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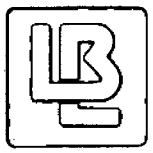

Lawrence Berkeley Laboratory

UNIVERSITY OF CALIFORNIA

To be presented at a Workshop on Future Directions in Transplutonium Element Research, Washington, DC, February 28-March 2, 1983

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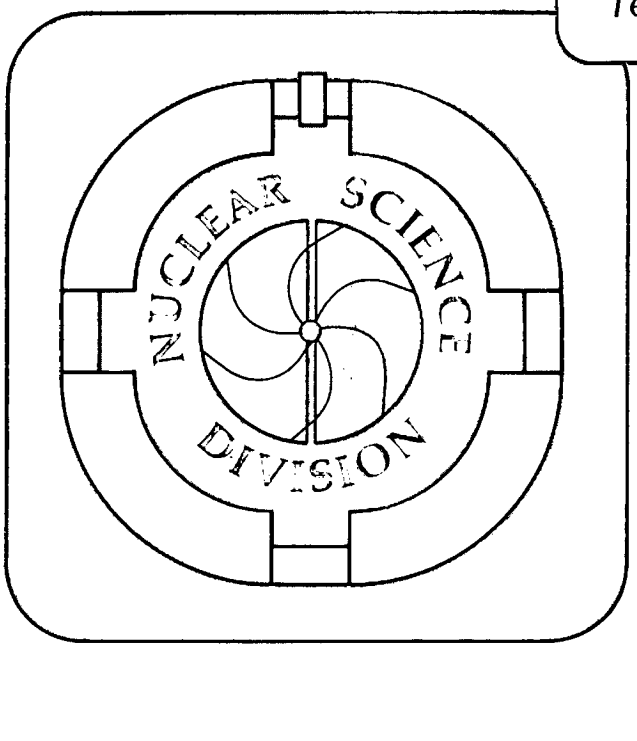

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NUCLEAR REACTIONS AND SYNTHESIS OF NEW TRANSURANIUM SPECIES

By Glenn T. Seaborg

At the time of this writing (late 1982) 16 transuranium elements are known (atomic numbers 93 to 107, inclusive, and number 109). Distributed among these 16 elements are nearly 200 nuclear species. Those with atomic numbers 93 to 101, inclusive, were first synthesized and identified (i.e., discovered) through the use of neutrons, deuterons or helium ions. The half lives become shorter and shorter with increasing atomic number, until at elements 107 and 109 these are in the range of milliseconds.

Three of the transuranium elements (numbers 104, 105 and 106) and a large proportion of the transfermium (beyond number 100) nuclear species discovered during the last 15 years have utilized actinide isotopes produced in the HFIR - TRU facilities at ORNL as the target material. Present indications are that most of the future discoveries of new transuranium nuclear species and the discovery of superheavy elements, if successfully accomplished, will utilize such target material.

In this short review, I shall describe the special aspects of heavy ion nuclear reaction mechanisms operative in the transuranium region, the role of new techniques, possible nuclear reactions for the production of additional transuranium elements and nuclear species and the importance of work in this region for the development of nuclear models and theoretical concepts. This discussion should make it clear that a continuing supply of elements and isotopes, some of them relatively short-lived, produced by the HFIR-TRU facilities, will be a requirement for future synthesis of new elements and isotopes.

In this short account, I shall not cover the historical aspects except for some brief references that are indispensable to an understanding of the theme. A compilation[1] of reprints of the original publications, accompanied by explanatory material, covers very thoroughly all aspects of the historical background of the transuranium elements through atomic number 106. This includes such information on the discovery of these elements, including accounts of the competing claims for the discovery of elements 104, 105 and 106 by scientists at the Dubna Laboratory in the Soviet Union, as well as original information on many isotopes, nuclear reactions for their production, radioactive properties, chemical properties of the elements, etc. The discovery of all elements beyond atomic number 100 (fermium) has been made on a one-atom-at-a-time basis and this will surely continue to be the case. It has been crucial to the claim of discovery that the atomic number of the reaction product be clearly identified using chemical or physical techniques. Criteria for establishing that such an identification has been made have been published[2] by an international group of experts and these criteria will guide the discussions in the review.

Nuclear stability considerations and radioactivity decay properties, which are of central importance in devising experiments for the production and identification of new elements and new isotopes, are the subject of another paper[3] prepared for this workshop.

However, for the convenience of the reader, I shall briefly summarize some salient information about superheavy elements (SHE). Simple extrapolations of radioactive decay properties for elements beyond atomic number 109 suggest that the half life, especially that for decay by spontaneous fission, shall become so short as to make detection very difficult and soon impossible. However, in the period from 1966 to 1972, a number of calculations[4] based on modern theories of nuclear structure showed that in the region of proton number Z = 114 and neutron

number N = 184 the ground states of nuclei should be stabilized against decay by spontaneous fission. This stabilization is due to the complete filling of major proton-and nuetron-shells in this region and is analogous to the stabilization of chemical elements such as the noble gases by the filling of their electronic shells. Such superheavy elements are predicted to form an island of relative stability extending both above and below Z = 114 and N = 184 and separated from the peninsula of known nuclei by a sea of instability.

Some more recent calculations,[5] based on careful consideration of the effect of mass asymmetry on the fission barrier and a reduced spin-orbit coupling strength, have indicated that the Z = 114 shell effect is not very large. These calculations do confirm the existence of a shell at N = 184, but also suggest less stability for species with N < 184; that is, the island of stability has a cliff with a sharp drop-off for N < 184. If these considerations are correct, it would become considerably more difficult to synthesize and detect the superheavy elements. A premium would be placed on producing a nucleus with N = 184 or very close to this, N = 183, in order that it might have a half life sufficiently long to make it detectable.

During the last 15 years numerous attempts have been made to synthesize and identify superheavy elements through the bombardment of heavy target nuclei with heavy ions. None of these experiments has been successful. A summary and analysis[6] suggests that this failure is not due to the failure to produce superheavy intermediate nuclei, but is due to the low survival probabilities of these superheavy precursors.

NUCLEAR REACTION MECHANISMS

Nuclear reactions designed to produce isotopes in the transplutonium region are dominated by the competing fission reaction, which diminishes the yield of

the desired products. This diminution in yield is usually very drastic and is greater the higher the atomic number and degree of excitation of the intermediate nucleus. The tendancy toward undergoing this unwanted fission also increases with increasing angular momentum imparted to the intermediate nucleus. (Angular momentum increases with the increasing mass of the bombarding projectile nucleus and with the increasing magnitude of the impact parameter, the lateral distance between the centers of the target and projectile nuclei). Thus, the name of the game is to produce intermediate nuclei with a minimum of excitation (cold nuclei) and a minimum of angular momentum so they can reach the ground state (usually through the emission of neutrons and gamma-rays) with the minimum of loss from the competing fission reaction.

There are a number of heavy ion reaction mechanisms that might lead to the identification of new isotopes, and in one case possibly new elements, through the production of cold nuclei in sufficient yields to be detectable. The compound nucleus mechanism, known and understood for the longest time, offers the best, and probably the only, hope for the production and identification of new elements; this seems to be operable with heavy element target nuclei (lead and bismuth) only for heavy ion projectiles with atomic number up to 26 (iron) - for transplutonium targets, this limitation produces no roadblock to the possible production of Superheavy Elements.

The other heavy ion nuclear reaction mechanisms cover a range of categories whose definition is not always unambiguous and which tend to merge at the boundaries of the definitions one into the other. An oversimplified categorization into two types will serve our present purposes. One type is well described as simple transfer reactions and is applicable for our purposes (transplutonium targets) with light heavy ions - up to atomic number 10 (neon) or somewhat higher. The other can be described as deep inelastic transfer reactions, whose importance

increases with the increasing atomic number of the heavy ion projectile, with uranium projectiles the most interesting for transplutonium targets.

