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# RARE PARTICLES*

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Abstract

The use of Accelerator Mass Spectrometry (AMS) to search for hypothetical particles and known particles of rare processes is discussed.



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## 1. Introduction

To call a particle rare is usually more than just a quantitative statement about its abundance. It somehow expresses the expectation that its detection will tell us something fundamental about nature. In this sense the term can be strongly time dependent since a "rare" particle at the time of its discovery can quickly lose this attribute as more efficient production and detection methods are developed. This is particularly evident for many "man-made" particles (e.g. antiprotons). On the other hand, if one considers only natural production of these particles, a more stable frame of reference is obtained and at least the quantitative aspect is less time dependent. With one exception only naturally produced particles will be discussed in this paper. They will be separated into two distinctly different classes. In section 2 searches for hypothetical particles are described, whereas in section 3 the detection of known particles produced in very rare processes is discussed.

For an actual experiment with rare particles it makes a big difference whether the particle one is trying to detect has well-known properties or is a hypothetical one with usually some uncertain physical properties. In the first case one wants to use a technique which is very selective to the properties of the particle. In the search for hypothetical particles, on the other hand, one would like to be as broad-ranged as possible for parameters which are not well defined in the theoretical predictions (e.g. the mass of free quarks). Accelerator Mass Spectrometry (AMS) has the potential to adjust to these different conditions which makes it a particularly versatile instrument in experiments with rare particles. In the following a number of AMS experiments are discussed with emphasis on those which are very new. In addition to completed and ongoing work a few suggestions for possible future experiments in this area are presented.

## 2. Search for hypothetical particles

In general, hypothetical particles can be searched for in two quite different ways. One can attempt to produce them in accelerator experiments under well defined but always somewhat limited conditions. The other possibility is to search for their occurrence in nature. Here the problem is the supposedly very low concentration in normal matter and the question where to search for them. Depending on the specific particle one is looking for the source of production can be quite different. For example, superheavy elements may have been produced in some stage of stellar evolution whereas free quarks and anomolously heavy baryons could actually be remnants of the big bang. In any case the particle will eventualy get involved in some chemical processes whose nature can only be guessed. Besides the mere discovery of a new particle it is a very intriguing aspect of this field to find possible fingerprints of the early universe.

### 2.1 Fractionally charged particles

The confinement of quarks in hadrons is one of the most fascinating aspects of modern particle physics. Since it does not follow as a rigorous consequence of the theory of strong interaction (QCD), it can still be considered an open question. Certainly, when the first reports from Stanford were published [1] showing evidence for fractional charges on superconducting Niobium spheres there was great hope to find them somewhere else too. However, neither in accelerator experiments nor in cosmic-ray or in stable matter searches any further evidence for free quarks has been seen. The Stanford group continued their searches [2] confirming the earlier results and finally published a paper [3] presenting very strong evidence for the observation of fractional charges. Despite great efforts, nowhere else have they been found in well over hundred experiments since the concept of quarks was proposed two decades ago.

AMS seems a natural method to get involved in these searches but only limited effort was devoted to it in the past. Recent experiments attempt to cover a much larger range of possible fractional charges and masses and some of the present results will be discussed here.

The Stanford experiments [1-3] found fractional charges on a macroscopic object, superconducting Nb spheres of about $10^{-4}$g (~ $10^{18}$ Nb atoms). It is interesting to note that Millikan in his work on oil drops [4] measured electron charges on objects of only about $10^{-11}$g, where the charge dominates all other electric forces (e.g. dipole interactions) and gravitation is of the same order of magnitude. For the $10^{7}$ times heavier Nb spheres the comparatively huge gravitational force needs to be precisely balanced by a magnetic levitation, and electric higher-moment background forces are of similar strength as the force on the charge. Therefore it seems difficult to extract the residual fractional charges. Whether these charges are due to fractionally charged particles is still uncertain. However, the frequent changes of fractional charges observed on some of the Nb balls suggest a rather high mobility of whatever the object is. For an AMS experiment one can therefore try to evaporate these objects and accelerate them. This was the basis of the recent quark search at Argonne [5]. A more rigorous method is to atomize the material by sputtering, a line pursued in ongoing quark searches at Caltech [6], Rochester [7], and Toronto [8]. Compared to Millikan-type experiments the AMS methods have the virtue of probing directly and unambiguously the fractionally-charged nature of atomic particles.

