

MASTER

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The Search for New Isotopes at Brookhaven

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The study of radioactivities has always occupied an important place in nuclear physics research at Brookhaven National Laboratory. In a very large measure this interest originated and has been due over the years to the inventiveness and stimulation of Maurice Goldhaber. Upon his arrival at Brookhaven in 1949, Maurice was literally bubbling over with ideas for experiments - and he has continued to "bubble" during his long and still highly productive career.

As a young physicist in the early 1950's I was eager to participate in some of the exciting research concerned with experimental tests of the nuclear shell model and with sorting out the systematics of isomeric transitions. After four years at Brookhaven (1948-1952), working largely on problems in collaboration with Maurice Goldhaber and others in his group, the award of a National Science Foundation fellowship and an invitation from Kai Siegbahn gave me the opportunity of spending a year at the Nobel Institute of Physics in Stockholm, Sweden. The instrumentation at the Institute was the best in the world for β -ray spectroscopy and the research staff was excellent, assuring a favorable outcome on the main problem, an experimental and theoretical shell model study¹ of the decay of ^{206}Bi , done in collaboration with M. H. L. Pryce, then at Oxford University.

Just before I left for Sweden, Maurice happened to mention one of the outstanding problems in isomerism that was worrying him at that time and which he thought could be solved at the Nobel Institute. This concerned a very high-spin isomer of 7-hour half-life in molybdenum whose decay scheme was thought to be understood, but whose mass number was not known. Soon after arriving in Stockholm, and in collaboration with Sigvard Thulin², the 7-hour activity was produced by bombarding niobium with 25-MeV deuterons from the Nobel Institute cyclotron. The active sample was placed in the Institute's isotope separator and after only 2 hours of collection it was clearly seen that the 7-hour activity was appearing at the mass number 93 position. In the shortest letter I have ever written the news back to Brookhaven was,

"Dear Maurice, Definitely Mo-93!"

Of course, a more complete account followed a few days later.

Upon returning to Brookhaven in the Fall of 1953, I found the Goldhaber group generating a great deal of interesting research with reactor and cyclotron-produced radioactive sources. Also the 3.5-MV Van de Graaff was nearing the end of its reconstruction. During the following years the work at the Van de Graaff was a mix of nuclear reaction studies and β -ray spectroscopy on short-lived radio-isotopes. A big boost to this research came in 1960 with the introduction of a helium-3 beam. Before that time the cost of 1 cc atm. of He-3 gas was quoted at \$2000, but when it dropped precipitously to 15 cents per cc it became feasible to use it in an ion source even without a gas recovery system. Q-values of $^3\text{He},p$ and $^3\text{He},n$ reactions

are often high and many interesting nuclear states could be studied. Meanwhile the instrumentation for nuclear physics was improving rapidly with such developments as large-channel two-dimensional pulse-height analyzers and Ge(Li) detectors for the detection of γ rays with high resolution.

The next major development at the 3.5-MV Van de Graaff came in 1968 when a facility for accelerating a triton beam was installed. Tritium acceleration was already being done in a small Van de Graaff at Los Alamos but there was no place where nuclear spectroscopy was being investigated using triton reactions. For quite a while there was a unique opportunity to do many new things with (t,p), (t,n), and (t, α) reactions on light elements. Among these were studies of some short lived radioactivities that are easily produced with a triton beam, but difficult to make otherwise. These included ^{11}Be , ^{13}B , ^{17}N , ^{20}O , ^{21}F , ^{25}Na , ^{28}Mg , and ^{29}Al , all made via (t,p) reaction, whose half lives and decay schemes were investigated. In all cases but ^{20}O , made in the $^{18}\text{O}(t,p)^{20}\text{O}$ reaction and having a half life of 12 sec, new spectroscopic information was obtained. A number of states of ^{20}F were energetically available to ^{20}O decay and the object was to search for weak β -ray branches to some of these states by looking for the subsequent γ radiation. In this way it was hoped to learn something about β -decay rates as well as the properties of the ^{20}F excited states. However, the sensitivity of the search was severely limited by the γ rays due to the direct production of ^{20}F in the competing reaction $^{18}\text{O}(t,n)^{20}\text{F}$. ^{20}F is known to decay 100% by β -ray emission to a 1.63-MeV γ -ray emitting state in ^{20}Ne . Although ^{20}F

is actually the daughter in the decay of ^{20}O itself and one cannot escape from some 1.63-MeV γ -ray background, the major part of the background came from the direct production of ^{20}F . This obscured the search for weak γ -ray lines at the lower energies expected in ^{20}O decay.

For the time being the ^{20}O problem was dropped, but meanwhile the MP Tandem Van de Graaff facility, proposed by the author in 1962, came into operation at Brookhaven in 1970 under the very capable direction of Harvey Wegner. While the initial proposal for the Tandem mentioned nothing about the production and study of short-lived radioactivities it became clear from results being obtained elsewhere that a general program of this type would be worthwhile. Therefore plans were made, together with David Goosman, to build a target bombardment-and-transfer facility. At just the time (1970) that this "rabbit" system was under construction, there was a paper published by Mak, Spinka, and Winkler³ at the California Institute of Technology on the study of ^{20}O produced in an EN Tandem using the 2-neutron transfer reaction $^{18}\text{O}(^{18}\text{O}, ^{16}\text{O})^{20}\text{O}$ with ^{18}O ions of 25-35 MeV. They reported a good yield of ^{20}O and noted that the direct production of ^{20}F was negligible. This seemed like the answer to the background problem of ^{20}F encountered earlier in the triton bombardment. The Cal Tech group, incidentally, reported seeing a number of weak γ -ray peaks, not all of which were identified and assigned to known contaminant activities.

With the rather mundane problem of studying ^{20}O as a start to this program, and believing that improvements could be made over the Cal Tech work, ^{18}O targets were fabricated by oxidizing Ta foils in

an atmosphere of enriched ^{18}O gas. These samples were clamped in a "rabbit" for bombardment by a beam of ^{18}O ions from one of the MP tandems. Figure 1 shows a sketch of the rabbit system, the heart of which is a stainless steel tube with square internal dimensions of $1 \times 1 \text{ cm}^2$. The rabbit, fabricated from the light-weight plastic Delrin, fits loosely inside the tubing and is propelled back and forth by short bursts of helium gas. Helium was chosen because no background activities are produced in it, regardless of the nature of the beam. At each end of the transfer tubing the rabbit comes to a stop by striking a bumper consisting of a length of polyethylene tubing. The lengths of the bumpers are cut so that at the bombardment position the collimated beam strikes the exact center of the target, while at the detector end the activated spot on the target is at the center of a Be foil exit window, in case one wishes to measure β rays emerging from the sample. At both ends there are glass windows cemented on the top of the transfer tubing allowing the position of the rabbit to be monitored on TV as indicated by a dark line scribed on the top of the rabbit immediately above the target center line.

For light ion beams such as protons or deuterons a window made from Ni foil of 1 mg/cm^2 thickness separates the rabbit system from the high vacuum of the beam line thus allowing the rabbit to be cycled back and forth in any way desired. But heavy ions lose too much energy in window so it is usually necessary to use a "windowless" arrangement. This means that upon the return of the rabbit to the bombardment position, and after the valve furnishing the returning

burst of helium gas has been closed, one must wait until the vacuum pumps have reduced the pressure in the rabbit line low enough to open a main gate valve to the accelerator beam line. Under the best conditions the delay introduced by the pump-out time could be as short as 2 sec. Three such rabbit lines actually exist, two at the Tandem and one at the 3.5-MV Van de Graaff. Their lengths are 4-5 meters, sufficient to carry the rabbit through a shielding wall to a region of low background. Plastic water bottles or metal shot fills the hole in the shielding wall to insure low background. Depending on the pressure of the helium gas burst the transit time of the rabbit can be as short as 0.1 sec, although 0.3 sec is a more reasonable figure that insures the survival of both the rabbit and its target under the shock of stopping.

Another necessary component in the system is a chopper that allows the beam to pass to the target after the gate valve has been opened, but then cuts off the beam before the gate valve is closed. This keeps the beam from striking the gate valve itself, or in the arrangement using a foil window and no gate valve, the beam does not hit other parts of the rabbit. The chopper consists of a cylindrical frame having a Ta foil wrapped around through which two holes are cut on opposite sides. By means of a rotating type solenoid (rotation angle 45°) the holes in the Ta line up for beam on but prevent passage of the beam in its other position. Typical of such electro-mechanical devices the chopping time is ~ 50 msec.

The entire sequence of functions for operating the gate valve, chopper, and rabbit transfer is precisely controlled by a

timer-programmer designed by Schwender, Goosman, and Jones.⁴ For a number of years there was no comparable device commercially available and it has been one of the most useful pieces of home-made equipment in the Tandem project. Ten independent functions can be controlled with set and re-set times specified, all deriving their timing from a 10 kC quartz crystal oscillator. In addition to controlling the rabbit functions, other channels of the timer-programmer can be used for such things as routing data in the Σ -7 computer or starting and stopping multi-scaling devices for half-life measurements.

