed.

The particle monitor energy axis was roughly calibrated using the known energy of the oxygen beam. The kinematics of the different charge states of the heavier beams were determined by the 90° magnet NMR frequency and the Rutherford formula for elastic scattering from gold. Correction for energy loss in the gold foil was then done using tables [NO 70]. Comparison to the
approximate energies of the spectrum peaks determined by the oxygen beam calibration identified the charge state and energy of each peak. The peak centroids and their corresponding calculated energies were then fit with a linear least squares program to determine the calibration gain and baseline. According to these calibrations, the detector gain slowly decreased during successive runs. This gain loss was mainly due to decreasing bias on the particle detector as the leakage current increased with radiation damage. The gain shift between successive runs was not large enough to confuse the identification of the peaks, and therefore did not alter the results.

The relations between the monitor and multiparameter ADC's for both energy parameters were determined from a set of pulser runs taken during the experiment. From these the multiparameter ADC calibrations were determined.

4. Data Analysis

The number of characteristic \( x \)-rays of the incident particle escaping the particle detector per incident particle is

\[
N_x(E_x, E_p) = \frac{N_c(E_x, E_p)}{N_p(E_p) \cdot L \cdot E_d(E_x) \cdot T(E_x) \cdot \left( \frac{N_x}{4 \pi} \right)}
\]

where \( N_c(E_x, E_p) \) is the number of x-ray/particle
coincidences observed having the incident particle's characteristic x-ray energy $E_x$ for particles of energy $E_p$; $N_\rho(E_p)$ is the number of counts in the particle monitor peak at $E_p$; $L$ is the ratio of the particle monitor ADC live time to the multiparameter ADC live time; $E_d(E_x)$ is the x-ray detector efficiency at $E_x$; $T(E_x)$ is the x-ray vacuum window transmission at $E_x$; and $\Omega_x$ is the average solid angle in steradians subtended by the x-ray detector for a particle incident on the surface-barrier detector face.

a. Particle Monitor

The computer program used to calculate peak centroids in the particle monitor was also used to integrate each peak to ± 2 standard deviations with no background subtraction. The error in this procedure (about 2%) was in all cases negligible relative to the statistical error in the total number of coincidence events accumulated for each data point.

b. Data Sorting

The limits used in the particle monitor peak integration determined the multiparameter sorting windows for the particle energy.

The x-ray monitor spectra failed to have enough statistics to show any peaks, so it was necessary to construct a simulated x-ray spectrum using the x-ray
source calibration. This was done by guessing the relative line intensities from a preliminary inspection of the data, and using these, the energies of the expected lines and their known relative intensities, and the detector resolution, to build a sum of gaussian distributions [FI 71]. The x-ray channel limits for sorting were then determined from this spectrum by dividing it into different regions of characteristic lines, as shown in Figure 16. The validity of this procedure is questionable, since there is evidence to indicate significant shifts in x-ray energies and intensities involving excitation by different heavy ions [BU 70]. Background sources, such as secondary electron bremsstrahlung, were not considered. Nevertheless, the low number of counts, even when all the spectra in the run were summed, prohibited identification of any x-ray peaks other than the Si and Cl K lines, so this method of x-ray identification was a reasonable approach.

Coincidence events were then sorted according to the windows defined by the procedure described above. No TAC window was applied since the accidental background appeared to be negligible when a TAC projection of each run was made. The TAC peak was very wide (70 nsec FWHM for the pulser with Ex = 4 to 8 v and Ef = .5 to 1 v, both ADC's being 8 v full scale). The primary effect of the particle detector bias drift was to change the
Figure 16

Simulated x-ray spectrum showing choice of sorting windows for the x-ray parameter

<table>
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<tr>
<th>Label</th>
<th>(Energy limits)</th>
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<tr>
<td>Br L</td>
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<tr>
<td>Si K</td>
<td>(1.63- 1.94 keV)</td>
</tr>
<tr>
<td>Au M</td>
<td>(1.94- 2.38 keV)</td>
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<tr>
<td>Cl K</td>
<td>(2.38- 3.01 keV)</td>
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<td>I L</td>
<td>(3.31- 5.32 keV)</td>
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<tr>
<td>Br K</td>
<td>(11.70-12.24 keV)</td>
</tr>
<tr>
<td>I K</td>
<td>(overflows)</td>
</tr>
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</table>

Also shown is the calculated x-ray detection efficiency for 25.4 μm mylar, 25.4 μm Be and a 3.49 mm Si crystal. The contribution of the 2.4 mm air gap was neglected. Mass absorption coefficients from [BE 70] were interpolated using the formula $\mu = CE_x^n$. 
rise time of the signal, so care was taken to see that the limits of the TAC range did not exclude any real events. The sorted x-ray events and corresponding particle monitors are presented in Table 1.

The difference between monitor and coincidence ADC live times was in all cases less than 1%. For simplicity their ratio was assumed to be 1.

c. X-ray Detection Efficiency

The efficiency for x-ray detection was calculated for a 25.4 μm mylar vacuum window, a 25.4 μm Be window, and a 3.49 mm Si crystal. The calculations assumed simple exponential absorption by uniform materials. The actual values used for each x-ray line are shown in Figure 16. The value used for the x-ray solid angle was \( 17.0 \pm 0.9 \) millisteradians.

5. Results

Table 2 contains the calculated yield values in x-rays per particle corrected for detector efficiency and solid angle. Only points containing reasonable statistics are included. These results for characteristic x-rays of the incident particle and for the characteristic x-rays of the silicon detector are depicted as excitation functions in Figure 17. The error bars represent \( \pm 1 \) standard deviation in the number of detected events, which in all cases dominated the error. The upper limits
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Ep = Calculated energy of detected particle in MeV

Np = Number of detected particles (units of $10^3$)

BrL...BrK = Number of x-ray - particle coincidences detected in x-ray window indicated
TABLE 2 - X-ray Yields per Particle

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<th><em><strong>ClK</strong></em></th>
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</table>

(a) X-rays per particle emitted @ 45° from a silicon surface-barrier detector for particles incident @ 45°

(b) Standard deviation in (a)

(c) Values of (a) and (b) are in units of $10^{-3}$ x-rays per fragment

(d) Upper limits for Br and I K lines as defined in text, in units of $10^{-3}$ x-rays per fragment
Figure 17

X-ray yields

(a) Heavy ion characteristic x-rays emitted from a silicon surface-barrier detector @ 45° for heavy ions incident @ 45°

(b) Silicon characteristic x-rays emitted from a silicon surface-barrier detector @ 45° for heavy ions incident @ 45°

Error bars represent 1 statistical standard deviation.
in Table 2 for Br and I K x-ray yields are estimates based on one count as the maximum yield where no counts were observed.

In Figure 17(b), it is notable that for all particle energies, the Si K x-ray yield for Cl lies below that for Br and above that for I. This observation can be explained in terms of electron promotion between closely lying levels in the quasi-molecule formed by the two heavy ions during the collision. Note from Figure 16 that the Si K shell is energetically closer to the Br L shell than to the Cl K shell, and closer to the Cl K shell than to the I L shell. In molecular orbital theory, closely matched energy levels in the separated atom system will form energetically close levels in the molecular system. At some point in the internuclear separation the levels may become degenerate. There is a high probability for electron transfer ("promotion") between energetically close levels. Generally one of the levels represents electronic motion localized on one of the atoms and the other level, localization on the other atom. Since electrons may find their way from one atom to another via transfer from one level to the other, it is possible that an extra electron may be trapped on one of the atoms when the energy barrier between the levels increases as the atoms move apart. One can thus expect the probability for ionization to follow two
general trends: 1) It should increase as level matching in the two collision partners improves, allowing the transfer of electrons from one atom to another to occur; and 2) It should increase the faster the electron "exchange portal" shuts; so as long as the portal is open long enough for electron transfer to occur (roughly the electron velocity in the shell involved divided by the radius of the shell), we would expect the ionization probability to increase with relative energy. This is consistent with what was observed.

The data point for O beam in Figure 17(b) must be explained by the fact that the yield was not corrected for secondary electron bremsstrahlung (from bound electrons). For O beam at these energies, the yield of this radiation in the Si K region is known to be very high.

In conclusion, the L shell yields for typical heavy ions at subfission energies are high enough to be used for Z identification. The yield for K lines was immeasureably lower, the upper limits set at $4 \times 10^{-3}$ K x-rays per particle for 39 MeV Br, and $10^{-2}$ K x-rays per particle for 46 MeV I. This suggests that use of the well resolved K lines in a Z spectrometer using a Si matrix for x-ray production would be very inefficient. Since the ionization cross section is increasing with particle energy, the possibility exists that the yields
are much higher for real fission fragments, since the fragment energies are higher than those used here. It is also possible that use of an x-ray production medium which is, on the average, a better electronic level match for heavy ions, will increase the x-ray yield. These possibilities are investigated in the following experiments.
B. X-ray Production by Fission Fragments Passing Through Foils of CH₂, Ni, and Au

1. Introduction

In order to study how x-ray production varies with target atomic number, fission fragments from a $^{252}$Cf source were passed through thin foils of polyethylene (CH₂), Ni, and Au. The average x-ray production cross section for fragments in each of the foil materials was measured by detecting x-rays in coincidence with the fragments. The results showed that the fragment characteristic x-ray yield drops rapidly for lower Z stopping media, which explains why the characteristic K x-ray yield in the previous experiment was immeasurably low. The average L x-ray yields were highest in the Ni foil: 0.39 L x-rays per light fragment and 0.18 L x-rays per heavy fragment. The average K x-ray yields were less than 0.01 per fragment.

