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THE APPLICATION OF ALPHA SPECTROMETRY TO THE DISCOVERY OF NEW ELEMENTS BY HEAVY-ION BEAM BOMBARDMENT*

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Abstract:

Starting with polonium in 1898, α-spectrometry has played a decisive role in the discovery of new, heavy elements. For even-even nuclei, α-spectra have proved simple to interpret and exhibit systematic trends that allow extrapolation to unknown isotopes. The early discovery of the "natural" α-decay series led to the very powerful method of "genetically" linking the decay of new elements to the well-established α-emission of "daughter" and "granddaughter" nuclei. This technique has been used for all recent discoveries of new elements including Z = 109. Up to mendelevium (Z = 101), thin samples suitable for α-spectrometry were prepared by chemical methods. With the advent of heavy-ion accelerators new sample preparation methods emerged. These were based on the large momentum transfer associated with heavy-ion reactions, which produced energetic target recoils that, when ejected from the target, could be thermalized in He gas. Subsequent electrical deposition or a He-jet technique yielded samples that were not only thin enough for α-spectrometry,

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but also for α- and β-recoil experiments. Many variations of these methods have been developed and will be covered in this paper. For the synthesis of element 106 an aerosol-based recoil transport technique was devised. In the most recent experiments, α-spectrometry has been coupled with the magnetic analysis of the recoils. The time from production to analysis of an isotope has thereby been reduced to 10^{-6} s; while it was 10^{-1}-10^{0} s for He-jets and 10^{1}-10^{3} s for rapid chemical separations. Experiments are now in progress to synthesize super heavy elements (SHE) and to analyze them with these latest techniques. Again, α-spectrometry will play a major role since the expected signature for the decay of a SHE is a sequence of α-decays followed by spontaneous fission.

I. Introduction

In 1898 P. and S. Curie published a paper "On a New Radio-active Substance Contained in Pitchblende" [1]. The last sentence of their paper reads, "Perhaps we may be permitted to remark that if the existence of a new element is confirmed, this discovery will be due solely to the new method of investigation which the Becquerel rays provide." Thus starts the story of the application of α-spectrometry to the discovery of new elements.

In 1898 the Curies reported the discovery of the two elements, polonium and radium. They also took the first steps from α-spectroscopy to α-spectrometry by developing the forerunner of the ionization chamber, which they called "parallel-plate apparatus". A few years later Rutherford determined the e/m ratio of the "α-rays" and concluded that their mass was of the same order of magnitude as the hydrogen atom [2], and his magnetic measurements showed that their specific energy was 3.2 MeV/A. Another important discovery made by Rutherford and Soddy was the radioactive decay series. This method of linking known to unknown elements via their α-decay sequences is still one of the most powerful means for the discovery and identification of new elements.

II. The Synthesis of New Elements

All elements between uranium and lead, with the exception of astatine were discovered as members of the radioactive decay series of thorium, actinium, and uranium/radium. Neptunium was the first transuranium element of the actinide series to be synthesized in a nuclear reaction with neutrons on a uranium target. In the same year (1940) astatine was discovered in the bombardment of bismuth with α-particles. Subsequently elements up to fermium (Z = 100) were produced in neutron or light ion (≤He) reactions on heavy

targets. All these discoveries involved steps to separate the new element from the target material by chemical methods based on the predicted chemical properties of the actinide series. The chemical procedures yielded samples thin enough for off-line α-spectrometry with grid chambers. A new element was in most cases characterized by a specific α-energy (or energies) and a partial half-life for α-decay.

Compared to α-spectrometry the study of β- and γ-decay plays only a minor role in the discovery of new elements even today. There are several reasons for this: (1) α-particles can be detected with nearly 100% efficiency using ionization chambers or surface barrier detectors (SBD); (2) the detectors are essentially background free which, combined with a 100% detection efficiency results in high sensitivity; (3) an SBD can function as a particle identifier if the sensitive depth is only slightly larger than the range of the most energetic α-particles; under these conditions the probability of absorbing high energy β- or γ-rays is very small; (4) for α-particles of 5 to 10 MeV energy the resolution of modern (cooled) SSD's approaches 10 keV or 0.1%, which is sufficient to reveal fine details in the level structure of the residual (daughter) nucleus.

On the other hand fission fragments that are frequently observed in the decay of heavy elements are detected with the same efficiency as α-particles. However, they rarely create confusion since their energy is about one order of magnitude higher.

The main problem that stands in the way of the effective use of α-spectrometry in heavy element research is the preparation of thin samples; this will be discussed in more detail in the next chapters. An occasional problem is the pile-up between $\beta^{\pm}$- and α-particles due to the finite time resolution of the detection system. This is, however, quite rare and results

in a high-energy tailing of an α-line, which is equivalent to a decrease in resolution.

Early in the study of the α-decay systematics of the heavy elements, pronounced correlations were found between half-lives, energies, N, and Z-values. We now know from theoretical models that for nuclei far from the magic numbers Z = 82 and N = 126 the shell corrections to the nuclear mass formula are smooth functions of Z and N, which, when added to the smooth behavior of the liquid-drop or droplet model, give a simple relationship between the α-decay Q-value (Q_α) and the half-life ($T_{1/2}$); $\log T_{1/2} = A/\sqrt{Q_\alpha} + B$; where the constants A and B depend weakly on the Z of the element. The data for even-even nuclei are fitted well by the semiempirical formula [3]:

$$\log T_{1/2} = 1.61\ (Z\ E_\alpha^{-1/2} - Z^{2/3}) - 28.9\ .$$

Here E_α is in MeV, Z refers to the daughter nucleus, and $T_{1/2}$ is calculated in years. E_α shows an approximately linear rise with increasing Z for a family of isobars as can be deduced from semiempirical mass formulae. This accounts for the fact that beyond uranium the partial α-decay half-life of heavier elements becomes shorter, and S.F. decay starts to become more prevalent. Decreasing half-lives and decreasing production cross sections are the two principal difficulties in synthesizing new elements and the reason that the relatively slow chemical procedures had to give way to purely physical identification methods as will be discussed in the next chapter.