All of the necessary equipment was finally ready for the ^{20}O experiment. A 42-MeV beam of ^{18}O at a current of ~ 50 particle-nAmp gave an excellent yield of ^{20}O activity as measured in a Ge(Li) detector by the presence of the 1.057-MeV γ ray that occurs in $\sim 100\%$ of the ^{20}O decays. Weak γ -ray peaks were then looked for that might be associated with low intensity β -ray branches of ^{20}O . The results on ^{20}O decay were eventually published⁵ (no new β -ray branches were found) but a great deal happened in between. As the Cal Tech group had reported there were many small peaks, and since competing reactions were evidently taking place the attempt was made to identify and assign every line in the spectrum. Identification of a given γ ray relies most heavily on its precise energy which can be measured with a Ge(Li) detector usually to ± 0.1 keV. But half-life information is also very useful and the most straightforward method is to sequentially store the entire spectrum in sections of the computer. Analysis of these data "bins" allows the decay half-lives of various lines to be extracted. If a given line is suspected

as belonging to a certain known radioactivity then all of the other γ rays of that activity must also produce peaks with the proper relative intensities. A fortunate circumstance is that the energies of the most obvious contaminant γ -ray peaks are often known with high accuracy so they can be used as internal calibrators to aid in the identification of less certain lines.

As the analysis progressed it was surprising to note that so many interesting reactions had been taking place. In fact the γ -ray peaks due to some 15 known radioactivities were present including ^{15}C , ^{16}N , $^{19,20}\text{O}$, $^{20,21}\text{F}$, $^{23,24}\text{Ne}$, ^{24}Na , $^{27,28}\text{Mg}$, $^{28,29,30}\text{Al}$, and ^{34}P . Compared with the ^{20}O most of them were weak. Although there was scant information at that time on heavy-ion reactions in general, it was possible to propose $^{18}\text{O} + ^{18}\text{O}$ reactions, or $^{12}\text{C} + ^{18}\text{O}$ reactions on contaminant carbon, that would produce all of these activities. For example, ^{30}Al can be made by the reaction $^{18}\text{O} + ^{18}\text{O} \rightarrow ^{30}\text{Al} + \alpha + p + n$ and ^{34}P by the reaction $^{18}\text{O} + ^{18}\text{O} \rightarrow ^{34}\text{P} + p + n$.

What was unsettling about the analysis was the presence of three mystery peaks, the strongest of which had an energy of 1848 keV, the others being at 1431 and 2538 keV. They did not correspond to the γ rays from any radioactivity reported in the literature. The first step was to measure their energies accurately, and then the half lives, by the procedures noted above. It turned out that the three γ rays all decayed with a half life of 6.3 sec, making it very likely that they were associated, but with what activity? Given the obvious fact that the compound nucleus in the ^{18}O bombardment of ^{18}O is ^{36}S , and assuming that a heavier contaminant nucleus in the

target was not responsible, this restricted the search. It was further known that many nuclear species were excited in nuclear reactions, whose energy level structures were known, but which were not necessarily populated by a known β decaying parent. Upon searching through the compilations of energy levels it was David Goosman who noted that the three mystery γ rays seemed to correspond fairly well with the energies of the first three excited states of ^{33}P , a nucleus that can be formed in several reactions including $^{31}\text{P}(t,p)^{33}\text{P}$, but not previously known to be populated by a β -ray emitter. The parent would evidently have to be the then unknown nucleus ^{33}Si .

At that time the accuracy with which the excitation energies of states in ^{33}P were known was only ± 3 keV, so the next task was to renew the study of the $^{31}\text{P}(t,p)^{33}\text{P}$ reaction at the 3.5-MV Van de Graaff and to remeasure the energies of the states in ^{33}P . The best techniques of γ -ray spectroscopy using a Ge(Li) detector were brought to bear and a factor of 10 to 20 improvement in accuracy was achieved; thus the energy of the ^{33}P first-excited state was obtained with ± 0.15 -keV accuracy. Since the delayed radioactivity measurements at the Tandem had been done with similar accuracy it became convincing from the agreement of the energies in the two cases that a new β decay⁶ was being observed, namely $^{33}\text{Si} \xrightarrow{\beta^-} ^{33}\text{P}$. It could further be stated that the reaction responsible for its formation was $^{18}\text{O} + ^{18}\text{O} \rightarrow ^{33}\text{Si} + 2p + n$.

Whenever β -ray spectroscopists are fortunate enough to discover a new β -decaying isotope they immediately want to know all of the details of the decay scheme such as the spin-parity of the parent,

the decay energy, and the branching ratios to various excited states in the daughter nucleus. All of these properties are important in relating experimental results to theoretical predictions. In the case of ^{33}Si decay, the shell model had already predicted that its spin-parity should be $J^\pi = 3/2^+$, and it was known from nuclear reaction experiments that the ground and first three excited states of ^{33}P have spin-parities of $J^\pi = 1/2^+$, $3/2^+$, $5/2^+$, and $3/2^+$, respectively. So it was reasonable that all four of these states should be populated by allowed β decays from ^{33}Si . The missing piece of information at that point was the decay energy which was needed to calculate the comparative half-lives or ft values of the various β -ray branches. ft values can help to confirm a suspected spin-parity assignment.

The decay energy of a β -ray emitting isotope is equal to the mass difference between the parent and daughter expressed in energy units, and it is also the end-point energy of the β rays, provided the β 's lead to the ground state. But for β branches to excited states one can measure the β end-point energy and add the energy of the following ground-state γ ray to get the total energy. Experimentally this is very convenient since there is usually no delay between the β - and γ -ray emissions and they can be measured in coincidence. This is especially important if many radioactivities are present in the source. One can then pick out the γ -ray peak belonging to a particular branch in a given activity, using the high resolution properties of a Ge(Li) detector, and measure the β -ray spectrum in coincidence in order to find its end-point energy. A magnetic spectrometer would

be the most accurate way to determine a β end-point energy, but these devices have absolute transmissions of at most a few per cent of 4π and can only measure a few per cent of the complete spectrum at a given magnetic field setting. On the other hand, a plastic scintillation detector responds to β rays, it can measure the entire spectrum simultaneously, and it can do this with a solid angle of as much as 30% of 4π with respect to the source. It also produces very short output pulses making it excellent for coincidence applications. The dimensions of the scintillator must be sufficient to completely absorb the β rays that are being measured. While it was expected that the extreme simplicity and other advantages of the plastic scintillator would be gained at considerable expense to the accuracy in determining end point energies, as compared with a magnetic spectrometer, this turned out not to be the case, as described below.

The rabbit schematic in Fig. 1 includes the arrangement for measuring decay energies by the β - γ coincidence method using a Ge(Li) for the γ rays and an NE102 plastic scintillator for the β rays.⁷ Both detectors are very close to the rabbit for high coincidence efficiency. With the computer techniques then available the procedure was to locate the γ -ray peak of interest and to place digital windows on the peak and on the background on either side of the peak as shown in Fig. 2 for the 1848-keV line occurring in ³³Si decay. β -ray spectra were recorded in coincidence with the various windows and in the later analysis the net β spectrum in coincidence

with the peak alone was obtained by subtracting the normalized background spectrum from the total. Figure 3 shows the net β spectrum in coincidence with the 1848-keV peak of ^{33}Si decay.

While the curve in Fig. 3 certainly looked like a good β -ray spectrum there was the problem of determining its end-point energy. With a magnetic spectrometer one can rely on the constancy of its transmission and the linearity of its momentum versus magnetic field setting to calculate a Kurie plot, obtaining the momentum calibration from a measurement of just one internal conversion electron line from a reference source. But with the Tandem experiment no such calibration sources were available, and even if they were there would have been uncertainty as to the energy linearity of the device. Here again the circumstance of other known radioactivities in the source came to the rescue because they were essentially built-in calibrators. There was enough computer capacity to do the same thing on several other γ -ray peaks as was done on the unknown ^{33}Si illustrated by Figs. 2 and 3. In this way a series of β -ray spectra were obtained, each of which corresponded to a known end-point energy. Whatever distortions were present in the scintillator spectrum of the unknown ^{33}Si , due to geometrical effects for example, would also be present in known spectra that were nearby in energy. Two ways of carrying out the analysis were developed, both of which gave comparable results. One was to make Kurie plots of the unknown and the known spectra, to find their end-points, and to interpolate to obtain the unknown energy. The other was to fit a nearby known spectrum with a functional form in the computer and then to stretch, or contract this spectrum

shape for a best fit, after normalization of the intensities, to the unknown. The stretching factor was then applied to the known end-point to calculate the end-point energy of the unknown. These procedures gave surprisingly accurate results. Thus, in collaboration with Cary Davids, the end-point energy of the ^{33}Si β rays in coincidence with 1948-keV γ rays was found to be 3920 ± 50 keV.⁸ Since practically the entire error in the sum $E_{\beta_{\text{max}}} + E_{\gamma}$ is in the β -ray energy, the same error applies to the total decay energy and therefore to the mass difference.

More modern computer techniques have by now made the old system of setting digital gates obsolete. The present procedure is to use the so-called "event mode recording" or EMR method in which all coincidence events are recorded on magnetic tape, the amplitudes or channel numbers of the γ -ray and β -ray pulses being specified. In a later computer play-back of the tapes there is complete freedom to select the windows on γ -ray peaks and backgrounds as desired, and to do this on any peak in the γ -ray spectrum for developing the coincidence β spectrum.

With the total decay energy of ^{33}Si known it was possible to construct the decay scheme shown in Fig. 4 and to draw some conclusions about the comparative half lives of the β -ray branches. Unfortunately, an important piece of information that was missing was the β -ray branching intensity of ^{33}Si to the ground state of ^{33}P . There did not seem to be any way to measure this branch since the method depended on coincidence events. With the large number of activities present in the source there was no hope at all of picking out the 6.3-sec ground-state β -ray component by a singles measurement.