Compound Nucleus Mechanism. This mechanism involves the complete amalgamation (complete fusion) of the heavy ion projectile with the target nucleus, followed by the de-excitation of this compound nucleus through the emission (usually) of neutrons and gamma-rays in competition with the more prevalent fission reaction. The most central collisions (smallest impact parameter) which impart only a small amount of angular momentum to the system, lead to such complete fusion of the projectile and target nuclei. The product nuclei are strongly focused in the forward (i.e., the projectile beam) direction due to momentum conservation. The classical method of preparing transuranium nuclei has been through the use of the complete fusion reaction. A typical example is the synthesis of element 106 by Ghiorso et al.[7] who bombarded ^{249}Cf with 95 MeV ^{18}O and observed the ^{249}Cf (^{18}O, 4n) ^{263}106 reaction with a cross section of $\sim$0.3nb. The ^{263}106 atoms ($t_{\frac{1}{2}} = 0.9 \pm 0.2$ sec) were identified by observing the previously known daughter, ^{259}Rf, and granddaughter, ^{255}No, through the decay sequence $^{263}106 \xrightarrow{\alpha} {}^{259}\text{Rf} \xrightarrow{\alpha} {}^{255}\text{No} \xrightarrow{\alpha}$. The ^{263}106 atoms were isolated using a helium jet and deposited on the rim of a wheel. The deposit was then rotated in front of a sequence of surface barrier detectors which then detected the primary α-decay of ^{263}106. These primary detectors were then moved to face another series of secondary detectors which detected the decay of the previously known daughter atoms implanted in the primary detectors by recoil in the initial decay. The time correlated decay information was recorded using an on-line computer. A total of 14 time-correlated events was observed.

However, despite successes such as this, if we look at the cross sections for complete fusion reactions such as the X (heavy ion, 4n) Y reaction, we find a sharp decrease in the magnitude of these cross sections as the (Z,A) of the

heavy ion projectile increases. As a further constraint, the limits on availability of target materials with Z > 98, presents great, but no insurmountable, difficulties in synthesizing new transuranium nuclei with complete fusion reactions. A number of workers, particularly those in the Soviet Union, have pointed out that if some heavy ion projectiles with $40 \leqslant A \leqslant 60$ are used in combination with tightly bound target nuclei containing closed shells of nucleons, it is possible to form extremely "cold" compound nuclei whose survival probabilities might be high enough to compensate for the decreased complete fusion cross sections.

A number of experimental studies involving the magic lead and bismuth nuclei have shown that the possibility of such "cold fusion" reactions is, in fact, a reality. Flerov et al.[8] observed the production of ^{252}No using the ^{206}Pb (^{48}Ca, 2n) reaction, while Nitschke et al.[9] found ^{254}No to be produced with a surprisingly large cross section of 3.4 ± 0.4 μb in the ^{208}Pb (^{48}Ca, 2n) reaction. In this same manner, Gäggeler et al.[10] observed the production of ^{244}Fm in the ^{206}Pb (^{40}Ar, 2n) reaction.

The principal development, however, that has pushed "cold fusion" reactions to the forefront in efforts to synthesize new transuranium nuclei was the work of Münzenberg et al.[11,12] using the velocity filter SHIP at GSI. They have observed unambiguous evidence for the occurrence of the ^{207}Pb (^{50}Ti, 2n) ^{255}Rf and ^{209}Bi (^{50}Ti, 2n) ^{257}Ha reactions at a bombarding energy of 4.85 MeV/u. Most interestingly at a ^{50}Ti energy of 235 MeV (4.70 MeV/A), they observed the ^{208}Pb (^{50}Ti,n) ^{257}Rf reaction. Since SHIP is a velocity separator, "transfer" and "deep inelastic" products are strongly suppressed since the products from these binary reactions have a wide angular distribution and do not move with the velocity of the complete fusion evaporation residues. The separator accepts recoils only from a limited range of angles near 0°; thus, these reactions are quite probably cold complete fusion reactions.

Of special interest is the use of cold fusion reactions to synthesize, and thus discover, element 107[11] and element 109.[13] For the former, ^{209}Bi was bombarded with 4.85 and 4.95 MeV/u ^{54}Cr and the ^{209}Bi (^{54}Cr,n) ^{262}107 reaction was observed. The identification of the ^{262}107 ($t_{\frac{1}{2}} = 4.7^{+2.3}_{-1.6}$ ms) was based upon the observation of a set of correlated alpha particle decays which end in the known ^{250}Fm decay. No complete decay chains were observed due to the small yield and the 50% efficiency of the detector system, but two partially complete chains ending in ^{250}Fm were observed and one chain ending in ^{254}Lr was observed. The velocity of the ^{262}107 atoms was determined two different ways, by the velocity separator itself and by a time-of-flight measurement. The energy of the evaporation residue was measured and agreed with expectations for the cold fusion mechanism. Similarly, element 109 was identified[13] as the result of the bombardment of ^{209}Bi with 5.15 meV/u ^{58}Fe ions according to the reaction ^{209}Bi (^{58}Fe,n) ^{266}109. A single atom of ^{266}109 (which decayed after a time interval of 5 ms) was identified through the observation of correlated alpha particle and spontaneous fission decays involving previously known products.

Transfer Reactions. In this type of nuclear reaction there is transfer of nucleons from the projectile to the target nucleus leading (of interest to us) to products with atomic numbers all the way from that of the target nucleus to that of the compound nucleus. (The transfer of just a couple of nucleons is also referred to as quasielastic scattering and the transfer of a large part of the projectile is alternatively referred to as "massive transfer" or "incomplete fusion"). These reactions seem to result from the more peripheral collisions (those with larger impact parameters) and higher angular momentum. The product nuclei are not so strongly focused in the forward direction as in the case of the compound nucleus mechanism. For light heavy ions and heavy target nuclei, of primary interest to us, the protons transfer preferentially from the projectile nucleus to the target nucleus, a tendency that can be understood by considering

the potential energies of the systems near contact.[14]

One of the first and more careful studies of these heavy ion "transfer" reactions involving production of transuranium nuclei was done by Hahn et al.[15] This remains one of the few studies in which kinematic measurements were attempted. Hahn et al. studied the excitation functions, recoil range distributions and angular distributions of the heavy transuranium recoil products. In particular, they studied the characteristics of the production of ^{245}Cf and ^{244}Cf via the transfer reactions ^{239}Pu (^{12}C, α2n) and ^{239}Pu (^{12}C α3n) and via the complete fusion reactions ^{238}U (^{12}C, 5n) and ^{238}U(^{12}C, 6n). As expected, the complete fusion reaction products are strongly forward focused with their angular distribution peaked at 0° and show Gaussian range distributions with mean ranges that increase with increasing projectile energy and whose values agree with the assumption of complete fusion. The same $^{244,\ 245}$Cf products when produced in the transfer reaction show angular distributions which peak near the complement of the grazing angle and show asymmetric range distributions whose mean value decreases with increasing projectile energy. The yields of $^{244,\ 245}$Cf are much larger in the transfer reactions compared to the complete fusion reactions. The yields of the transfer products are described by Hahn et al. with modest success using a modification[16] of the semi-empirical Sikkeland systematics of product yields in heavy fusion reactions.[17,18] These calculations indicate that the reason for the higher product yields in the transfer reaction is the relatively cold residual nucleus produced in this reaction compared to the complete fusion reaction.

Demin et al.[19] used multi-nucleon transfer reactions to produce ^{246}Cf, $^{251,\ 253}$Es, $^{250,\ 254}$Fm, and ^{256}Md from ^{249}Cf using ^{22}Ne projectiles. Perhaps the most significant of the recent "transfer" reaction studies as far as creating interest in these reactions as useful tools for transuranium nuclide synthesis is the work of Lee et al.[20] Lee et al. measured the yields of heavy actinides formed in the interaction of near barrier energy ^{16}O, ^{18}O, ^{20}Ne with ^{248}Cm. In a

follow up to this work Lee et al.[21] measured the excitation functions for the production of isotopes of berkelium through fermium in bombardments of ^{248}Cm with ^{18}O ions and of isotopes of berkelium through nobelium in bombardments of ^{249}Cf with ^{18}O ions.

Schädel et al.[22] measured the distributions of heavy actinide products from reactions of ^{18}O and ^{22}Ne with ^{254}Es and found greatly enhanced yields, by many orders of magnitude, especially for neutron-rich products, of mendelevium and nobelium isotopes as compared with those produced by the same projectiles with ^{248}Cm or ^{249}Cf targets. For example the yields of $\Delta Z = 2$ products - californium isotopes from ^{248}Cm and mendelevium from ^{254}Es - were approximately equal, indicating the tremendous advantage of using a ^{254}Es target for the production of the heaviest actinide isotopes.

Apparently, in one reaction mode, at the higher energies, for these transfer reactions the projectile breaks up elastically in the nuclear field with the breakup probability having a maximum for the grazing angle (the angle of deviation from the original direction for a projectile nucleus making a peripheral collision with a target nucleus). A fragment of the projectile fuses with the target nucleus, while the remainder of the projectile carries away a large amount of the available energy as kinetic energy, thus reducing the excitation energy of the target plus the absorbed fragment and producing a cold nucleus. The excitation functions appear to be consistent with calculations[23] based on simple energy balance considerations using the masses of the projectile, target nucleus, and products and assuming the energy of the projectile in excess of the Coulomb barrier is apportioned to the target nucleus according to the fraction of the projectile mass transferred. At energies close to the reaction barrier the quasielastic transfer mechanism apparently prevails.