Another reason for the popularity of α-spectrometry is that the interpretation of many α-spectra is straightforward, in particular for even-even emitters, where most of the transition strength goes to the ground state of the daughter nucleus, and the branching that occurs to the 2^+ first

rotational level is a smooth function of the 0^+-2^+ level spacing. As the level spacing decreases because of an increase in the distortion of nuclei farther removed from the N = 126 neutron shell, a significant amount of transition strength does, however, go to the first 2^+ excited state of the rotational band.

The spectra of odd-nucleon α-emitters are in general much more complex, and the transition to the ground state is hindered. It is, however, possible to establish some order through the concept of "favored" α-decay [4], in which the last odd particle is in the same Nilsson orbital in the daughter as in the parent nucleus. Hindrance factors for such transitions are close to unity, since they resemble ground-state transitions in even-even nuclei. The more complex the rearrangement of the nucleons between α-emitter and final nucleus is, the larger, in general, the hindrance factor for α-decay; this becomes plausible considering the overlap integrals of the nuclear wave functions.

The α-particle, when emitted from a heavy nucleus, carries away about 2% of the nucleus' mass and imparts to the daughter nucleus a recoil energy of about 100 to 150 keV. A nucleus of this energy is able to traverse 10-50 $\mu g/cm^2$ of material and in 50% of all decays will therefore be "kicked out" of the sample if it is thin enough. For <u>very</u> thin samples even the recoil from β-decay is sufficient to eject the daughter nucleus. Extensive use has been made of these recoil effects to produce thin, secondary samples and to establish genetic relationships between successive members of an α-decay chain.

III. Techniques Developed for α-Spectrometry in Conjunction with Heavy-Ion Reactions

All elements above einsteinium (Z = 101) were synthesized with heavy ion beams. There are distinct advantages to heavy ions compared to α-, d-, p-, or n-projectiles as used in earlier experiments. The heavy-ion fusion product acquires a large forward momentum, which can propel it through target thicknesses in the order of 1 mg/cm^2. While in earlier experiments the target material had to be dissolved to isolate the reaction products, the recoil nuclei can now be stopped separately in a catcher foil. Another advantage of the heavy-ion recoil method is that very exotic targets can be used since the material is not lost to the chemist after each bombardment. The discovery of element 101, mendelevium, illustrates this point. Starting with plutonium placed inside a high flux reactor, a total of 10^9 atoms of ^{253}Es with a half-life of 20 days was produced. A tiny electroplated target was formed and bombarded by a He beam. The recoils were caught in a gold catcher foil, which was subsequently dissolved, and element 101 was isolated by a chemical procedure, which in turn yielded thin samples for α- and spontaneous fission (SF) counting [5].

In general the recoil products are imbedded so deeply in the catcher foil (several hundred $\mu g/cm^2$) that these samples are not suitable for direct α-spectrometry. The problem of sample thickness can, however, be solved elegantly by the use of a stopping gas [6]. As the recoils are slowed to thermal velocities in a noble gas (or N_2) at about 1 atm pressure a large percentage retains a positive charge and can be guided electrostatically to a catcher [7-9]. Samples obtained in this fashion are often as thin as one atomic monolayer and make excellent sources for α-spectrometry. A modern version of this technique has been developed by a French group [10] and is

shown in Fig. 1. The thermalized recoils are guided along the electric field lines and are deposited directly on an SBD. The total transport time is 10 ms.

For the discovery of nobelium (No, Z = 102) Ghiorso and co-workers refined the target recoil method into a "double recoil" technique [11]. The nobelium atoms produced in the reaction of ^{12}C ions with a curium target were stopped in He gas and attracted electrically to a moving, metallic belt that in turn passed near a negatively charged foil. The ^{250}Fm from the alpha decay of $^{254}102$ was "kicked" off the belt with about 50% probability and captured by the foil. The discovery of element 102 was then based on observing the 30-min α-activity of ^{250}Fm on the foil and thereby linking the new element/isotope to a known one via the genetic sequence $^{254}102 \xrightarrow{\alpha} {}^{250}Fm \xrightarrow[30\ min]{\alpha} ({}^{246}Cf)$.

An accidental observation that was made during experiments with He as a stopping gas led to the development of a new technique [12], which was used extensively in the discoveries of elements 102, 104, 105, and 106, and several of their isotopes. Figure 2 shows an early version of what was later called a He jet system. The heavy-ion beam from the accelerator passes through a set of degrader foils to adjust its energy to the peak of the excitation function of the isotope to be studied. It impinges on the target, and the recoiling fusion products are thermalized in a gas chamber that is filled with He of about 1 atm pressure. A small orifice lets some of the gas escape into a vacuum chamber. The rapidly moving He entrains with it the radioactive recoils and deposits them on the periphery of a wheel. Since the velocity of the He gas is in the sonic regime the recoils do not penetrate the metal of the wheel and thus form a thin sample ideally suited for α-spectrometry. The wheel is rotated by a stepping motor so that the irradiated spots face several SBD's in succession, and spectra similar to those shown in Fig. 3 are obtained. The half-life of a new isotope is calculated from the observed decay rate at each counting station.

In the early days of gas-jet systems the recoil transport mechanism was not well understood. It was occasionally observed that a very clean system gave poor yields, while gas that was contaminated with diffusion pump oil had a good transport efficiency. Now we know that the charged recoils are attaching themselves via Van-der-Waals forces to aerosol particles, which can remain suspended in a gas stream if they are of the correct size [13]. Worth mentioning is that as early as 1907 Mme. P. Curie had already observed that the "dust" in a vial seemed to become radioactive and settle on the bottom under the influence of gravity [14]. Once the crucial role of aerosols was discovered, a wide variety of them was studied: water, acetone, sodium chloride, potassium chloride, carbon tetrachloride, oil, ethylene, trichlorethylene, ethanol, ethylenglycol, and others. To initiate the formation of aerosols, condensation nuclei of silver chloride or silver iodide have been used as well as irradiation with ultraviolet light. The aerosol technique has allowed the transport of radioactive species over tens of meters in a few seconds. It has, however, a serious drawback for high-resolution α-spectrometry: since a non-negligible amount of aerosol material is deposited together with the radioactive substance, the sample becomes progressively thicker, and long-lived α-activities exhibit poor energy resolution due to a "cemetery effect".