2. Apparatus

a. Chamber, Detectors, Source

Fission fragments from an open $^{252}$Cf source were allowed to pass through thin foils in the configuration shown in Figure 18. The distance from the source to the foil was 5.1 ± 0.2 cm. The particles passing the foil were detected by a surface barrier detector (Ortec 6-132D, 400 mm² x 50 µm, resistivity 425 ohm-cm, FWHM 144
Figure 18

Experimental Configuration

Ep = silicon surface-barrier particle detector
Ex = Si(Li) x-ray detector crystal
C = cryostat
Be = 25.4 μm Be detector entrance window
M = 3.175 μm mylar vacuum window
F = x-ray production foil
S = $^{252}$Cf open source
A = air gap
FF = fission fragment
X = x-ray
Br = brass
Al = aluminum
Lu = lucite
Figure 19

X-ray efficiency calculated for a 3.175 μm mylar vacuum window, a 25.4 μm Be detector entrance window, and a 3.49 mm Si crystal
3.175 μm mylar
+25.4 μm Be
+3.49 mm Si
keV @ the 6.117 MeV alpha-particle of $^{252}$Cf) located 3.7 cm downstream of the foil center. The liquid-nitrogen cooled Si(Li) x-ray detector (Ortec 3-576, 30 mm$^2$ x 3.49 mm, 25.4 μm Be window, FWHM 200 eV @ 5.895 keV, 370 eV @ 22.105 keV) observed x-rays from the region of the foil through a 3.175 μm mylar vacuum window. Figure 19 shows the x-ray detection efficiency calculated for this configuration. The Si(Li) crystal was located 4.0 ± 0.1 cm from the center of the foil. No limiting collimation was used for either detector, but care was taken to assure that any particle reaching the surface-barrier detector would pass through the foil at a position that could be seen by the x-ray detector, unobstructed by any of the support structure. X-rays produced in the particle detector itself could not reach the Si(Li) directly. Despite this, it is obvious from the free spectra that some Au L x-rays from the particle detector window electrode did reach the x-ray detector. A lead shadow was placed between the californium source and the Si(Li) crystal, reducing the number of x-ray background gamma-rays by roughly a factor of 2. The chamber was carefully sealed from light to reduce particle detector leakage current variations. The geometry was kept fixed for each foil.
b. Electronics

Fast timing between the x-ray energy (Ex) and the particle energy (Ef) pulses was achieved by generating delay-line shaped bipolar pulses directly from the preamplifier signals and taking time markers from the crossover points which then drove the TAC, Figure 20. The FWHM of the TAC peak was typically 16 nsec for x-ray energies between 15.6 and 41.5 keV (K lines) and fragments of 30 to 127 MeV. The time resolution was dominated by the relatively high noise on the x-ray signal, due to the low energy of the x-rays being detected (less than 40 keV). Since the TAC produced output pulses for each start pulse, a slow coincidence (1 μsec overlap) was required between the two time markers and the TAC output. The coincidence output was then used to gate the computer which stored three-parameter events (Ex, Ef, TAC) on magnetic tape. Spectra for Ex and Ef were accumulated simultaneously as monitors by gating separate ADC's with each of the time markers. ADC live times under each gate were recorded by gated clocks internal to the computer. Care was taken to terminate long cables in order to suppress oscillations, especially on the TSCA's. The slow timing was carefully tested to insure that no pulses were lost or distorted either by walk out of the coincidence circuit or the ADC acceptance window. System operation was periodically checked with the pulser.
Figure 20

Electronics schematic

Ex = Si(Li) x-ray detector
Ep = silicon surface-barrier particle detector
P = preamplifier
PS = preamplifier power supply and amplifier
HV = detector bias supply
DA = delay-line amplifier
TSCA = timing single channel analyzer
CTSC = constant fraction TSCA
TAC = time to pulse height converter
S = spectroscopy amplifier
PL = pulser
GDG = gate and delay generator
C = 1 μsec overlap coincidence
D = linear delay
ADC = analog to digital converter
Pa = attenuated pulser signal
Pd = direct pulser signal
BI = bipolar
3. Procedure

a. Foil Thickness Measurement

Foil thicknesses were measured in situ by measuring the energy loss of the 6.117 MeV $^{252}$Cf alpha particle with the foil in position and the foil out. The broadening of the peak was used to estimate the foil uniformity by the following method. Approximately,

$$W_1^2 = W_2^2 + W_3^2 + W_4^2$$

where $W_1$ is the width of the alpha peak with the foil in; $W_2$, with the foil out; $W_3$, the broadening due to foil thickness nonuniformity; and $W_4$, the broadening due to straggling. $W_3$ was observed to be much greater than $W_4$ in all cases, so $W_4$ was neglected. Hence

$$W_3^2 \approx W_1^2 - W_2^2$$

It must be pointed out that this thickness error represents not only the actual foil nonuniformity but also the geometric thickness variation due to the large solid angle subtended by the particle detector. Results of these estimations are summarized in Table 3.

b. Energy Calibrations

The x-ray energy was linearly calibrated by placing x-ray sources of $^{57}$Co, $^{55}$Fe, and $^{109}$Cd in the chamber
TABLE 3 - Foil Thicknesses

<table>
<thead>
<tr>
<th>FOIL</th>
<th>X</th>
<th>DX</th>
<th>A</th>
<th>P</th>
<th>dEL</th>
<th>dEH</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH₂</td>
<td>100</td>
<td>63</td>
<td>4.7</td>
<td>13.</td>
<td>11.0</td>
<td>9.8</td>
</tr>
<tr>
<td>Ni</td>
<td>820</td>
<td>94</td>
<td>58.7</td>
<td>8.4</td>
<td>22.9</td>
<td>18.9</td>
</tr>
<tr>
<td>Au</td>
<td>720</td>
<td>91</td>
<td>197.</td>
<td>2.2</td>
<td>11.4</td>
<td>10.0</td>
</tr>
</tbody>
</table>

X = Foil thickness in μg/cm²

DX = Standard deviation in X due to nonuniformities

A = Average atomic mass in g/mole

P = Foil thickness in units of $10^{18}$ atoms/cm²

dEL = Average energy loss of light fragments in foil in MeV

dEH = Average energy loss of heavy fragments in foil in MeV
### TABLE 4 - Run Durations

<table>
<thead>
<tr>
<th>RUN</th>
<th>$L_c$</th>
<th>$L_f$</th>
<th>$Y_f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{CH}_2$</td>
<td>.356</td>
<td>.307</td>
<td>49.5</td>
</tr>
<tr>
<td>Ni</td>
<td>.356</td>
<td>.307</td>
<td>49.6</td>
</tr>
<tr>
<td>Au</td>
<td>.364</td>
<td>.313</td>
<td>50.7</td>
</tr>
<tr>
<td>Open</td>
<td>.382</td>
<td>.328</td>
<td>53.1</td>
</tr>
</tbody>
</table>

$L_c$ = Live time of multiparameter ADC (units of $10^6$ sec)

$L_f$ = Live time of fragment monitor ADC (units of $10^6$ sec)

$Y_f$ = Total number of fragment monitor counts for 45 to 127 MeV initial fragment energy (units of $10^6$ counts)
center and accumulating free spectra. The fragment energy axis was linearly calibrated with the free spectrum of the $^{252}$Cf fission fragments with no foil in the chamber, assuming the peak centroids to have energies 79 and 104 MeV for the low and high energy peaks, respectively (see Appendix C). A series of pulser runs yielded linear relations between the monitor and multiparameter ADC's.

The x-ray calibration was stable to within the channel width of the ADC over the duration of the measurements. The gain on the fragment detector tended to drift due to small temperature fluctuations which caused large variations in the detector reverse leakage current. This problem was compensated for by periodically adjusting the detector bias supply as described in Appendix B.

The TAC was linearly calibrated with the pulser by varying the precision delays in the CFTSCA's. All ADC's were used on the 1024 channel scale.

c. Background Measurements

1) Random

The background due to random x-ray - fragment coincidences was observed for all data to be less than 1% of the real events, so no accidental subtraction was done.
2) In-flight Fragment Deexcitation

A large number of the events were observed to be due to in-flight deexcitation of the fragment nuclei. To estimate this contribution, a "background" run was made with only an empty foil holder; i.e., with only the foil itself missing. The validity of subtracting this run directly from the others is questionable, because the presence of the foil distorts this background: 1) Fragments losing energy in the foil slow down and spend more time in the field of view of the x-ray detector, increasing the chance of observation of an in-flight deexcitation event; 2) Fragments deexciting before reaching the foil will have some of the resulting x-rays absorbed in the foil, reducing the probability for observing a deexcitation event; 3) Compton scattering of fission gamma-rays from the foil will appear as coincidence events, and the relative intensity of these will depend on the Z and thickness of the foil. It is hoped that these processes have a small effect on the "background" yields. No correction for these effects was attempted since there were too many unknowns involved.

3) Californium Migration

Migration of the $^{252}$Cf into the chamber produces the problem of an increased yield of in-flight deexcitations since more fragments are observed earlier in the fragment
flight path. This is a time-dependent source of undesirable interference. Following the experiment, the source was removed. Short runs with the empty foil holder in and with each foil in place showed that most of the contamination was localized near the position of the source. There was little additional buildup on the foils and detector. The contribution to the coincidence data was in all cases at most 10%. A simple attempt to correct the data by assuming a linear buildup with exposure showed no major spectral feature changes, and only served to degrade the statistical quality of the spectrum due to the poorer statistics of the short background runs. For simplicity, the effect of migration is ignored in the following analysis.

4. Data Presentation and Analysis
a. Particle Monitor

The monitor spectra were summed over all runs for each foil, shown in Figure 21. The resolution loss due to bias fluctuations and electronic drift was comparable to the inherent detector resolution, which is about 1 MeV FWHM for fission fragments (Appendix D). This was much less than the broadening due to foil thickness nonuniformities.
Figure 21

Monitor spectra

Horizontal axis is channel number (energy). Vertical axis is counts per channel.

(a) Particle monitors: The strong low energy peaks are the 6.117 MeV alpha and its pileup peaks.

(b) X-ray monitors: Some of the peaks have been identified and labeled with element and line symbols. Many of these x-rays are generated by the $^{252}$Cf alpha particles.
b. Sorting Procedure

1) Ex-TAC Windows

The multiparameter events were sorted and displayed as pulse height vs. TAC. The sorting program allowed the application of two-dimensional windows for real events. This allows one to take advantage of the good timing obtained with large Ex pulse heights. The majority of random events were due to the high alpha particle count rate. Figure 22 shows the Ex-TAC display for fission fragments in Ef. The accidental background is very low, with the region of early events due to late start (Ex) pulses, mainly internal conversion x-rays and gamma-rays from delayed in-flight deexcitation by the fragments. A real window only was placed on the Ex-TAC display as shown in Figure 22. For each data set, the time position of the real window relative to the real TAC peak was maintained constant in order to treat all true time-delayed effects equivalently. The data was then again sorted requiring events to lie within the Ex-TAC real window and displayed as Ex vs. Ef, Figure 23.

2) Ex-Ef Windows

Windows were then placed on the Ex-Ef data of Figure 23 to define the K and L x-ray regions for fragments and exclude characteristic foil lines where possible. Using the detector calibrations described above, the
Figure 22

Ex-TAC displays showing the two-dimensional windows for identifying real events.

The area darkened in each cell is linearly proportional to the number of counts in the cell. The projections on each axis consist only of the events lying within the two-dimensional envelope shown. The TAC axis shown is only part of the TAC range used, which was ± 500 nsec from the real TAC peak.
(a) Ex-Ef displays and projections

The area darkened in each cell is linearly proportional to the number of counts in the cell. The projections on each axis consist only of the events lying within the two-dimensional envelope shown. The label on the Ef axis is the fragment energy before passing through the foil. The smooth curves are drawn along the valley of isotopic stability for x-ray lines corresponding to the average Z of the mass determined by the fragment energy. A fragment mass scale on the Ef axis and a Z scale on the Ex axis (for the x-ray line indicated) are to aid visual orientation along these curves.