At Argonne we were intrigued by the fact that the Stanford experiment was performed at very low temperature with Nb in its superconducting state. Consequently, our AMS search was designed to come as close as possible to the conditions at Stanford. Our experiment was based on the hypothesis [9] that cryogenic temperatures reduce the mobility of + 1/3 e charged particles to such an extent that an observation becomes possible in a Millikan-type experiment, whereas at room temperature they move freely through matter never spending enough time on a macroscopic object to be detected. The observed distribution of + 1/3 e and - 1/3 e charges on the Nb balls [3] may then be explained as a random distribution of + 1/3 e charges only, - 1/3 e charges being equivalent to two + 1/3 e charges on the same ball (plus one electron charge). Therefore our experiment was designed to search for + 1/3 e charged particles. Here only a short summary of the results are given. Details can be found in ref. [5].

A cryogenic ion source was built (fig. 1) in which a Nb filament could be cooled to liquid He temperature for several hours. The source was mounted in the terminal of a Cockcroft-Walton accelerator operated at + 700 kV. Any positively charged particle emerging from the Nb filament was accelerated to an energy corresponding to its charge and measured in a Si surface barrier detector mounted at ground potential. The Nb filament could be heated in fractions of a second from 4.2 °K to room temperature or several hundred degrees above. It was expected that fractionally charged particles trapped in the cold filament would be released at the elevated temperature and would show up as a burst of particles of corresponding energy. An example of a typical run showing cold and hot conditions is given in fig. 2. It clearly shows that there is not the slightest indication of the expected effect. The result of our experiment can be summarized in saying that we have not observed any excess events when the Nb metal was heated from 4.2 °K to 500 °K, above a randomly distributed background of about one count per minute. A mass range from 0.01 to 100 amu was covered, where the lower limit was determined by residual field deflection and the upper limit by pulseheight defects in the Si detector. Compared to the Stanford Nb balls our filament had an area which was larger by a factor of 160 and a volume larger by a factor of 1250. Thus, assuming equal trapping probability we should have seen a burst of several hundred fractionally charged particles in the first few seconds after the heating. We clearly have seen more. The interpretation of a negative result is always difficult and our result is no exception. However, we believe that the goal to test a hypothesis which may have explained the Stanford results has been achieved.

The efforts currently underway at Caltech, Rochester and Toronto cover a wide range of fractionally charged particle properties (charges and masses). They all start with atomizing certain materials in an ion source, accelerating the ions in tandems and using various charge-changing processes combined with energy and electric rigidity measurements to filter the desired particles from the background. These searches should be capable of reaching sensitivities of around $10^{-18}$ fractionally charged particles per "normal" atom, approximately the concentration seen in the Stanford experiment [3]. Although, based on the current experimental evidence, the hope of finding free quarks is slim, it is a question of such fundamental nature that it is certainly justified to pursue it as long as one can significantly improve on the detection limit.

## 2.1 Anomalously heavy isotopes

The term anomalously heavy isotope is used for stable isotopes of known elements with masses far beyond what can be formed by simply adding neutrons. The anomalous mass results from binding a hitherto unknown heavy particle to the nucleus. A number of papers have been published on this subject [10-14], with the general conclusion that the existence of such particles cannot be excluded from our current understanding of fundamental theories. These particles may be neutral [10-12] or integrally charged [13-14] with masses up to $10^5$ amu. It is conjectured that these particles may have been created in the big bang and that some fraction survived the particle-multiparticle annihilation. Primordial abundances of anomalously heavy isotopes of some light elements with $Z > 1$ were estimated [11,12] to lie in the range around $10^{-10}$, with concentrations of anomalous Be isotopes as high as $10^{-6}$ [13]. Such high abundances are well within the reach of AMS measurements.

AMS searches at the FN tandem in Philadelphia on anomalously heavy isotopes of O [15], He, Li and Be [16] have established abundance limits several orders of magnitude below the predictions [11,12], with the exception of Li. Mass ranges of 20 to 54 amu (O), 3.06 to 8.12 (He), 11 to 32 (Li), 16 to 93 (Be) were covered by incrementing the magnetic elements of the accelerator system in small, mass-overlapping steps.

