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Search for Fractionally Charged Particles*

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We have performed a geophysical search for fractionally charged particles in seawater and rock samples. The samples were run through cation exchange columns to separate most of the material from particles of charge $\pm\frac{2}{3}$ and $\pm\frac{1}{3}$. The purified samples were converted into azides and evaporated in an electron gun, which accelerated the negatively charged particles to a solid-state detector. Quarks with charge $+\frac{2}{3}$ (q_1) would probably evaporate from azides as (q_1+e) and would thus have an effective charge of $-\frac{1}{3}$. The pulse-height spectra from the detector were scanned for peaks corresponding to particles of a charge between -1 and 0; no such particles were found. Limits on the concentration of q_1 quarks are $\lesssim 10^{-23}$ quarks/nucleon in the rock samples and $\lesssim 10^{-24}$ quarks/nucleon in the seawater samples. These limits depend, of course, on the assumptions concerning the chemical behavior of quarks made in the text.

I. INTRODUCTION

In 1964, Gell-Mann[1] and Zweig[2] independently suggested that the success of $SU(3)$ in high-energy physics could be due to the existence of a fundamental triplet of particles, quarks or aces, and their antiparticles. The most unusual property of quarks is their fractional charge. A few charge-independent searches for massive quarks have been reported.[3] The other searches have been direct or indirect determinations of charge. The ultraviolet solar spectrum has been scanned for lines that could be attributed to transitions in atoms whose nuclei contain quarks[4]; however, all candidates could be explained in terms of transitions from normal elements.[5] Hunts for quarks at high-energy accelerators[6] have put a limit on the cross section for quark production of 10^{-32}–10^{-36} cm^2 for a quark mass m_q, less than 2–7 GeV/c^2 and a charge Q of $\pm\frac{1}{3}$ or $\pm\frac{2}{3}$. Cosmic-ray experiments have put a limit on the flux of relativistic quarks at the earth's surface of 10^{-8}–10^{-10}/cm^2 sr sec for $Q=\pm\frac{2}{3}$ and $\pm\frac{1}{3}$ [7] and 1.3×10^{-10}/cm^2 sr sec for $|Q|\geq\frac{4}{3}$.[8] Results of other geophysical searches[9–12] are listed in Table I.

The motivation for the geophysical searches is that a detectable concentration of quarks could exist in various media if a steady flux of quarks has hit the earth's surface for a long time. Adair and Price[13] have made Monte Carlo calculations of the production and diffusion of quarks through an infinite ideal atmosphere. Using Fig. 6 from their paper and assuming $m_q=5$ GeV/c^2 and a quark flux at the earth's surface of 10^{-9}/cm^2 sr sec, we estimate a quark concentration of the order of 10^{-20}–10^{-23} quarks/nucleon for surface rock and 10^{-23}–10^{-28} quarks/nucleon for seawater. If $m_q=10$ GeV/c^2, these estimates are reduced by about one order of magnitude. The above estimates were made assuming that no dilution or enhancement processes, such as geochemical processes, were occurring. The concentrations would be larger if the cosmic-ray flux had been higher in the past or if quarks are left over from the formation of the solar system. Cosmological estimates[14] based on a Friedmann model vary from 10^{-9} to 10^{-18} quarks/nucleon.

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We have performed a geophysical search for fractionally charged particles in seawater and rock samples. The samples chosen were two 3-liter seawater samples and ten rock samples, which are believed to have been within a few tens of meters of the earth's surface for a time of the order of 10^6 yr. The samples were run through cation exchange columns to separate most of the material from particles of charge $\pm\frac{2}{3}$ and $\pm\frac{1}{3}$. The purified samples were converted into azides and evaporated in an electron gun, which accelerated the negatively charged particles to a solid-state detector. Quarks with charge $+\frac{2}{3}$ (q_1) would probably evaporate from azides as (q_1+e) and would thus have an effective charge of $-\frac{1}{3}$. The pulse-height spectra from the detector were scanned for peaks corresponding to particles of a charge between -1 and 0; no such particles were found. Limits on the concentration of q_1 quarks are $\lesssim 10^{-23}$ quarks/nucleon in the rock samples and $\lesssim 10^{-24}$ quarks/nucleon in the seawater samples.