(b) Expanded Ex-Ef displays and projections

Same as (a) but the x-ray energy axis has been expanded for examining the L x-ray region in detail.
mass of the fragment was calculated from its energy (see Appendix C), and the locus of characteristic x-ray energies for nuclear species in the isotopic stability valley (defined by the naturally occurring isotopic abundances) were overlayed on Figure 23. Note that these loci do indeed pass through the observed regions of intensification.

For fragment K x-rays, a window was drawn on the open run, since the K x-ray data for this run had the highest statistics. For the same reason, the Ni foil run was used to draw the L x-ray window. Using a linear gain shift on the fragment energy axis, determined from the two centroid positions of each Ef monitor spectrum, these windows were shifted in fragment energy for each foil in order to encompass equivalent Ex-Ef regions. This was necessary in order to compare the x-ray yields over an equivalent fragment mass range for different foils. The windows used are shown on each plot in Figure 23.

c. Calculations

The data in Figure 23 was sorted again, applying the Ex-Ef windows and projecting on the x-ray axis. These projections \( \chi_{\text{R}(E_x)} \) were then corrected for the x-ray efficiency \( \chi_{\text{E}(E_x)} \) shown in Figure 19, and summed over separate light and heavy fragment regions
to obtain the total number of x-rays produced for each region,

$$Y_x = \frac{1}{L_c \left( \frac{\Omega_x}{4\pi} \right)} \sum_{E_x} \frac{X_{R}(E_x)}{X_{E}(E_x)}$$

where $L_c$ is the live time of the coincidence ADC's and $\Omega_x$ is the x-ray solid angle in steradians. Note that this is a total yield, not an average. The low statistics at the high end of the x-ray spectrum where the efficiency is low may very easily dominate the error. Considering the $X_{E}(E_x)$ as exact quantities, the statistical error in $Y_x$ is

$$\sigma_{Y_x} = \frac{1}{L_c \left( \frac{\Omega_x}{4\pi} \right)} \frac{1}{\sqrt{N}} \sum_{E_x} \frac{\sigma_{X_{R}}(E_x)}{X_{E}(E_x)}$$

where

$$\sigma_{X_{R}}(E_x) = \sqrt{X_{R}(E_x)}$$

and $N$ is the number of terms in the sum. Table 5 contains the results of this procedure. The value of $\Omega_x$ used was 18.8 millisteradians.

The average yield of x-rays per fragment is then

$$Y_{x/F} = Y_x \cdot 2 \cdot \left( \frac{L_F}{Y_F} \right)$$
<table>
<thead>
<tr>
<th>RUN</th>
<th>LIGHT FRAGMENT L lines 1.92-3.70keV (a)</th>
<th>(b)</th>
<th>HEAVY FRAGMENT L lines 3.70-5.96keV (a)</th>
<th>(b)</th>
<th>LIGHT FRAGMENT K lines 13.72-24.71keV (a)</th>
<th>(b)</th>
<th>HEAVY FRAGMENT K lines 24.71-40.23keV (a)</th>
<th>(b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH₂</td>
<td>5.7</td>
<td>.1</td>
<td>4.08</td>
<td>.09</td>
<td>10.7</td>
<td>.2</td>
<td>26.9</td>
<td>.4</td>
</tr>
<tr>
<td>Ni</td>
<td>35.0</td>
<td>.3</td>
<td>18.3</td>
<td>.2</td>
<td>10.7</td>
<td>.2</td>
<td>28.0</td>
<td>.4</td>
</tr>
<tr>
<td>Au</td>
<td>75.4*</td>
<td>.4</td>
<td>12.1</td>
<td>.2</td>
<td>11.0</td>
<td>.2</td>
<td>28.1</td>
<td>.4</td>
</tr>
<tr>
<td>Open</td>
<td>3.35</td>
<td>.08</td>
<td>3.76</td>
<td>.08</td>
<td>10.8</td>
<td>.2</td>
<td>27.5</td>
<td>.4</td>
</tr>
</tbody>
</table>

(a) X-rays emitted per second
(b) Standard deviation in (a), same units

* Includes some contribution from Au M line
where \( Y_F \) is the total number of fragments in the monitor falling within the \( E_F \) limits defined for the coincidence events. \( L_F \) is the particle monitor ADC live time. The statistical error in \( Y_F \) is much less than that for \( Y_X \), so

\[
\sigma_{Y_{X/F}} = \sigma_{Y_{X}} \cdot 2 \cdot \left( \frac{L_F}{Y_F} \right)
\]

Values of \( Y_{X/F} \) are assembled in Table 6. The results of the run with no foil (fragment in-flight deexcitation) were then subtracted from the result of each foil run and the total error is given by

\[
\sigma_{Y_{X/F}} = \sigma_{Y_{X/F}}^{\text{(foil in)}} + \sigma_{Y_{X/F}}^{\text{(foil out)}}
\]

Values in Table 6 for which only the standard deviation is given indicate that the yield was negative or smaller than the standard deviation.

The average x-ray production cross sections (Table 7) were calculated using

\[
\bar{\sigma}_X = Y_{X/F} \cdot \frac{1}{\rho}
\]

where \( \rho \) is the foil thickness in atoms/cm\(^2\) from Table 3. The standard deviation in foil thickness due to nonuniformities dominated the error in the cross section.
<table>
<thead>
<tr>
<th>RUN</th>
<th>LIGHT FRAGMENT</th>
<th>HEAVY FRAGMENT</th>
<th>LIGHT FRAGMENT</th>
<th>HEAVY FRAGMENT</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>L lines 1.92-3.70keV</td>
<td>L lines 3.70-5.96keV</td>
<td>K lines 13.72-24.71keV</td>
<td>K lines 24.71-40.23keV</td>
</tr>
<tr>
<td>CH₂</td>
<td>.070 .001</td>
<td>.051 .001</td>
<td>.132 .002</td>
<td>.333 .005</td>
</tr>
<tr>
<td>Ni</td>
<td>.433 .001</td>
<td>.227 .002</td>
<td>.132 .002</td>
<td>.346 .005</td>
</tr>
<tr>
<td>Au</td>
<td>.932* .001</td>
<td>.150 .002</td>
<td>.137 .002</td>
<td>.347 .005</td>
</tr>
<tr>
<td>Open</td>
<td>.041 .001</td>
<td>.046 .001</td>
<td>.133 .002</td>
<td>.340 .005</td>
</tr>
<tr>
<td>CH₂ -Open</td>
<td>.029 .002</td>
<td>.004 .002</td>
<td>- .004</td>
<td>- .010</td>
</tr>
<tr>
<td>Ni -Open</td>
<td>.391 .001</td>
<td>.180 .003</td>
<td>- .004</td>
<td>- .010</td>
</tr>
<tr>
<td>Au -Open</td>
<td>.891* .002</td>
<td>.103 .003</td>
<td>- .004</td>
<td>- .010</td>
</tr>
</tbody>
</table>

(a) X-ray yield, in characteristic x-rays per fragment

(b) Standard deviation in (a), same units

* Includes some contribution from Au M line
<table>
<thead>
<tr>
<th>RUN</th>
<th>LIGHT FRAGMENT (a) L lines 1.92-3.70keV (b)</th>
<th>HEAVY FRAGMENT (a) L lines 3.70-5.96keV (b)</th>
<th>LIGHT FRAGMENT (a) K lines 13.72-24.71keV (b)</th>
<th>HEAVY FRAGMENT (a) K lines 24.71-40.23keV (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH\textsubscript{2} - Open</td>
<td>2. 1.</td>
<td>.3 .3</td>
<td>- .29</td>
<td>- .75</td>
</tr>
<tr>
<td>Ni - Open</td>
<td>47. 6.</td>
<td>22. 3.</td>
<td>- .45</td>
<td>- 1.2</td>
</tr>
<tr>
<td>Au - Open</td>
<td>400.* 50.</td>
<td>47. 6.</td>
<td>- 1.7</td>
<td>- 4.5</td>
</tr>
</tbody>
</table>

(a) X-ray production cross section, kilobarns
(b) Standard deviation in (a), same units

* Includes some contribution from Au M line
values.

5. Discussion

The domination of the K x-ray yields by the in-flight fragment deexcitation, as seen in Table 6, made it only possible to assign upper limits to the contributions due to impact ionization in the foil. However, the nuclear deexcitation had less effect on the L shell, and the L x-ray yields due to impact ionization reveal general trends, despite some interference by characteristic foil lines. For both fragment groups, the x-ray cross sections (Table 7) increase as a function of foil material Z. The cross section for light fragment L x-ray production increases a factor of more than 20 between CH$_2$ and Ni, but only a factor of 9 between Ni and Au. This can be interpreted as L-M level matching between light fragments and Au. The cross section for heavy fragment L x-ray production increases roughly a factor of 2 between Ni and Au. This reflects the small change in the average level matching when going from heavy fragment L shell - Ni K shell to heavy fragment L shell - Au L shell.

The indication is that the x-ray production by impact ionization for fission fragment L x-rays is enhanced when level matching in the quasi-molecule formed by the ion-atom system occurs. The highest L x-ray yield was observed with Ni. Since this level matching effect
occurs for the L shell, it might also be true for the K shell since the process of electron promotion will be even more important for K shell ionization. Also, the cross section drops off rapidly for lower Z's, and this is why the silicon particle detector gave such poor x-ray yields in the previous experiment. A better K shell match for all fragments would require a higher Z particle detector material for x-ray production. Ge might be sufficient. Estimating from the observed data,

\[ \overline{\sigma}_L(Si) \sim \frac{1}{\alpha_0} \cdot \overline{\sigma}_L(Ge) \]

A similar behavior in the cross section for K x-ray production is expected.
C. X-ray Production by Fission Fragments Stopping in a
Silicon Surface-barrier Detector

1. Objective

The average fragment characteristic x-ray yield for
$^{252}$Cf fragments stopping in a silicon surface-barrier
detector was measured for the purpose of evaluating the
efficiency of the method for atomic number spectrometry
of heavy ions. No x-ray peaks characteristic of fragments
could be identified in the fragment K line region. The
average x-ray yield in the fragment K line region due
to impact ionization of fission fragments when stopping
in a silicon surface-barrier detector was determined to
be $0.018 \pm 0.004$ K x-rays per fragment.

