Attempts to Use Accelerator Mass Spectrometry to Detect Fractionally Charged Particles in Nature [and Discussion]

R. D. McKeown, W. Kutschera and K. Ledingham

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Attempts to use accelerator mass spectrometry to detect fractionally charged particles in Nature

By R. D. McKeown

W. K. Kellogg Radiation Laboratory, California Institute of Technology, Pasadena, California 91125, *U.S.A.*

The search for fractionally charged particles (FCP) in Nature is ultimately motivated by the belief that the fundamental constituents of the atomic nucleus are quarks, which have charge in integral units of $\frac{1}{3}$ of the electronic charge. The reported observation of fractional charge in niobium by a group at Stanford University in 1981 has motivated many new efforts to detect FCP in the past few years. The techniques of accelerator mass spectrometry (AMS) have been successfully applied to this problem at several laboratories. The method generally involves the use of electrostatic analysis systems to separate the FCP from integrally charged ions, since the mass of the FCP is not known *a priori*. A variety of materials have been searched in these experiments and the most sensitive limits are at concentration levels of less than 10^{-18} FCP per atom of host material.

INTRODUCTION

One of the goals of modern particle physics is to identify the fundamental constituents of matter, and to explore their properties. Near the beginning of this century, Millikan (1910) performed pioneering experiments that showed electric charge to be quantized in units of a fundamental value, e. Until the 1960s, it was generally believed that the ultimate constituents of matter would be particles that contained electric charge in integral multiples of e and all experimental evidence was consistent with this belief. In 1964, Gell-Mann (1964) and Zweig (1964) independently proposed that the observed hadrons (particles that are subject to the strong nuclear force) were composed of more fundamental particles which Gell-Mann called 'quarks'. The idea was motivated by experimental evidence that the proton (one of the hadrons that is present in ordinary atomic nuclei) has a finite size, and that the hadrons came in groups with similar properties called multiplets. The pattern of the multiplets can be explained if all of the many known hadrons are composed of quarks in various combinations allowed by certain rules. In addition, the quarks have fractional electric charge in units of $-\frac{1}{3}e$ or $+\frac{2}{3}e$, but the group theoretic rules invoked by Gell-Mann and Zweig only allow combinations with integral charge to be associated with the observed particles. It was later shown that the structure of the proton as measured in high-energy electron scattering was in fact well described by a model with 'point-like' constituents, which could be identified with quarks.

This very successful description of the structure of hadronic particles in terms of fractionally charged quarks motivated many experiments to search for evidence of the existence of 'free' quarks (not confined inside hadrons of integral charge). These experiments were generally of three types: production at high-energy accelerators; searches in cosmic rays or their interaction products; and searches in bulk, stable matter. The high-energy and cosmic-ray searches were based on the possibility that perhaps very high-energy collisions would liberate a quark for observation. Stable-matter searches covered the possibility that the quarks might be left over from the 'big bang' at the beginning of the Universe in some small concentration. Stable-matter searches also represent integrals over billions of years of cosmic-ray exposure, and so complement the other searches. Experiments were performed over a period of many years, with no convincing evidence for the observation of free fractionally charged particles (FCP). A rather comprehensive review of the situation was written by L. W. Jones (1977), and the reader is referred to that article for further information on the subject during that period. Nevertheless, some experimental efforts have continued to the present in all three categories, and some very interesting results were obtained in one of the stable-matter searches that stimulated further work in this field in the 1980s.

Using a technique that involves magnetic levitation of superconducting niobium spheres, LaRue *et al.* (1981) observed that the behaviour of some of the spheres under the application of an electric field indicated the presence of fractional electric charge. Their published results are shown in figure 1 and would seem to indicate a concentration of FCP per niobium atom of at least 2×10^{-18}. Similar experiments by another group on iron samples showed no such effects (Marinelli & Morpurgo 1982), but it is of course possible that the chemistry of FCP might lead to higher concentrations in certain materials. The results obtained by LaRue *et al.* (1981) motivated many new experiments attempting to obtain additional evidence for or against the existence of FCP.