Searches for anomalously heavy isotopes of hydrogen were performed with a variety of methods, including mass-spectrometric [17,18] and time-of-flight techniques [19] at keV energies, and AMS measurements at MeV energies with a cyclotron [20] and a Van de Graaff accelerator [21]. The most stringent limits were established by the group at Rutherford Laboratory [19] which used a water sample enriched in heavy isotopes by a factor of about $10^{11}$. This allowed to set abundance limits of $10^{-28}$ to $10^{-29}$ for a mass range of 12 to 1200 amu. Limits of $2 \times 10^{-19}$ were measured for the mass range 3 to 8.2 amu [20] and $3 \times 10^{-18}$ for 6 to 16 amu [17] using deuterium as source material.

In summary, the searches for anomalously heavy isotopes have established limits well below the abundance estimated from cosmological models. However, it is difficult to predict whether these primordial abundances will be preserved in matter we find on earth. It is conceivable that depending on mass an appreciable depletion of these heavy isotopes may have occurred through their geophysical and geochemical history. The uncertainty as to where to search for these species seems to have discouraged further AMS attempts in recent years.

## 2.3 Superheavy elements

Compared to anomalously heavy isotopes discussed in the previous section, predictions about superheavy elements are based on a relatively straightforward extrapolation of known nuclear and atomic properties. They are expected to occur around $Z \approx 114$ and $A \approx 300$, however predictions about half-lifes vary by many orders of magnitude. To find primordial superheavy elements with AMS techniques requires half-lives of at least $1 \times 10^8$ years. A rather limited AMS effort has so far been devoted to this subject. The most thorough AMS experiment [22] was performed with the FN tandem in Philadelphia in a search for naturally occurring element $Z = 110$, $A = 294$ in its supposedly chemical homolog platinum. Using a platinum nugget from Alaska as the source material and a time-of-flight technique for identifying mass 294, a limit on the presence of $^{294}110$ in platinum of 1 part in $10^{11}$ was established.

Searches of similar kind could certainly be performed for other superheavy elements. The continuous improvement of TOF techniques should encourage further experiments. In order to develop a reliable technique for this heavy mass region one might consider to start with an AMS attempt to confirm the existence of $^{244}Pu$ ($T_{1/2} = 8.5 \times 10^7$ y) in nature, originally detected with a mass spectrometric measurement by Hoffman et al. [23]. This group found a $^{244}Pu$ concentration of 2.4 x $10^6$ atoms per kg of Precambrian bastnatite, a rare earth fluocarbonate mineral. As the authors pointed out [23] a confirmation of this singular result which gave an unexpectedly high $^{244}Pu$ concentration would be important for our understanding of the possible origin of very heavy elements.

## 3. The detection of known particles produced in rare processes

In the context of this paper a rare process is one which produces a very low number of atoms in any reasonable amount of target material. For example, the inverse β decay induced by solar neutrinos on earth produces typically about one atom per ton of target material per year. The most extreme example is the predicted proton decay where about 1000 tons of material are needed to give the rate of one decay per year. A comparatively frequent process is the double β decay which requires about 100 g of material for one decay per year. The very recently observed spontaneous emission of $^{14}C$ from $^{223}Ra$ [24] is another process one might call a rare one. $^{223}Ra$ is part of the $^{235}U$ decay series and about 0.1 g of natural uranium would give one $^{14}C$ emission per year. In fact, of all the examples given above the last one is the most performable one using AMS techniques. Before going into more detail about these examples a recent AMS experiment from Argonne to search for doubly charged negative ions [25] will be discussed.

### 3.1 Search for doubly-charged negative ions

Although once thought to be produced in nA quantities [26] doubly-charged negative ions have escaped their detection in any subsequent measurements. AMS is well suited to search for these apparently rare ions with great sensitivity. At Argonne [25] we looked for the production of $^{11}B^{2-}$, $^{12}C^{2-}$ and $^{16}O^{2-}$ from an inverted Cs-beam sputter source by tuning the FN tandem accelerator system in such a way that only ions fully stripped at the terminal could reach the final detection device, an Enge split-pole magnetic spectrograph. Since doubly charged negative ions gain twice as much energy on their way to the terminal as compared to singly charged negative ions, the final energy of fully stripped ions delivers an unambiguous signature. No singly-charged negative ions of the same element can gain as high an energy as the doubly negative ones. Figure 3 shows a spectrum measured in the spectrograph focal plane detector when the accelerator system was tuned to accelerate $^{12}C^{2-}$ ions. Clearly, no counts corresponding to $^{12}C^{2-}$ ions were found. The following limits for ratio of doubly-charged to singly charged ions were measured: $^{11}B^{2-}/^{11}B^{-} < 1 \times 10^{-15}$, $^{12}C^{2-}/^{12}C^{-} < 2 \times 10^{-15}$, $^{16}O^{2-}/^{16}O^{-} < 2 \times 10^{-14}$. It is possible that doubly-charged ions could be more easily produced in a charge exchange ion source, similar to metastable $He^{-}$ ions. Searches with such a device at facilities set up for free quark searches (Caltech, Rochester, Toronto) should have a good chance to find these elusive ions.

