EXPERIMENTAL SEARCH FOR STABLE, FRACTIONALLY CHARGED PARTICLES*

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Various samples of matter were examined to search for stable quarks, particles of charge $\frac{1}{3}e$ or $\frac{2}{3}e$. The three materials examined were iron meteorites, air, and sea water; the concentrations of quarks were less than 10^{-17}, 5×10^{-27}, and 3×10^{-29} per nucleon.

Following a suggestion by Gell-Mann[1] that particles with fractional charges (quarks) may be the basic constituents of nucleons and that some form of quarks would be stable, we have tried a series of experiments designed to observe such particles in nature. Most experiments reported so far in the literature have attempted to recognize such particles immediately after their production, by the anomalously small ionization they would cause in the relativistic limit. Experiments using accelerators[2] and cosmic rays[3] can be summarized as setting a probable limit $M_q \gtrsim \mathrm{BeV}/c^2$ on the quark mass. The present experiment is an effort to exploit the stability of quarks, and the property of fractional charges that they cannot be neutralized in ordinary substances. In particular, negative quarks of charge $-\frac{1}{3}e$ would be captured in ordinary atoms in a Bohr orbit, which for such a heavy particle would be inside the nucleus. Such atoms then would be fractionally charged and remain so indefinitely. The possibility of the negative quark annihilating or recombining into a nucleon will be vanishingly small. It is possible that the nucleus-quark system will undergo beta decay; but the end product must still have fractional charge. Such an atom will have chemical properties somewhat different from the ordinary element with the same atomic number, but with the number of electrons being the same, the properties will tend to be qualitatively similar.

It is somewhat more difficult to speculate about positive quarks. Upon capturing an electron, they would behave somewhat as oversize, very electropositive and reactive hydrogen atoms—chemically more like the alkali metals. A quark with charge $+\frac{1}{3}e$ would have an ionization potential of 1.51 eV, and would exist as a hydrated ion in water solution, evaporating predominantly as a positive ion (possibly associated with a neutral molecule), and would best be sought as such. A quark of charge $+\frac{2}{3}e$ would have an ionization potential of 6.04 eV, would exist as a hydrated ion in water solution, and under most conditions would evaporate predominantly in a tight association with an electron or a negative ion; the $+\frac{2}{3}$ quark thus is probably best sought as a negatively charged species. Our experiments have been concentrated on these.

It is amusing to note that Millikan, in his first published report on measurements of the electron charge on water droplets in a cloud chamber, remarks: "In the third place I have discarded one uncertain and unduplicated observation apparently upon a single charged drop, which gave a value of the charge on the drop some 30% lower than the final value of e."[4] It may even be argued that later measurements of the electron charge with oil drops were less likely to turn up quarks because oil-bearing strata are at such depths as to be shielded from any quarks produced by cosmic rays, and that the chemical properties of a fractionally charged atom would be sufficiently different to result in elimination of the atom from the refined oil used in the Millikan experiments. The results of Ehrenhaft[5] on subelectronic charges are in no way consistent with charges of $\frac{1}{3}e$ or $\frac{2}{3}e$. The measurement of Hillas and Cranshaw,[6] in which limits were set on the possible deviation from equality of the electron and proton charges, may at first sight be thought to set limits on quark concentration as well. However, the purification of the gases they used may have eliminated fractional charges; and more importantly, their apparatus included an ion trap designed to retard any atoms with large charges. Thus any atoms with fractional charge would not have contributed to the measurement. Recent proposed modified versions[7] of the Millikan experiment, which in effect increase the size of the droplets, may be able to detect quarks in graphite particles if the concentration is $\geqslant 10^{-19}$. It seems unlikely that cosmic-ray-induced quarks would exist in graphite in anywhere near this large a concentration.

Each of our experiments makes use of the unique feature that atoms with quarks could never become electrically neutral. The sample being studied is allowed to pass in gaseous form through an electric field sufficiently strong to extract fractionally charged atoms or molecules. With this simple method large quantities of material can be studied and, in principle, molar concentrations as small as 10^{-30} could be detected. In most of the experiments, any atoms collected by the electric field were further concentrated onto a small platinum filament which was placed at a positive potential to retain negatively charged species and then heated for ~10 sec in air to ~600°C in order to drive off impurities. A larger heated, negatively charged filament was positioned so as to ensure that quark-containing atoms which volatilize in the much less probable form of positive ions would be collected, re-evaporated, and returned to the small platinum filament. This small filament was then put into a device which accelerated any negative ions through 15 kV and onto the first dynode of an electron multiplier. Pulses from this multiplier were recorded on a multiscaler as a function of time. A 100-in. mass spectrometer[8] was used in some of the measurements in an attempt to identify masses. The materials tested were iron meteorites, atmospheric air, and sea water. Since we are quite uncertain regarding the chemical properties of the hypothetical atoms for which we conducted this search, it is important to examine samples in their natural state.