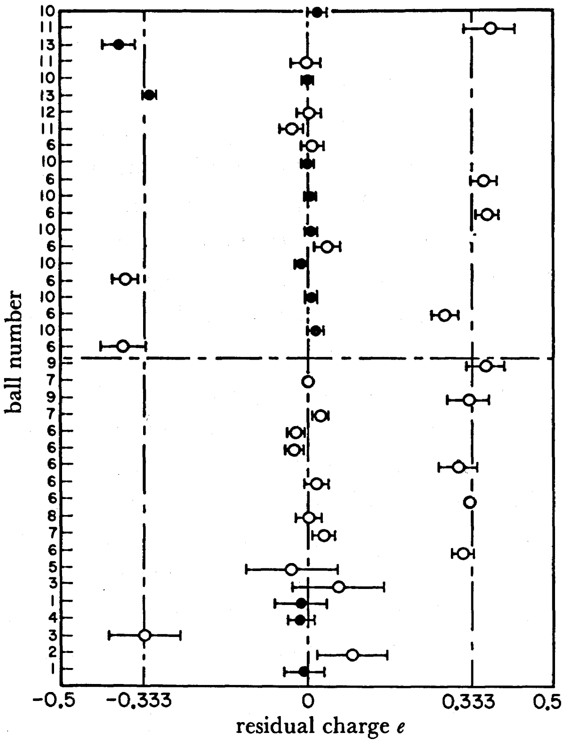

FIGURE 1. Residual charges on niobium spheres reported by the Stanford group (LaRue *et al.* 1981).

During the last decade, the theoretical understanding of quark confinement in hadrons has developed considerably. A renormalizable gauge theory, quantum chromodynamics (QCD), has been very successful at describing a wide variety of strong-interaction phenomena at high energies and has become accepted as the correct theory of the strong interaction between quarks. Numerical calculations with this theory strongly indicate that the quarks are perfectly confined and cannot exist as free isolated particles (Creutz *et al.* 1983). Nevertheless, it is true that the fundamental constituents of matter are now believed to possess fractional charge in integral multiples of $\frac{1}{3}e$, and even if the quarks cannot exist in isolation one might find other new particles by searching for FCP. Such a discovery would probably be even more interesting than a free quark.

Many of the new efforts to detect FCP have used the recently developed technology of accelerator mass spectrometry (AMS), which is the main subject of this paper. In the next section, I will describe some of the general principles and techniques applied to this problem. The following section describes the recent history of this subject summarizing the work at various laboratories. A subsequent section presents our work at Caltech in some detail. Finally, I will conclude with some results from experiments not using AMS, to update the status of the whole field of FCP searches.

Description of techniques

The use of AMS to search for FCP is attractive for a variety of reasons. In general, many different materials can be searched (most other techniques are more selective in this regard). If a positive signal were obtained, further information (e.g. the mass of the particle) could be measured and the particles could be concentrated for further study.

There are four basic elements to the apparatus used in FCP searches of this type. The first element extracts the FCP from the host sample, the second accelerates the particles, the third performs some electrostatic (and sometimes magnetic) analysis, and finally the particles are detected in a particle-detection system. Whereas many variations of each element have been employed by different experiments, I will try to discuss the general features of each element in turn and leave specific comments on particular systems for the next section.

The first element is the source of particles used in the experiment, and the choice will depend on the material to be searched and possibly some hypothesis about the FCP and their properties. There seem to be two general classes of FCP sources used: normal ion sources and specially designed sources. Normal ion sources are generally designed to maximize the output of ions, and therefore generate large background beams of integrally charged ions. The advantage is that few assumptions about the efficiency of extraction of FCP are required because the ion source efficiently ionizes all atoms in the sample. Specially designed sources for extracting FCP can often enhance the signal-to-background by eliminating or reducing the integrally charged ion flux through the apparatus, but the extraction efficiency for FCP is sometimes difficult to estimate.