### 3.2 Solar neutrino detection

The detection of solar neutrinos on earth by Ray Davis, Jr. et al. [27,28] from Brookhaven is an astounding achievement of radiochemical methods. About 40 $^{37}Ar$ atoms, produced over several months through the reaction $^{37}Cl\ (\nu,e^{-})\ ^{37}Ar(T_{1/2} = 35.0\ d)$ in 550 tons of $C_2Cl_4$ ($\sim 10^{31}$ atoms) are extracted and counted through their radioactive decay. This can hardly be accomplished by any other technique. Only the method of Resonance Ionization Spectroscopy (RIS) [29,30] seems to have the potential of counting such a small number of atoms directly. The radiochemical method works of course only for relatively short-lived atoms, since essentially all of them have to decay in a reasonable time. Both RIS and AMS are independent from half-life limitations which opens the possibility of using other radioisotopes. At present, the overall efficiency for detecting an atom in a sample with AMS is at the very best a few percent. Often it is several orders of magnitude lower. This means that a detector experiment involving only a few tens of atoms like the one by Davis is not possible with AMS. However, there are several proposals for so-called geological solar neutrino experiments with long-lived radioisotopes which can accumulate over a very long time span in minerals. The following systems were suggested: $^{41}K(\nu,e^-)^{41}Ca(T_{1/2} = 1.0 \times 10^5$ y) ref. [31]; $^{81}Br(\nu,e^-)^{81}Kr(2.1 \times 10^5$ y) ref. [32]; $^{98}Mo(\nu,e^-)^{98}Tc(4.2 \times 10^6$ y) ref. [33]; and $^{205}T\ell(\nu,e^-)^{205}Pb(1.4 \times 10^7$ y) ref. [34]. Depending on the particular system equilibrium concentrations of solar neutrino products of the order of $10^5$ to $10^7$ atoms per ton of mineral can be expected. With its great power of discrimination AMS could indeed be used for the final stage of counting the neutrino-produced atoms. An interesting attempt to develop the AMS technique for detection of $^{205}Pb$ is underway at the UNILAC accelerator at GSI [35]. From Rochester first attempts to study the mass-98 system were reported [7].

It is well known that every solar neutrino experiment is extremely difficult. An important question for the geological experiments is whether one can reliably estimate background reactions producing the same radioisotope. Much work needs to be done in this respect for any of the above mentioned systems. However, AMS is developing towards becoming a viable tool for the detection aspect of the problem. In view of the fundamental importance for our understanding of stellar processes it would be nice to see that some of these attempts develop into real projects.

### 3.3 Spontaneous emission of $^{14}C$ from $^{223}Ra$

In a recent experiment at Oxford [24] a branching ratio of $(8.5 \pm 2.5) \times 10^{-10}$ for the emission of $^{14}C$ nuclei relative to α-particles from $^{223}Ra$ has been measured. The experiment was performed with a Si ΔE-E telescope detector system by direct counting of carbon ions. A confirmation of this interesting result could be either done with an improved detector system (e.g. or magnetic spectrograph to suppress the α particles) or by looking for $^{14}C$ accumulated in a uranium mineral. Since $^{223}Ra$ is part of the $^{235}U$ decay series one can easily estimate that the $^{14}C$ equilibrium concentration should be $1.3 \times 10^5$ $^{14}C$ atoms per gram of uranium. If one assumes a 1 ppm carbon content of the uranium mineral one would get a $^{14}C/^{12}C$ ratio of $2.6 \times 10^{-12}$ approximately twice the concentration of $^{14}C$ in the biosphere. If the production of $^{14}C$ from other sources, in particular (α,n) reactions on light elements of the uranium mineral, turns out to be negligible a verification of the Oxford result with AMS should be quite feasible.