Figure 4 shows the experimental arrangement that was used in the discovery of element 106. It incorporates many of the experimental developments that occurred before 1974 [15]. The intense heavy-ion beam from the SuperHilac enters the target chamber through a gas-cooled window. The $^{263}106$ atoms together with other transmutation products recoil from the target into a stopping gas chamber and are swept by a flow of helium (containing NaCl aerosols) through teflon tubing into an adjoining counting area. Here the recoils are deposited onto the periphery of a wheel with seven detector

stations that examines the samples for α- and S.F. decay. The main difference to the wheel system shown in Fig. 2 is that each detector station consists of two fixed and two movable SBD's instead of one fixed detector (cf. inset of Fig. 4). This permits the observation of the decay sequence $^{263}106 \xrightarrow[0.9s]{\alpha} {}^{259}104 \xrightarrow[3.0s]{\alpha} {}^{255}No \xrightarrow[3.1\ min]{\alpha} ({}^{251}Fm)$ using a double recoil method, which works in the following way: the set of seven detectors monitoring the wheel shuttles every six seconds to a low background position facing seven stationary detectors while another set of movable detectors resumes the monitoring of the wheel. In the case where the daughter ($^{259}104$) and granddaughter (^{255}No) nuclei remain on the wheel their α-decays are observed by subsequent detectors facing the wheel. In the event that a $^{259}104$ daughter recoils from the wheel onto a detector and the α-decay of this daughter is later observed with this detector in the off-wheel position, this detector is not returned to the on-wheel position until 10 minutes have elapsed. This time period permits an adequate opportunity for detecting the subsequent decay of the 3-min granddaughter, ^{255}No. A total of 22 atoms of recoil-transferred ^{255}Rf were observed to decay in the off-wheel detectors. Shortly after these daughter events, granddaughter ^{255}No α-particles were detected from four off-wheel daughter decays, thus establishing a genetic link from ^{245}No to the new element/isotope $^{263}106$.

Starting with element 101 (mendelevium) and culminating with the discovery of element 109, new elements have been synthesized and identified one atom at a time. This has been possible to a large extent because of the high detection efficiency, low background, and good resolution achieved in α-spectrometry.

IV. The Combination of Alpha Spectrometry with Other Analytical Tools

The discovery of a new element requires the unambiguous determination of its Z-value [16]. In the previous chapter the method of genetic linking via α-decay chains to determine Z was discussed. This method has worked well up to the latest discoveries of elements 107 [17] and 109 [18].

The classical way of determining the Z-value of a new element has, however, been through chemical experiments; Z = 104 is the last element with which this was possible [19]. The short half-life of $^{261}104$ (65 s) later required the development of the computerized, fast chemistry system shown in fig. 5 [20]. This system allows the study of the anionic-chloride complexes of element 104 on a "one-atom-a-time" basis. The final samples, obtained after chemical processing, were examined by twenty SBD's for the characteristic α-group of element 104 at 8.28 MeV. A major drawback of the chemical apparatus shown in Fig. 5 is its batch processing mode. Continuous gas-chemical methods that yield samples thin enough for α-spectrometry have been developed at GSI [21]. Gas chromotographic methods for elements up to 107 [22] were developed at Dubna. These methods do not yet lend themselves to on-line α-spectrometry and are only applicable to spontaneous fission emitters. They are, however, about one to two orders of magnitude faster than rapid solution chemistry.

A very powerful method of Z-identification is based on an α-x-ray coincidence technique [23]: The parent nucleus decays by α-emission to an excited state in the daughter nucleus, which subsequently deexcites by the internal conversion process. This process yields characteristic K-series x rays of the daughter element in coincidence with α-particles from the parent and provides an unambiguous Z-identification for both elements. While this method is very elegant, it suffers from the reduced efficiency of most coincidence experiments.

Being aware of the difficulties of Z-determinations by chemical means and x rays, experimentalists have designed an entire class of instruments for the somewhat easier task of determining the mass of unknown isotopes. Among these are: (1) α-recoil time-of-flight mass spectrometers [24], (2) He-jets coupled with ion sources and magnetic mass spectrometers [25], (3) direct mass analysis of singly charged, thermalized recoils [26], (4) conventional on-line isotope separators, (5) gas-filled magnetic spectrometers [27], and (6) velocity filters. While all these instruments have been more or less useful in the study of new isotopes, so far only the velocity filter SHIP [28] has contributed to the discovery of new elements. Details will be covered in another contribution to this conference. Relevant to the subject of α-spectrometry and the discovery of new elements is, however, a technique that was developed at GSI in connection with SHIP [29] and has also been used in Berkeley with the gas-filled separator SASSY [30] in the search for super heavy elements. The principle is shown in Fig. 6: The evaporation residues are implanted several μm deep in a position-sensitive surface barrier detector. In cases where the evaporation residues are α- or S.F. emitters the decay sequences of mother-daughter and granddaughter nuclei can be observed in $>2\pi$ geometry, whereby the position coincidence requirement results in a drastic reduction of "background" counts. This very powerful evolution of conventional α-spectrometry has been a key component in the discovery of elements 107 and 109 and has also been used to study several new isotopes of known heavy elements as will be discussed in another contribution to this conference.

V. Conclusion and Outlook

The previous chapters have given a very limited overview of the role α-spectrometry has played in the discovery of new elements up to 107 and 109. In 1978 attempts were made in Dubna to synthesize element 108 in the reaction $^{226}Ra(^{48}Ca,xn)^{274-x}108$ [31]. Only upper limits for α- and S.F. decay down to 1 ms half-life could be obtained. The SHIP velocity filter at GSI and the gas-filled spectrometer SASSY in Berkeley could in the future extend the detectable half-life range for element 108 down to the region of microseconds.