A general aspect of most FCP searches of this type is the use of primarily electrostatic extraction, acceleration, and analysis systems. By using non-relativistic mechanics, one can show that the trajectory of a particle starting from rest in purely electrostatic fields is independent of the mass of the particle. Because one would like to search for FCP with a large range of masses (usually from 0.3 GeV c^{-2} up to 100 GeV c^{-2} or higher), it is important that the apparatus be set up to avoid selecting a narrow mass range. The use of electrostatic systems conveniently allows this feature. Both single-ended and tandem electrostatic accelerators have been used in these experiments.

The use of electrostatic analysis following acceleration is typical of all of these experiments for the reason stated above. Often this is followed by a magnetic analysis to eliminate integrally charged background beams (especially if a normal ion source is used); one then scans between the peaks of these background beams searching for particles with anomalous charge : mass ratios.

There are mass-dependent relativistic effects in electrostatic analysers that can be an important consideration in these experiments. The relativistically correct expression for the radius of curvature of the orbit of a particle with charge q, mass m, and kinetic energy K, in a transverse electrostatic field ϵ is

$$\rho = \frac{2K}{q\epsilon}\frac{1+K/2mc^2}{1+K/mc^2}. \tag{1}$$

For $K \ll mc^2$, ρ is independent of m. However, if $K = 1$ MeV and $m = 250$ MeV c^{-2} one finds that ρ differs from that of heavy particles by 0.2%. Many experiments are performed with $K \geqslant 1$ MeV, so a high-resolution analysing system can limit the mass range of the measurement.

The final element of the apparatus associated with these experiments is the particle-detection system, which measures K, the kinetic energy of the detected particles. Thus, the electrostatic analyser selects $\rho \propto K/q$ and the detector measures K yielding a unique value for q. Again there may be mass-dependent effects in the kinetic-energy measurement, such as the well-known pulse-height defect (Kaufman *et al.* 1974). The sensitive mass range of silicon detectors for FCP has been studied in some detail by Lewin & Smith (1985). For FCP with $m < 100$ GeV c^{-2} one expects the response of detectors to be similar for FCP and normal incident particles with kinetic energies of megaelectronvolts, so a reasonably accurate correction for mass-dependent effects can be applied. Very light mass FCP ($m \leqslant 1$ GeV c^{-2}) can be very penetrating; this necessitates designing detector systems capable of handling ranges for stopping particles from *ca.* 10 μg cm^{-2} to *ca.* 1 g cm^{-2}. This is generally accomplished by using stacks of solid-state silicon detectors and/or gas-ionization chambers.

Finally, I would like to point out an advantage of using a tandem accelerator in these experiments. All particles (including FCP) emitted from the source of a single-ended electrostatic accelerator emerge with the same ratio of kinetic energy per unit charge, and thus the same (non-relativistic) trajectory in an electrostatic deflector (see (1)). Thus one must generally use a specially designed low-background source or some magnetic analysis with these accelerators to avoid prohibitively high count rates in the detectors. However, a tandem accelerator can produce FCP beams at unique values of the ratio.