Even with this branch unknown it was still possible to place lower limits on the ft values of the branches to the first three excited states. In all cases the results were consistent with allowed β decays, thereby offering confirming, but not conclusive evidence that ^{33}Si indeed has $J^\pi = 3/2^+$.

The discovery of ^{33}Si was quite an exciting event, and along with the large number of other activities produced in $^{18}\text{O} + ^{18}\text{O}$ reactions, it gave the first indication of the great potential of heavy-ion reactions for producing new species of nuclei. But why should one want to engage in a program of "nuclear prospecting"? With respect to shell-model predictions the answer was clear, since confirming evidence of the model's prediction of the spin-parity of ^{33}Si had already been obtained. Other spectroscopic properties are also involved in model predictions so it is always very valuable to establish the decay properties of new isotopes. It then occurred to us that ^{33}Si is an "exotic" nucleus of $T_2 = +5/2$, (exotic nuclei are arbitrarily defined as being 3 or more neutrons away from the nearest stable isotope of the same element. In this case ^{30}Si is the closest stable silicon isotope) and since ^{33}Si is far from the valley of stability there might be theoretical predictions as to its mass value. Work along this line had been done by Garvey, Kelson, and others⁷ who had devised a semi-empirical mass formula for relating the masses of nuclear species. When the calculation was made for ^{33}Si its mass differed from the experimental result by 420 keV, far greater than the accuracy with which the mass of ^{33}Si

had been established. The confirmation of shell-model predictions of spectroscopic properties was not nearly so exciting as the discrepancy between the predicted and measured masses.

An examination of the chart of nuclides showed that none of the series of eight $T_z = + 5/2$ nuclei in the s-d shell, including ^{21}O , ^{23}F , ^{25}Ne , ^{27}Na , ^{29}Mg , ^{31}Al , ^{33}Si , and ^{35}P , had been found and studied, although the particle stabilities of some of them had been demonstrated by measurements in Dubna. The clear mission was to search for these activities by using heavy-ion reactions together with the techniques that were successful in the ^{33}Si experiments. Essentially there were double unknowns, first the decay properties of the radioactive products, and secondly the untested nuclear reactions to produce them. As for the decay properties we were not working completely in the dark since the fortunate (and essential) fact was that in every case the energy level structure of the daughter nucleus was reasonably well known, i.e., excitation energies, spins and parities, and γ -ray branching ratios of the excited states. Furthermore, the shell-model predictions of the spin and parity of the unknown nucleus, together with the Garvey-Kelson prediction of the mass, could be used to make crude guesses as to what β -ray branches might take place, the γ rays to look for, and even what half-life might be expected. The half-life estimate was particularly important in setting up the experiment since the timing of the irradiate and count cycles could then be adjusted to optimize the efficiency for finding a particular weak activity in the presence of many others.

Heavy ion reactions to produce the new activities were not difficult to think of. Although some theoretical work had been done on predicting the cross sections of fusion-evaporation reactions, there were not enough systematic experimental measurements at that time to have confidence in the calculations. Table I shows the list of the eight $T_z = + 5/2$ isotopes in the s-d shell together with the suggested reactions to produce them, the ^{33}Si , of course, already having been found. While commonly available targets were to be used in most of the proposed reactions, several involved targets of ^{10}Be , i.e., for producing ^{21}O and ^{23}F . This came about through the long-term project by David Goosman in fabricating high-quality targets enriched in ^{10}Be to 94%.

Since targets of ^{18}O were already available, the first of the intentional searches for a new isotope was ^{35}P , requiring only a switch to a beam of ^{19}F ions from the Tandem. Actually the reaction $^{18}\text{O}(^{18}\text{O},p)^{35}\text{P}$ was considered, but at the high excitation energy of the ^{36}S compound system it was thought to be extremely unlikely that only a single nucleon would be emitted. Certainly the $^{18}\text{O}(^{19}\text{F},2p)^{35}\text{P}$ reaction would be expected to occur at ^{19}F beam energies of ~ 45 MeV. ^{35}P was predicted to have a spin-parity of $1/2^+$ and to decay mainly to the 1572-keV $1/2^+$ first-excited state of ^{35}S followed by a γ ray of that energy.

The Tandem experiment on ^{35}P turned out to be relatively easy. A γ ray of 1572 keV was observed¹⁰ having a half-life of 48 sec, and β - γ coincidences gave the β end-point energy and mass excess of ^{35}P to an accuracy of ± 75 keV. A β branch to only one excited state

was found, as anticipated from the level structure of ^{35}S , and the log ft of that branch was in accord with shell-model predictions. These results were confirmed later by producing the ^{35}P activity at the 3.5-MV Van de Graaff using the reaction $^{36}\text{S}(t,\alpha)^{35}\text{P}$.

The next search, this time for ^{25}Ne , carried out in collaboration with David Goosman and John Hardy,¹¹ presented much greater challenges, not only in detecting the activity, but in the analysis of the results. Actually there had meanwhile been an unpublished report by a group at Dubna attributing an activity of 0.64-sec half-life and 7.1-MeV β end-point energy to ^{25}Ne decay. Here again the energy level scheme of the daughter nucleus ^{25}Na was known and it was expected that there would be γ rays of 90 and 980 keV following β -ray branches of ^{25}Ne to the first two excited states of ^{25}Na , with perhaps other higher levels also being fed. But when the $^9\text{Be}(^{18}\text{O},2p)^{25}\text{Ne}$ reaction was tried, using a Be foil target clamped in the rabbit, the resulting γ -ray spectrum was so strongly dominated by the lines from the decay of ^{25}Na , formed in the competing reaction $^9\text{Be}(^{18}\text{O},pn)^{25}\text{Na}$, that no peaks could be seen at all that could be ascribed to ^{25}Ne decay. ^{25}Na has a half-life of 61 sec which is a factor of 100 longer than that expected for ^{25}Ne (later found to have $T_{1/2} = 0.602$ sec), so the build-up of the ^{25}Na activity in the target presented a serious problem.

The way out of this dilemma was to dispense with the rabbit and to construct a gas transport system as sketched in Fig. 5. The Be foil serves as both the target and the entrance window to a gas cell for stopping the product particles recoiling out of the target. Helium gas flowing through the target cell sweeps the activity out through a transfer tubing to a counting cell. By taking advantage of the fact

that neon is a noble element, the transport of other activities including ^{25}Na was inhibited by passing the gas through a liquid nitrogen trap. This allowed the ^{25}Ne to pass to the counting cell but most of the ^{25}Na was trapped. Background activities that were able to get through were pumped out of the counting cell on each cycle, except for those atoms that stuck to the walls of the cell. The improvement in the selective sensitivity of the gas system in detecting ^{25}Ne compared with ^{25}Na was a dramatic factor of about 400 over the rabbit system and this made the peaks due to ^{25}Ne decay stand out very clearly. Of the several configurations of counting cell used, the one shown in Fig. 6 was for the measurement of β - γ coincidences. The front face of the 10-cm dia. NE102 β detector forms one side wall of the cylindrical counting cell, with a layer of Al foil inserted for good light collection by the 8575 photomultiplier tube. A large NE102 scintillator had to be used because of the energetic β rays of ^{25}Ne which have end points of up to 7.3 MeV.

It was with ^{25}Ne that the first real difficulties were encountered in the analysis and this was because there were 9 γ rays and 4 β -ray branches taking place. The close geometry and large size of the β detector resulted in substantial β - γ and γ - γ real summing probabilities. In order to isolate the β -ray spectrum leading to a given excited state in ^{25}Na , as measured by the net spectrum in coincidence with a peak in the Ge (Li) spectrum, it was necessary to establish the additional contributions to that peak. The procedure consisted of measuring the absolute efficiency and response function of the NE102 detector for specified γ -ray energies, obtaining the β spectrum shape as a function of end-point energy, and then constructing the coincidence contributions based on the

β and γ branching ratios in the decay scheme. All of this could be done on the computer and checks on the method could be made by seeing if the same total decay energy was obtained for β -ray branching to various excited states. Calibrations from built-in reference standards were used in all of the analyses as described above. Good consistency was found for several β branches and the mass excess of ^{25}Ne was established to an accuracy of ± 300 keV. While the predicted spin-parity of $1/2^+$ for ^{25}Ne was confirmed by the constructed decay scheme the mass excess not only differed from the Garvey-Kelson prediction by 600 keV but the difference was in the opposite direction to that of ^{33}Si . The ^{25}Ne mass result was later confirmed by a more accurate reaction measurement made at Berkeley.

Returning to the rabbit technique ^{31}Al was found¹² by using the original $^{18}\text{O} + ^{18}\text{O}$ combination, (see Table I) but this time speeding up the irradiate-count cycle so that this short-lived activity ($T_{1/2} = 0.64$ sec) could be detected. The mass of ^{31}Al was measured with an accuracy of 100 keV. In collaboration with Cary Davids ^{27}Na , which had meanwhile been discovered by a group at Orsay, was then studied.¹³ The half-life and mass measurements agreed with those of Orsay, but additional details were obtained on the decay scheme.