2. Apparatus

The experimental configuration is shown in Figure 24.
Fission fragments from a $^{252}$Cf source located 28.3 ± 0.1
cm from the center of the chamber entered the chamber
and were stopped in a silicon-surface barrier detector
(Ortec 11-325A, 600 mm$^2$ x 60 µm, resistivity 745 ohm-cm,
FWHM 100 keV @ the 6.117 MeV alpha particle of $^{252}$Cf).
The detector was located at the center of the chamber
at an angle of 45° with respect to the incident axis.
X-rays produced in the detector were observed by the
same detection system and geometry as described in section
III.B. For determination of the fragment
Figure 24

Experimental Configuration

\[ \text{Ep(a)} = \text{surface-barrier detector positioned for data run} \]
\[ \text{Ep(b)} = \text{surface-barrier detector positioned for background (open) run} \]
\[ \text{Ex} = \text{Si(Li) x-ray detector crystal} \]
\[ \text{C} = \text{cryostat} \]
\[ \text{Be} = 25.4 \, \mu\text{m Be detector entrance window} \]
\[ \text{M} = 3.175 \, \mu\text{m mylar vacuum window} \]
\[ \text{A} = \text{air gap} \]
\[ \text{S} = ^{252}\text{Cf source} \]
\[ \text{F} = 170 \, \mu\text{g/cm}^2 \text{ Ni foil} \]
\[ \text{FF} = \text{fission fragment} \]
\[ \text{X} = \text{x-ray} \]
\[ \text{Br} = \text{brass} \]
\[ \text{Al} = \text{aluminum} \]
\[ \text{Lu} = \text{lucite} \]
deexcitation contribution the surface-barrier detector was positioned on the downstream wall of the chamber, far enough that x-rays from the detector could not reach the Si(Li) crystal directly.

The $^{252}\text{Cf}$ source was covered by a 170 $\mu\text{g/cm}^2$ Ni foil which eliminated the migration problems (Appendix C) encountered in the previous experiment.

Detector gain drift was stabilized by cooling the chamber as described in Appendix B. The associated TAC drift was concurrently eliminated.

The electronics were identical to those used in section III.B.

3. Procedure

Energy calibrations were determined by the same procedure as used in section III.B.

Count rates were approximately 30 times lower than in the experiment of section III.B. due to the larger source distance necessary to reduce the fragment deexcitation contribution. Most random events came from the strong alpha particle peak. As before, random x-ray - fragment coincidences were negligible and no accidental subtraction was done.

Once again, nuclear deexcitation of the fragments long after fission resulting in x-rays contributed to the data. A background (open) run was taken with the
surface-barrier detector mounted downstream out of sight of the x-ray detector. One expects this to give a low estimate of the deexcitation background, since the fragment stopped in the detector spends all of its subsequent lifetime in the field of view of the x-ray detector whereas the fragment in the background run only spends several nanoseconds in this same field of view. The observed deexcitation rate decreases roughly exponentially as a function of distance from the source (see below), with a half-life distance of about 6 cm. Since fragment velocities are around 1 cm/nsec, this suggests a half-life of about 6 nsec, and a time dependence like

\[ A e^{-\left(\frac{t-t_0}{\ln 2 / t_{1/2}}\right)} \]

The x-ray detector field of view in the background run was about 6 cm so if the number of x-rays observed in the background run was \( B \),

\[ B = \int_{-3}^{3} A e^{-\left(\frac{t-3}{\ln 2 / t_{1/2}}\right)} dt \]

This gives \( A = B / 8.66 \). Assuming the exponential law holds as time increases, the contribution from deexcitation of the stopped fragments will be
\[ \int_{-3}^{\infty} \frac{B}{8.66} e^{-(t-3) \cdot \ln 2/6} \, dt = 2.8. \]

Actually the deexcitation rate decrease has a slower than exponential time dependence. However, the TAC window will limit the acceptable delay in the x-ray signal to about 60 nsec. At worst the deexcitation rate is constant, so the real contribution will be less than \( 10 \cdot B \) (but greater than \( 1 \cdot B \)). The calculations employed simply subtracted \( 1 \cdot B \) from the data.

4. Data Presentation and Analysis

The monitor spectra are shown in Figure 25. The slight gain drift on the cooled detector caused considerably less pulse height resolution broadening than with the uncooled detector.

Identification of real events was done by applying an Ex-TAC window as in section III.B. Figure 26 shows the Ex-TAC displays for fission fragments in the particle detector with the time window superimposed.

The K and L line windows determined in section III.B. were transformed using a linear relation in \( Ef \) to apply to the calibrations of this experiment, Figure 27. The lines on the same plot indicate the position of characteristic x-ray lines for nuclear species lying in the valley of beta-stability. The same windows were
Figure 25

Monitor spectra

Horizontal axis is channel number (energy). Vertical axis is counts per channel.

(a) Particle monitors: The strong low energy peaks are the 6.117 MeV alpha and its pileup peaks. Pileup in this experiment is much less due to the lower count rate resulting from the larger source to detector distance.

(b) X-ray monitors: Some of the peaks have been identified and labeled with element and line symbols. Many of these x-rays are generated by the $^{252}$Cf alpha particles.
(a)
Figure 26

Two-dimensional Ex-TAC displays showing sorting envelope identifying real events.

The area darkened in each cell is linearly proportional to the number of counts in the cell. The projections on each axis consist only of the events lying within the two-dimensional envelope shown. The TAC axis shown is only part of the TAC range used, which was ± 500 nsec from the real TAC peak.
Figure 27

(a) Ex-Ef displays and projections

The area darkened in each cell is linearly proportional to the number of counts in the cell. The projections on each axis consist only of the events lying within the two-dimensional envelope shown. The label on the Ef axis is the fragment energy before passing through the foil. The smooth curves are drawn along the valley of isotopic stability for x-ray lines corresponding to the average Z of the mass determined by the fragment energy. A fragment mass scale on the Ef axis and a Z scale on the Ex axis (for the x-ray line indicated) are to aid visual orientation along these curves.

(b) Expanded Ex-Ef displays and projections

Same as (a) but the x-ray energy axis has been expanded for examining the L x-ray region in detail.
applied to both data and background runs since the calibrations were the same for both.

X-ray yields per fragment were calculated by the procedure described in the previous experiment. Pertinent results are listed in Table 8 (run durations), Table 9 (x-rays emitted per second), and Table 10 (x-rays per fragment).

5. Discussion

The in-flight background yield from this experiment can be compared to that for the source much closer and the activity can be plotted as a function of distance from the source, Figure 28. It is not valid to extrapolate a single time constant from these two points, but it is apparent that the in-flight background is decreasing at about the same rate for all four x-ray - fragment regions, with a "half-distance" (analogous to half-life) of about 6 cm. Extrapolation shows that the K line deexcitation yields drop below 1% of the observed impact ionization yields in silicon at a distance of less than 1 meter.

It is interesting to note that the K x-ray yields are roughly the same for light and heavy fragments. This may mean that either the K shells are equally unmatched for both fragment peaks, or that the yield is dominated by Compton scattering of fission gamma-rays from the particle detector structure.
## TABLE 8 - Run Durations

<table>
<thead>
<tr>
<th>RUN</th>
<th>Lc</th>
<th>Lf</th>
<th>Yf</th>
</tr>
</thead>
<tbody>
<tr>
<td>Detector</td>
<td>.875</td>
<td>.862</td>
<td>14.1</td>
</tr>
<tr>
<td>Open</td>
<td>.871</td>
<td>.856</td>
<td>15.0</td>
</tr>
</tbody>
</table>

Lc = Live time of multiparameter ADC (units of $10^6$ sec)

Lf = Live time of fragment monitor ADC (units of $10^6$ sec)

Yf = Total number of fragment monitor counts for 45 to 127 MeV (units of $10^6$ counts)
<table>
<thead>
<tr>
<th>RUN</th>
<th>LIGHT FRAGMENT</th>
<th>HEAVY FRAGMENT</th>
<th>LIGHT FRAGMENT</th>
<th>HEAVY FRAGMENT</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>L lines 1.92-3.70keV</td>
<td>L lines 3.70-5.96keV</td>
<td>K lines 13.72-24.71keV</td>
<td>K lines 24.71-40.23keV</td>
</tr>
<tr>
<td>Detector</td>
<td>(a) 6.27* (b) .07</td>
<td>(a) .57 (b) .02</td>
<td>(a) .21 (b) .01</td>
<td>(a) .30 (b) .03</td>
</tr>
<tr>
<td>Open</td>
<td>(a) .032 (b) .005</td>
<td>(a) .031 (b) .005</td>
<td>(a) .086 (b) .009</td>
<td>(a) .15 (b) .02</td>
</tr>
</tbody>
</table>

(a) X-rays emitted per second

(b) Standard deviation in (a), same units

* Includes some contribution from Au M line
**TABLE 10 - X-ray Yield per Fragment**

<table>
<thead>
<tr>
<th></th>
<th>LIGHT FRAGMENT</th>
<th></th>
<th>HEAVY FRAGMENT</th>
<th></th>
<th>LIGHT FRAGMENT</th>
<th></th>
<th>HEAVY FRAGMENT</th>
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<td>K lines</td>
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<td>L lines</td>
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<td>K lines</td>
<td></td>
</tr>
<tr>
<td>RUN</td>
<td>(a)</td>
<td>(b)</td>
<td>(a)</td>
<td>(b)</td>
<td>(a)</td>
<td>(b)</td>
<td>(a)</td>
<td>(b)</td>
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<tr>
<td>Detector</td>
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<td>.009</td>
<td>.069</td>
<td>.002</td>
<td>.026</td>
<td>.002</td>
<td>.037</td>
<td>.003</td>
</tr>
<tr>
<td>Open</td>
<td>.0036</td>
<td>.0006</td>
<td>.0035</td>
<td>.0006</td>
<td>.010</td>
<td>.001</td>
<td>.017</td>
<td>.002</td>
</tr>
<tr>
<td>Detector - Open</td>
<td>.76*</td>
<td>.01</td>
<td>.066</td>
<td>.003</td>
<td>.016</td>
<td>.003</td>
<td>.020</td>
<td>.005</td>
</tr>
</tbody>
</table>

(a) X-rays per fragment emitted @ 45° from a silicon surface-barrier detector for fragments incident @ 45°

(b) Standard deviation in (a), same units

* Includes some contribution from Au M line
Figure 28

Internal conversion x-ray yield as a function of distance from the $^{252}$Cf source

(a) Light fragment L line region
(b) Heavy fragment L line region
(c) Light fragment K line region
(d) Heavy fragment K line region
(e) Intensity equivalent to 1% of fragment characteristic K x-ray yield in Si
(f) Intensity equivalent to 1% of fragment characteristic L x-ray yield in Si
Judging from the x-ray projection in Figure 27, the contribution of the Au M line to the light fragment L yield is not more than 50%. So the heavy fragment L yield is more than an order of magnitude lower than the light L yield. Since the light fragment L shells match the Si K shell much better than the heavy L shells this again substantiates that at fission fragment energies, the inner-shell ionization mechanism is predominately electron promotion.