$$R \equiv K/q. \tag{2}$$

If we consider a terminal voltage V_0, integrally charged ions emerge with the values $R = 2V_0, \frac{3}{2}V_0, \frac{4}{3}V_0\ldots$ (I assume only charge $-e$ injected). FCP injected with charge $-\frac{2}{3}e$ will emerge with $R = 3V_0, \frac{3}{2}V_0, \frac{9}{7}V_0\ldots$, and those injected with $-\frac{1}{3}e$ have $R = 2V_0, \frac{3}{2}V_0, \frac{4}{3}V_0\ldots$ as for the integral charges. Note that the value $R = 3V_0$ is unique to FCP with nuclear charge $Z = n+\frac{1}{3}$, where n is an integer $n \geqslant 0$. Therefore these FCP will have unique trajectories in the electrostatic analyser resulting in large background suppression and very clean experiments. In reality there are particles, even 'beams' in the vicinity of the $R = 3V_0$ trajectories that are generally caused by rare charge-exchange processes in the vacuum system of the acceleration–transport system. Of course, the injection energy has been neglected in the above treatment, but it is straightforward (and necessary) to include it, and the basic conclusions remain unaffected. One must also estimate the charge-state fractions for FCP in the stripper of the tandem to compute the experimental sensitivity, but this can be done by using the known stripping properties of integrally charged particles. Use of a tandem also requires formation of negative FCP in the source before injection.

SURVEY OF TECHNIQUES AND RESULTS

The first published work in which AMS was used to search for FCP is by Elbert *et al.* (1970) and contains many of the key ideas exploited in more recent work. This effort made use of a single-ended 1 MV accelerator, an RF ion source, an electrostatic analysis system followed by a magnetic analyser and a silicon detector. Various gas samples were searched, and their results for air are shown in figure 2 where it can be seen that sensitivities of order 10^{-13} per nitrogen molecule were attained.

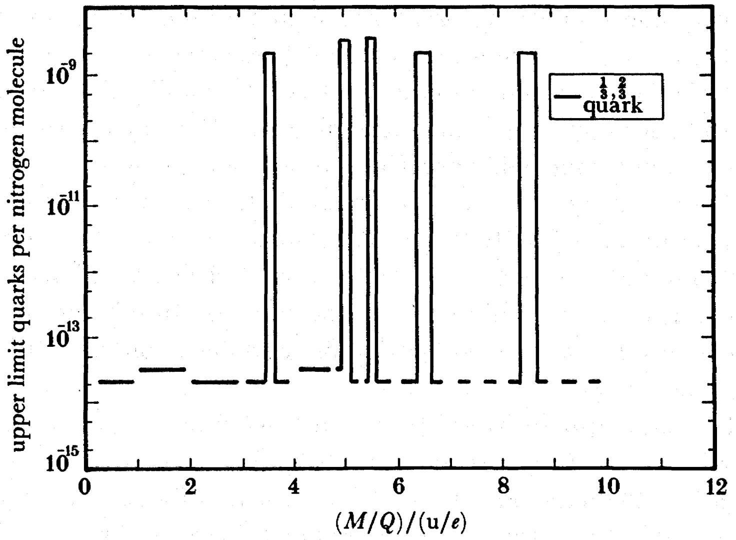

FIGURE 2. Upper limits obtained for FCP search in air reported by Elbert *et al.* (1970).

In 1977, a search for fractionally charged tungsten ions was performed (Boyd *et al.* 1978) with the University of Rochester MP tandem with a sputter source and normal magnetic analyser followed by low-resolution electrostatic analysis and a position-sensitive silicon detector. The result was an upper limit of 10^{-12} per W atom for FCP with $Z = 73\frac{2}{3}$ and mass between 182 and 192 u.† This work concentrated on tungsten because there were some indications that the early results from the Stanford niobium levitation experiments were associated with the annealing of niobium on a tungsten substrate (LaRue *et al.* 1977).

Schiffer *et al.* (1978) used a single-ended dynamitron of 1 MV and an electrostatic analyser followed by a silicon detector to search for $Z = \frac{1}{3}$ FCP in Nb, W and Fe. Metal filaments of each material were heated in the high-voltage terminal of the accelerator, and the emitted positive charges were focused and accelerated. The attained upper limits were of order 10^{-20} per atom for FCP that would diffuse out of the metals at 600 °C. A similar experiment (Kutschera *et al.* 1984) was performed with a 700 kV single-ended accelerator and a cryogenic Nb filament. The hypothesis was that FCP would be cryogenically trapped in 4.2 K niobium and then released when heated to *ca.* 500 K. The emitted particles were accelerated into a silicon detector without deflection, and no positive signal was observed over the sensitive region $10\ \mathrm{MeV}\ c^{-2} < m < 100\ \mathrm{GeV}\ c^{-2}$.