A discussion of the discovery of new elements would be incomplete without mention of super heavy elements (SHE). Many attempts have been made at their discovery in nature and their synthesis in heavy-ion bombardments, and an entire conference was dedicated to this subject [32]. At the time of this writing (April 1983) an intense effort is under way to try again to synthesize SHE's in the reaction $^{248}Cm(^{48}Ca,xn)^{296-x}116$. A collaboration of scientists from LBL and GSI recently performed a large number of experiments at both laboratories using SHIP, SASSY, and chemical methods to find an α-S.F. decay signature that would indicate the existence of elements beyond the known region. A possible decay sequence for element 116 formed in the above cold fusion reaction with x = 2 is shown in Fig. 7. Again, α-spectrometry will play a decisive role in identifying a SHE "event". Closer inspection of Fig. 7 shows that the expected α-energies are not extraordinarily high, and the observation of α-particles with say 10 MeV energy and a few seconds half-life would <u>by itself</u> not indicate an unusual event since it could belong to a high-spin isomer like ^{212}Po or another isotope near the target that was formed in a transfer reaction. The observation of <u>two</u> correlated α-particles <u>and</u> a correlated S.F. event with high total kinetic energy would, however, be an indication that the decay of a heavy parent nucleus was observed. Very

preliminary results from several months of experiments indicate that such an event was seen neither in the physical nor in the chemical experiments in either laboratory.

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Figure Captions

Fig. 1. Schematic view of an "in beam" recoil thermalization and collection system. The target recoils are thermalized in N_2 or He gas near the field line labeled "mean path" and are accelerated by a potential gradient towards the α-detector. The total transport time from the target to the detector is about 10 ms (from [10]).

Fig. 2. He-jet system coupled with a vertical wheel that transports the activity deposited by the He-jet to several counting stations along its periphery. This apparatus was used in the discovery of element 104 radioactivities [33].

Fig. 3. α-spectra observed at consecutive detector stations of a system similar to the one shown in Fig. 2. The spectrum labeled SUM is the sum of all five detectors. Note in spectrum 1 through 5 that several α-lines show a decay while others remain constant. This information was used to determine the half-life of a new isotope of element 105 (from [34]).

Fig. 4. Experimental setup used in the discovery of element 106. The principal new features compared to Fig. 2 are the use of an aerosol loaded He jet and the incorporation of three additional detectors at each station to observe three generation-decay sequences (from [15]).

Fig. 5. Schematic diagram of the computer-automated fast-chemistry equipment used in studying chloride complexation of element 104. The numbers refer to individual steps in the procedure (from [20]).

Fig. 6. Implantation of evaporation residues in a position-sensitive surface barrier detector and α-decay of the implanted nuclei (from [29]).

Fig. 7. Radioactive decay of the hypothetical compound nucleus $^{294}116$ formed in the reaction $^{248}Cm(^{48}Ca,2n)$. Half-life predictions and α-energies, which are shown in parentheses, were taken from [35]. Percent figures are branching ratios for α-decay and electron-capture (E.C.) decay.