The average K x-ray yield for impact ionization of $^{252}$Cf fission fragments when stopped in a silicon surface-barrier detector is $0.018 \pm 0.004$ x-rays per fragment. The implications of this are discussed further in following sections.
IV. Evaluation

A. Further Development

This study has revealed several requirements that must be met for the proposed x-ray Z spectrometer to be of practical use for fission fragments.

1. Detector Location

The distance of the detectors from the point of nuclear reaction should be made as far as possible to reduce the effect of nuclear deexcitation coincidences. This would also help reduce background due to nuclear processes in the support materials, and it would facilitate insertion of shielding material between the x-ray detector and the reaction site. If fission fragments are to be analyzed from a spontaneously fissioning radioactive source, the source should be covered with a thin foil to prevent contamination, as discussed in Appendix C.

2. X-ray Production Efficiency

The x-ray production medium, the particle detector, must be made of a material with Z higher than silicon in order to increase the average inner-shell vacancy production cross-section. The best average level matching would be around Z = 50, between the light and heavy peaks. However, detectors of good energy resolution are not available in such substances. Use of a Ge (Z = 32)
detector would probably provide sufficient improvement. The amount of Compton scattered gamma-rays from the heavier material will also increase with Z so one must be wary of how great this contribution will be. In charged particle-induced reactions, there will be a high random coincidence rate due to characteristic target x-rays Compton scattered from the particle detector.

3. X-ray Detection Efficiency

The efficiency of the Si(Li) detector for fragment K x-rays is about 20%. The efficiency could be improved by use of a Ge detector, which is nearly 100% efficient in the K x-ray region. Ortec has the technology for 200 mm$^2$ x 5 mm crystals having resolution of 300 eV (FWHM) @ 5.9 keV, 600 eV @ 122 keV.

The x-ray solid angle must be maximized but there must be some method of shielding the x-ray detector from particles and delta-ray electrons. A low Z shield could accomplish this as well as cut out the high yield of low energy x-rays. Using the simple configuration depicted in Figure 29(a), a solid angle of about 3 steradians can be achieved.

The arrangement in Figure 29(b) having a solid angle for x-ray detection of almost $2\pi$ steradians would require a layer of low Z material to stop charged particles such as secondary electrons and the elastically scattered
Figure 29

Detector arrangements for Z spectrometer

$Ep = 200 \text{ mm}^2$ Ge particle detector (13 $\mu$m of Ge is sufficient to stop 100 MeV Br)

$Ex = 200 \text{ mm}^2$ Ge x-ray detector

$W =$ low $Z$ window for shielding $Ex$ from particles

$A =$ anticoincidence guard ring

$S =$ shielding material

$HI =$ heavy ion

$X =$ x- or gamma-ray

$P =$ proton

$e =$ electron or delta-ray

(a) Solid angle for x-ray detection is about 3 steradians.

(b) Solid angle is almost $2\pi$ steradians.

(c) Annular x-ray detector: Solid angle is still almost $2\pi$. 
primary beam in a charged particle-induced experiment. It would also suffer from a high bremsstrahlung background and a very high direct x-ray background from the target in charged particle-induced reactions.

The situation could be improved if it were possible to construct an annular Ge x-ray detector for the configuration shown in Figure 29(c). The x-ray solid angle would still be close to 2π. The light particle shield would no longer be necessary since most of these will go through the particle detector. The direct x-rays could be shielded out. Also, bremsstrahlung should be lower with the x-ray detector behind the bremsstrahlung production site since most of the energetic secondary electrons go forward. An anticoincidence guard ring would be helpful in reducing the number of Compton-escape photons.

Since the Ge x-ray detector must be liquid nitrogen cooled, these detectors would require special fabrication to be mounted in such close proximity. The average K x-ray yield expected in the detection system of Figure 29(c) above is 0.18 x-rays per fragment, calculated from the average yields in Si and an estimated average ionization cross section for Ge of 20 times that for Si. Thus a detection efficiency of 18% might be expected, an improvement of more than 10^4 over the efficiency for the system used in the experiments of section III.
B. Time-of-Flight Mass Spectroscopy for Heavy Ions in the Fission Fragment Mass and Energy Range

Measurement of the energy of a particle and its time-of-flight over a known distance determines its mass from the nonrelativistic formula

\[ m = \frac{2Et^2}{s^2} \]

or, in user's units;

\[ m(\text{amu}) = \frac{E(\text{MeV}) \cdot t^2 (\text{ns})}{(71.9)^2 \cdot s^2 (\text{meters})} \]

Since measurements of \( E \), \( t \), and \( s \) are independent and uncorrelated, the error in \( m \) is

\[ \sigma_m^2 = (\frac{\delta m}{\delta E})^2 \sigma_E^2 + (\frac{\delta m}{\delta t})^2 \sigma_t^2 + (\frac{\delta m}{\delta s})^2 \sigma_s^2 \]

or

\[ \frac{\sigma_m}{m} = \left[ \left( \frac{\sigma_E}{E} \right)^2 + 4 \cdot \left( \frac{\sigma_t}{t} \right)^2 + 4 \cdot \left( \frac{\sigma_s}{s} \right)^2 \right]^{1/2} \]

For a silicon surface-barrier detector, energy resolution for unchanneled heavy ions in the fission fragment range is typically 1 MeV FWHM (Appendix D). This becomes worse for heavier ions [SC 66] and a heavier detector material like Ge [HA 66].
Ordinarily the flight path variation is very small. For a 600 mm² detector oriented perpendicular to the flight path and s = 1 meter,

$$\frac{\sigma_s}{s} = 2 \times 10^{-7}$$

which is negligible compared to the other quantities in (4). Note, however, that the same detector mounted at 45° to the incident direction will cause a 1% dispersion in flight path length for a path length of 1 meter. For the following, the flight path variation will be neglected and equation (4) becomes

$$\frac{\sigma_M}{M} = \left[ \left( \frac{\sigma_E}{E} \right)^2 + 4 \cdot \left( \frac{\sigma_t}{t} \right)^2 \right]^{1/2}.$$

Figure 30(a) depicts contours of dM (FWHM) on the E vs. M plane calculated for the experimental limitations: dE = 1 MeV (FWHM), dt = 1 nsec, and s = 10 meters. The positions of the heavy and light fission fragment peaks are indicated for reference points. Note that only some of the light fragments are resolved (dM less than 1 amu FWHM). In Figure 30(b), the time dispersion has been eliminated by setting dt = 0. The difference from Figure 30(a) is small, showing that the dominant contribution to the mass dispersion is the poor energy resolution. Figure 30(c) shows the effect on Figure 30(a)
Figure 30

Mass resolution of an E-TOF mass spectrometer as a function of m and E

Each contour label is the value of the mass resolution (FWHM) for that contour in amu. The label H indicates the region of typical heavy fission fragments. The label L indicates the region of typical light fission fragments.

(a) Performance of the spectrometer using a silicon energy detector with a path length of 10 meters and a time dispersion of 1 nsec

(b) Same as (a) but with time dispersion of 0

(c) Same as (a) but with a factor of 2 improvement in energy resolution
if the energy resolution is improved by a factor of 2. Light fragments become well resolved, heavy fragments partially resolved. The energy dispersion still dominates the mass resolution. Apparently the energy resolution will limit the mass resolution for fission fragments to at best 1% if a silicon surface-barrier detector is used for the energy determination.

As pointed out in Appendix D, the resolution properties of silicon detectors suffer from a basic physical limitation when used with fission fragments, that being the obnoxious ability of the fragments to transfer a large amount of energy to a detector atom in a single elastic collision. Any detection medium will have this problem. The solution is to use a different detector that has more of a mismatch between particle and detector nuclear masses.

Recent work has been done using proportional counters as energy loss detectors for the mass identification of heavy ions [AJ 72, BA 75B, FO 75, GA 74, HI 70]. However no work was found in which a proportional or pulse ion chamber was used as a total energy detector for heavy ions in the fission fragment region. If the energy resolution of a 100 MeV fragment in silicon is about 1 MeV (FWHM) or 1%, the mass resolution is limited to $dM \text{(FWHM)} = M \times (dE/E) = 100 \times 1\% = 1 \text{ amu}$. Assuming 100% charge collection, the statistical resolution broadening
for an ion chamber is 0.13% @ 100 MeV if 30 eV is required to produce an ion pair. This would contribute to the mass dispersion about

$$\Delta m = m \left( \frac{\Delta E}{E} \right) = 100 \times 0.0013$$

or 0.13 amu FWHM, which is encouraging.

Use of a low mass detector gas such as deuterium in an ion chamber would reduce the average energy transfer in the non-ionizing collisions. So even if the total energy lost was the same, the variance in it should improve with the reduction of the average energy transfer per collision. Since energy loss in nuclear collisions is roughly proportional to $\frac{Z^2}{M}$ of the medium, the use of deuterium in an ion chamber can reduce the average energy loss to a nucleus of the detector medium a factor of 14 with respect to silicon. The ratio of nuclear energy loss to electronic energy loss is proportional to $\frac{Z}{M}$ of the medium, so the total energy lost to nuclear collisions should be about the same for deuterium and silicon. Consequently the standard deviation of energy lost to nuclei of the detector might be improved by almost a factor of 4. This would correspond to an energy dispersion of $1\% \times 0.25 = 0.25\%$ and the mass resolution would be
\[ \Delta m = 100 \times (0.0013^2 + 0.0025^2)^{1/2} \sim 0.3 \text{ amu (FWHM)} \]

which should be sufficient to easily resolve masses in the fission fragment range. Experimental investigations are needed.

Additional advantages of a gas counter would include insensitivity to radiation damage, detector homogeneity, little restriction due to small angle scattering, and the possibility of reducing recombination losses by adjusting the pressure of the gas (and thereby the ionization density and carrier mobility).

The disadvantages of ion chambers are well known: long term instability, low count rates, electronic noise limitations, large size, low solid angle, window thickness, and sensitivity to electrode surface effects. However, if the higher resolution is required, these might be partially surmounted. One expects low count rates in heavy ion experiments, and a 3 meter flight path would require TAC dead times of a few \( \mu \text{sec} \) anyway. Electronic noise can be reduced to a negligible amount (300 eV). Low solid angle would be a disadvantage on a long time-of-flight arm, but heavy ion experiments often require a small solid angle to minimize kinematic energy spread, especially at forward angles. Windows can be made very thin and wire grids have been used successfully to support
large thin windows. Ions scattering from the grid could be discriminated against by requiring coincidence between an initial electrode wire and all succeeding wires. A fast TOF stop pulse could be taken from the window scintillation, or from secondary electrons produced from the window. The range of a primary fragment in 0.1 atm of hydrogen may be as much as 120 cm. The length of the detector required by this could be reduced by using a stack of the recently developed thin film detectors (TFD's) [LA 75, CL 75, MU 75, BA 75C, BA 75D] in front of the ion chamber, and the chamber itself for the measurement of the residual energy only. This might reduce window losses and the path length required in the gas counter, but the average energy loss in the TFD is roughly 1 keV per secondary electron detected using 6 \( \mu \text{g/cm}^2 \) carbon foils [CL 73]. Z information would be a byproduct of this method. Alternatively the gas counter size may be tolerated and a multi-wire ion chamber with the wires aligned perpendicular to the incident direction might be used, yielding a series of energy loss measurements along the path of the fragment.