II. EXPERIMENTAL PROCEDURE

A. General Description

The basic idea of the experiment is to evaporate quarks from a sample, accelerate them through a voltage V, and measure their kinetic energy T. Since $QV=T$, the charge Q is determined. A charge of $\pm\frac{1}{3}n$, where n is any integer not divisible by 3, would be evidence for the existence of quarks.

In order to collimate most of the fractionally charged particles which might evaporate from the sample, a Pierce cathode type of electron gun was used (see Fig. 1). Besides collimating and focusing, it served to accelerate the particles through a voltage V, where V was 50 kV. The energy T was measured with a solid-state particle detector (Ortec silicon surface-barrier detector TMCJ-025-500), which was coupled to a RIDL pulse-height analyzer through a charge-sensitive preamplifier and a linear amplifier. In an ideal case the particle loses all of its kinetic energy to ionization, the output voltage of the amplifier is proportional to T, and the pulse-height spectrum should exhibit peaks corresponding to the different charge states present in the beam. Figure 2 is a spectrum taken with the electron gun-detector system and using thermionic electrons, generated by heating a Ta foil (O in Fig. 1). The resolution is about 4 keV, the electronic noise extrapolates to zero at about 8 keV, and the electron peak corresponds to an energy of 50 keV. Negative particles of charge $-\frac{1}{3}$, such as (q_1+e), should give a peak at 16.7 keV. The relatively flat background between about 8 and 45 keV is mainly due to scattered electrons.

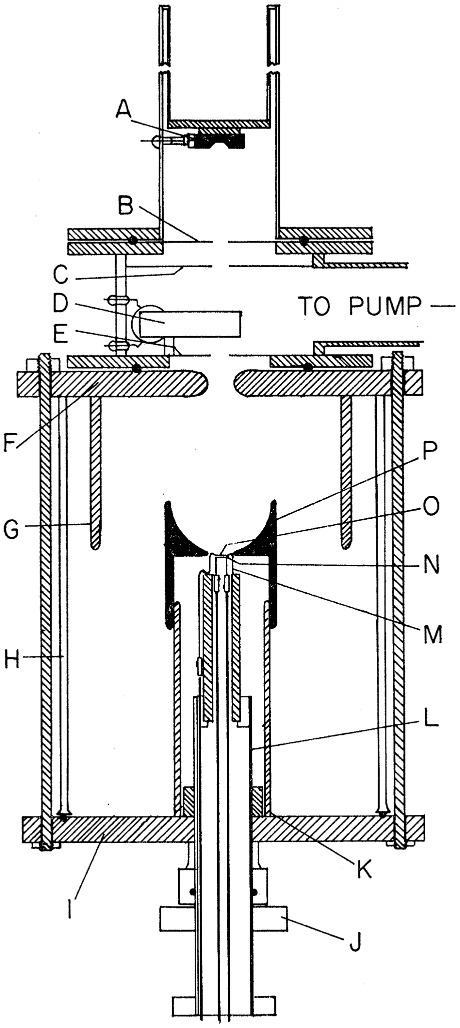

FIG. 1. The electron gun. A—detector; B, C, E—collimator; D—electromagnet; F—anode; G—focusing shield; H—glass cylinder; I—bottom HV plate; J—adjustable collar; K—guide; L—source tube; M—source holder; N—spiral heater; O—source foil; P—Wehnelt cylinder.

TABLE I. Summary of geophysical searches.

Method	Sample	Concentration limit (quarks/nucleon)	Reference
Surface evaporation	Air	5×10^{-27}	
	Seawater	3×10^{-29}	Chupka et al.[a]
	Meteorite	10^{-16}	
Magnetic levitation	Graphite	10^{-16}	Gallinaro and Morpurgo[b]
Magnetic levitation	Iron	4×10^{-19}	Stover et al.[c]
Oil drop	Oil	10^{-20}	Rank[d]
Spectroscopic	Seawater	10^{-18}	
	Seaweed, oysters, and plankton	10^{-17}	Rank[e]

[a] Reference 9.
[b] Reference 10.
[c] Reference 11.
[d] Reference 12.
[e] Reference 12.