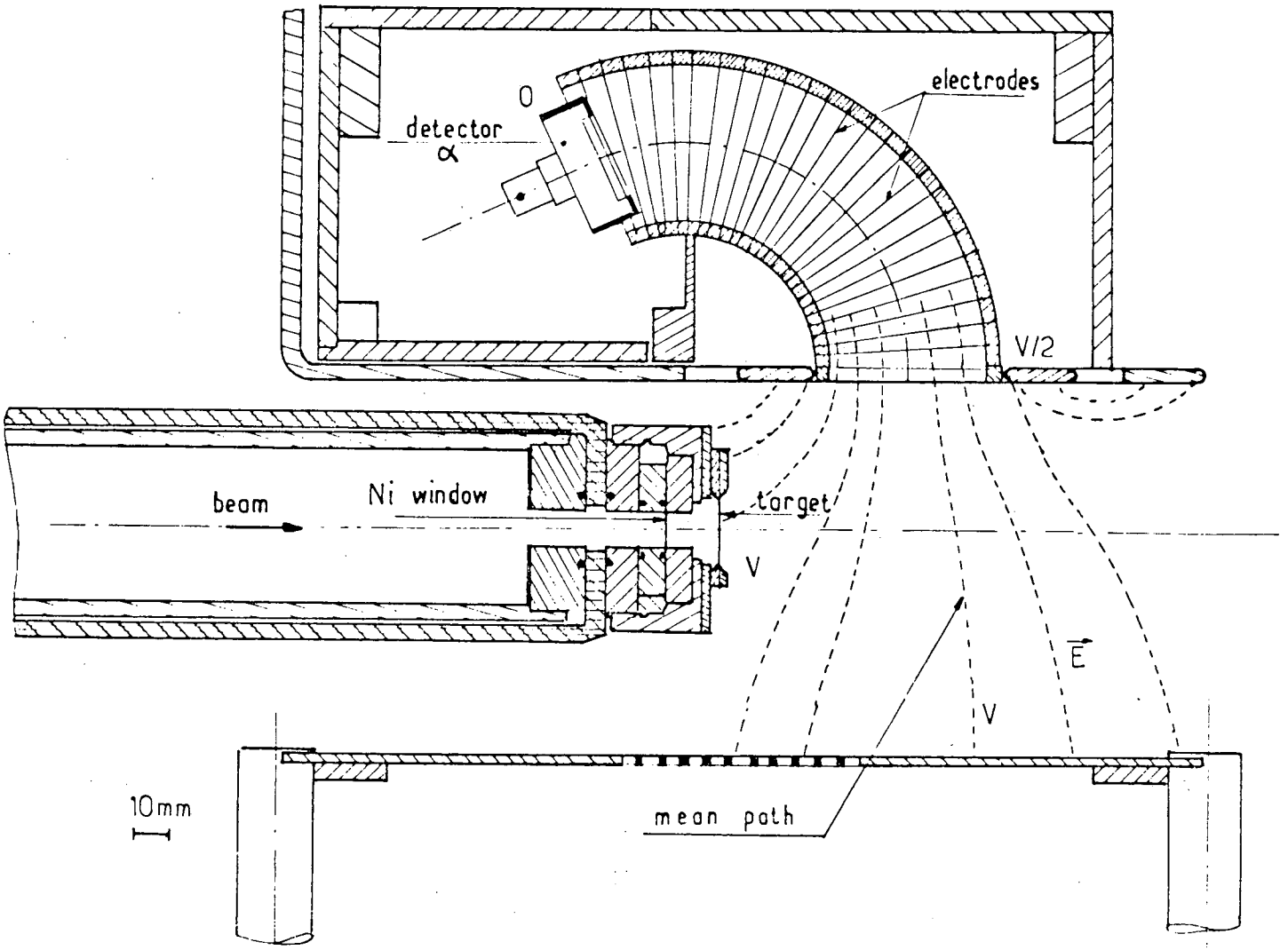

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Fig. 1

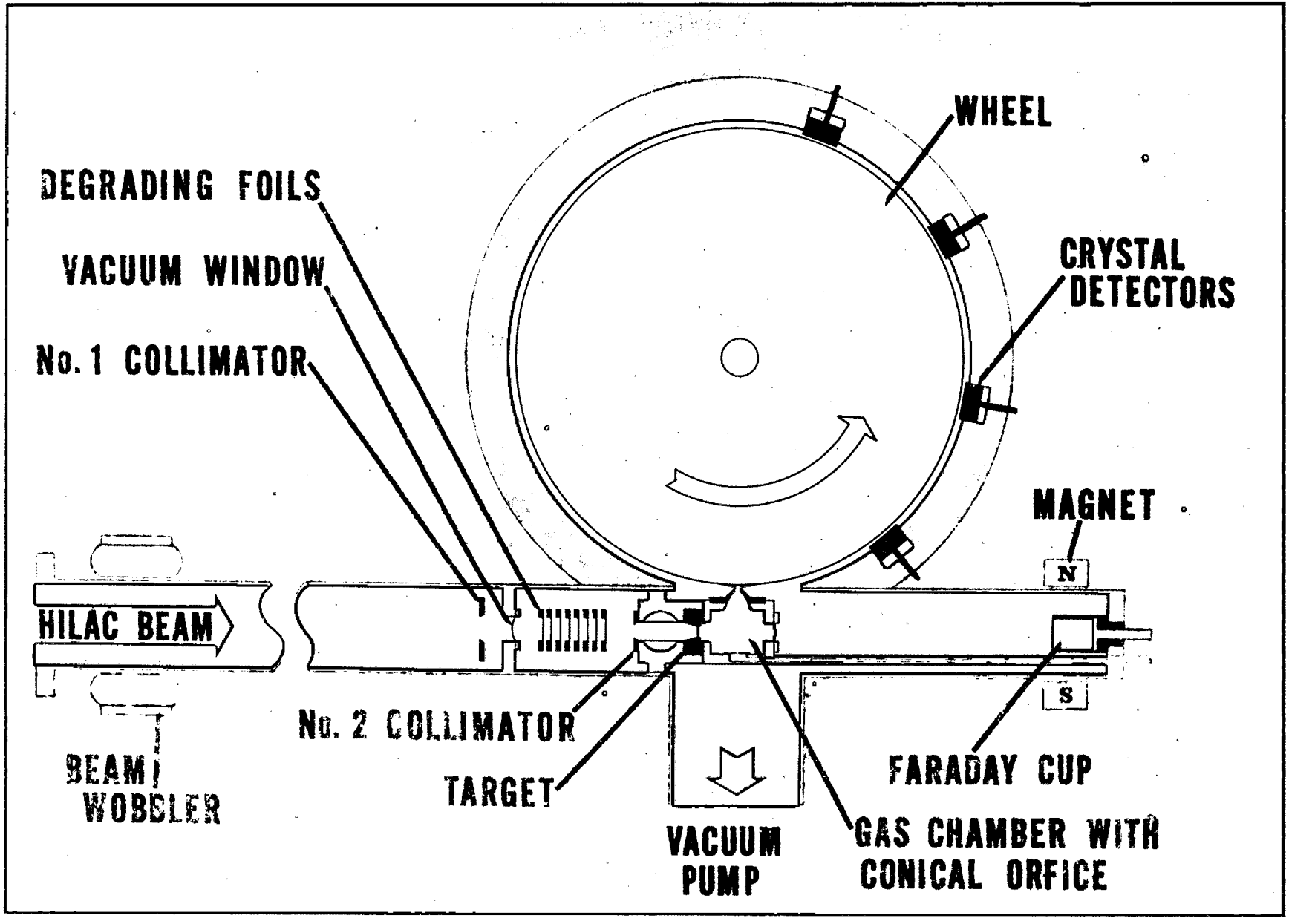
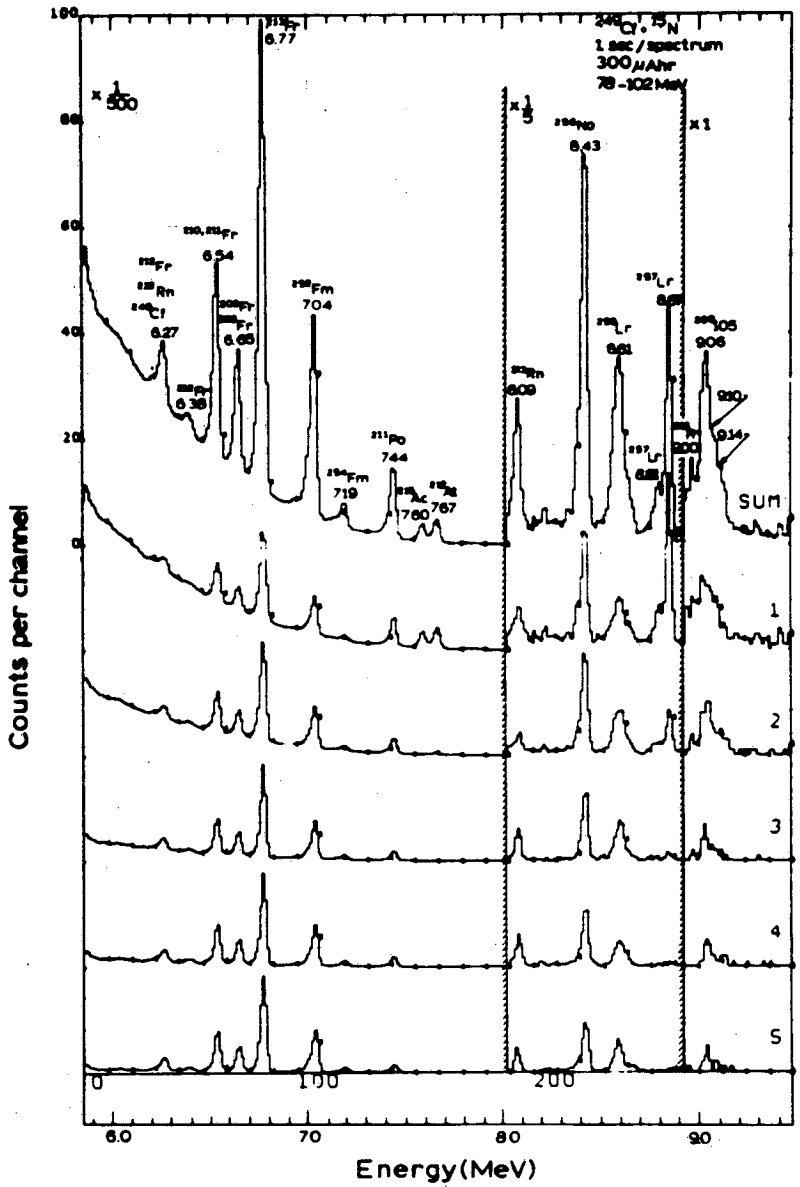