## 4. Conclusion

AMS has grown into a very powerful tool for trace-isotope measurements in many different areas of research. Its potential is, however, not restricted to species with well-known properties. The steadily increasing computerization of heavy-ion accelerators certainly supports searches in an unknown parameter space of particle properties. Although the search for hypothetical particles always has the great risk of ending with a negative result, the importance of such a result grows in proportion to the confidence in the measuring technique. In this sense the search for the unknown with improved AMS techniques should continue. The history of physics tells us that surprising results very often showed up by mere improvement of the equipment.

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## Figure Captions

Fig. 1 Simplified layout of the cryogenic source and the acceleration structure used in the Argonne search for fractionally charged particles in Niobium [5].

Fig. 2 Two-dimensional display of the events measured in a Si detector when the Nb filament shown in fig. 1 was subject to temperature changes shown in the lower part of the figure. The event threshold allows for single counts to be visible. The trace of particles around 680 keV is due to $H^+$ ions condensed on the filament at 4.2 °K (from ref. [5]).

Fig. 3 Two-dimensional spectrum measured in the AMS search for doubly-charged negative carbon ions at Argonne [26]. The expected location of $^{12}C^-$ and $^{12}C^{2-}$ ions accelerated to maximum energy is indicated. The $^{12}C^{5+}$ ions originate from $^{12}C^-$ background ions injected into the tandem. $^6Li^{3+}$ ions show up as an intense background since they are ion-optically identical to $^{12}C^{2-}$ ions.