Extrapolations of the yield curves suggest that higher energy transfer

reactions may provide a means of producing new neutron-rich isotopes of known elements. In this process, at least formally, such transfer reactions provide a source of exotic projectiles. For example, ^{22}Ne could give rise to ^{21}F capture and ^{18}O give rise to ^{17}N capture. This mechanism, with heavier projectiles such as ^{48}Ca, might offer another route to superheavy elements.

Somerville et al.[24] have produced a number of interesting isotopes, that decay via spontaneous fission, by bombarding heavy transplutonium nuclei with light heavy ions. These include an isotope with a half-life of about 100 milliseconds, produced with the unexpectedly large cross section of a microbarn by the bombardment of ^{254}Es with ^{18}O ions. Another interesting isotope, presumably ^{260}Rf (element 104), produced by the reaction of ^{15}N ions with ^{249}Bk nuclei and by other reactions, has been shown by Hulet et al.[25] (when produced by the reaction of ^{15}N ions with ^{249}Bk nuclei) to decay with an unusual symmetric distribution of fission products. This may indicate that the reactions and decay of nuclei in this region may be governed by a single fission barrier, in contrast to the doubly humped fission barrier characteristic of lighter heavy transplutonium nuclei.

Deep Inelastic Transfer Reactions. For heavy targets and heavy ions with $A \leq 40$, it has been observed that the complete fusion cross section is a very important part of the reaction cross section. By extrapolation, it was felt that this situation would continue with projectiles as heavy as krypton. Thus, the reaction of ^{209}Bi with ^{84}Kr was thought to be a possible avenue for producing transuranium nuclei. However, it was discovered in the course of this attempt to make new transuranium nuclei, that the complete fusion cross section was negligibly small and that a new type of reaction, deep inelastic scattering, was occurring.[26] This failure of heavy nuclei to fuse is due to the fact that the Coulomb repulsive forces between the touching nuclei exceed the nuclear attractive

forces leading to fusion, resulting in the inability of the nuclei to interpenetrate inside the fission saddle point.[27] However, it was soon realized that this new reaction with its characteristic extensive exchange of nucleons between projectile and target nuclei during the reaction could lead to significant production of trans-target species. The reaction product mass distributions are bimodal with centroids near the target and projectile masses[28] and with distribution widths larger for high mass target and projectiles. Also the emission directions of the products are more complicated than for the other reaction mechanisms. Thus, deep inelastic transfer reactions involving uranium or heavier targets with heavy projectiles would be expected to lead to the production of transuranium nuclei with atomic numbers ranging well above that of the target nuclei.

The most significant use and understanding of deep inelastic transfer reactions to produce transuranium nuclei has been in the studies of the ^{238}U + ^{238}U reaction at the UNILAC at GSI. The first realization of the unusual potential of this reaction for transuranium nuclide synthesis was in the work of Hildenbrand, Freiesleben and co-workers.[29,30] who found, from reconstructed primary Z and Q value distributions, more particle transfer at a given energy loss than in other systems, i.e., the diffusion process seems to proceed colder in this system. Cold transfer is, of course, just what is needed to make the fragile transuranium species. Radiochemical studies by Schädel et al.[31] confirmed the coldness of the products from the ^{238}U + ^{238}U reaction and its implications. A somewhat expanded version of these studies has been reported by Gäggeler, et al.[32] The distribution of target-like fragments from the deep inelastic reaction peaks at Z = 91 rather than Z = 85 (as found in the Xe + U reaction[33] or Z = 79 (as found in the Kr + U reaction[34]). Thus, the "goldfinger" (as this feature was dubbed in Reference 34) had become the "Protactinium finger". This upward shift of Z of the peak of the survivor distribution and its broadening are further indications of the colder

diffusion occurring in this system. Reconstruction of the primary target-like fragment distribution led to an estimation of the production cross section of Z = 70 fragments in this reaction of 10^{-28} cm^2 which, under the assumption of a binary process, must also be the estimate of the primary fragment yield of the superheavy element 114 in this reaction.

The fact that the transuranium element distributions have the same general shape in the U + U and Xe + U reactions and the fact that the centroids and widths of the distributions change little with projectile energy[35] can be understood in terms of the fact that despite changes in the primary distributions with projectile Z, A and E, only those few nuclei in the low excitation energy, low J angular momentum tails of the primary distributions will survive fission. The principal advantage of the U + U reaction is that because of the generally broader primary product distributions, the number of nuclei in the tails of the distributions increases enormously.

As a logical followup to the work with the $^{238}U + ^{238}U$ reaction, Gäggeler *et al.*[32] and Schädel *et al.*[36] have reported the results of attempts to produce transuranium nuclei in the reaction of 7.4 MeV/u ^{238}U with ^{248}Cm. The shapes and centroids of the isotopic distributions are similar to those observed in the $^{238}U + ^{238}U$ reaction but the magnitudes of the yields are much greater in the $^{238}U + ^{248}Cm$ reaction. For example, the $^{238}U + ^{248}Cm$ reaction gives $\sim 10^4$ times more Cf, and 10^3 times more Fm than the $^{238}U + ^{238}U$ reaction. Gäggeler *et al.*[32] extrapolate to a 10^2 fold enhancement in the Md and No yields in the $^{238}U + ^{254}Es$ reaction and a $\sim 10^3$ fold enhancement in the Lr yields.

Unfortunately, however, no products beyond Z = 101 were observed even in the $^{238}U + ^{248}Cm$ reaction, indicating that the deep inelastic transfer will not provide a route to new transuranium elements, although it should provide a route to new transuranium isotopes.

TECHNIQUES FOR IDENTIFICATION OF TRANSURANIUM REACTION PRODUCTS

General. In the study of heavy ion reactions resulting in transuranium products, it is of paramount importance to be able to isolate and uniquely identify the products as to their atomic number (Z), mass number (A) and formation cross section. Indeed the claim to discovery[2] of a new element must involve identification of Z while the claim of discovery of a new nuclide must involve measurement (and/or deduction) of both Z and A. Nitschke[37] has classified the commonly used techniques of isolating transuranium reaction products by the half life ($t_{\frac{1}{2}}$) of the products and the minimum detectable cross section. His classification scheme is shown in Figure 1. Some of the isolation techniques shown in Figure 1 such as chemistry, magnetic spectrometers, etc., can also serve as methods of establishing the Z and/or A of the species involved.

Chemical Methods. For reaction products with the longest half-lives, chemical separation techniques offer a convenient method of isolating individual reaction products and establishing their atomic numbers. These techniques offer the greatest sensitivity of all methods because of the large amounts of target material that can be used.

A typical example of the use of chemical techniques to study heavy ion reactions is the effort of Kratz, Herrmann and their co-workers at GSI[38,39] to study the production of trans-target actinides and possible superheavy elements formed in the reaction of ^{238}U with ^{238}U. The chemical problems involved in these studies are formidable. Because of the large cross sections for deep inelastic scattering and the high fissionabilities of the transuranium nuclei, the sought after actinide (Fm, Md) production cross sections are approximately 10^7 less than those of interfering Ra, Ac and Th activities. The separation scheme used is illustrated in Figure 2 and involved the use of four linked chromatographic columns, three of which involved High Performance Liquid Chromatography techniques. A chemical yield of 80-90% with a separation factor of greater than

10^7 was achieved. Similar chromatographic techniques were used by Unik et al.[40] to study actinide production in proton-irridated U targets while ion exchange procedures devised by Kratz, Liljenzin and Seaborg[41] and Lee et al.[20] have been widely used in heavy ion reaction studies at Berkeley.