By this time there were enough examples of $T_z = + 5/2$ masses to look for systematic trends. Included in the survey was an experimental measurement of the mass of ^{29}Mg by Scott et al. who had obtained the Q-value of the 3-neutron transfer reaction $^{26}\text{Mg}(^{11}\text{B}, ^8\text{B})^{29}\text{Mg}$. Unfortunately, the differences between the experimental masses and those predicted by the Garvey-Kelson formulation, when plotted against the mass number, showed only large fluctuations which were far greater

than the statistical accuracies of the measurements and appeared to be random.

It was important to check the ^{29}Mg mass, so together with Cary Davids the effort was concentrated on finding this isotope and measuring its properties. The search was successful,¹⁴ using the $^{18}\text{O}(^{13}\text{C}, 2p)^{29}\text{Mg}$ reaction, but the mass measurement was difficult because of the low yield relative to background. However, even with the fairly large error of 400 keV on the ^{29}Mg mass there was a significant discrepancy of 1.7 MeV with the result of Scott *et al.* Somewhat later a Berkeley group did the reaction experiment over and got a mass value for ^{29}Mg very close to the Brookhaven National Laboratory result, but with much greater accuracy (± 60 keV).

The final case was the discovery¹⁵ of ^{23}F , activity of 2.23-sec half-life, and the determinations of its decay scheme and its mass to an accuracy of 170 keV. This result confirmed a consistent picture that had begun to emerge with the ^{29}Mg mass determination. As may be seen in the upper part of Fig. 7 the differences between the experimental masses and those calculated from the Garvey-Kelson formulation showed a systematic behavior, almost an S-shaped curve. Probably the most significant contribution of mass measurements of this series, including the work at Brookhaven,¹⁶ was that the departures from the Garvey-Kelson formulation stimulated other theoretical calculations. One of these, a shell-model calculation due to Wilcox and others¹⁷ at Berkeley, resulted in the data shown in the lower part of Fig. 7. As is apparent the agreement with experiment is very much better than in the upper curve, with the possible exception of the ^{21}O mass excess which had been determined at Dubna in a nuclear reaction measurement.

Four serious attempts to find ^{21}O at Brookhaven were all fruitless and the discovery of this activity has yet to occur. However, in a very nice experiment the Chalk River group of Ball et al.¹⁸ measured the ^{21}O mass excess from the Q-value of the 3-neutron pick-up reaction $^{208}\text{Pb}(^{18}\text{O}, ^{21}\text{O})^{205}\text{Pb}$. On both of the plots shown in Fig. 7 their result brings the ^{21}O point down by about 1 MeV, and in the shell model case it comes into very close agreement with the average of the seven other cases.

Having had favorable results with 7 out of 8 of the $T_z = +5/2$ series in the s-d shell, it was decided to pursue other cases including nuclei in the same region but even farther from the valley of stability. Three examples of unknown $T_z = +3$ nuclei (one neutron farther from the valley of stability than those of $T_z = +5/2$) were ^{26}Ne , ^{30}Mg , and ^{34}Si . The first two searches were unsuccessful, but in collaboration with Alan Nathan ^{34}Si was found and its mass was measured¹⁹ using the $^{18}\text{O}(^{18}\text{O}, 2p)^{34}\text{Si}$ reaction. In other work ^{14}B was found using the $^{10}\text{Be}(^6\text{Li}, 2p)^{14}\text{B}$ reaction²⁰ and ^{55}V using the reaction $^{48}\text{Ca}(^9\text{Be}, np)^{55}\text{V}$. The latter was done in collaboration with Alan Nathan, John Olness, and Earnest Warburton.²¹ Table II shows the unsuccessful searches, a list of isotopes that remains as a challenge for future work both at Brookhaven and elsewhere.

With experiments becoming more and more difficult in the neutron-excess nuclei of the s-d shell it was decided to look at other interesting regions of the chart of nuclides, in particular the neutron-deficient nuclei in the region of mass $A \sim 80$. As may be seen from Fig. 8 one can make use of the curvature of the valley of stability

in the chart of nuclides (a chart in which proton number Z is plotted against the neutron number N) to reach this region by heavy-ion reactions such as $^{40}\text{Ca} + ^{40}\text{Ca}$. At the time this project was being formulated there were two technical obstacles preventing the study of activities induced by $^{40}\text{Ca} + ^{40}\text{Ca}$ reactions. The terminal voltage on the MP-7 Tandem at Brookhaven was not high enough to generate ^{40}Ca beams of sufficient energy to overcome the Coulomb barrier, and it was furthermore notoriously difficult to produce the required negative Ca ions from the sources then available. Fortunately both of these problems were solved a few years ago. Up-grading of the MP-7 Tandem raised the reliable upper limit of terminal voltage from 9-10 MV to 13-14 MV. Meanwhile at the University of Pennsylvania Roy Middleton made the important discovery that copious beams of the hydrides CaH^- and CaH_2^- were produced when ammonia gas was introduced into his UNIS ion source. This technique, which was also successful in making negative ion beams of hydrides from Be, Al, Mg, and many other elements, has literally revolutionized the Van de Graaff research field. When an accelerated CaH^- ion passes through the foil stripper at the center of a Tandem the molecule is broken up and Ca in a high positive charge state emerges for the next stage of acceleration.

Initial experiments, together with John Olness, consisted of clamping a calcium foil in the rabbit and bombarding it with a Ca beam of ~ 140 MeV. After transferring the target the γ radiations were measured in a Ge(Li) detector. An enormously complex γ -ray spectrum was observed and it was evident that many different radioactivities were being produced. The job of sorting things out looked as though it

was going to be very difficult. γ - γ coincidence measurements helped to some extent, but it was concluded that the build-up of some of the strong long-lived activities was going to hamper the search for short-lived isotopes. This situation was reminiscent of the ^{25}Ne problem described earlier.

Instead of the ^{25}Ne system (Fig. 5) a more modern and sophisticated type of gas transfer method is the so-called helium-jet already developed and in use at other laboratories. For several years a device of this type had been under consideration, not only as a general research tool for the Tandem, but to solve a problem²² carried over from work with Denys Wilkinson in 1971. This was an investigation of the alpha particles occurring in the β decay of ^{11}Be , an activity of 14 sec half-life that can be made in the small Van de Graaff using the $^9\text{Be}(t,p)^{11}\text{Be}$ reaction. ^{11}Be has sufficient decay energy so that it is possible for it to β decay to excited states in ^{11}B that are above $^7\text{Li} + \alpha$. In fact heavy particle emission was found in 1971 (presumably α 's) from a 3% branch in ^{11}Be decay, but the detailed structure of the spectrum could not be resolved due to the fact that the ^{11}Be nuclei, recoiling into the target backing, form a thick source so far as the emerging α particles are concerned, and this smears the structure. The obvious way to solve this problem was to transfer the activity in a He-jet device in which the particles move and are deposited on a tape at sonic velocities. Because of the small amount of material involved in the deposit and the negligible penetration of the particles into the collecting surface the effective thickness of the source is very small, so the energy resolution is preserved.

The helium-jet system shown in Fig. 9 includes a source-deposition and tape-transport unit²³ developed together with Ted Robinson. Radioactivity produced in the target cell is transported in a mixture of helium plus an aerosol through a capillary of 1 mm i.d. and is then sprayed through a skimmer, which removes much of the helium transport gas but allows the heavy aerosol particles (to which the activity particles are attached) to pass undeflected onto the tape. After activity is deposited for a certain time in a spot ~ 1.5 mm dia. the tape is quickly moved a distance of 30.5 cm to the detecting region where any combination of devices can be used for measuring β , γ , or delayed particles in singles or in coincidence. Figure 9 shows the set-up for γ - γ coincidence measurements using two Ge(Li) detectors. Novel features of this system include the use of a single 5-foot long loop of aluminized mylar tape (rather than supply and take-up reels) and a unique method of moving the tape a fixed distance. This is done by punching a continuous series of sprocket holes in the tape (spacing exactly 10 to the inch) and using a circuit such that the reading head counts 120 holes and then applies a fast acting brake. The source spot is moved reproducibly within an accuracy of about 0.3 mm and, because of the speed of the tape reader, the time for moving the source from the jet to the detector is 0.2 sec. With a particular length of tape 29 source spots can be measured without repeating and there is adequate shielding against long-lived backgrounds from spots other than the one in front of the detectors.

Even before the ¹¹Be problem could be started, the He-jet system was called into service at the Tandem early in 1979 to begin

the program of studying neutron-deficient nuclei in the mass region $A \sim 80$. Instead of trying the $^{40}\text{Ca} + ^{40}\text{Ca}$ reaction itself as originally planned, (because of target difficulties) it was easier to begin with the reaction $^{58}\text{Ni} + ^{24}\text{Mg}$ which forms the compound nucleus ^{82}Zr (^{80}Zr is formed in the $^{40}\text{Ca} + ^{40}\text{Ca}$ case). The operational quality of the system exceeded expectations. Even in the first few runs, in collaboration with Peter Haustein, Kim Lister, and John Olness, two new short-lived isotopes were established, $^{24}\text{80}\text{Y}$ of 34-sec half-life and ^{79}Sr of 2.3-min half-life (previous reports of having observed ^{79}Sr appear to be erroneous). Decay schemes and masses were measured. Numerous other radioactivities were also observed and the analysis and experimental work are still continuing.

In the meantime the ^{11}Be problem, in collaboration with Denys Wilkinson, was solved²⁵ by the use of the helium-jet apparatus. Complete resolution of the $\alpha + ^7\text{Li}$ spectrum in singles and in coincidence with γ rays made it possible to show that the 9875-keV state of ^{11}B is fed in ^{11}Be decay and that this state emits α 's to both the ground and 478-keV states of ^7Li . The result is particularly interesting because it agrees reasonably well with a theoretical prediction by Millener of the α branches from the 9875-keV ^{11}B state to the states of ^7Li .