The contour plots of Figure 31 contrast the mass resolution using a silicon surface-barrier detector with that estimated for the deuterium ion chamber. Figure 31(a) shows the E-TOF mass resolution performance using the deuterium ion chamber as the energy detector, with \( s = \)
Figure 31

Mass resolution of an E-TOF mass spectrometer as a function of m and E

Each contour label is the value of the mass resolution (FWHM) for that contour in amu. The label H indicates the region of typical heavy fission fragments. The label L indicates the region of typical light fission fragments.

(a) Performance expected with an ion chamber as an energy detector: Flight path is 10 meters, time-of-flight dispersion is 0.5 nsec

(b) "Best possible" performance using a silicon energy detector: Path length is 10 meters, time dispersion 0

(c) Realistic experimental situation for fission fragments: Same as (a), but path length is 3 meters
10 meters, time resolution of 0.5 nsec (FWHM), and energy resolution calculated using

\[ \sigma_E = 0.25 \text{ MeV} + \left[ 30 \times 10^{-6} E(\text{MeV}) \right]^{1/2} + 300 \text{ eV (FWHM)} \].

All fragments are well resolved. Figure 31(b) contrasts the best mass spectroscopy possible using a silicon energy detector by setting \( \Delta t = 0 \) and estimating

\[ \sigma_E = 1. \text{ MeV} + \left[ 4 \times 10^{-6} E(\text{MeV}) \right]^{1/2} + 300 \text{ eV (FWHM)} \].

As before, only some light fragments are resolved. Figure 31(c) shows the effect on Figure 31(a) if a flight path of 3 meters is used. The energy resolution still dominates the mass resolution for fission fragments, and the entire range of fragments remains well resolved. Consequently the use of an ionization chamber will allow an increase in solid angle by shortening the flight tube.

C. Conclusions

The conclusions to be drawn from this work concerning charge and mass identification of fission fragments by characteristic x-ray detection and energy-time-of-flight measurements are summarized here.

The highest probability for correct Z identification
is obtained when characteristic K x-rays are detected.

Background sources in true coincidence must be eliminated to remove ambiguity, especially nuclear deexcitation processes such as internal conversion.

Reproducible inner-shell vacancy production may be obtained by impact ionization at low energy (below the coulomb barrier for particle-detector atom collisions) long after the nuclear event.

Experimental studies of K x-ray production in a silicon surface-barrier detector for fission fragment energies and below show a prohibitively low x-ray yield from impact ionization.

Experimental studies of L x-ray production by fission fragments in different materials show a level matching behavior in the x-ray yield as a function of the Z of the material, consistent with the electron promotion mechanism of inner-shell ionization. The enhanced x-ray yield observed when shells are matched suggests that higher efficiency can be achieved by choosing a material close to the fission fragment Z range.

Matching Z means matching mass. For equal ion-detector atom masses, nuclear energy straggling effects are greatest so a compromise must be made between x-ray production efficiency and mass resolution if a simultaneous time-of-flight mass determination is to be performed.
The estimated efficiency of the Z spectrometer in the most favorable situation considered is 18%.

The best mass resolution obtainable with a solid-state energy detector due to nuclear straggling effects is greater than 1 amu FWHM for fission fragment time-of-flight mass spectroscopy. Considerable improvement might be gained if a low mass gas ion chamber is used as an energy detector.
V. Appendices

A. Study of Heavy Ion Beam Composition from a Tandem Van de Graaff Accelerator Utilizing a Gas Stripper

1. Introduction

Work has been done on the behavior of tandem accelerators utilizing a foil stripper for production of heavy ion beams [FR 73, YN 74, TH 74], but at the time of this work no published results were found involving the use of a gas stripper. The existing data of charge-changing in gasses, reviewed more recently by Betz [BE 72], was not necessarily directly applicable to this accelerator, since the true pressure in the various parts of the vacuum system was not known. Furthermore, the composition of the stripper gas was unknown, since it has long been used without replenishment of the gas in the stripper bottle, indicating possibly a leak from the insulating gas. Insulating gas is mainly a mixture of $N_2$ and $CO_2$ (4:1). The most direct approach was to observe the actual output of the tandem as a function of the stripper pressure and terminal voltage.

2. Production of Negatively Charged Halogen Beams

Increasing interest in heavy ion nuclear reactions has prompted study and development of heavy ion sources [SY 71, LA 73]. Negative heavy ion sources have received
some of this attention, both the exchange and direct extraction types [MI 74, HE 68, DA 72, DA 69, HE 69, PO 73]. The McKibben direct extraction duoplasmatron negative ion source [LA 65, MO 59, WI 63] is normally used to produce negative proton and deuteron beams for the tandem, see Figure A-1. On occasion $^{16}\text{O}^-$ and $^{16}\text{OH}^-$ is obtained by bleeding an oxygen:helium (2:1) mixture into the hydrogen arc. Similarly, beams of several μA of Cl$^-$, Br$^-$, and I$^-$ were produced without any modification. The elemental form of each was introduced to the source gas mixture. The low vapor pressures of Br and I made it necessary to gently heat the gas lines to prevent condensation and constriction. Addition of He to the H$_2$ used to support the arc helped to extend filament life, which was typically 10 hours for a BaO coated Pt mesh. Source failure was primarily due to the deterioration of the oxide coating. The original H.V.E.C. 20° magnet was replaced with a larger one capable of bending the heavier beams without overheating. Attempts to produce a beam with HBr and the less corrosive CH$_2$I$_2$ in the source gas mixture failed. Only qualitative features of the source performance were investigated.

3. Experiment

The heavy ion beam leaving the tandem passed through a chamber mounted on the beam line immediately in front
Figure A-1

Direct extraction duoplasmatron negative ion source

Pd = palladium leak
B = needle bleed valve
R1 = regulator
R2 = corrosion service regulator
F = BaO coated Pt mesh filament cathode
A = copper plate anode
E = extraction cone
X = negative heavy ion beam
M = 20° analyzing magnet
Figure A-2

Rice EN Tandem Van de Graaff schematic

IS = 100 keV negative ion source
M = $20^\circ$ analyzing magnet
L1 = einzel lens
P = oil diffusion pump
PG = high vacuum discharge gauge
for monitoring stripper pressure
L2 = magnetic quadrupole lens
D = x-y deflector pair
C = chamber
S = stripper tube
FC = quartz viewer and faraday cup
Figure A-3

Scattering chamber

C1 = 3.18 mm diameter Ta collimator
C2 = 1.59 mm diameter Ta collimator
T = Au scattering foil
E = silicon surface-barrier charged particle detector
HI = heavy ion beam from tandem
of the analyzing magnet, Figure A-2. Scattered particles from a thin Au foil mounted in the center of the chamber were detected at 45° by a Si surface-barrier detector, Figure A-3. The beam was stopped on the chamber wall. The measurements were taken with the quadrupole and deflector off, to avoid selective alteration of the beam charge state distribution. Tuning was done by varying ion source parameters to obtain maximum current in the high energy faraday cup.

Single parameter energy spectra were taken, for $^{16}$O, Cl, and Br beams for various stripper settings and terminal voltages in the range 3 to 5 MV. The terminal voltage was determined by the generating voltmeter reading, and was observed to drift less than ± 100 KV at 4 MV, by surveillance of the meter. This is consistent with the observed width of the spectrum peaks. The stripper pressure was measured with an ionization gauge at the high energy end of the tandem. This gauge was calibrated to read one gauge unit per $0.57 \times 10^{-6}$ Torr at the time of the experiment. The relation of this to the true pressure in the stripper tube was not determined, since it would have involved disassembling the terminal, inserting a gauge and running in air. The drive motor would have overheated in the time it would take to bring the stripper up to pressure. The alternative was constructing a gauge unit for operation in high pressure.
This was considered more effort than necessary, since the objective was to produce and identify the components of the heavy ion beam. The pressure measured with the high voltage on and beam in was observed to decrease, and this effect increased as electron loading increased. Electron loading was monitored using an x-ray detector near the terminal and also by the amount of up charge supplied to the charging belt. The pressure decrease is mainly due to the increased ionization of residual gas with higher electron loading and the subsequent self-pumping action of the high energy tube. The spectra showed more low energy background under these conditions.

4. Data Reduction

a. Data

The typical spectra obtained are shown in Figure A-4. The effect of correcting for the energy dependence of the Rutherford cross section is shown in Figures A-4 to A-6, where the spectra have been multiplied by the channel number squared.

b. Calibration

The detector was initially calibrated by injecting a proton beam at a particular terminal voltage. The energy of different charge state peaks in the spectra were then calculated for elastic scattering from Au at the terminal voltage used.
Figure A-4

Typical spectra

TV = terminal voltage
ST = stripper pressure in gauge units (see text)

(a) Raw data

The peaks are labeled with the charge state (in parentheses) and energy (MeV) of the detected particle. The small peaks on the high side of the primary peak in the Cl spectrum are kinematically identified as \( M = 14 \).

(b) Spectra of (a) after being corrected for the energy dependence of the Rutherford cross section by multiplying the counts in each channel by the channel number squared and dividing by \( 10^4 \).

(c) Same as (b) but the vertical scale is linear, clearly illustrating the variation of the energy spectra with beam type
OH⁻ injected
TV = 5 MV
ST = 9.5

Cl⁻ injected
TV = 5 MV
ST = 9.0

Br⁻ injected
TV = 5 MV
ST = 9.5
OH$^{-}$ injected
TV = 5 MV
ST = 9.5

Cl$^{-}$ injected
TV = 5 MV
ST = 9.0

Br$^{-}$ injected
TV = 5 MV
ST = 9.5

Channel Number
Figure A-5

Effect of terminal voltage on the energy spectra

TV = terminal voltage
ST = stripper pressure in gauge units (see text)

The peaks are labeled with the charge state (in parentheses) and energy (MeV) of the detected particle. Spectra have been corrected for cross section energy dependence.
OH⁻ injected
TV = 3 MV
ST = 9.0

0.0
100
200
300
400

Counts per Channel (thousands)
CHANNEL NUMBER

OH⁻ injected
TV = 5 MV
ST = 9.5
Cl⁻ injected
TV = 3 MV
ST = 9.