Since the concentration of quarks in terrestrial samples is probably less than 10^{-20} quarks/nucleon, large samples such as liters of seawater or grams of rock are required. However, large samples are impractical sources

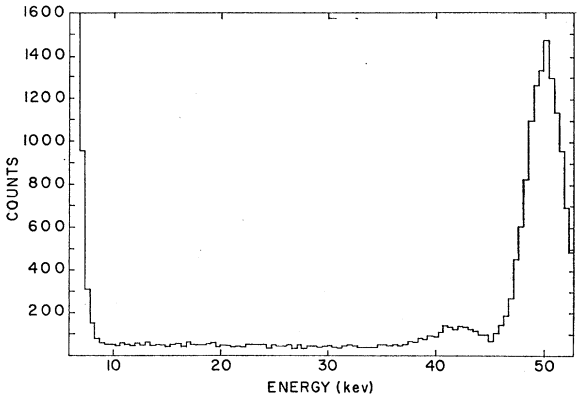

FIG. 2. Typical pulse-height spectrum for a Ta foil at 600°C.

for the electron gun-detector system. The materials were, therefore, run through cation exchange columns to separate most of the bulk from particles of charge $\pm\frac{2}{3}$ and $\pm\frac{1}{3}$. This procedure yielded solid samples weighing on the order of 10^{-3} g.

The solid-state detector is not always an ideal device for measuring the kinetic energy T of a charged particle. Only for a "high-energy" charged particle does the entire kinetic energy go into ionization. If the particle is traveling "slowly," part of its energy is lost through elastic nuclear scattering[15] and the observed peak is shifted down in energy and broadened with respect to the full-energy peak. The dead layer of gold on the detector (about 200 Å thick) has a similar effect on the spectrum. We estimated that, if quarks were evaporated bound to all but the lightest elements, then the corresponding peak(s) would be shifted down into the electronic noise. Fortunately though, if $m_q \lesssim 10$ GeV/c^2, the peak(s) corresponding to free quarks or quarks plus electrons would be easily observable. Since for most types of samples the evaporated quarks would be bound to atoms or molecules and therefore not be observable, $\mathrm{LiN_3}$ was used as a host material. When such a sample was heated to 100–300°C, the nitrogen was evaporated leaving behind mainly lithium. If quarks were present in the $\mathrm{LiN_3}$, they would presumably not be evaporated with the nitrogen, but would remain in the lithium. Upon increasing the temperature, the quarks would be evaporated with bound electrons or lithium atoms, such as (q_1+e) or $(\mathrm{Li}q_1)$. Therefore, the corresponding peak(s) should not be shifted into the electronic noise.

Since the evaporation of many $\mathrm{LiN_3}$ samples in the electron gun-detector system would coat the detector with a large layer of lithium, the $\mathrm{LiN_3}$ samples were evaporated in an auxiliary system. Any quarks present in the $\mathrm{LiN_3}$ sample would be transferred to a foil. An electric field between the $\mathrm{LiN_3}$ sample and the foil guided most of the evaporated charged particles of one sign of charge to the foil. This foil was then transferred into the electron gun. Pulse-height spectra were taken as the temperature of the foil was increased and then scanned for a peak which might be due to quarks.

B. Sample Preparation

Application of the cation exchange method[16] requires that the quarks be hydrated in an electrolytic solution. This requirement is met by seawater samples, but not by solid samples. The fate of positively charged quarks, generated by cosmic-ray collisions and stopped in the oceans, is an open question. It has been suggested[17] that q_1 would probably pick up an $\mathrm{OH^-}$ molecule and form $(q_1\mathrm{OH})^{-1/3}(aq)$. We have considered this possibility and, also, that a more favored state might be $(q_1+e)^{-1/3}(aq)$ or $q_1^{+2/3}(aq)$. However, owing to the uncertainty in our estimate of the hydration energy of $q_1^{+2/3}(aq)$, we are unable to say which possibility is the most likely. The negative quarks are likely to be captured by an oxygen nucleus, producing $\mathrm{H_2O}^{-n/3}(aq)$. Therefore, both positively and negatively charged quarks are likely to become hydrated. The fate of quarks in surface material such as rock is even more difficult to estimate. From a comparison of ionization potential, electron affinity and electronegativity of the elements with those estimated for q_1, it can be argued that q_1 behaves similarly to $\mathrm{Li^+}$. However, for purposes of applying the cation exchange method, a weaker assumption is sufficient. Since the alkalis and most of the other metals can be leached out of the surface material, it is only necessary to assume that the same will be true for quark species. Therefore, the solid samples were treated by a standard geochemical method for the extraction of alkalis from rocks.