As we enter the 1980's the future projections for Tandem research are optimistic. Many nuclear species remain to be discovered on both sides of the valley of stability in regions accessible to reactions induced by heavy-ion bombardment with the Tandem Van de Graaff facility at Brookhaven. With the instrumentation on hand,

including the rabbit and helium-jet systems and a variety of beam-chopping and pulsing devices, many interesting research problems are anticipated. If a requested energy booster for the Tandem beam comes into being it will be possible to move all the way up the chart of nuclides to the heaviest elements in an even greater expansion of this type of study. All of this is certainly not just to have the considerable satisfaction of discovering new isotopes. There is currently a vital need to establish the properties of nuclei as far as possible from the valley of stability. Theorists need this information to help in the as-yet unsolved problem of predicting accurately, and in detail, the spectroscopic properties and masses of nuclei from nuclear structure theory. The field of radioactivity studies has progressed enormously since Maurice Goldhaber started guiding our way 30 years ago, but there is very much more to be done.

Collaborators in the various searches for new isotopes described in this article included the following, in chronological order: D. R. Goosman, J. C. Hardy, C. N. Davids, A. M. Nathan, J. W. Olness, E. K. Warburton, C. J. Lister, and P. E. Haustein.

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Table I. Compound reactions making $T_z = + 5/2$ nuclides in the s-d shell

Nuclide	Reaction	Nuclide	Reaction
^{21}O	$^{10}\text{Be}(^{13}\text{C}, 2\text{p})^{21}\text{O}$	^{29}Mg	$^{18}\text{O}(^{13}\text{C}, 2\text{p})^{29}\text{Mg}$
	$^9\text{Be}(^{18}\text{O}, \alpha 2\text{p})^{21}\text{O}$		$^{14}\text{C}(^{18}\text{O}, 2\text{pn})^{29}\text{Mg}$
^{23}F	$^{18}\text{O}(^7\text{Li}, 2\text{p})^{23}\text{F}$	^{31}Al	$^{18}\text{O}(^{18}\text{O}, \alpha\text{p})^{31}\text{Al}$
	$^{10}\text{Be}(^{18}\text{O}, \alpha\text{p})^{23}\text{F}$		$^{15}\text{N}(^{18}\text{O}, 2\text{p})^{31}\text{Al}$
^{25}Ne	$^9\text{Be}(^{18}\text{O}, 2\text{p})^{25}\text{Ne}$	^{33}Si	$^{18}\text{O}(^{18}\text{O}, 2\text{pn})^{33}\text{Si}$
^{27}Na	$^{11}\text{B}(^{18}\text{O}, 2\text{p})^{27}\text{Na}$	^{35}P	$^{18}\text{O}(^{19}\text{F}, 2\text{p})^{35}\text{P}$

Table II. Searches not yet successful.

Isotope	Reaction	Measurement
^{15}B	$^{10}\text{Be}(^7\text{Li}, 2\text{p})^{15}\text{B}$	n
^{17}C	$^{10}\text{Be}(^9\text{Be}, 2\text{p})^{17}\text{C}$	γ
^{19}N	$^{10}\text{Be}(^{11}\text{B}, 2\text{p})^{19}\text{N}$	n, γ
^{21}O	$^{10}\text{Be}(^{13}\text{C}, 2\text{p})^{21}\text{O}$	γ
	$^9\text{Be}(^{18}\text{O}, 2\text{p}\alpha)^{21}\text{O}$	γ
^{26}Ne	$^{10}\text{Be}(^{18}\text{O}, 2\text{p})^{26}\text{Ne}$	γ
^{30}Mg	$^{14}\text{C}(^{18}\text{O}, 2\text{p})^{30}\text{Mg}$	γ

Figure Captions

- Fig. 1 Schematic diagram of the "rabbit" target-transfer system.
- Fig. 2 Portion of the γ -ray spectrum resulting from activities induced by the $^{18}\text{O} + ^{18}\text{O}$ reaction showing the 1848-keV peak due to the new isotope ^{33}Si . Digital windows were set so as to measure the net β -ray spectrum in coincidence with the peak.
- Fig. 3 β -ray spectrum of ^{33}Si from an NE102 scintillator in coincidence with the 1848-keV γ -ray peak shown in Fig. 2.
- Fig. 4 Decay scheme proposed for ^{33}Si .
- Fig. 5 Schematic diagram of the gas transport system used for measurements on ^{25}Ne activity. The sequence of valve operations is shown in the lower part of the figure.
- Fig. 6 Diagram of the gas cell used for β - γ coincidence measurements on ^{25}Ne . The front face of the large NE102 scintillator forms one wall of the cell.
- Fig. 7 Differences between experimental masses and those predicted by the Garvey-Kelson formulation, upper curve, and a shell-model calculation, lower curve, both plotted versus mass number A. Points labeled LBL were measured at Berkeley.
- Fig. 8 Simplified representation of the chart of nuclides showing the curvature of the center line of the valley of stability. This illustrates how the region of neutron-deficient nuclei of $A \sim 80$ can be reached in the $^{40}\text{Ca} + ^{40}\text{Ca}$ reaction at beam energies above the Coulomb barrier. The box represents the compound nucleus ^{80}Zr .

Fig. 9 Schematic diagram of the BNL helium-jet system as used for studies of radioactivities induced by $^{58}\text{Ni} + ^{24}\text{Mg}$ and $^{58}\text{Ni} + ^{28}\text{Si}$ reactions.