ANL-P-17,088

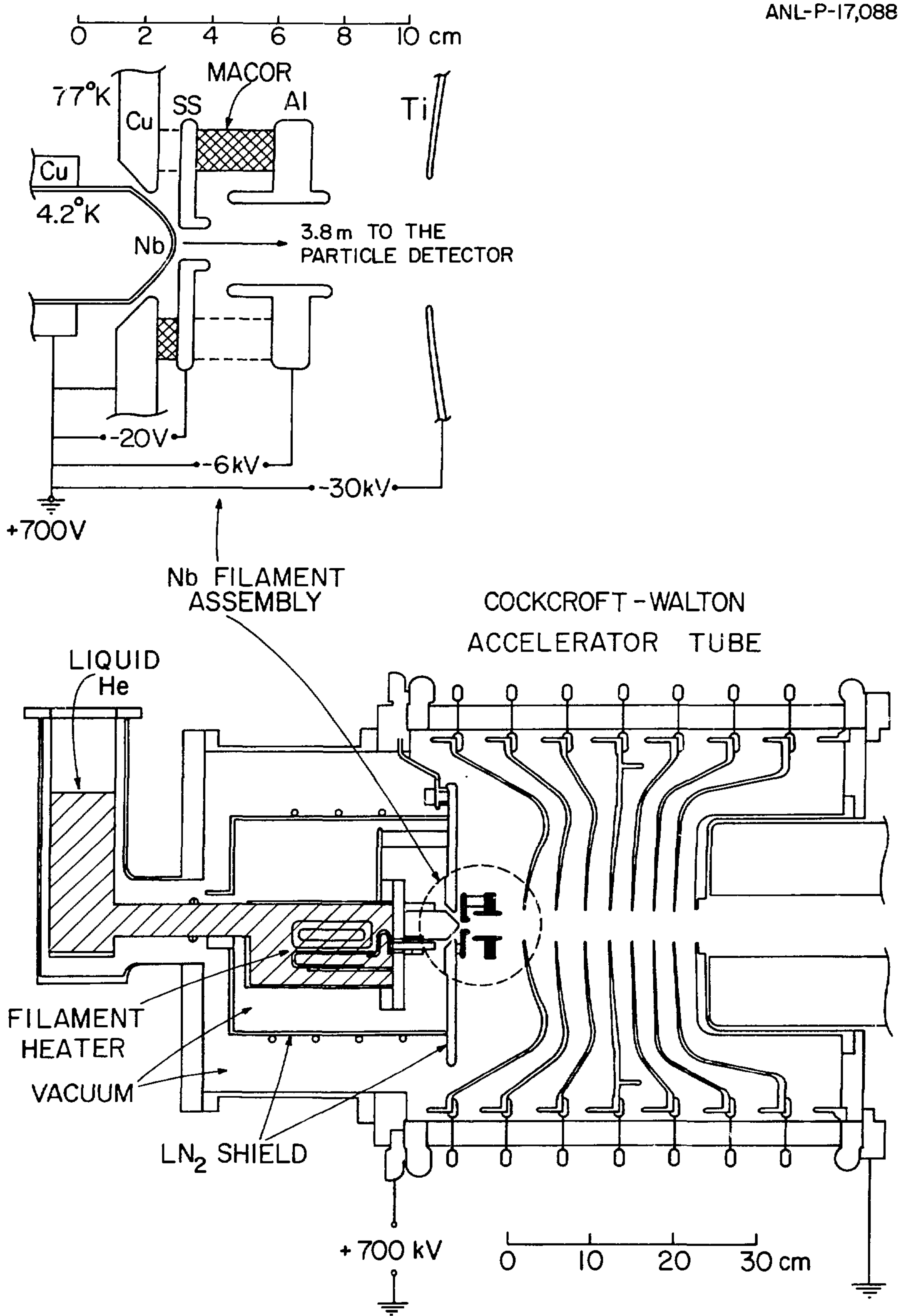


Fig. 1

ANL-P-17,099