The Helium Jet, Drums, Tapes and Wheels. For species with half-lives in the range from $0.1 \leq t_{\frac{1}{2}} \leq 10$ Sec, the helium jet is a superior method of isolating reaction products, as witnessed by its use in the discovery of new elements.[7] In this method, first developed by Ghiorso et al.,[42] Friedman and Mohr,[43] and McFarlane and Griffoen,[44] reaction products recoiling from the target are thermalized in helium gas at approximately one atmosphere pressure which leaves the target chamber via a connection to a low pressure area, creating a "jet" or stream of helium,[45] (Figure 3). The helium gas stream impinges upon a collection device such as a tape or wheel or drum which moves the activities to the detectors. The selectivity of the jet system may be improved by performing a gas phase chemical separation in the jet during transport of the stopped recoils.[46]

Identification of the collected reaction products can be made with a variety of techniques. Perhaps the most important of these techniques is the "mother-daughter" or "double-recoil" method which establishes a genetic link between the unknown reaction product and known daughter and/or grand-daughter activities. In this technique (see Figure 3), the recoil heavy atom produced by the alpha-decay of the collected initial reaction product strikes and imbeds itself in a "mother crystal". The mother crystal is then moved in front of a "daughter crystal" which can detect the alpha-decay of the imbedded atom in the mother crystal. The procedure can be extended to detect additional descendents in the alpha-decay chain. If the alpha-particle decay characteristics of the daughter, grand-daughter, etc., nuclei are known, then a genetic link is established and the (Z, A) of the parent are established. This technique was used in the discovery

of several elements and isotopes.[47,48,7,49]

A newer technique of exceptional power to identify the Z of collected reaction products is the X-ray method.[50] In this technique the coincidences between the alpha-particles emitted by the decay of the collected recoils and the K X-rays of the daughter nuclei (produced as a result of internal conversion decay in the daughter) are observed. The energies and relative intensities of the X-ray lines,[50,51] serve to identify the Z of the daughter, and therefore the parent nucleus.

For species whose half-lives are in the range 1 ms $\leq t_{\frac{1}{2}} \leq$ 100 ms, the product collection device is placed in close proximity to the irradiated target and catches the recoils emerging from the target directly. In such systems, the heavy ion beam after passing through the target will strike the collection surface (drum, tape, etc.). Schematic diagrams of two such collection devices, are shown in Figures 4 and 5.[52] Unfortunately, such devices offer no selectivity as to which reaction products are collected, the recoils are usually implanted so deeply that alpha spectra are those obtained from a very thick source, and it is difficult to detect the radioactive decay of the reaction products amidst a high beta-particle background. Therefore these devices are used frequently to detect new spontaneously fissioning nuclides. Since spontaneous fission cannot, in general, be used to identify the Z and A of the fissioning system, experimenters frequently resort to arguments based upon nuclear reaction energetics, systematics and excitation functions to identify the collected products. Such identifications are generally considered unreliable and make up the bulk of those identifications classified as E, F and G by the Table of Isotopes compilers.[53]

Magnetic Spectrometers, Velocity Filters. The principal problem with the isolation devices discussed above (tapes, jets, etc.) is that the reaction pro-

duct must be stopped and mechanically transported to radiation detectors before product identification can occur. This restricts their use to studies of nuclei whose $t_{\frac{1}{2}} \geq 1$ ms. For detection and identification of species whose $t_{\frac{1}{2}} \geq 1$ μs, an instrument based upon magnetic and/or electrostatic deflection of target recoils can be employed. One of the most successful of these devices in recent years is the velocity filter SHIP (Separator for Heavy Ion Reaction Products) located at the UNILAC at GSI.[54,11] A schematic diagram of this separator is shown in Figure 6. Evaporation residues produced in a nuclear reaction emerge in the forward direction from the target and pass through a thin carbon foil which has the effect of equilibrating the ionic charge distribution of the residues. The ions then pass through two filter stages consisting of electric deflections, dipole magnets and a quadrupole triplet for focusing. The solid angle of acceptance of the separator is 2.7 msr with a separation time for the reaction products of approximately 2 microseconds. Since complete fusion evaporation residues have very different velocities than target-like transfer and deep-inelastic products, the separator with its ± 5% velocity acceptance range effectively separates the evaporation residues from other reaction products. Following separation, the residues pass through a large area time of flight detector and are stopped in an array of seven position-sensitive detectors. From their time of flight and their energy deposit in the position-sensitive detectors, a rough estimate of their mass may be obtained. The final genetic identification of the residues is made by recording the correlations between the recoil position in the detector and subsequent decay signals from alpha or spontaneous fission decay or even signals from gamma or X-ray detectors placed next to the position sensitive detector. This device has been used in the discovery of element 107[11] and element 109[13] and the identification of the new nuclides ^{247}Md, ^{243}Fm and ^{239}Cf.[12]

Separators like SHIP are quite expensive and represent major instrumentation projects. A less sophisticated spectrometer which costs considerably less and provides the capability to measure the formation cross sections, recoil range distributions and angular distributions of short-lived ($t_{\frac{1}{2}} \geq 1$ ms) alpha emitters formed in heavy ion reactions has been described by Dufour et al.[55]

Another type of device used to isolate and identify transuranium nuclei ($t_{\frac{1}{2}} > 1$ μs) is the gas-filled mass separator, typified by the separator SASSY (Small Angle Separator System) in use at the Lawrence Berkeley Laboratory.[56] In this system (Figure 7) the heavy product recoils from a nuclear reaction enter a helium-filled (1 torr pressure) magnetic spectrometer. The time of flight and energy of the recoil nuclei are measured, giving a rough determination of the product mass number. The recoil nuclei which are imbedded in the focal plane detectors are identified by their α-particle decay and the decay of their daughters. SASSY has been used to discover a new $_{86}$Rn isotope produced in the bombardment of $_{50}$Sn nuclei with $_{86}$Kr projectiles[57] and its efficacy in the transplutonium region has been demonstrated by its use to observe the production of ^{254}No from the ^{208}Pb (^{48}Ca, 2n) reaction.[58]

Time of Flight (TOF), Decay in Flight (DIF) and Blocking Techniques. To detect species whose lifetimes are substantially less than 1 microsecond, special techniques must be employed. They include time of flight (TOF) techniques which when combined with a measurement of the product energy will give information about the product mass number. For suitable mass resolution, the time of flight must be approximately 10 - 100 nanoseconds. When searching for rare events, some selection process (like SHIP or SASSY) must be employed to reduce the "background" levels in the apparatus. The decay in flight technique whose use is described in Reference 59, and the crystal blocking technique,[60] ($10^{-18} \leq t_{\frac{1}{2}} \leq 10^{-14}$ sec) give very little information about the identity of a reaction product other than its existence and its approximate lifetime.

FUTURE DIRECTIONS

New Technical Developments. Of the chemical isolation and identification techniques to be used in the future, those capable of being used for short half-lives are likely to find the most use. The aqueous chemistry and volatility separation techniques[61] that have been used in off-line experiments will need to be further adapted to on-line operations to make it possible to detect short-lived products. The use of volatility separation techniques is of particular interest because some of the superheavy elements have been predicted to be very volatile (number 112) or quite volatile (number 114) in the elemental (metallic) form.[62] In addition, chemical isolation and identification techniques will probably use the more modern physical chemical techniques. An example of these techniques is the use of lasers[63] to do single atom detection as in the work of Bemis et al.[64] to measure the optical isomer shift for the 1 ms spontaneously fissioning isomer ^{240}Am. The other traditional identification techniques involving genetic identification via observations of decay chains or direct measurement of product Z by X-ray or photoelectron detection appear to be quite applicable to short half-life species, especially with improvements in detector efficiency. Clearly the fast isolation techniques such as the magnetic spectrometers or velocity separators will have special importance particularly if adapted to study transuranium nuclide production by a variety of different reaction mechanisms. For a number of the transuranium production methods of the future, new target technologies, similar to those currently used at the ISOLDE facility for p-nucleus reactions,[65] to allow the use of higher intensity, heavy ion beams, will have to be developed.

Light Heavy Ion Transfer and Deep Inelastic Transfer Reactions. As indicated above, current research in the use of light heavy ion transfer[20,21,22] and deep inelastic transfer[32,36] reactions has progressed far enough to indicate that transfer reactions involving heavier targets such as ^{249}Cf or ^{254}Es could lead to

the production of more neutron rich heavy actinides. It should be possible to produce and identify important new isotopes of transactinide elements. In addition to the near transactinide elements, the possibility might extend as far up as the superheavy elements. Hoffman[66] has speculated that in the $^{48}Ca + ^{248}Cm$ reaction one might be able to produce $^{288}112$ or $^{291}113$ in a relatively "cold" manner.