By virtue of their charge, the hydrated quark species of charge $\pm\frac{1}{3}$ or $\pm\frac{2}{3}$ will come before sodium in the elution of the cation exchange column and possibly before lithium.[18] Since the natural lithium content of the bulk samples selected was small, everything before the appearance of sodium in the elution was collected, including lithium. If this method was to be successful in separating quark species from bulk samples, the sodium and lithium bands had to be well resolved. By choosing the right resin, size of columns, flow rate, and normality of solution this condition can be met.[19] The resin that

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[15] J. Lindhard, M. Scharff, and H. Schiott, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **33**, No. 14 (1963).

[16] General discussions of the theory and operation of cation exchange columns can be found in *Handbook of Analytical Chemistry*, edited by L. Meites (McGraw-Hill Book Co., New York, 1963); *Trace Analysis*, edited by J. Yoe and H. Koch, Jr. (John Wiley & Sons, Inc., New York, 1955); O. Samuelson, *Ion Exchange in Analytical Chemistry* (John Wiley & Sons, Inc., New York, 1963).

[17] M. Anbar (private communication).

[18] Since quark species always have a net charge there is a hydration energy which favors their staying in solution. However, if the quarks are in the form of $q_1\mathrm{OH}^{-1/3}$ or $\mathrm{H_2O}^{-n/3}$, then this waterlike molecule might substitute for water of hydration in insoluble compounds, such as $\mathrm{CaSO_4\cdot H_2O}\cdot q_1\mathrm{OH}^{-1/3}$. We have assumed that the quarks were not removed by this process.

[19] L. Reichen, Anal. Chem. **30**, 1948 (1958); R. Sweet, W. Rieman III, and J. Beukenkamp, *ibid.* **24**, 952 (1952); J. Riley and M. Tongudai, Deep-Sea Res. **2**, 563 (1964).

was chosen is a polystyrenesulfonic acid (Dowex 50W-X8, 50–100 mesh size, H^+ form, Dow Chemical Co.). Two sizes of columns were used. The large-sized one had a diameter of 10 cm and was filled with 6 kg of resin. The small-sized one had a diameter of 2.5 cm and was filled with 450 g of resin. The elution for both columns was started with $0.1N$ HCl and was continued with $0.2N$ HCl until the appearance of sodium in the elution. The output collected before the appearance of sodium from a large column was 60–70 liters, and for a small column, about 3 liters. Tests run indicated that at least 80% of the lithium and sodium input can be collected; a corresponding efficiency for collecting quarks was assumed. Collection of the eluent stopped when sodium was first detected. The eluent was evaporated to dryness, the residue was dissolved in water and run through the column again. The residue was then fumed to dryness with $HNO_3(c)$, $HClO_4(c)$, and $H_2SO_4(s)$ to drive off any organic material, such as contamination from the resin. Deionized water was added, and any insoluble part was filtered off and discarded. The sample was then fumed to dryness with $H_2SO_4(c)$ to convert it into sulfate form, mainly $Li_2SO_4(s)$. The final residue in most cases weighed less than 5 mg. It was dissolved in water, and a dilute solution of $Ba(N_3)_2$ was added. The main reaction is

$$Li_2SO_4(aq)+Ba(N_3)_2(aq)\rightarrow 2\,LiN_3(aq)+BaSO_4(s)\,.$$

The $BaSO_4(s)$ was eliminated by repeated vacuum filtering. The solution was slowly evaporated, yielding mainly $LiN_3(s)$ as the host material for the quarks.

The $LiN_3(s)$ sample was pressed into a thin 3-mm-diam pellet and placed in a Mo crucible in an evacuated evaporation chamber. The source tube of the electron gun was attached to a transfer rod and positioned in the evaporation chamber such that the Ta foil was 2 cm above the crucible. The foil and crucible were part of a capacitor plate arrangement consisting of two horizontal plates; a potential of 300 V was applied between the plates. At least 90% of the properly charged quark species evaporated from the heated crucible would be accelerated to the foil. The solid angle was such that about 3% of the lithium was deposited on the foil. Therefore, the evaporated quarks are likely to be embedded in a thin lithium film on the Ta foil. The evaporation was done so that the pressure never exceeded 5×10^{-5} mm Hg.[20] $LiN_3(s)$ decomposes[21] between 115 and 298°C, leaving lithium in the crucible.[22] The temperature was further increased until the lithium started to evaporate. After the lithium was evaporated, the temperature was raised to about 700°C for 2 min. The temperature was then decreased to room temperature. If q_1 and (q_1q_1) were evaporated, they would be evaporated with an electron attached, since their ionization potential is greater than the work function of lithium. Other combinations are less likely to occur and the polarities were set so that negative particles would be accelerated.