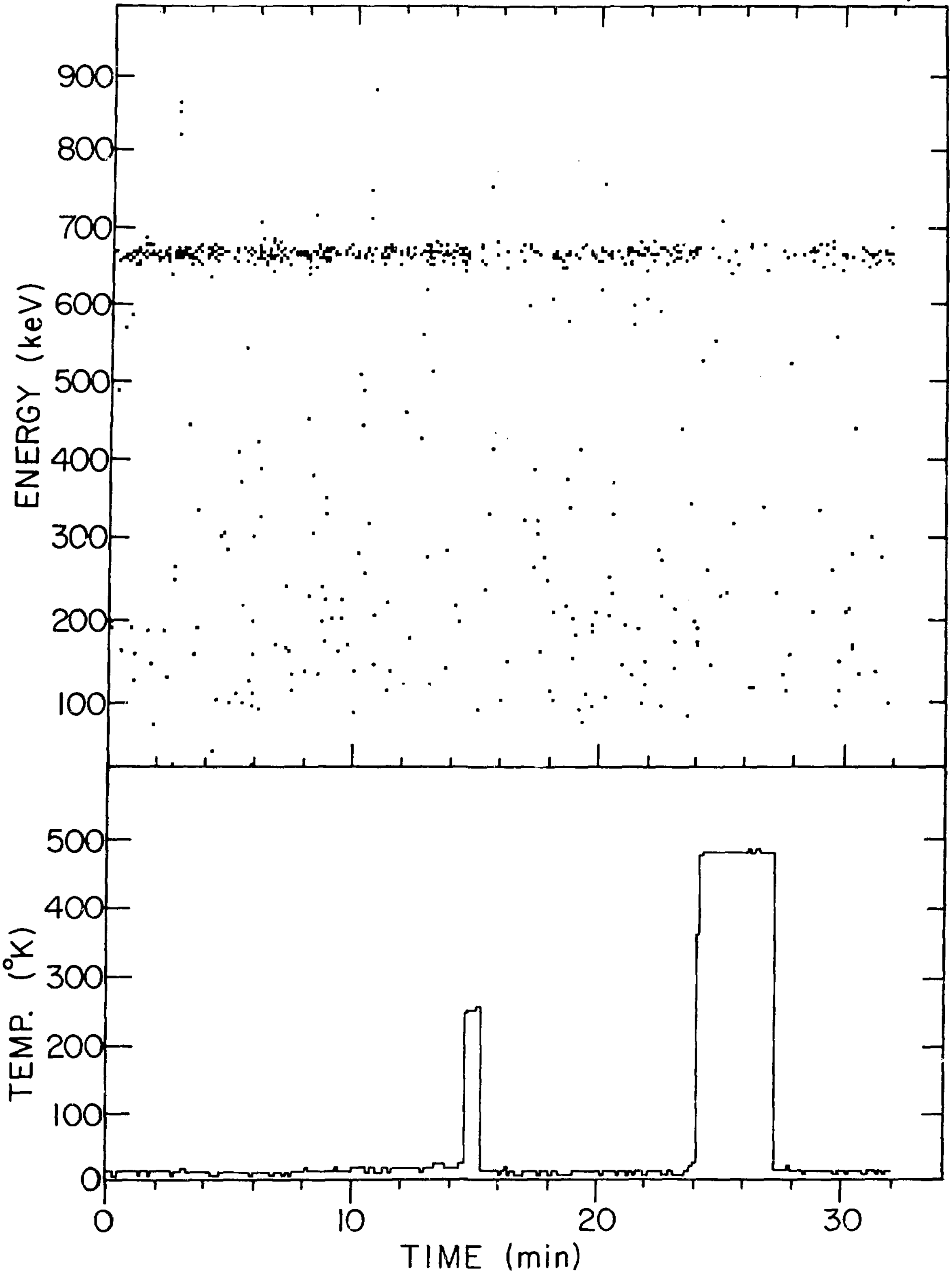


Fig. 2

ANL-P-17,036

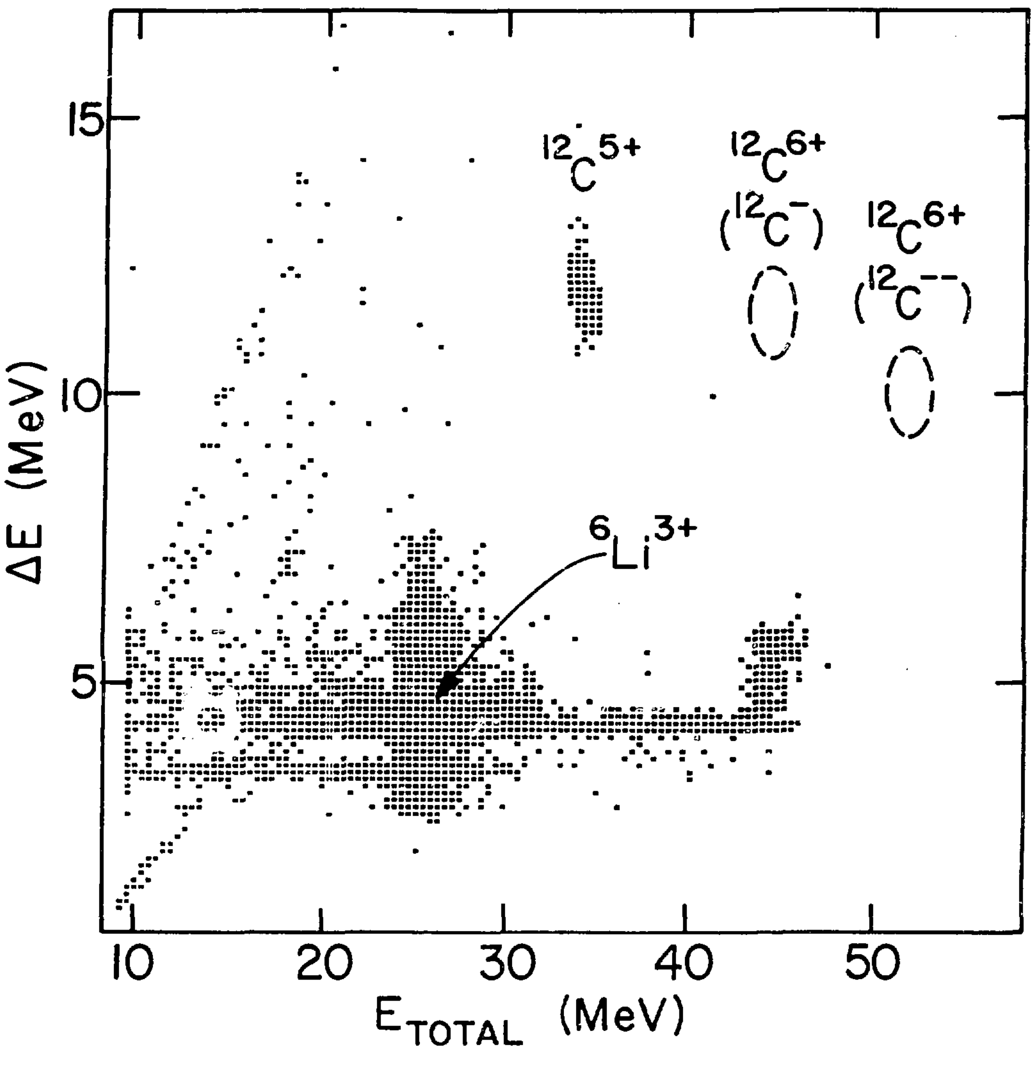


Fig. 3