Cold Fusion Reactions. The success in synthesizing elements 107 and 109 using cold fusion reactions has revived interest in the use of the ^{48}Ca and ^{248}Cm reaction to make superheavy elements. Current attempts,[67] still in progress, to synthesize superheavy nuclei with the $^{48}Ca + ^{248}Cm$ cold fusion reaction, are using bombarding energies closer to the interaction barrier. These collaborative experiments are using off-line chemical, including volatility, procedures and the on-line detection devices SASSY at LBL and SHIP at GSI. Previous experiments[68,69] that give negative results had been carried out at approximately 20 MeV above the barrier because theoretical estimates indicated that beyond a certain critical size of projectile and target an extra energy above the interaction barrier would be required to fuse the nuclei.[70-72] The critical nuclear sizes, above which this "extra push" phenomenon would set in, were not known at the time. If the $^{48}Ca + ^{248}Cm$ system turned out to be beyond the critical size, then an extra bombarding energy would be needed for fusion, but whether this would do more harm than good was an open question. Experimental confirmation of the extra push phenomenon is accumulating in recent studies,[73-78] and estimates of the critical nuclear reaction sizes are becoming available. They indicate that the $^{48}Ca + ^{248}Cm$ reaction is, indeed, close to the critical condition, but the qualitative description of the extra push phenomenon and of the subsequent fission vs. neutron emission competition is not sufficiently precise

to enable one, even now, to decide whether an extra bombarding energy would or would not be advantageous as regards the final probability of making a superheavy nucleus. It is still necessary to make a careful experimental study of the relevant cross sections as a function of energy, in a range from somewhat below to somewhat above the interaction barrier.

Secondary Beams. One interesting idea for using heavy ion reactions to synthesize transuranium nuclei has been put forth by Dufour, Fleury and Bimbot.[79] The basic idea is to use a heavy ion reaction to create an "exotic" secondary beam with which to do the actual synthesis reaction. The principal difficulty is that such processes involve the product of the probabilities of two events, the initial beam generation reaction and the synthesis reaction. However, as Dufour, Fleury and Bimbot show, there are some particularly attractive secondary beam possibilities. For example, with the use of projectile fragmentation reactions in the intense 10^{12} part/sec 86 MeV/n heavy ion beams at the SC synchrocyclotron at CERN, one might expect to produce ^{16}C beams with an intensity of approximately 10^{9} part/sec. The use of such neutron-rich secondary beams in complete fusion reactions might lead to the production of 10-100 atoms/hr of $^{260,261}No$. Similar studies with proton-rich secondary beams are estimated to produce 10^{1}-10^{2} atoms/hr of $^{237,238,239}Bk$. These examples are only illustrative of the many possibilities.

Exotic Targets. Because of its large number of neutrons (155) the isotope ^{254}Es may be the best route to the synthesis and identification of the superheavy elements. This is the case if nuclear species containing 184 neutrons or very close to 184 neutrons must be produced in order to have a half-life sufficiently long to be detectable. To reach this goal ^{48}Ca would have to be used as the projectile. This combination might produce a nuclear species containing 182 neutrons by a reaction utilizing the one neutron channel (cold nucleus intermediate) as follows:

$$^{254}_{99}Es + ^{48}_{20}Ca \rightarrow ^{301}119 + n$$

The isotope ^{254}Es is difficult to produce even in microgram quantities, but it is probably the best available target material. Other possibilities are more difficult to realize. The isotope ^{255}Es as target material could lead to an odd-odd nuclear species with 183 neutrons (even more desirable) but it would be very difficult to produce more than nanogram quantities reasonably free of its intensely radioactive precursor (^{253}Es) in the chain of neutron capture reactions required for its production; the 40-day half-life of ^{255}Es is more difficult to deal with than the 275-day half-life of ^{254}Es.

Another desirable material is ^{250}Cm, with 154 neutrons, but this could be made available only be recovering it from the debris of underground nuclear explosions, an expensive undertaking. Here the one neutron channel with the reaction:

$$^{250}_{96}Cm + ^{48}_{20}Ca \rightarrow ^{297}116 + n$$

would produce a product of smaller atomic number (presumably an advantage) with 181 neutrons.

IMPORTANCE OF TRANSPLUTONIUM RESEARCH

There is much to be learned about nuclear reactions and their products by continuing and extending the study of the reactions of heavy ions with transplutonium target nuclei. Especially through the use of the heaviest available target nuclei, such as ^{248}Cm, ^{249}Cf, and ^{254}Es, and possibly ^{250}Cm and ^{255}Es, it will be possible to produce interesting new high Z actinide and transactinide isotopes. The detemination of their decay properties will make it possible to determine the role of the 152 neutron shell in this region.

With the competing fission reaction playing a role as a monitor of nuclear

temperature it should be possible to reach a better understanding of the puzzling mechanism of transfer reactions initiated by light heavy ions. Similarly, the competing fission reaction should provide an additional dimension in helping to measure the distribution of excitation energy as a function of the number of nucleons transferred in the deep inelastic transfer reaction initiated by the heaviest heavy ions, and in studying the degree of coldness of heavy nuclei produced in fusion reactions. Such information should also throw light on the behavior of nuclei as a function of angular momentum.

If it should prove possible to produce and identify superheavy elements, we will learn much about fission barriers and the nature of closed shells in a region of proton and neutron numbers well beyond where we now have an understanding of these quantities. The nuclear decay properties of superheavy elements are impossible to predict with any degree of accuracy and thus an experimental knowledge of these properties, and the decay sequences of genetically related nuclei, would provide information to put our understanding of the nuclear structure of the very heaviest nuclei on a sounder basis.

The chemical properties of the superheavy elements will be of extraordinary interest because of the importance of relativistic effects in determining their electron configuration.[62] For example, the six 7p electrons are predicted to be split into two groups, four $7p_{3/2}$ and two $7p_{1/2}$ electrons, with the splitting between their energies being such that the filled $7p_{1/2}^2$ subshell will act as a closed shell and additonal $7p_{3/2}$ electrons will act as electrons outside a closed shell. As an example of this effect, element 115 (ekabismuth) is predicted to have its valence electrons in the configuration $7p_{1/2}^2\ 7p_{3/2}$ with a stable +1 oxidation state, in contrast to the stable +3 oxidation state of its homologue bismuth. Thus chemists are excited about this possibility of studying "relativity in a test tube". The chemical properties of the near transactinide elements which have not been studied so far (nos. 105, 106, etc.)

should be compared with those of their homologues (tantalum, tungsten, etc.).

Research concerned with the limits of the periodic table of elements has been growing more and more demanding as the lifetimes of the isotopes in question decrease into the millisecond range or less, and the cross sections for producing them plunge into and below the nanobarn regime. Past progress in this field has already relied heavily on having available the right targets and projectiles. There are many examples where the substitution of one combination for another can improve the cross sections for making the desired isotope by orders of magnitude. This situation will become ever more drastic in the future, when it will be more and more common that a new element or isotope can only be made in a single, highly specific reaction, involving a unique combination of target and projectile. In particular, it is clear that reaching the predicted island of superheavy elements will not be easy, and if this quest is eventually crowned with success, it will most likely involve exotic transplutonium targets such as ^{248}Cm, ^{250}Cm, ^{254}Es or ^{255}Es. A whole area of research at the extreme limits of the periodic table may then hinge on the availability of some such exotic target material: only the laboratory fortunate enough to have access to such a target will be able to explore this field of research. If on an expedition you come to a crevasse 20 feet wide and want to explore the land beyond, you either have a 20+ foot plank available or you don't reach that land. A 19 foot plank will not do.

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FIGURE CAPTIONS

Figure 1. Classification of techniques used to isolate transuranium reaction products by minimum detectable $t_{\frac{1}{2}}$ and production cross section. See text for a discussion of these techniques. From (Reference 37).

Figure 2. Schematic diagram of chemical procedures used by Schädel *et al.*[36] to isolate actinide elements from heavy ion irradiated U targets.