The Ta foil was then transferred into the electron gun. However, the transfer could not be made in air, since the lithium film would quickly convert into LiOH. Quarks evaporated from LiOH are likely to be evaporated as hydroxides and would not produce a signal above the electronic noise. The transfer, therefore, was accomplished by using an evacuated transfer chamber. To check that in the evaporation and transfer procedure the lithium did not appreciably react before it was heated in the electron gun, test samples of Li and LiN_3 were evaporated from the Mo crucible. A quartz crystal oscillator was used to monitor the reaction rate of the evaporated lithium film as a function of pressure and time. The results of these measurements implied that less than 10% of the lithium film had converted into LiOH by the time it was heated in the electron gun.

C. Detection

The electron gun (Fig. 1) was of the Pierce cathode type[23] and consisted of a particle source, a Wehnelt cylinder, and an anode. The Wehnelt cylinder was a highly polished stainless steel hemisphere[24] with a 13-mm-diam hole, into which the source was placed. The source was a 1-mil-thick Ta foil of diameter 10 mm, which fitted into a stainless steel holder. A tungsten spiral heater was about 2 mm below the foil. The source was at a potential V_s and the Wehnelt cylinder at a potential V_W relative to the grounded anode. Since the initial kinetic energy of an evaporated particle is much less than the potential difference near the foil, the path of the particle is independent of the particle's charge and mass.[25] Therefore, the focusing of fractionally charged particles should be approximately the same as for electrons.

The focusing properties of the gun were checked by using thermionic electrons as a source and a fluorescent screen in place of the solid-state detector. The Ta foil O was replaced by a foil with pin holes; electrons from the spiral heater N passed through these holes and the pattern could be observed on the screen as a function of $|V_W-V_s|$ and the vertical position of the foil. The efficiency of the gun was checked by replacing the fluorescent screen by the detector, which had a sensitive area of 25 mm². It was found that 60% of the emitted electrons were observed in the full-energy peak (Fig. 2).

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[20] Most azides are explosive if heated too rapidly. Bondar *et al.* (Ref. 22) have reported successfully obtaining alkali films from alkali azides, with the exception of $LiN_3(s)$, which they judged too explosive to work with. We had a few minor explosions, but if the heating was done very slowly, then this problem did not arise.

[21] *Handbook of Chemistry and Physics* (The Chemical Rubber Co., Cleveland, Ohio, 1964), p. B-187.

[22] A. Bondar, V. Karev, and A. Klyucharev, Instr. Exp. Tech. (USSR) 4, 761 (1961).

[23] J. Pierce, J. Appl. Phys. **11**, 548 (1940); G. Brewer, *ibid.* **28**, 7 (1957).

[24] M. Bricka and H. Bruck, Ann. Radioel. **3**, 339 (1948).

[25] P. Grivet, *Electron Optics* (Pergamon Press, Inc., New York, 1965), pp. 55–57, 61–62.

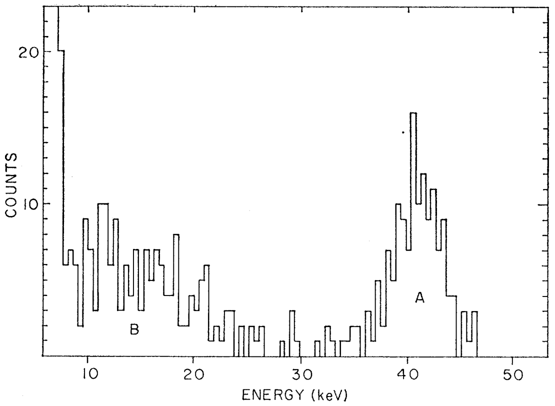

FIG. 3. Typical 30-sec pulse-height spectrum for a sample temperature of 530°C with electromagnet on. The peak A and the hump B are produced by negative ions.

Since the focusing is approximately independent of the particle's charge and mass, the efficiency for collecting fractionally charged particles was assumed to be the same.