Another hypothesis for the Stanford results was explored by Boyd *et al.* (1979). At the cryogenic temperatures of the Stanford experiments, some He should be absorbed on the

† $1\ \mathrm{u} \approx 1.6605655 \times 10^{-27}$ kg.

niobium spheres, and perhaps the FCP are contained in helium gas. By using an RF source in a single-ended accelerator, a mass scan was taken with a magnetic analyser and a silicon detector. It was assumed that ions would be extracted with unit efficiency from the source bottle with the RF power off (no plasma). Because the electric fields in the extraction region are orders of magnitude greater and shaped very differently when the plasma is present, it is unlikely that the above assumption is valid. Nevertheless, Boyd *et al.* (1979) claim that an upper limit of *ca.* 10^{-15} per He atom was obtained in this experiment.

A search for FCP in gallium was performed at the University of Toronto (Chang 1984) by using a tandem accelerator (3 MV) with a liquid-gallium positive-ion source. Electron attachment for negative injection was accomplished with a lithium charge-exchange canal. It should be noted that electron attachment to a charge $+\frac{1}{3}e$ FCP followed by electrostatic analysis yields unique trajectories before injection to the accelerator assuming that no charge $-2e$ ions are formed. (This can be seen by using similar arguments to those used in the previous section regarding electrostatic analysis following acceleration in a tandem accelerator). The Toronto experiment utilized an electrostatic analysis system followed by a silicon detector, and obtained an upper limit of 1.2×10^{-14} per Ga atom for $Z = n + \frac{1}{3}$ with $0 \leqslant n \leqslant 100$, $1\ \text{GeV}\ c^{-2} \leqslant m \leqslant 300\ \text{GeV}\ c^{-2}$, $Z/m \leqslant 1(\text{GeV}\ c^{-2})^{-1}$, and electron affinity, EA, such that $|\text{EA} - 5.39\ \text{eV}| \geqslant 1\ \text{eV}$. The various limitations are the result of considering the efficiencies for positive-ion formation, electron attachment, stripping, and detection in addition to the effect of stray residual magnetic fields.

An electrostatic beamline apparatus has been constructed at the University of Rochester Nuclear Research Laboratory (Elmore *et al.* 1985). It has been used with a 0° caesium sputter source and the Rochester MP tandem (at 6.67 MV) to search for FCP in the gaseous residue of a xenon fractional-distillation process. An enrichment factor of 10^6 was assumed for this purification process and the search was limited to FCP with mass greater than 80 u. The upper limit obtained was 10^{-18} per normal xenon atom. Recently, the Rochester group has also run searches in air samples (with pre-enrichment by ionization and sweeping long-lived ions onto titanium discs) and copper samples from a Fermilab beam dump (T. Hemmick, personal communication 1986), but the analysis of these runs is not complete.

The Caltech FCP search

At Caltech, we designed a special source for searching for FCP with a 3 MV tandem accelerator. The source allowed sampling relatively large amounts of host material while minimizing the integrally charged ion yield, which resulted in a very low background experiment. This source was successfully used to search in a variety of materials with very high sensitivity.

A schematic diagram of the source is shown in figure 3. A 30 keV Ar^+ beam (usually *ca.* 25 μA) was incident on the sample. The sputter yield was generally *ca.* 5–10 per incident Ar ion, and the ion yield was typically less than a few nanoamps (negative ions were a factor of *ca.* 10 lower than positives). The secondary ions were accelerated and electrostatically deflected into the accelerator. For FCP with electron affinity higher than about 4 eV, negative particles were directly extracted and transported to the accelerator. For FCP with electron affinity lower than *ca.* 4 eV, a rubidium charge-exchange canal was used to attach an electron (the efficiency of this process is quite high for the mass range 0.1–100 GeV c^{-2}). Both configurations were usually run for each type of sample.