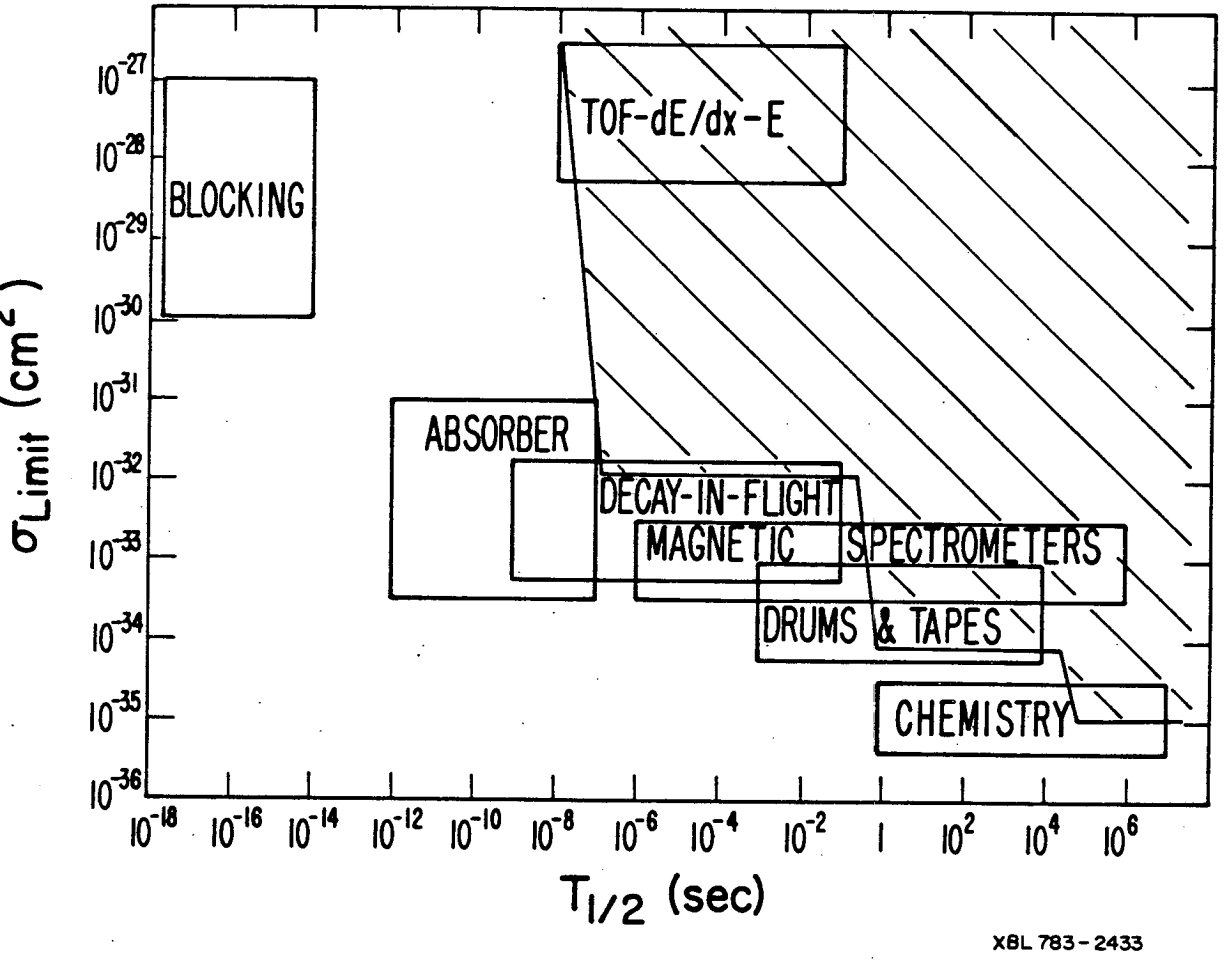
Figure 3. Schematic representation of a "gas-jet" recoil transport assembly. Thermalized product atoms are transported in the He gas stream and collected on the periphery of a wheel or other suitable collection device. Periodically, the wheel is moved to position the spot in front of the detectors. A "mother-daughter" detector assembly is illustrated in the lower portion of the figure and is used to establish a genetic link. From Bemis (Reference 45).

Figure 4 Rotating and scanning drum system for the detection of short-lived spontaneously fissioning nuclei. From (Reference 52)

Figure 5 Details of a tape system for the collection and detection of short-lived spontaneously fissioning nuclei. From (Reference 52).

Figure 6 A schematic diagram of the velocity filter SHIP at GSI.

Figure 7 A schematic diagram of SASSY at LBL.



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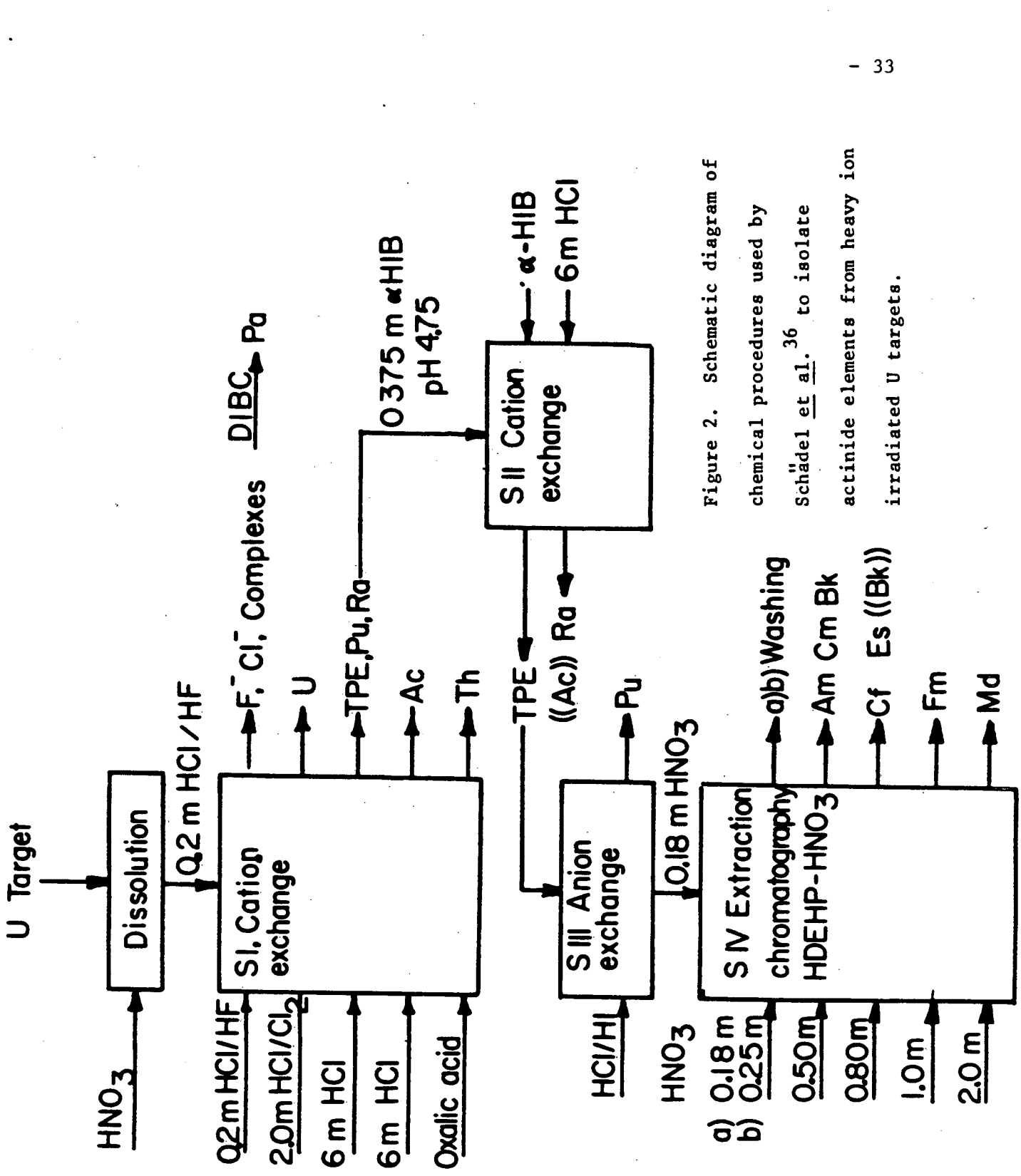