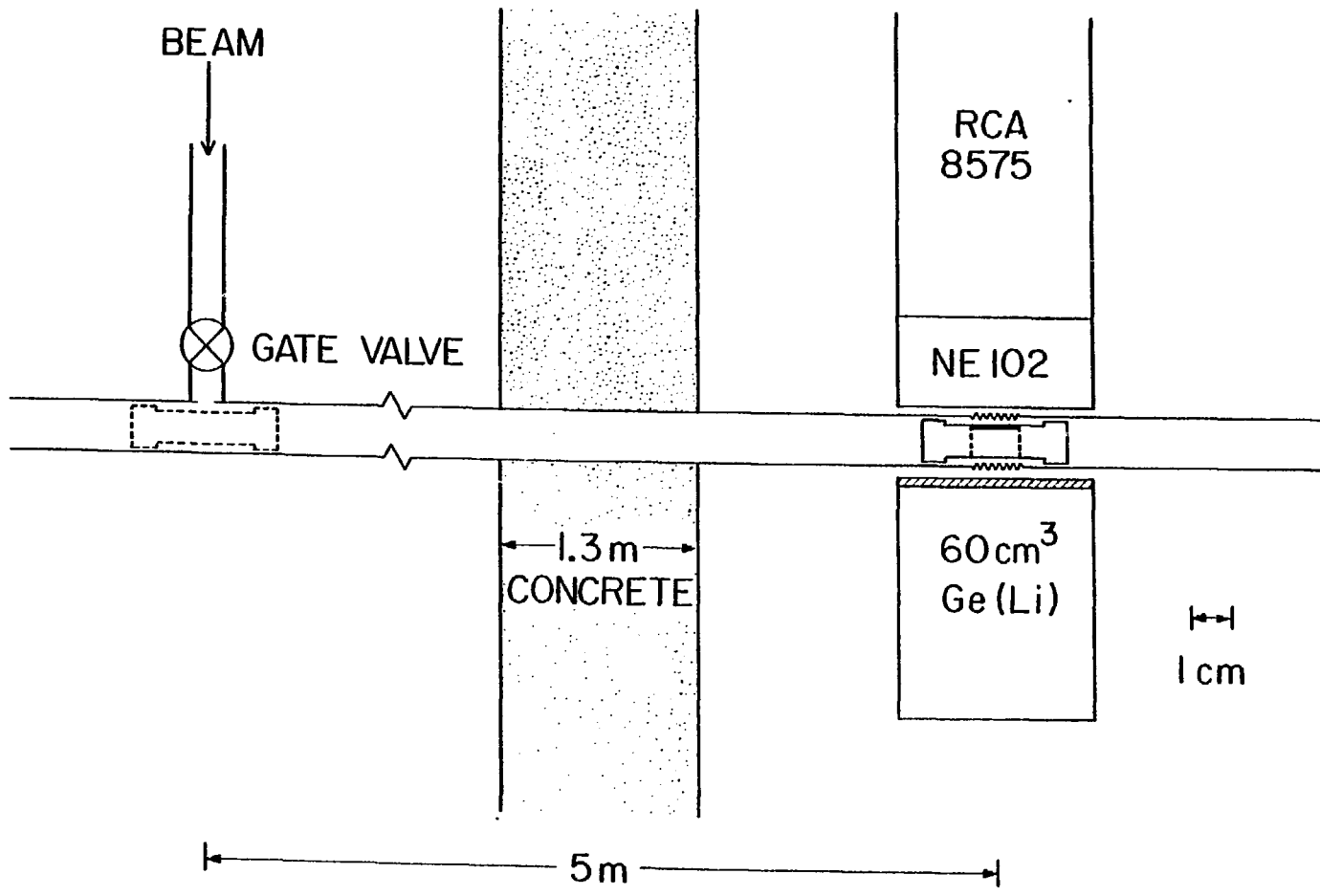


Figure 1

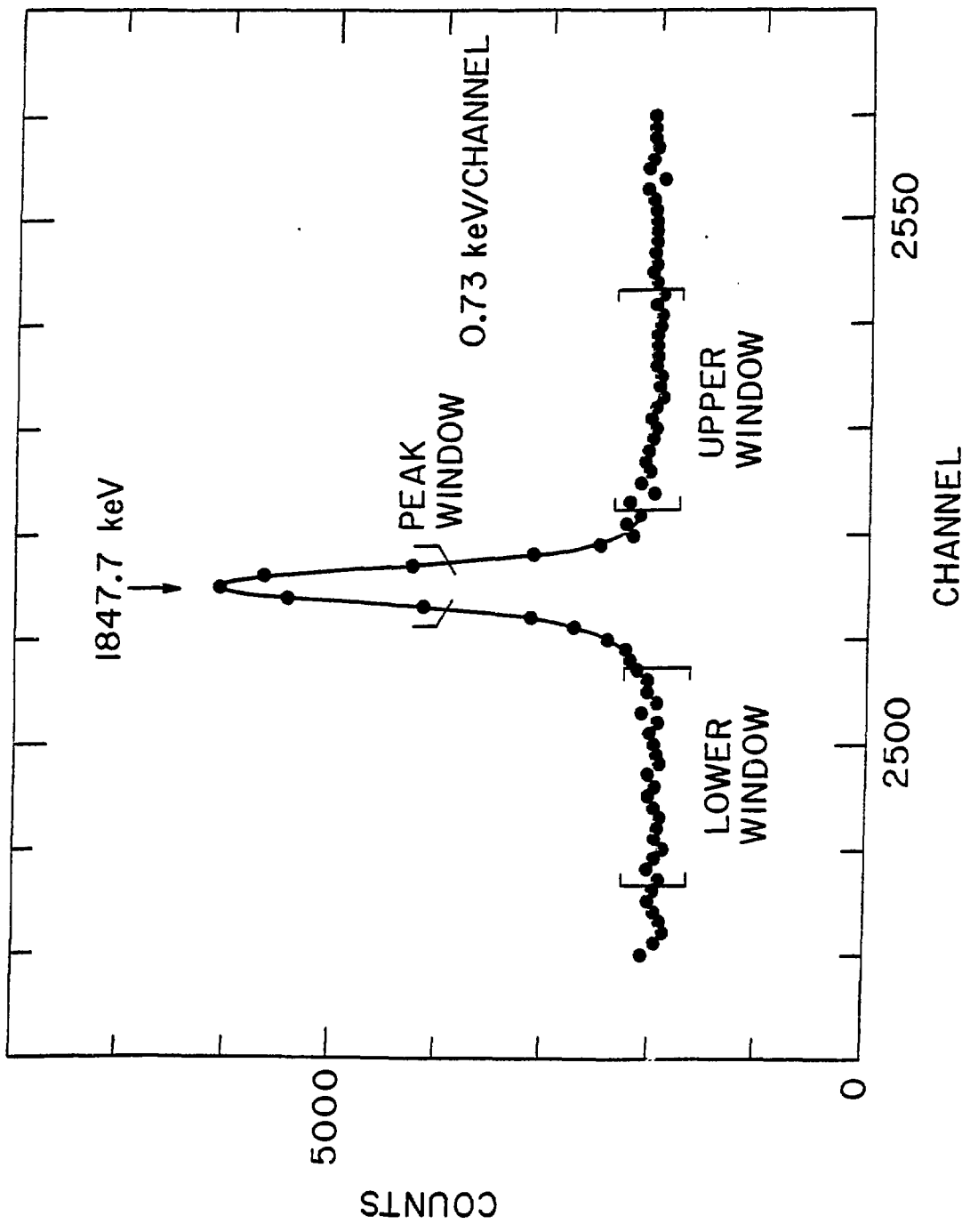


Figure 2