Fig. 2

CBB 689-5712



XBL 704 6171

Fig. 3

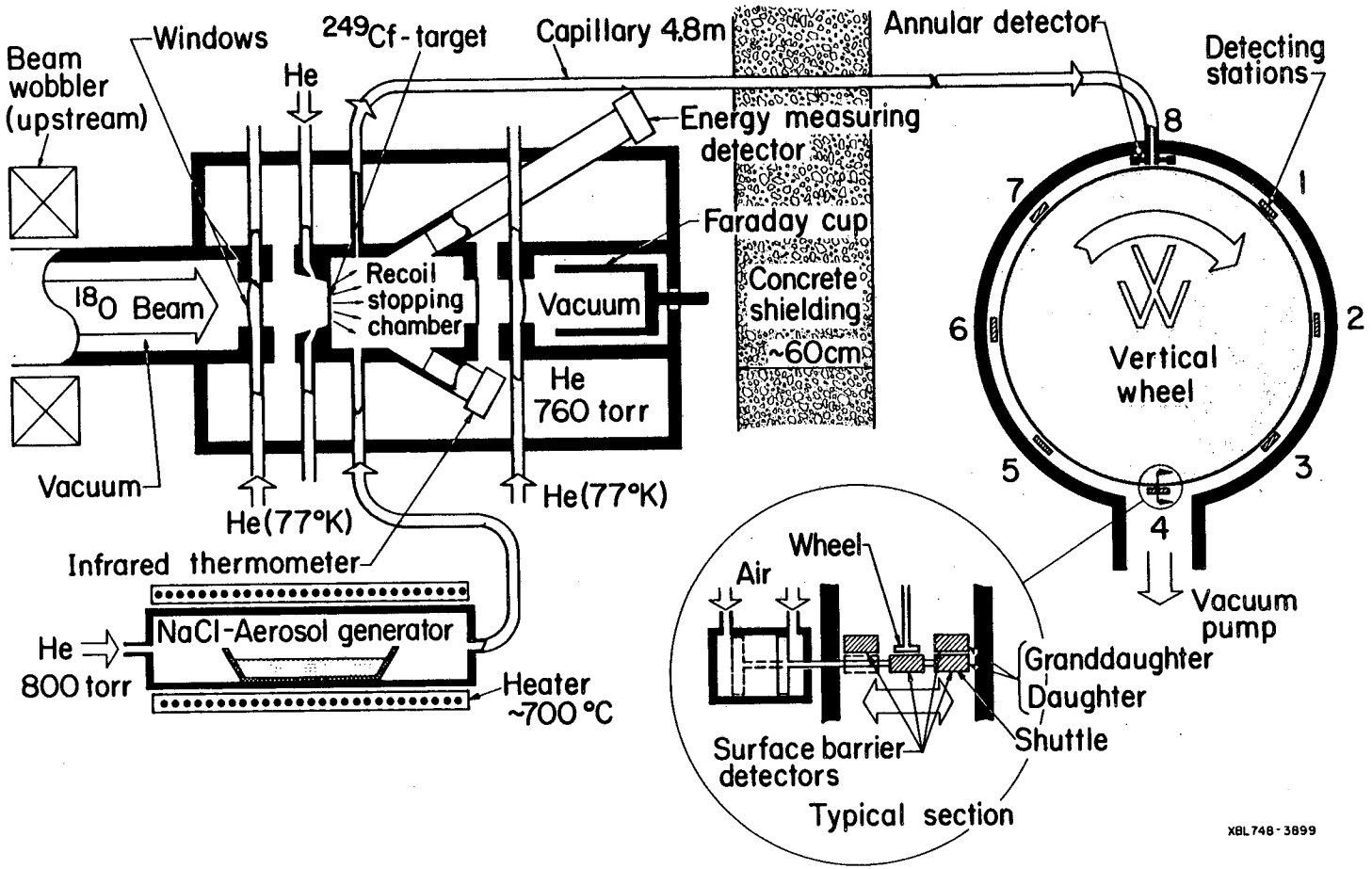

Beam wobbler (upstream)
Windows
He
249Cf-target
Capillary 4.8m
Annular detector
Detecting stations
Energy measuring detector
Faraday cup
Concrete shielding ~60cm
18O Beam
Recoil stopping chamber
Vacuum
He 760 torr
Vacuum
He(77°K)
He(77°K)
Infrared thermometer
He 800 torr
NaCl-Aerosol generator
Heater ~700 °C
Vertical wheel
1
2
3
4
5
6
7
8
Vacuum pump
Wheel
Air
Granddaughter
Daughter
Shuttle
Surface barrier detectors
Typical section
XBL748-3899