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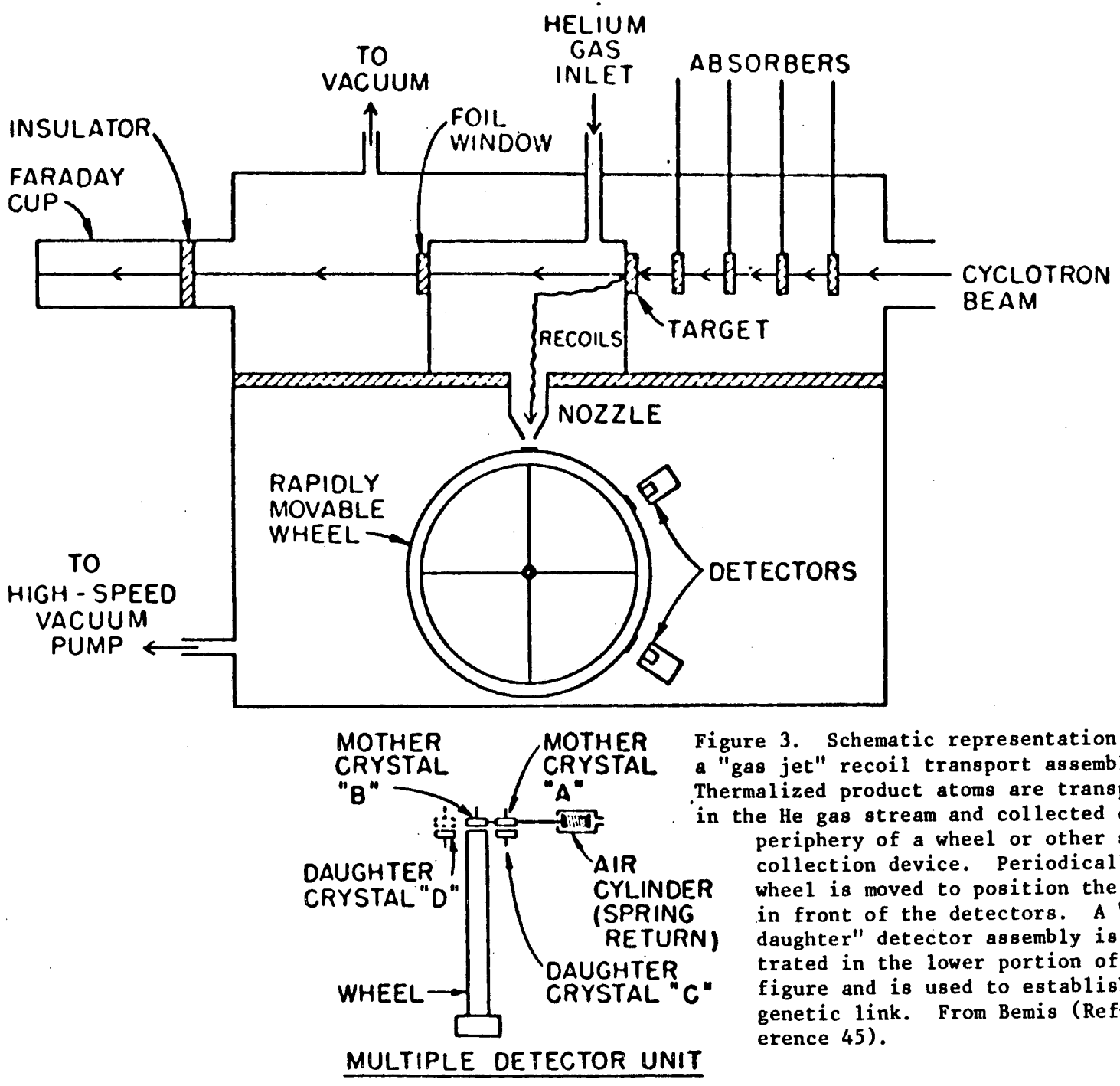

Figure 3. Schematic representation of a "gas jet" recoil transport assembly. Thermalized product atoms are transported in the He gas stream and collected on the periphery of a wheel or other suitable collection device. Periodically, the wheel is moved to position the spot in front of the detectors. A "mother-daughter" detector assembly is illustrated in the lower portion of the figure and is used to establish a genetic link. From Bemis (Reference 45).

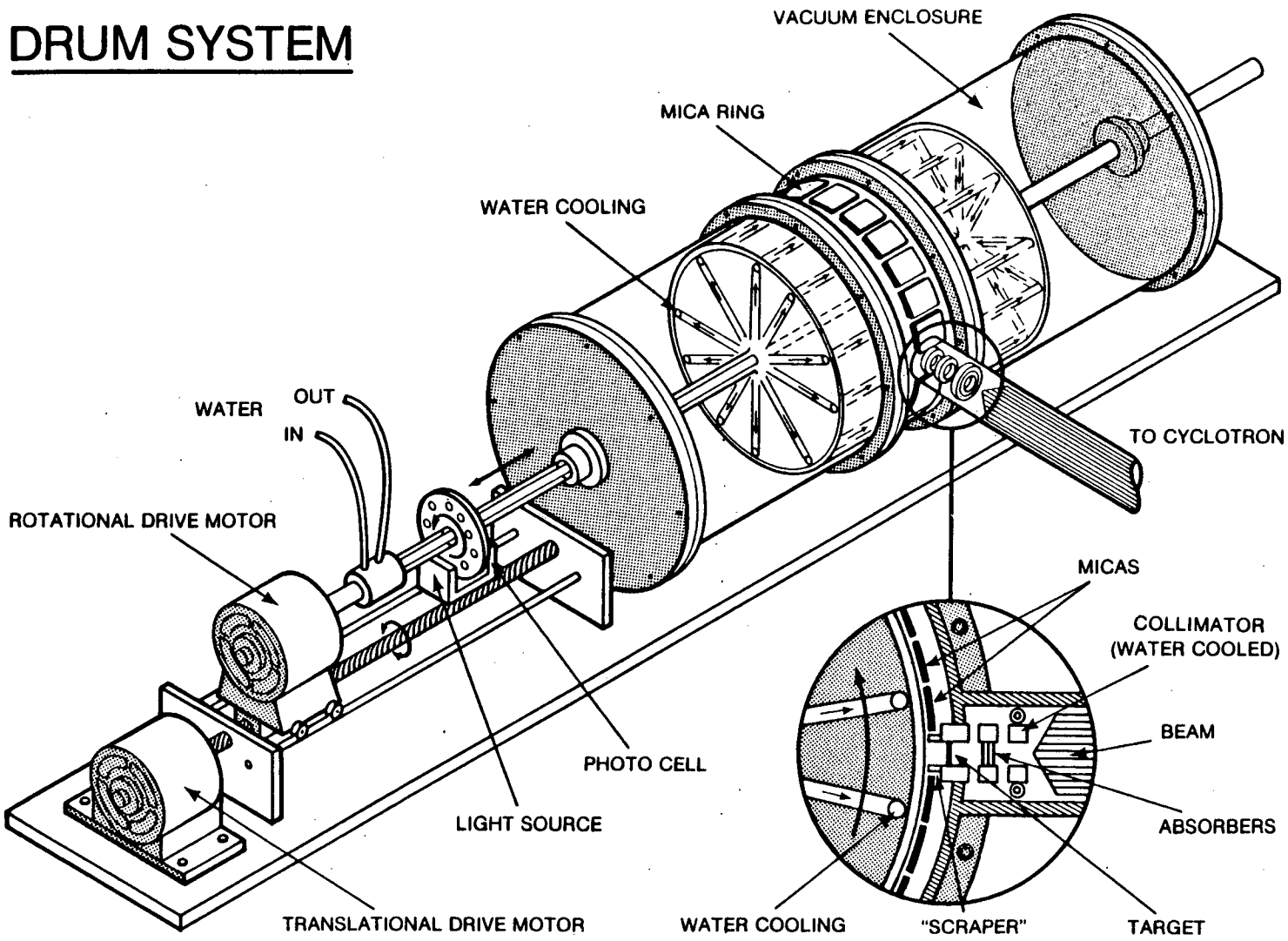

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Figure 4. Rotating and scanning drum system for the detection of short-lived spontaneously fissioning nuclei. From (Reference 52).