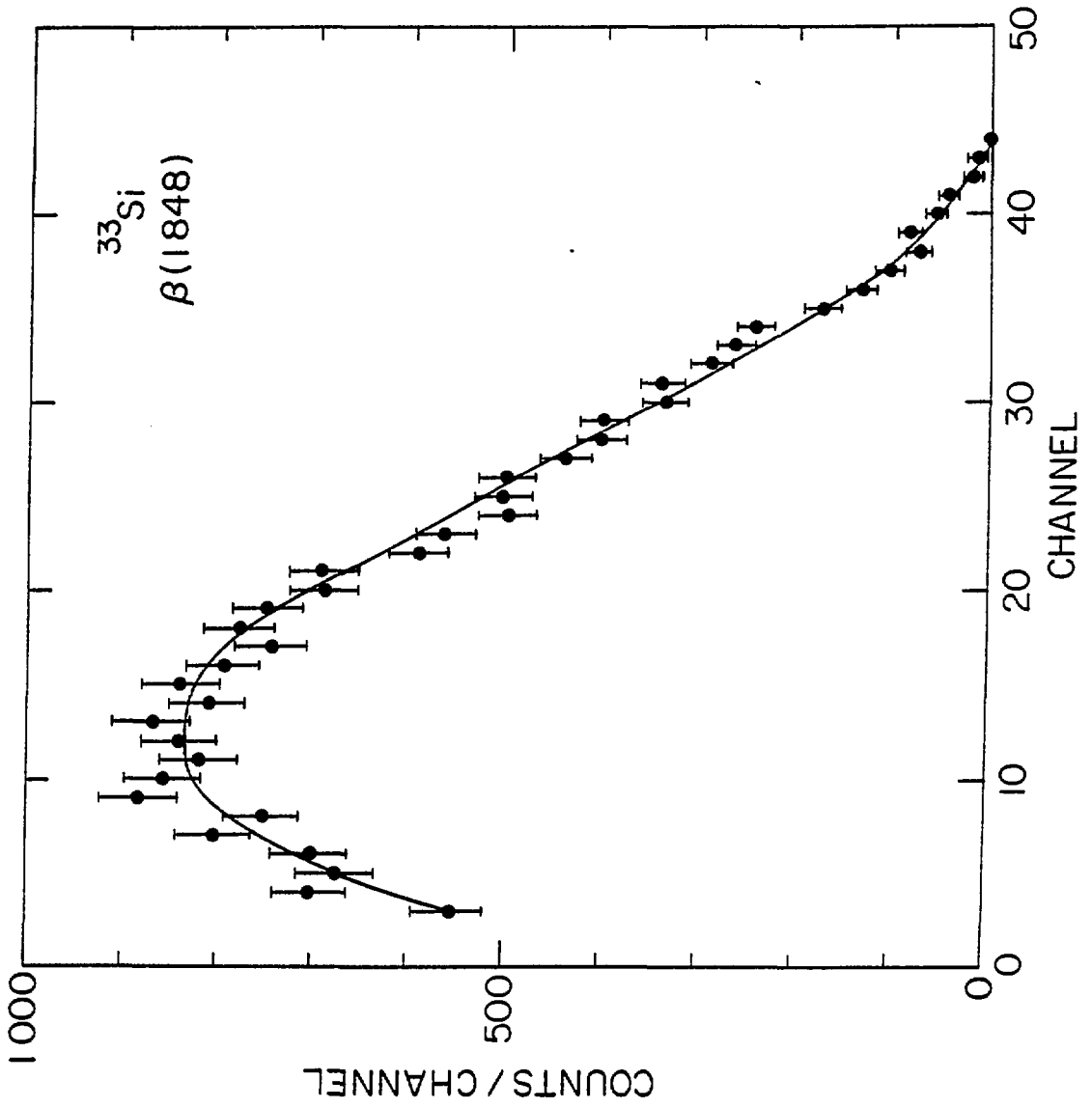


Figure 3

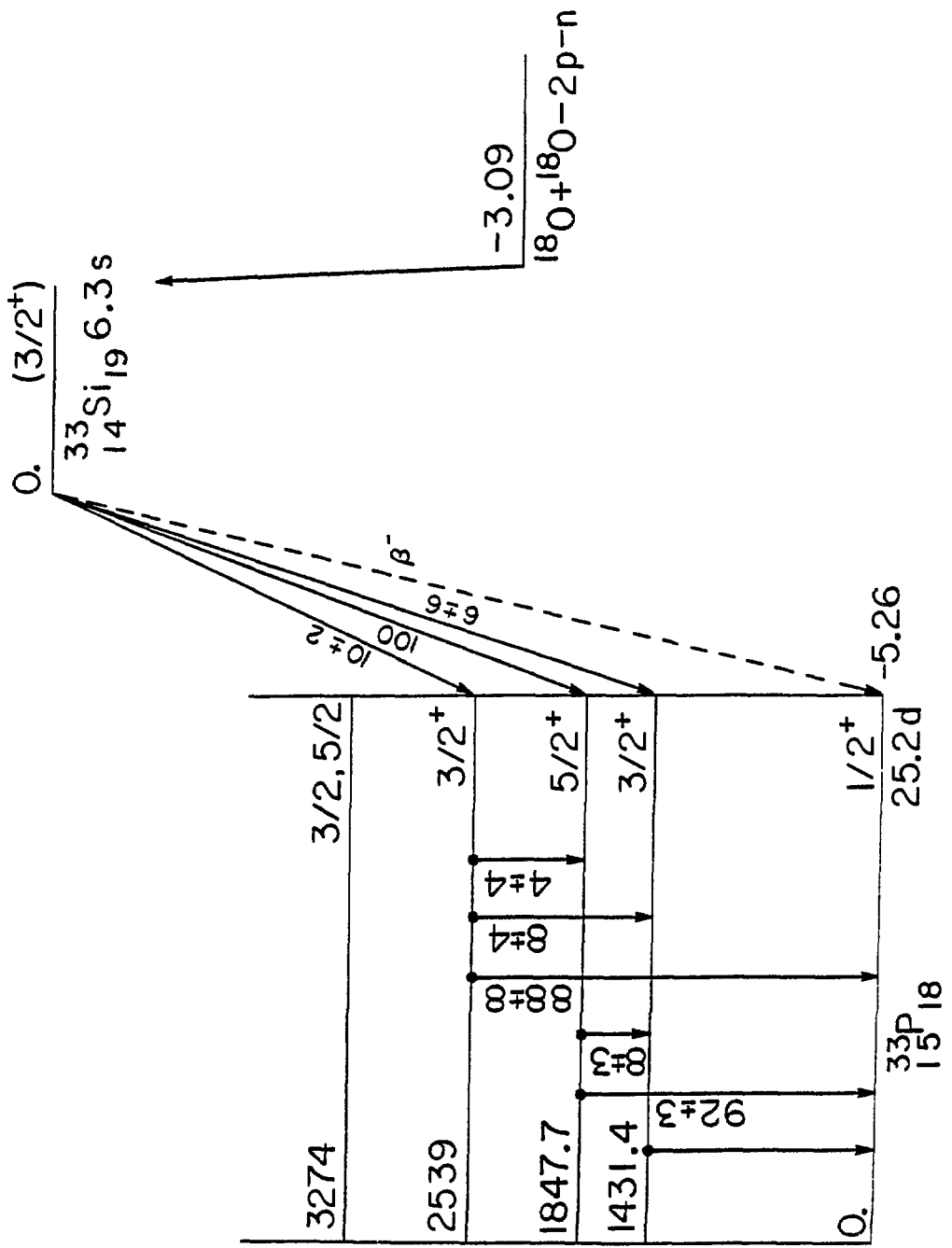
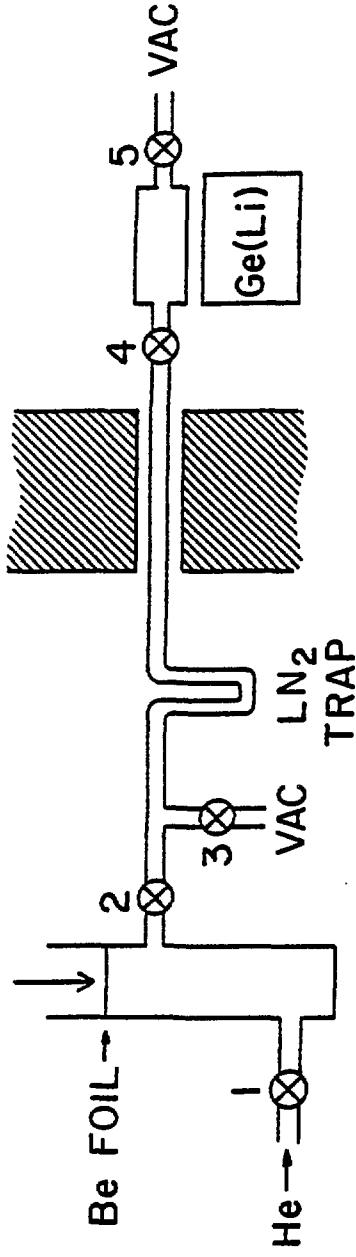