Cl⁻ injected
TV = 5 MV
ST = 9.
Figure A-6

Effect of stripper pressure on the energy spectra

TV = terminal voltage
ST = stripper pressure in gauge units (see text)

The peaks are labeled with the charge state (in parentheses) and energy (MeV) of the detected particle. Spectra have been corrected for cross section energy dependence.
OH$^-$ injected
TV = 4 MV
ST = 5.5

OH$^-$ injected
TV = 4 MV
ST = 13.5
Cl⁻ injected
TV = 4 MV
ST = 6.

Cl⁻ injected
TV = 4 MV
ST = 14.
c. Charge State Identification

The energy of each charge state was calculated for elastic scattering from Au and corrected for energy loss in the scattering foil. The energy resolution of the system was not sufficient to separate the Cl or Br isotopes, though both were injected. The proton calibration was used to identify the energy of the prominent peaks, allowing for some pulse height defect. The energy difference between each adjacent peak was constant, so moving in intervals of this energy difference, the charge state of each peak could be decremented towards low energy. The validity of the peak identification was tested by requiring that the charge state 0 peak be the last available spot on the positive energy axis. It was invariably found that this last energy spot was somewhat less than the terminal voltage used, indicating the presence of pulse height defect.

d. Peak Integration

Having identified the true energies of the beam components in the spectra, the peaks were then integrated. No background subtraction was done. In most cases the background contribution was less than 5%, except for charge states less than 2. In the case of overlapping peaks, a division was made at the minimum point between the peaks. The results from this procedure are contained
in Table A1.

e. Calculations

The number of counts in each peak are listed in Table A1. The elastic cross section $\sigma_R$ is proportional to $E^{-2}$ and $E = V(Q+1)$, where $E$ is the incident beam energy, $V$ is the terminal voltage, and $Q$ is the charge state leaving the stripper. The number of particles in the incident beam of a given charge state is proportional to $N_0/\sigma_R$ where $N_0$ is the number detected of that charge state at the given angle. Normalizing to charge state +3, the relative yield is

$$N = \frac{N_0/\sigma_R}{N_{+3}/\sigma_{R+3}} = \frac{N_0}{N_{+3}} \cdot \frac{(Q+1)^2}{16}.$$

No correction is made for possible dependence of the cross section on the effective charge state which may change with ion energy. The effect is probably small since a 45° scattering angle indicates a close nuclear collision.

The yields were then normalized to the total beam and are shown as fractions of total beam in Figures A-7 to A-11, as a function of stripper and terminal voltage for each beam.

5. Discussion

Certain general trends are shown by this data. The
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* = Counts in each spectrum peak; charge state +1 to +5 are in units of \(10^3\) counts

TV = Terminal voltage in MV

ST = Stripper tube pressure in gauge units (see text)

+N = Charge state of spectrum peak
Figure A-7

Charge state distribution vs. terminal voltage

The straight lines connect data points belonging to the same experimental conditions as LABEL indicates:

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Figure A-8

Charge state distribution vs. stripper for O beam

The straight lines connect data points belonging to the same experimental conditions as LABEL indicates:

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4. gauge units
5.
9.
13.
5.5
9.
13.5
5.5
8.
14.
8.
9.5
14.5
Figure A-9

Charge state distribution vs. stripper for Cl beam

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Figure A-10

Charge state distribution vs. stripper for Br beam

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6. gauge units
Figure A-11

Charge state distribution vs. beam type

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<tr>
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<td>Br</td>
<td>4.</td>
<td></td>
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<tr>
<td>5</td>
<td>O</td>
<td>4.</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>Cl</td>
<td>4.</td>
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<tr>
<td>7</td>
<td>Br</td>
<td>5.</td>
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</tr>
<tr>
<td>8</td>
<td>O</td>
<td>5.</td>
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</tr>
<tr>
<td>9</td>
<td>Cl</td>
<td>5.</td>
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</table>
average charge state increases with terminal voltage, stripper pressure, and the mass of the beam particle. There is a large background yield of low energy particles that decreases with increasing terminal voltage but increases with stripper pressure and the mass of the beam. The appearance of the low energy background with increasing stripper indicates that the bad vacuum produced by the stripper gas causes the ions to change charge during the high energy acceleration process. This has the effect of producing ions in the beam with an energy that is a non-integral multiple of the terminal voltage. The low energy background near charge state 0 and below is due to stripper gas accelerated in the high energy tube. Increased background at higher energies with higher stripper is due to increased charge change of the primary beam in the high energy tube, producing non-integral multiples of terminal voltage for beam energies.

The charge-changing cross section [BE 71] is rapidly decreasing with particle velocity in the region 0.1 to 1 MeV/amu. Lower energy or higher mass, for a given charge state, would cause the dominant term in the charge changing cross section to increase rapidly, indicating that more ions will change charge for the same amount of residual stripper gas in the high energy tube, and this is compatible with the observations.

Judging from the uniformity of the widths of the
peaks in Figure A-4 for different charge states, the broadening of the peaks is dominated by the detector resolution and/or terminal voltage drift, not by energy loss straggling in the Au foil or charge change in the high energy acceleration tube. The increased background under the peaks for lower energy and higher mass is due to charge change in the high energy tube. For Cl this "whitening" of the beam is not serious enough to render the regulation system ineffective, as it does for Br and heavier ions.

Freeman et al. [FR 73], using a foil stripper, attribute contaminants in their beam to ionization of residual gas near the terminal and consequent acceleration. In the case of a gas stripper this would be considerably worse. This is seen in the spectra. For example, the secondary peaks in Figure A-4(b) are kinematically identified as $M = 14$. These peaks have the correct energy for the main peak charge state minus 1 for nitrogen ions being scattered from gold. This means that some of the nitrogen stripper gas is being ionized and accelerated down the high energy tube. Since the elastic cross section is proportional to $Z^2$, the actual amount of nitrogen beam is about $(35/7)^2 = 25$ times what it appears to be. For $^{16}O$ and Br these stripper peaks are not separated from the main peaks. Corrections for their contributions were not performed.
Although the higher stripper pressure produces more of the higher charge states (greater than +6), these constitute such a small fraction of the beam as to be useless. Stated simply, the gas stripper is just not thick enough to produce an abundance of higher charge states at pressures that do not cause degradation of the beam by charge exchange in the high energy tube. It is now understood why the Br and I charge states passing the analyzing magnet had lower energies than the same charge state leaving the tandem. The ions were being further stripped between the tandem and the analyzing magnet. When this occurred at higher energy (compared to the energy of the ions at the stripper tube), the average charge state increased. The energy continuum produced by stripping during acceleration assured that there would always be some of each charge state having the right energy to pass the analyzing magnet.
B. Gain Stabilization of Silicon Surface-barrier Detectors

Experience with fission fragment surface-barrier detectors has shown that the leakage current on the detector increases rapidly due to crystal damage long before the resolution deteriorates. The voltage loss across the series resistor in the preamplifier can become significant if the leakage current becomes large enough, causing the bias across the detector to decrease. If the decrease is large enough the pulse height (gain) will decrease.

The saturation characteristic of the Ef detector used in the foil measurements is shown in Figure A-12. Even at the recommended bias, small fluctuations in the applied bias caused significant changes in the pulse height (0.3% per volt).

The leakage current was observed to fluctuate periodically each day which, later in the run, would have caused the detector bias to effectively be turned on and off. This periodic variation was caused by ambient temperature changes of a few degrees Centigrade between day and night.

Attempts to stabilize the gain after the pulse has been digitized by corrections imposed by the computer will result in periodic fluctuations in population between adjacent channels due to round-off truncations in channel number. Two ways to successfully conduct such an after-
Figure A-12

Saturation curve for silicon surface-barrier detector 6-132D

Manufacturer's (Ortec) recommended bias was 50 V.
the-fact correction are: 1) To use an ADC channel width much smaller than necessary for the analysis of the final spectra; or 2) To apply a smoothing routine which utilizes a real number spectrum and calculates a fractional (e.g. gaussian) contribution to all channels in the corrected spectrum from each channel in the uncorrected spectrum.

Another alternative is to do analog gain corrections during the run. A continuous feedback device on the bias supply for maintaining constant detector bias was needed but not available. Instead it was decided to correct for bias changes by periodically adjusting the bias supply. Close supervision kept the deterioration in the fragment pulse height resolution to values comparable to the inherent resolution of the detector for fission fragments (about 1 MeV), sufficient for the accuracy of the experiment.

As the daily fluctuations became more extreme with increasing detector damage, the bias adjustments became more frequent, to the point where a large portion (about 10%) of exposure time was spent in adjustments.

This predicament was finally overcome by cooling the chamber. The problem was to isolate the detector from the environmental temperature changes and couple it to a constant temperature source. Since the detector was not designed to be cooled to liquid nitrogen temperatures and neither were the seals on the chamber,
a more mild form of cooling was chosen.

A bucket of chopped ice was placed on top of the chamber and the system was enclosed with a garbage bag filled with styrofoam chips, Figure A-13. Access to the chamber was accomplished easily enough by using a vacuum cleaner to remove the styrofoam chips, and then the bucket could be removed from the chamber top. The melt water rising above an experimentally determined level in the bucket was drained off since too much water was observed to degrade the thermal coupling between the ice in the bucket and the chamber resulting in a rise of temperature at the detector. Best cooling was achieved with a packed chopped ice-water mixture in contact with the floor of the bucket at all times. Block ice or ice cubes did not provide enough surface area to enable melting to keep pace with the heat transfer required. Later in the experiment addition of force fed dehumidified air reduced condensation and slowed the heat transfer to the surrounding room.

The effect on the leakage current of cooling the chamber in this manner is illustrated in Figure A-14. The leakage was reduced by almost an order of magnitude, and more importantly, the constant 0°C heat sink provided by the ice bucket stabilized the detector leakage current sufficiently so the bias need only be adjusted to compensate for the effect of radiation damage. This was
Figure A-13

Illustration of equipment used to cool chamber and detector

C = chamber
P = particle detector preamplifier
X = x-ray window
I = chopped ice
W = melt water level
D = melt water overflow drain
S = styrofoam chips
DH = cool dehumidified air
V = vacuum system
Figure A-14

Effect of cooling detector to 0° C

Leakage current for detector 6-132D is shown as a function of elapsed time (t) with ice applied at $t = 0$. Temperature change at the detector was about 20° C. The actual bias on the detector was maintained at 50 v.
easily done when the ice was replenished.