A charged particle of mass m and nuclear charge Z, which is accelerated to the detector, will lose its kinetic energy mainly through ionization and elastic nuclear scattering in the detector. If its kinetic energy is less than $\frac{1}{2}m(vZ^{2/3})^2$, where $v=e^2/\hbar$, a non-negligible amount is lost in elastic nuclear scattering.[15] This critical kinetic energy is 26 keV for H$^+$, 810 keV for Li$^+$, and 148 keV for $(q_1+e)^{-1/3}$, assuming $m_q = 10$ GeV/c^2 and $Z=+\frac{2}{3}$. In this experiment the kinetic energy of a quark species with charge $\frac{1}{3}$ would be about 17 keV, which is much less than its critical kinetic energy. Therefore, estimates of the amount of ionization produced by such species were made by applying the theory of specific energy loss of low-energy particles[15,26] to determine if the pulse E_e generated by the ionization process, is larger than the electronic noise, 8 keV. The result is that if $m_q \lesssim 10$ GeV/c^2, $E_e \gtrsim 8$ keV for $(q_1+e)^{-1/3}$, and therefore $(q_1+e)^{-1/3}$, could be resolved from the electronic noise.

Because experimental data on the stopping of low-energy particles in solid-state detectors are scarce, we measured E_e as a function of T for Li$^+$ and H$^+$ beams. Li$^+$ ions were generated in the source region of the electron gun by heating $Li_2O \cdot Al_2O_3 \cdot 2SiO_2$ and were accelerated to kinetic energies of 20–50 keV. The Li$^+$ peak in the pulse-height spectrum was shifted on the order of 20–30%, compared to the theoretical estimate of 45%. H$^+$ ions were generated by ionizing the residual gas in the source region of the electron gun. The H$^+$ peak was shifted about 4% for kinetic energies of 20–50 keV, as compared with the theoretical estimate of about 10%. On the basis of these experiments, we conclude that the species $(q_1+e)^{-1/3}$ would produce a peak that is resolvable from the electronic noise if the quark mass m_q were less than about 10 GeV/c^2. Similarly, $(q_1+\mathrm{Li})^{-1/3}$ would be observable for a quark mass of less than 5 GeV/c^2.

In the actual search for quarks, foils prepared as described in Sec. II B were placed in the electron gun. Spectra were taken for 30-sec intervals as the temperature of the foil was increased. After each interval, the pulse-height spectrum was automatically transferred onto magnetic tape, the memory of the analyzer was erased, and the analyzer was restarted. The lost time between spectra was 1 sec. After the foil had been heated to over 600°C, the 20–30 spectra obtained were transferred from magnetic tape to punched paper tape and then to IBM cards.

III. RESULTS

Figure 2 shows a typical 30-sec spectrum for a blank foil at 600°C. Most of the counts between the electronic noise and the electron peak were due to stray electrons, which scattered into the detector with less than the full energy. A small electromagnet was stationed between the detector and anode (see Fig. 1). For the actual runs the electromagnet was set to produce about 250 G between the poles, which was sufficient to sweep the electrons out of the beam and to reduce the above background substantially. The (q_1+e) in the beam for which $m_q \gtrsim 1$ GeV/c^2 would not be appreciably deflected, and most would hit the sensitive area of the detector. Figure 3 is a typical spectrum for a "quark foil" at 530°C with electromagnet on. The background still present was probably due to negative-ion emission from the source region. Separate experiments with a large electromagnet indicated that the peak A at 40 keV was produced by particles with a charge-to-mass ratio between e/m_p and $e/3m_p$, where m_p is the proton mass. Likely candidates for this peak are H$^-$ and H$_2^-$. H$^-$ can be ruled out by considering the energy: The peak is shifted by 20% from the full-energy value, but we have indicated in Sec. II B that the shift for H$^+$ ions is only 4%. Deflection measurements on the hump B at about 15 keV gave a charge-to-mass ratio between $e/10m_p$ and $e/20m_p$. This hump could have been caused by O$^-$ and OH$^-$. The ratio of the height of hump B to that of peak A was approximately constant during a run, but varied from sample to sample. The ratio on the basis of 12 quark-sample runs was 0.8±0.4. Seven blank foils were heated in the same manner as the quark foils. They yielded similar spectra, but the ratio B/A for these runs was 2.2±0.9. Three runs were made in which pure LiN$_3$ was used instead of the LiN$_3$ which was prepared from the eluent believed to contain quarks. These samples were run in the same manner as the quark samples except the polarity in the evaporation chamber was such that no negative quark species could be evaporated to the foil. The spectra for these runs resembled those of the quark runs. The ratio of the peak height of B to A was 0.8±0.2, which is in agreement with that of the quark samples.