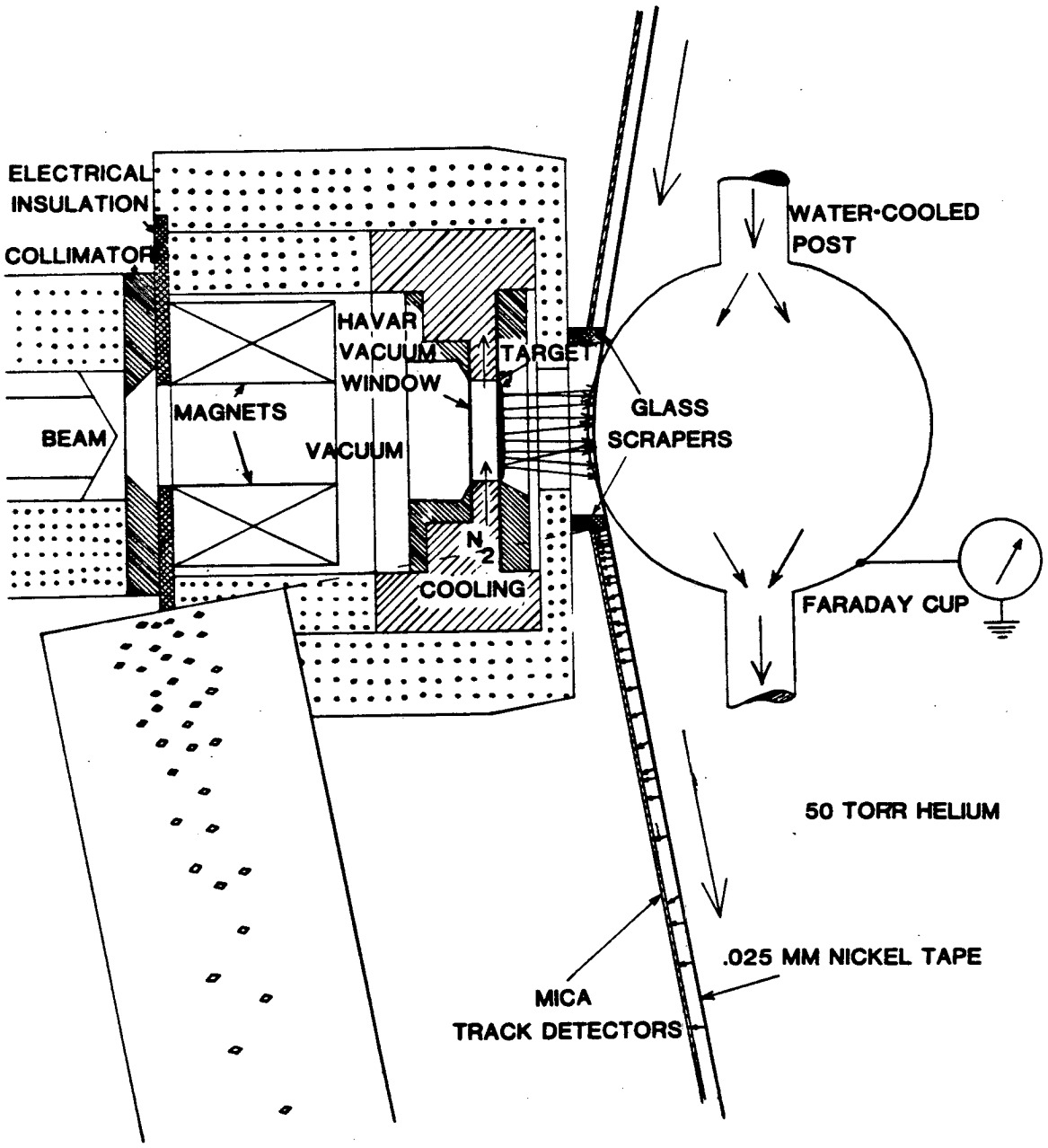


Figure 5. Details of a tape system for the collection and detection of short-lived spontaneously fissioning nuclei. From (Reference 52).

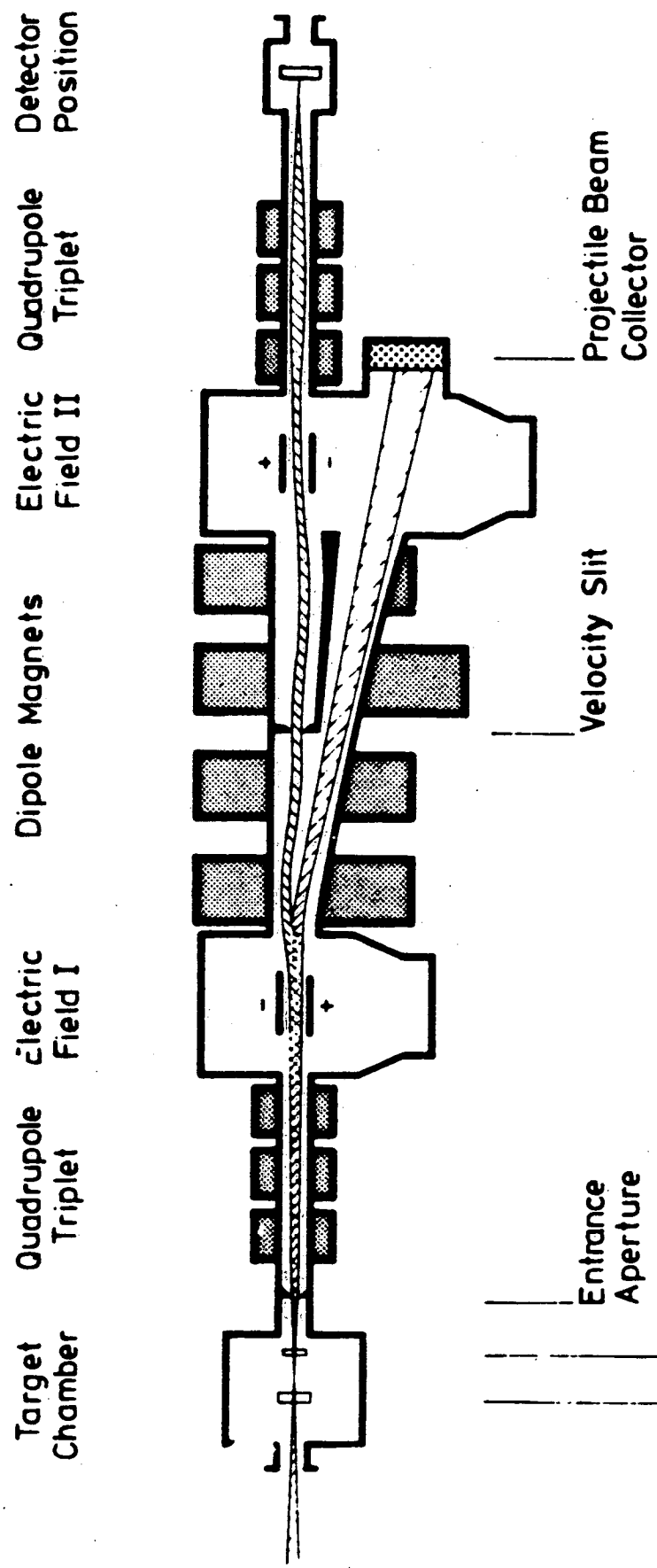
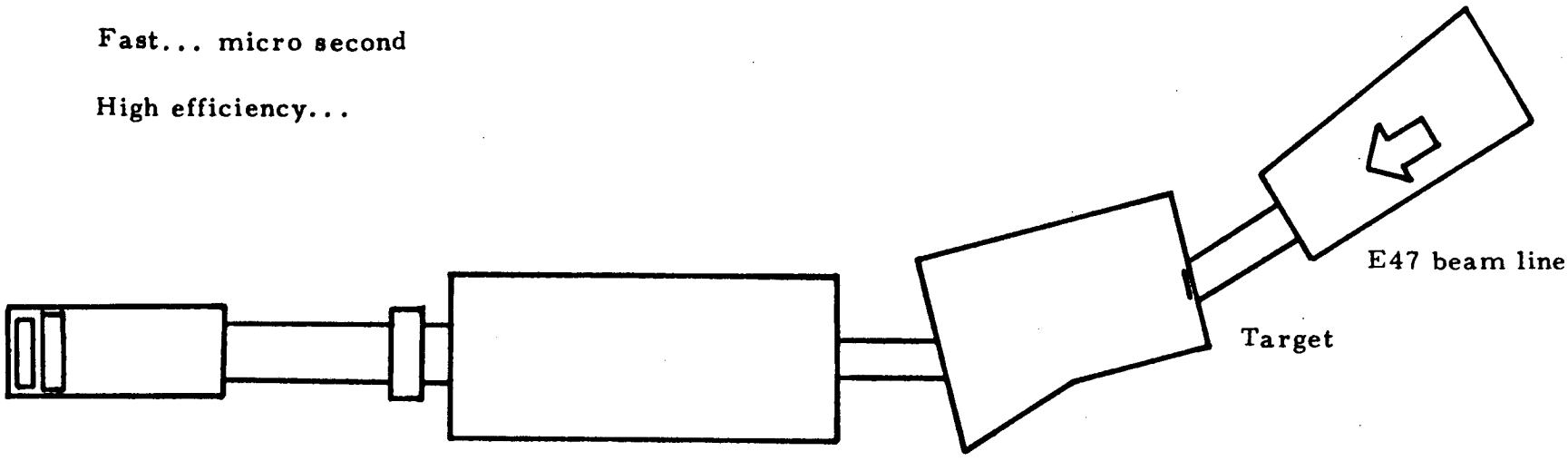


Figure 6. A schematic diagram of the velocity filter SHIP at GSI

SASSY (Small Angle Separator SYstem)

Fast... micro second

High efficiency...



Detector Array

Quadrupole Magnet

Bending Magnet

a. 2 position sensitive proportional counters

b. 10 Si crystal array

Recoil energy

α, S.F

Time of flight

a. Suppression of beam.

b. Mass separation.

14 feet from target to focal plane

Operating in 1 torr He

Figure 7. A schematic diagram of SASSY at LBL

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