It is interesting to note that the high reverse current sensitivity of the damaged detector to small temperature fluctuations suggests the possible application of imperfect or ion-implanted semiconductors as highly sensitive electronic thermometers.
C. Elimination of Californium-252 Migration

The large mass of fission fragments makes possible the transfer of a large portion of the fragment momentum to the surrounding nuclei via elastic collisions. The energy of the recoil nucleus can be large enough to enable it to leave the thin source deposit. In the case of an open spontaneously fissioning source the result is migration of the source material. This poses the problem of having a constantly varying source geometry. Tests showed that the recoils have enough energy to imbed themselves in the chamber walls, so the chamber exposed to such an open source will become activated, and cannot be cleaned. To eliminate the contamination, a thin foil was placed over the source which was thick enough to stop most of the recoils while causing a minimum of energy loss to the fragments. To determine the best foil type and thickness needed, it was necessary to know the energy distribution of the recoil $^{252}$Cf nuclei. Data concerning the source used in the experiments is contained in Table A2.

The elastic cross section in the center-of-mass is

\[ \sigma_{cm} = 1.296 \left( \frac{Z_1 Z_2}{E'} \right)^2 \left( \frac{m_1 + m_2}{m_2} \right)^2 \frac{1}{\sin^4 \left( \frac{\theta_i}{2} \right)} \]

where \( E' \) is the total center-of-mass energy and \( \theta_i \).
is the center-of-mass angle of $m_1$. One needs to know the energy distribution of the recoils $\sigma_{1ab}(E_2, E_1)$, where $E_2$ is the lab energy of the $^{252}$Cf recoils and $E_1$ is the lab energy of the incident fragment. If $N(E_1)$ is the lab energy spectrum of fission fragments, the energy distribution of primary recoils from this spectrum of fragments should be

$$ (2) \quad \sigma_{1ab}(E_2) = \frac{\int \sigma_{1ab}(E_2, E_1) \cdot N(E_1) \, dE_1}{\int N(E_1) \, dE_1}. $$

Equation (1) can be converted to (2) using the formulae:

$$ (3) \quad E' = \frac{m_2}{m_1 + m_2} E_1, $$

$$ (4) \quad \sin^4\left(\frac{\Theta_1}{2}\right) = \left(\frac{1 - \cos \Theta_1}{2}\right)^2 $$

$$ (5) \quad \cos \Theta_1 = 1 - 2 \cos^2(\Theta_2) $$

($\Theta_2$ is the lab angle of the recoil)

$$ (6) \quad \cos^2(\Theta_2) = \frac{(m_1 + m_2)^2}{4 m_1 m_2} \cdot \frac{E_2}{E_1}. $$
\[ (7) \quad \frac{\sigma_{1ab}}{\sigma_{cm}} = 4 \cos \theta_2 \]

\[ (8) \quad \sigma_{1ab} = \sigma_{cm} \cdot 2 \cdot \sqrt{\frac{(m_1+m_2)^2}{m_1 m_2} \cdot \frac{E_2}{E_1}} \]

Substituting (3), (4), (5), and (6) into (1) gives \( \sigma_{cm} \)

and (8) gives \( \sigma_{1ab} \). For each \( E_2 \) one integrates over

\( E_1 \) to get the probability of a recoil having energy

\( E_2 \). To do this, \( m_1, m_2, z_1, z_2, \) and \( E_1 \) must

be evaluated. The experimentally observed energy spectrum

of the fragments is shown in Figure A-15. The linear

energy scale was determined using 79 and 104 MeV for the

light and heavy fragment peak centroids, respectively.

For \( ^{252}\text{Cf} \) recoils, \( m_2 = 252, \) and \( z_2 = 98. \)

\( m_1 \) can be estimated as a function of \( E_1 \) in

\( ^{252}\text{Cf} \) fission. The requirements of momentum, energy and

mass conservation are:

\[ m_1 v_1 \approx m_2 v_2 \]

\[ m_1 v_1^2 + m_2 v_2^2 = 340 \text{ MeV} \]

\[ m_1 + m_2 \approx m_1 + m_2 + \sqrt{V} = 252 \]

which reduce to the formula
TABLE A2 - Californium-252 Source Data

$^{252}\text{Cf}$ electrodeposited to 0.38 $\mu$g/cm$^2$ in a 0.3175 cm
diameter spot on a 1.27 cm diameter x 0.0127 cm Pt disk*

Rated activity: 15 $\mu$Ci (0.03 $\mu$g)

Observed activity: 21 $\mu$Ci (0.041 $\mu$g)

$(1.85 \times 10^4$ SF/sec, $6.0 \times 10^5$ $\alpha$/sec)

$^{252}\text{Cf}$ Data

Average estimated fragment Z: heavy, 60-67; light, 41-46

M: 151 amu 101 amu

E: 79 MeV 104 MeV

Activity: 500 $\mu$Ci/$\mu$g; 97% $\alpha$, 3% SF

Effective half-life: 2.65 years

Alpha particle energy: 6.117 MeV

Average number of neutrons/fission: 3.76

Average neutron energy: 2.35 MeV

Gamma emission rate: $1.3 \times 10^7$ photons/sec/$\mu$g

Neutron emission rate: $2.34 \times 10^6$ n/sec/$\mu$g

Alpha decay rate: $14.6 \times 10^6$ $\alpha$/sec/$\mu$g

Fission rate: $4.5 \times 10^5$ SF/sec/$\mu$g

* Manufacturer: Isotope Products Labs, Burbank, California
Figure A-15

\(^{252}\text{Cf}\) fission fragment energy spectrum
\[ m = 252 - 1.482 \cdot E_1 \]

\[ Z_1 \] can be roughly estimated from \( m \), by plotting a straight line to the isotopic stability curve (\( Z \) vs. \( N \)) in the fragment region as follows: for \( Z = 35, M = 80, N = 45 \); for \( Z = 53, M = 127, \) and \( N = 74 \). This gives, using \( M = Z+N \),

\[ Z_1 = 0.383 m + 4.33 \]

The above relations were used to generate the recoil spectrum from 0.1 to 50 MeV, Figure A-16. The percentage breakdown of this spectrum is shown in Table A3. Range tables [NO 70] were extrapolated to low energy for \( ^{252}\text{Cf} \) in various substances. Of the available materials, thin Ni foils would cause the least energy degradation to the fission fragments. Available Ni foils of 115, 170, and 230 \( \mu \text{g/cm}^2 \) thick would stop the \( ^{252}\text{Cf} \) recoils with energies 2.0, 2.7, and 3.5 MeV, respectively. It was decided to use the 170 \( \mu \text{g/cm}^2 \) foil. In practice it proved sufficiently thick to completely eliminate the migration problem.
Figure A-16

Energy distribution of $^{252}$Cf recoils

(a) Recoils below 2.0 MeV (82% of total) are stopped by a 115 $\mu$g/cm$^2$ Ni foil
(b) Recoils below 2.7 MeV (84% of total) are stopped by a 170 $\mu$g/cm$^2$ Ni foil
(c) Recoils below 3.5 MeV (87% of total) are stopped by a 230 $\mu$g/cm$^2$ Ni foil
TABLE A3 - Recoil Energy Distribution

Fraction of all $^{252}\text{Cf}$ Recoils Between 0.1 and 50 MeV in Energy Range

<table>
<thead>
<tr>
<th>Fraction</th>
<th>Energy Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>.738</td>
<td>0.1 to 1 MeV</td>
</tr>
<tr>
<td>.125</td>
<td>1</td>
</tr>
<tr>
<td>.041</td>
<td>3</td>
</tr>
<tr>
<td>.042</td>
<td>5</td>
</tr>
<tr>
<td>.052</td>
<td>10</td>
</tr>
<tr>
<td>10</td>
<td>50</td>
</tr>
</tbody>
</table>
D. Pulse Height Resolution of Solid-state Particle Detectors for Heavy Ions

The causes of the nonlinearity of the pulse height response of solid state surface-barrier detectors to slow heavy ions and the related degradation in energy resolution have been studied for some time [WI 71, MO 66, KA 71B, NA 66, SC 66, FI 73A, FI 73B, FO 72, KA 74, BE 74]. The general consensus is that deviations from linear response is due to the combined effects of: 1) Electrode and dead layer losses; 2) Incomplete charge collection; and 3) Nonionizing atomic collisions.

A fission fragment entering the detector (Figure A-17) loses some of its energy by electronic collisions in the thin window electrode. Additional energy is lost in the silicon oxide layer immediately below the electrode. Most of the ionization produced (about 0.5 MeV) is not collected due to the absence of sufficient electric field in this region. The variance in the energy lost is small (at most 10 keV FWHM) since the energy is lost to many electronic collisions in which the energy transfer is small.

The fragment then enters the active volume of the detector, producing a dense column of ionization (plasma). Most of the electron-hole pairs produced are separated and swept to the electrodes by the electric field to produce the output pulse, but some recombine and are not
Figure A-17

Path of a fission fragment stopping in a solid-state detector

W = Au window electrode and silicon monoxide dead layer
I = ionization cone (width of the cone is intended to reflect the density of ionization)
A = rear electrode, usually Al
R = region of high recombination probability
N = region of nonionizing collisions
collected (representing about 2 or 3 MeV). The amount of recombination will depend on the field strength, number of trapping centers, plasma density, and carrier mobility. The density of the plasma is greatest at the beginning of the range, since the energy loss to electrons for fission fragments is an increasing function of energy. Most of the recombination occurs in a thin layer near the surface. Again, the variance in the total energy not collected is small (at most 7 keV FWHM) since the energy lost in an individual ionization event is small.

As the fragment loses energy it gains electrons and the energy loss to electrons decreases due to increased nuclear charge screening. The probability of a close nuclear encounter with a nucleus of the detector medium is at the same time increasing as $E^{-2}$. Near the end of its range, below about 6 MeV, the fragment is near neutrality and loses relatively little energy to ionizing events. Most of the remaining energy is lost to nuclear collisions with detector atoms in which a relatively large amount of energy can be transferred to the recoil nucleus, either elastically or inelastically. The elastic recoils have on the average very low energy (1 MeV or less for silicon) and will similarly lose most of their energy in close nuclear events. By this process the energy rapidly degrades into lattice vibrations without
having produced any appreciable ionization. The net result can be thought of as a sharp increase in the energy loss per ion pair produced near the end of the fragment range. One can estimate the number of events involved. If the energy resolution is 1 MeV (FWHM) resulting from a net 2 MeV lost in nuclear collisions (below 6 MeV), the average energy transfer per nuclear collision is then about 100 keV, consistent with the above explanation. It is the large statistical variation in the total non-ionizing energy lost (2 to 3 MeV) in these collisions that is the dominating contribution to the pulse height dispersion for heavy ions in silicon surface-barrier detectors.

The above conclusion is corroborated by the observation that fission fragments moving between the planes in a silicon crystal lattice show much better energy resolution \([\text{MO 66}]\) but attempts to utilize this effect to achieve 1 amu mass resolution \([\text{SU 74}]\) have failed.
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