Figure 4

^{18}O BEAM



VALVES

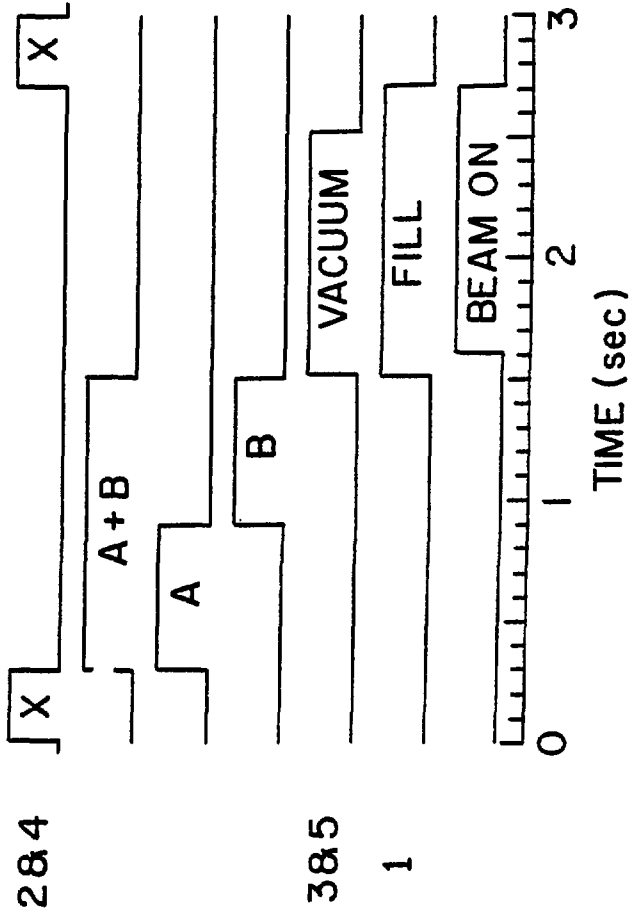


Figure 5

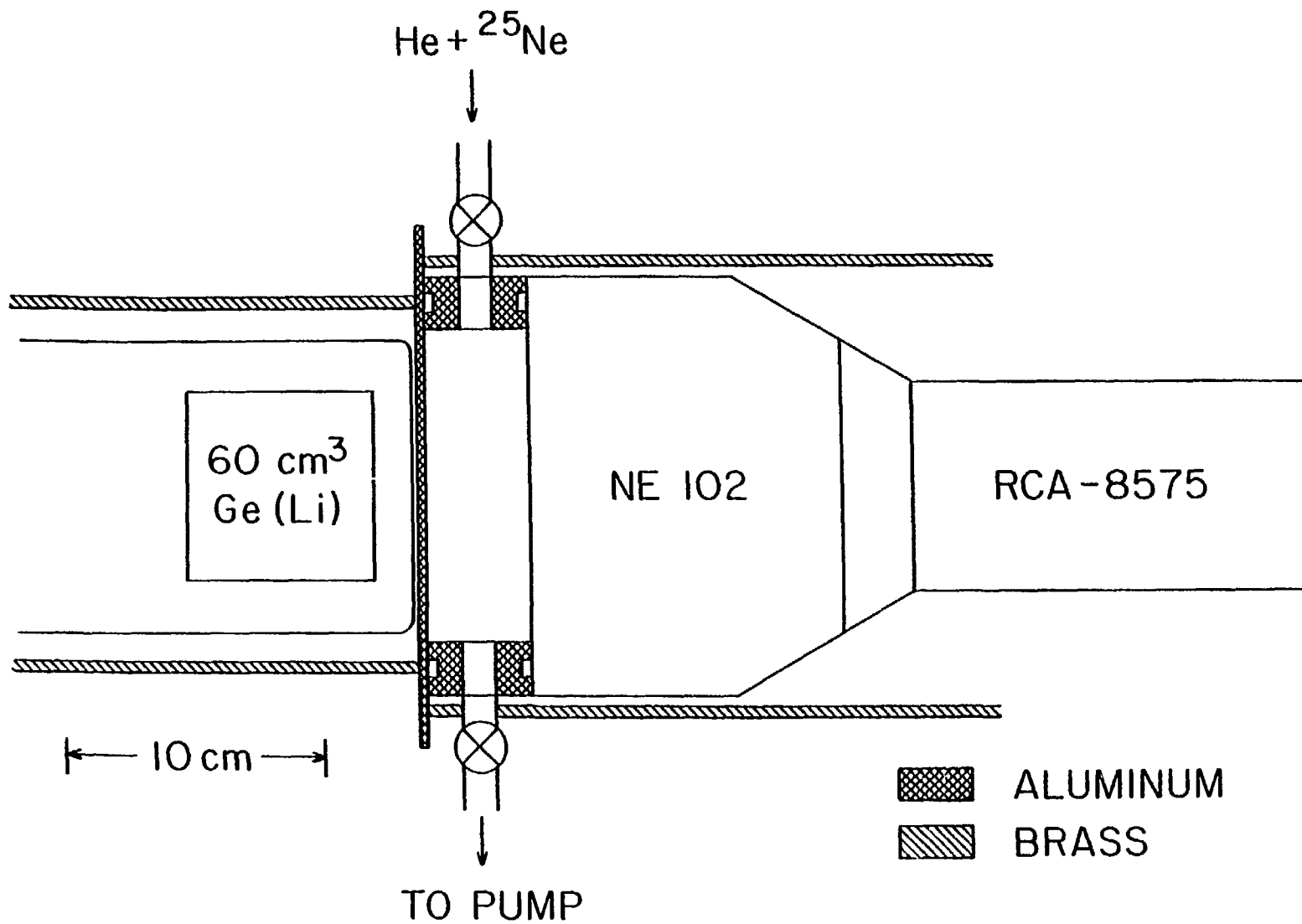
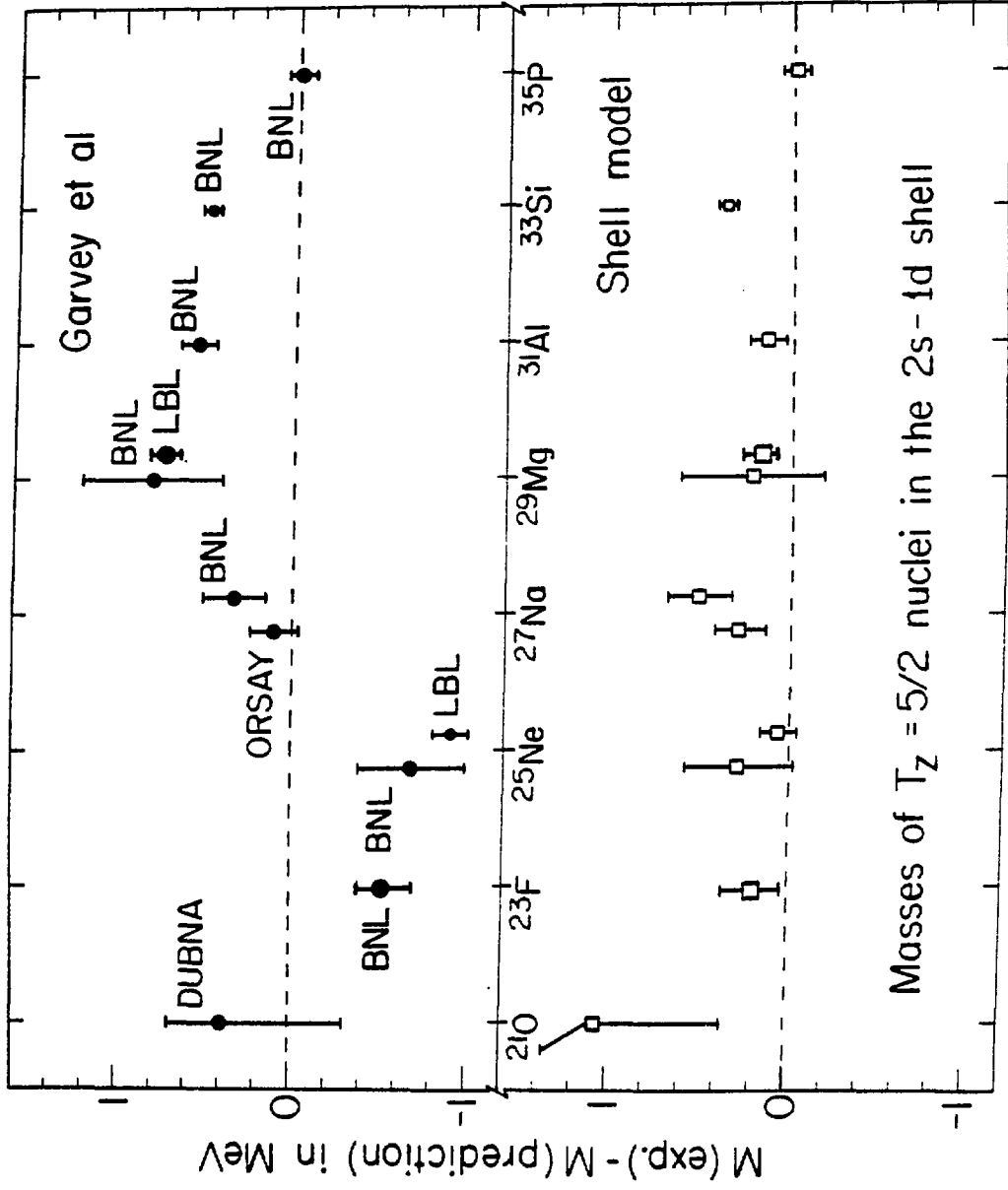
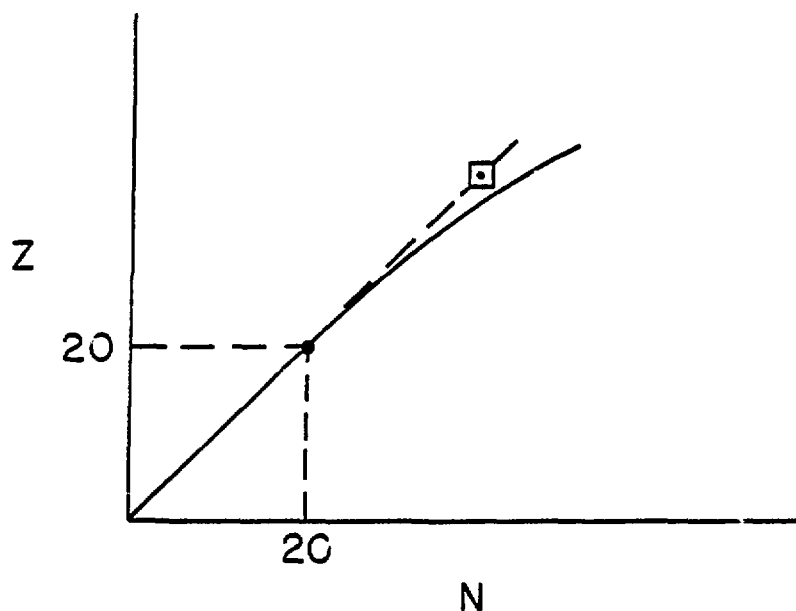


Figure 6



XBL738-3742

Figure 7



$$E_{\text{th}} \cong 110 \text{ MeV}$$

REQUIRE ~13 MV ON TERMINAL 2-STAGE,
OR 3-STAGE.

Figure 8

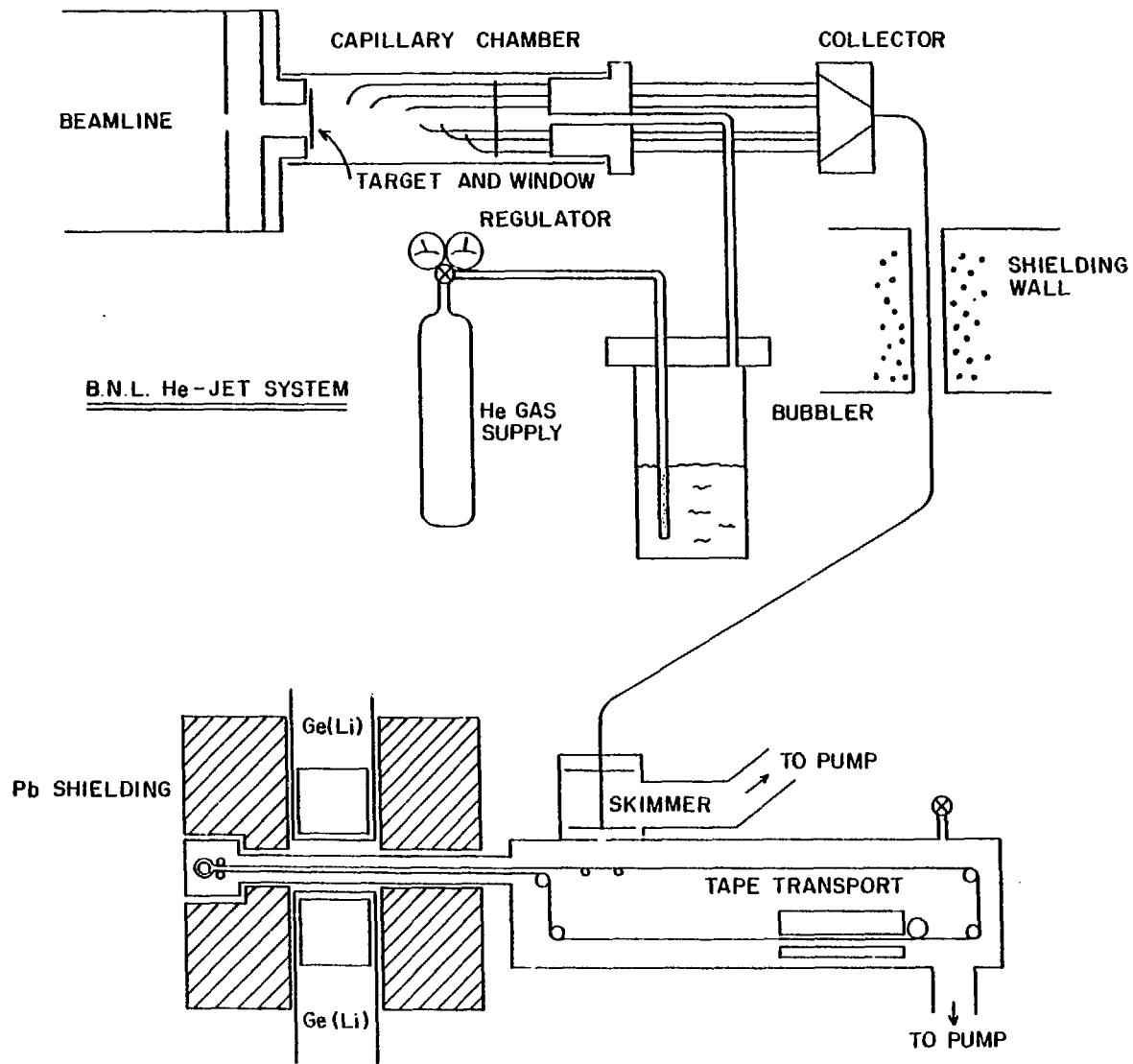


Figure 9