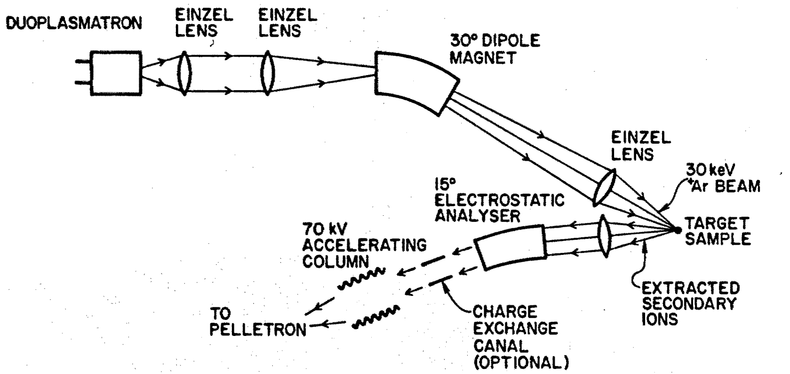

FIGURE 3. Schematic diagram of source of particles for Caltech FCP search.

The FCP sputter source generally yielded negative ions of fluorine, chlorine, and hydrogen. The yield of fluorine and chlorine was compared with the concentration in the niobium sample determined by a chemical analysis. By using the measured niobium sputter yield, the yields of these impurities indicated that they were emitted as negative ions with essentially 100% probability. This also verified that the extraction efficiency of the source optics was near unity.

The general layout of the experiment is shown in figure 4. The low-energy transport system was carefully shielded with Mu metal and the sputter chamber had bucking coils to eliminate the Earth's magnetic field (to a few milligauss) at the sputter side where the ions are moving rather slowly. The high-energy transport system consisted of an electrostatic quadrupole doublet followed by a two-stage electrostatic deflector system and had a resolution of 0.2%.

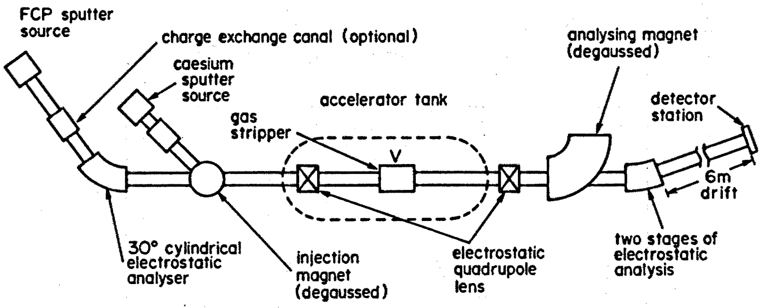

FIGURE 4. Layout of source, accelerator, and electrostatic analyser used in Caltech FCP search.

Hydrogen, oxygen, and copper beams from a Cs sputter source were generally used to establish a mass-independent tune, and slight magnetic steering was usually required to offset the effect of stray magnetic fields. In figure 5 the mass independence of the system is displayed in a ΔE–E spectrum showing protons and heavy ions that were detected simultaneously in a detector telescope with this system. (The FCP source and a niobium sample were used to obtain this spectrum; the argon sputter-beam intensity was reduced to obtain reasonable count rates and the electrostatic analyser was set for charge state +1.) Fortunately, a hydrogen beam was always present with each sputter sample, allowing universal use of this procedure.

Several detectors were used, including an ion chamber and silicon detectors of various thicknesses that were optimized for different FCP searches. In figure 6*a* the pulse-height spectrum of a silicon detector from a run on a niobium sample with negative extraction is shown.