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[26] E. Haines and A. Whitehead, Rev. Sci. Instr. **37**, 190 (1966).

All spectra were scanned for peaks that could not be explained by the above background fluctuations and that might indicate particles of charge between -1 and 0. No evidence for the existence of such fractionally charged particles was found.

The concentration limits set by this experiment depend upon what is assumed for the evaporation rate of quarks. The evaporation rate of lithium is such[27] that the lithium film should be evaporated within the first 4 min or 8 runs. However, since the evaporation rates of various quark species are unknown, two concentration limits are calculated from the data, which are representative of two extremes: a low-temperature limit L assuming all quarks were evaporated within the first 3 min or 6 runs (room temperature to 300°C), and a high-temperature limit H assuming all quarks were evaporated within the first 9 min or 18 runs (room temperature to 530°C). For temperatures much higher than 530°C, the electron and ion background was very large, the light output of the foil increased the noise level of the detector, and no meaningful limit could be given.

If quark species of charge $-\frac{1}{3}$ were evaporated, they would introduce counts in the region below about 17 keV of the spectrum. Therefore, the number of such quarks must be less than the integrated number of counts below 17 keV. Since the shift in the quark peak from 17 keV and the shape of the peak is unknown, all counts between the cutoff of the electronic noise, 8 keV, and 20 keV were summed in calculating concentration limits. If the quark peak were shifted very little, the high-energy side of the peak would extend beyond 17 keV. Therefore, 20 keV was chosen as a reasonable cutoff. The lower limit of 8 keV implies that the calculation is not valid for very massive quark species. If N is the total number of counts between 8 and 20 keV and N_q is the total number of quarks in the bulk sample, then $N_q\epsilon_1\epsilon_2\epsilon_3 \lesssim N$. ϵ_1 is the efficiency for separating quarks from the bulk sample by the cation exchange procedure. Since samples were cycled through the cation exchange columns up to three times and the efficiency for one cycle was greater than 80%, $\epsilon_1 \cong 0.5$ is reasonable. ϵ_2 $(=0.9)$ is the fraction of quarks that are likely not to be in a LiOH environment and ϵ_3 $(=0.6)$ is the efficiency of the electron gun. The concentration limit, therefore, is $(6.2\times10^{-24}N/M)$ quarks/nucleon, where M is the mass of the bulk sample in g. The concentration limits for 12 samples are presented in Table II. These limits depend on the validity of the assumptions made in Sec. II.

TABLE II. Limit on the concentration of quarks in various terrestrial samples. Concentration limit L was calculated assuming all quarks would be evaporated during the first 3 min, and concentration limit H was calculated assuming all quarks would be evaporated during the first 9 min.

Sample	Mass (g)	Concentration limit (quarks/nucleon) L	H
Lava	32	5×10^{-24}	4×10^{-23}
	50	5×10^{-24}	2×10^{-22}
	50	3×10^{-24}	2×10^{-23}
Limestone	30	5×10^{-24}	4×10^{-23}
	65	4×10^{-24}	6×10^{-23}
Grand Canyon rock	45	6×10^{-24}	5×10^{-23}
	100	3×10^{-24}	2×10^{-23}
Australian rock	31	1×10^{-23}	3×10^{-22}
Seawater	3000	8×10^{-26}	1×10^{-24}
	3000	7×10^{-26}	9×10^{-25}
Lava[a]	300	6×10^{-25}	3×10^{-23}
	500	3×10^{-25}	1×10^{-23}

[a] Assuming the hydrated quark species to be negative.

Our concentration limits are lower than those of other geophysical searches, except for those of Chupka *et al.*[9] However, their and our lowest limits were obtained with seawater and air samples and it has been pointed out that the accumulation time for quarks in seawater and air could be reduced to the order of years or less by the electric field of the earth.[28] We therefore consider our concentration limit $\lesssim 10^{-23}$ quark/nucleon, obtained with rock samples, as the most meaningful result.

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[28] M. R. C. McDowell and J. B. Hasted, Nature **214**, 235 (1967); W. I. Axford, J. Geophys. Res. **73**, 6855 (1968).