Fig. 4

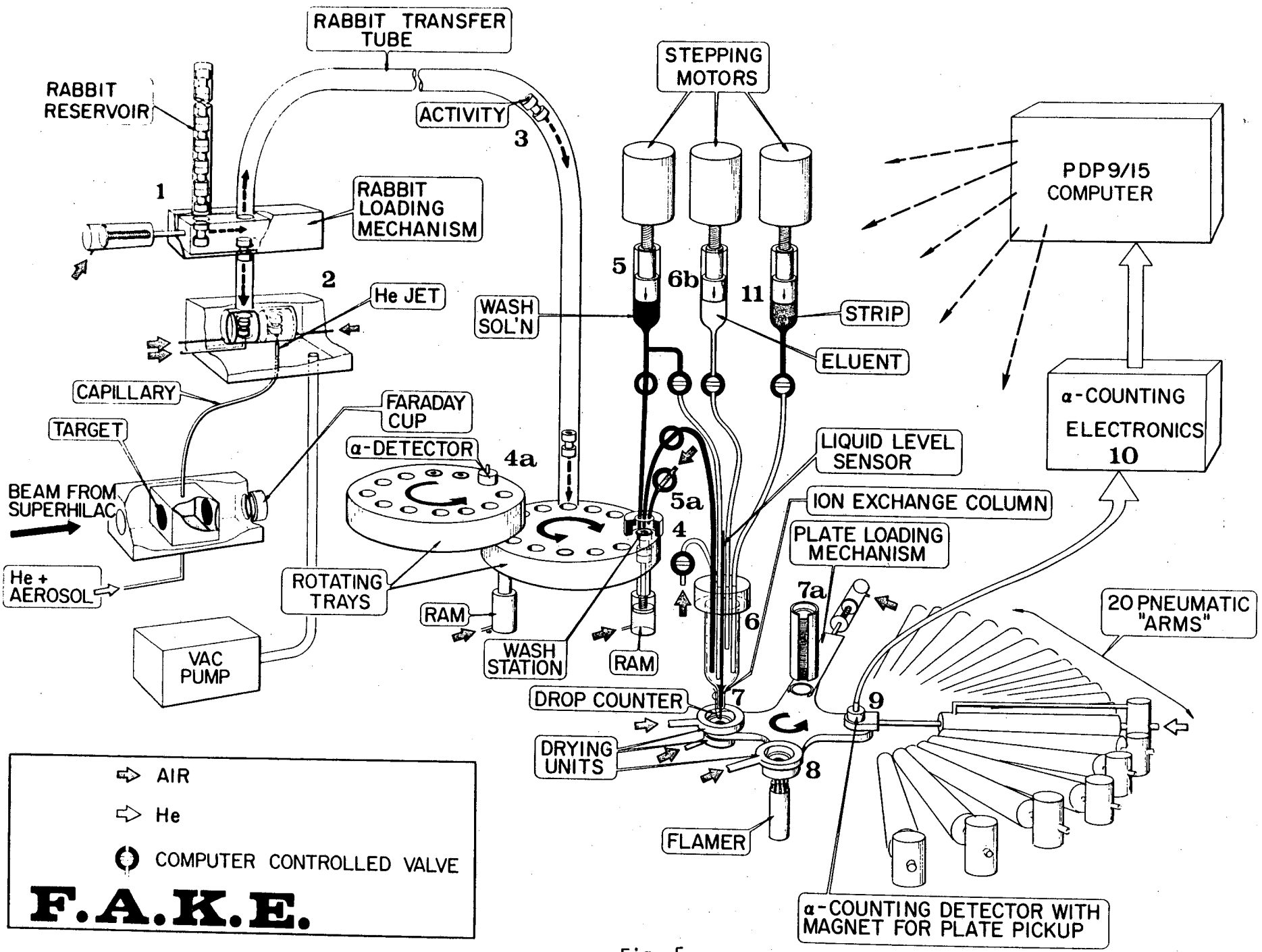

RABBIT TRANSFER TUBE
RABBIT RESERVOIR
ACTIVITY
3
STEPPING MOTORS
PDP9/15 COMPUTER
1
RABBIT LOADING MECHANISM
2
He JET
5
6b
11
WASH SOL'N
STRIP
ELUENT
CAPILLARY
FARADAY CUP
TARGET
α-DETECTOR
4a
LIQUID LEVEL SENSOR
α-COUNTING ELECTRONICS
10
BEAM FROM SUPERHILAC
5a
ION EXCHANGE COLUMN
4
PLATE LOADING MECHANISM
He + AEROSOL
ROTATING TRAYS
7a
20 PNEUMATIC "ARMS"
RAM
6
WASH STATION
RAM
VAC PUMP
DROP COUNTER
7
9
DRYING UNITS
8
FLAMER
α-COUNTING DETECTOR WITH MAGNET FOR PLATE PICKUP
AIR
He
COMPUTER CONTROLLED VALVE
F.A.K.E.