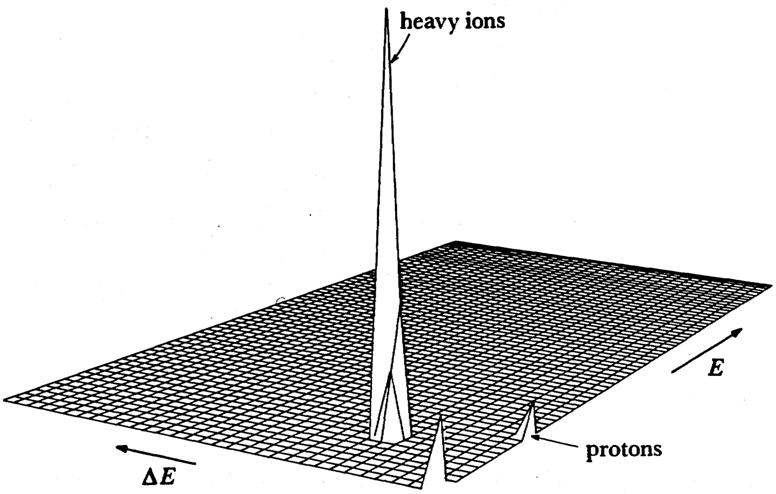

Figure 5. Perspective plot of charge-state +1 particles sputtered from niobium in the Caltech experiment (terminal voltage 2 MV). The ΔE counter was a gas ionization chamber and the E counter was a silicon detector. The heavy ions are primarily fluorine and chlorine. This plot demonstrates the mass independence of the trajectories of particles through the accelerator and beam-transport system.

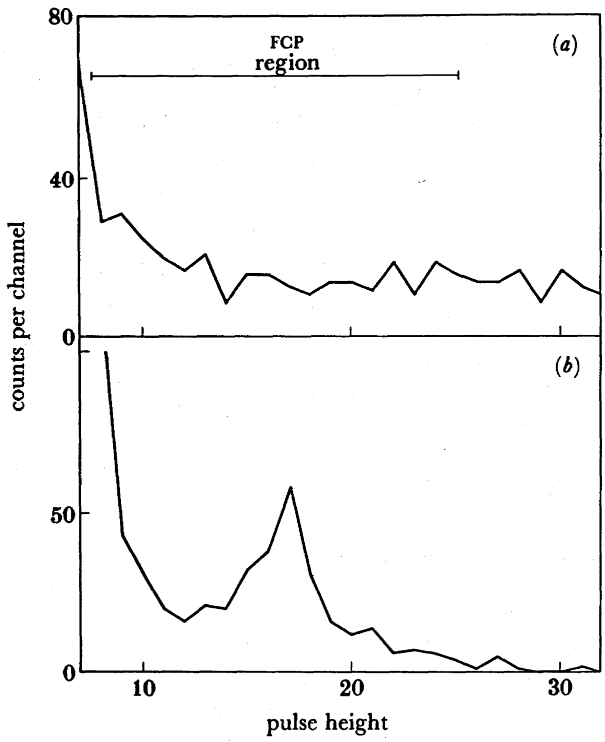

Figure 6. Pulse-height spectra in a silicon detector obtained in (*a*) the FCP search, and (*b*) a calibration run with 2 MeV Nb particles.

Hydrogen stripper gas was used to maximize the $+\frac{1}{3}$ charge state resulting from single-electron removal. A flat background due to scattered particles can be seen in the region where FCP would be expected to show a peak as in figure 6*b*, which is the response of the detector to niobium particles at the expected FCP energy. The spectrum in figure 6*a* was obtained in over 70 h of running, and can be used to set an upper limit of 1.5×10^{-18} per niobium atom for $Z = n + \frac{1}{3}$ FCP ($n > 1$) and electron affinity greater than 4 eV.

For $Z = \frac{1}{3}$, positive extraction was used because of the low electron affinity for these FCP, and nitrogen stripper gas was used to fully strip to the $+\frac{1}{3}$ charge state. A three-element detector telescope consisting of a gas ΔE counter followed by a 500 μm totally depleted silicon detector and then a 60 μm partly depleted silicon veto detector was used. (All known stable particles would stop before the veto counter.) Reduction of cosmic-ray-induced background was accomplished with plastic scintillator veto paddles surrounding the detector system. Counts that were not beam related still dominated the background in this type of experiment. An upper limit of 1.0×10^{-18} per niobium atom was obtained for the mass range $1 < m < 250$ GeV c^{-2}, and the limit for $0.2 < m < 1$ GeV c^{-2} is 2.5×10^{-19} per Nb atom.

Various other configurations were run to search for $Z = \frac{2}{3}, \frac{4}{3}$, and $\frac{5}{3}$ and the initial results on niobium and tungsten have been published (Milner *et al.* 1985). We have also searched in selenium for the types of FCP discussed by Milner *et al.* (1985) and have obtained upper limits of *ca.* 10^{-15} per host atom. In addition, various meteorite samples with isotopic anomalies were searched with typical upper limits of *ca.* 10^{-17} per nucleon of host material. Details of these latter searches will be published in the near future.

Conclusion

The experiments performed to search for FCP by using AMS techniques have been quite varied and have all returned negative results to date. In the last few years, much progress has been made in exploiting these methods to improve the sensitivity by many orders of magnitude. Experiments on niobium and tungsten have been sensitive enough to test the validity of the Stanford results and impressive results have been obtained on a variety of other materials.

Other techniques to search for FCP have also been improved in the recent past. In another levitation experiment, Smith *et al.* (1985) searched niobium coated with iron with negative results. The upper limit quoted in that work is 10^{-19} per niobium atom. The Stanford group has reported no new results since 1981.

Whereas the FCP searches represent an application of AMS techniques, the sensitivity reached in these experiments has been extended to extremely low concentrations by the implementation of various new and clever ideas. Hopefully, some of these ideas (or variations of them) will be useful in other applications of AMS. In this way, perhaps the quark hunters can repay their colleagues for the AMS technology that has made these very sensitive experiments possible.

I acknowledge my co-workers on the Caltech experiment, particularly Richard Milner, Kai Chang and Barbara Cooper, for a fruitful and enjoyable collaboration. Their advice and criticism have been very valuable to me in pursuing this subject, and their persistent efforts resulted in the very sensitive limits ultimately obtained in that experiment. I also mention George Zweig for his encouragement and for useful conversations in the early stages of this work. The support of the National Science Foundation (grant PHY82-15500) and Caltech Sloan Fund is gratefully acknowledged.

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Discussion

W. Kutschera (*Argonne National Laboratory, Illinois, U.S.A.*). Should not one of the key questions be where to look for suitable material? Niobium was chosen because of that Fairbank experiment but there could be many other materials that might be better. One needs to make a good educated guess where to look, because one cannot possibly look through the whole Earth.

R. D. McKeown. That is right. We thought that perhaps meteorites that showed isotopic anomalies would be interesting as they can be well-preserved old materials and so we tried that. Many people have lots of ideas and the problem is that the experiments are not easy. We had to run for 70 h on the niobium, so these experiments are very time consuming and difficult. Substantial improvements in the technique to improve the sensitivity would be very desirable.

K. Ledingham (*University of Glasgow, Glasgow, U.K.*). Could Professor McKeown's group do their experiment with one of the original Fairbank spheres?

R. D. McKeown. We could, but we could not achieve a sensitivity of 10^{-18}. The basic problem is that the efficiency of the system is of the order of 5–15% depending on the type of experiment you are doing. So we would need about twenty spheres to see one quark and Fairbank does not have twenty containing quarks. Our original hope that we could look at the original material is not therefore feasible.