Fig. 5

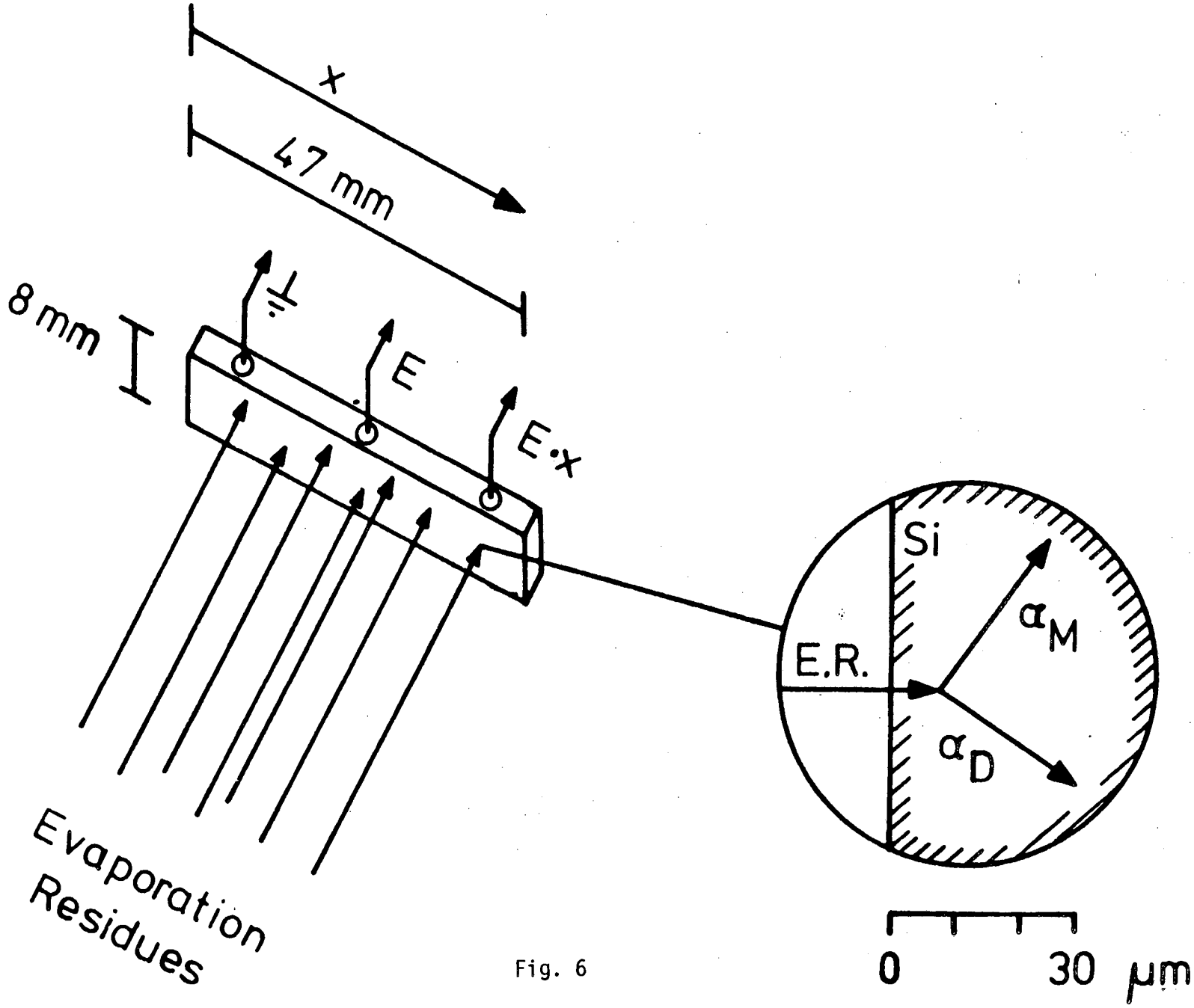


Fig. 6

XBL 834-9308

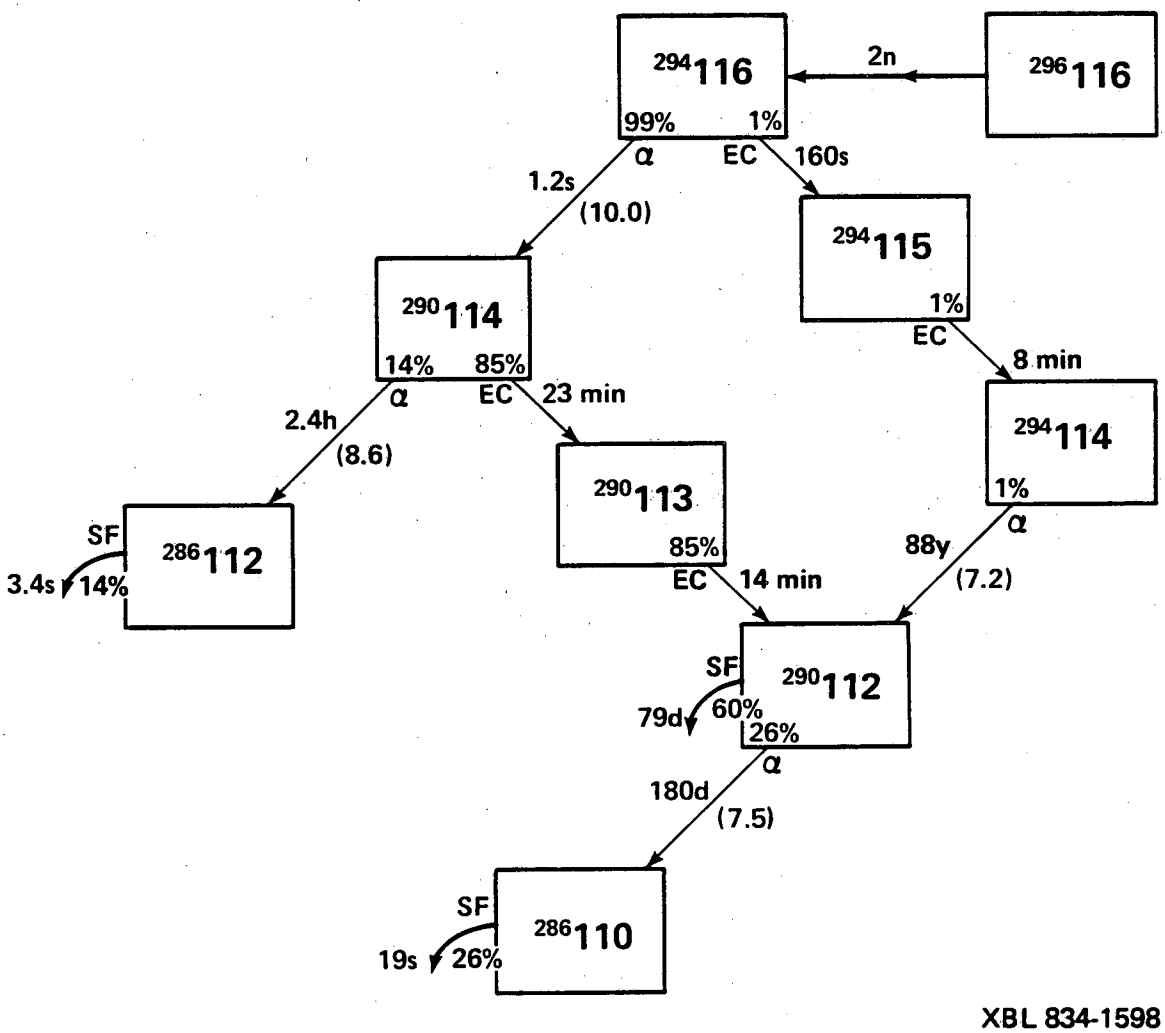

294116
99%
1%
α
EC
2n
296116
1.2s
(10.0)
160s
290114
14%
85%
α
EC
294115
1%
EC
8 min
2.4h
(8.6)
23 min
294114
1%
α
290113
85%
EC
88y
(7.2)
SF
286112
3.4s
14%
14 min
SF
290112
79d
60%
26%
α
180d
(7.5)
SF
286110
19s
26%
XBL 834-1598

Fig. 7

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