In the case of meteorites,[9] we depended on the fact that the ages of many meteorites are determined by measuring activities induced by cosmic radiation. Thus we need only to assume that the cosmic-ray spectrum has remained unchanged in the past approximately 500 million years. We have used samples of the Arispe, Henbury, Carbo, Grant, Toluca, and Canyon Diablo meteorites. In each case we vaporized samples of the meteorites in the ion source of the 100-in. mass spectrometer where an electric field extracted any negatively charged particles. Positively charged particles were also sought in separate experiments. Most of the masses observed were very low in intensity and could be identified with known atoms or molecules. Although the measurements on meteorites could have been improved considerably, this was not done. The reason was that the small physical size of most meteorites is insufficient to stop a quark with $M_q \gtrsim 5$ BeV/c^2 produced in the meteorite. On the assumption that the mean free path for collisions with nucleons is about 100 g/cm^2, one can estimate the amount of material needed to stop a quark with a given mass M_q. The energy loss per collision is quite insensitive to the inelasticity of the collision if $M_q \gg 5$ BeV/c^2, the reason being that the center-of-mass energy is a small fraction of the total energy of the quark. The Canyon Diablo meteorite would have been of sufficient size before impact, but nothing is known about which part of the original meteorite a particular fragment came from. If one uses 10^6 tons for the initial mass, then for $M_q \approx 10$ BeV/c^2 the quarks would have been stopped in a surface layer which constitutes only about 15% of the total mass of the meteorite, and a random fragment is unlikely to contain any quarks.

We next made measurements on samples of the atmosphere. Here the amount of material available for stopping an energetic quark can be quite large if one considers paths almost tangential to the earth's surface. The difficulty is that chemical processes may preferentially remove fractionally charged oxygen or nitrogen atoms from the atmosphere. One can assume, however, that argon atoms with $-\frac{1}{3}e$ quarks attached will retain the chemical properties of a noble gas. One can readily calculate by interpolation of the isoelectronic series K^+, Ar, Cl^- that the ionization potential of an argon atom with a $-\frac{1}{3}e$ quark attached would be about 11.3 eV, intermediate between the values for xenon and radon, and hence one expects similar chemical reactivity. The closed electronic shell would remain closed and chemical compounds probably would not be formed. The only significant difference between the properties of such an argon atom and ordinary argon would be that the fractional charge would increase the solubility in water and the physical adsorption on surfaces. One can then estimate that atmospheric turbulence would cause a fractionally charged argon atom in the atmosphere to have a mean life of the order of days.

We have sampled large volumes of atmospheric air by the use of an electric fence, consisting of 0.6-cm-diam aluminum tubing, at 20 000 V with respect to another fence approximately 25 cm away, so that the minimum field between fences was ~350 V/cm. From the wind velocity one can calculate that the amount of air sampled in a 24-h period is between 10^{10} and 10^{11} liters. Any charges accumulated by this fence were first collected at room temperature and next by baking the aluminum tubing at ~200°C. Any material driven off was flushed by an inert gas flowing through an electric field. It can easily be calculated that argon atoms with $-\frac{1}{3}e$ quarks attached will readily be desorbed from even ionic surfaces under these conditions. In order to eliminate the possibility that fractionally charged atoms in the atmosphere might adhere to dust particles, we also studied quantities of atmospheric dust taken from the air-intake filters of the Argonne physics building. The dust was baked at temperatures up to 400°C and flushed with an inert gas; then the material with the inert gas was passed through an electric field and any charged particles were collected.

At room temperature no particles (other than a very low background) were observed in these experiments. On heating to 350°C, between 10^3 and 10^5 particles were observed in each run for both the air and dust experiments. The particles observed on heating the platinum filament sometimes showed a rather peculiar behavior which is illustrated in Fig. 1. If particles adsorbed in low concentration on the surface of a filament are driven off by heating, their intensity will ideally decrease exponentially because of the decreasing concentration on the filament's surface. In the general case, for several types of particles evaporating from a heterogeneous surface, the decay will be a sum of such exponentials. The decay time, of course, is governed by the desorption time of the particular material at that temperature. The detecting system used in this exper-

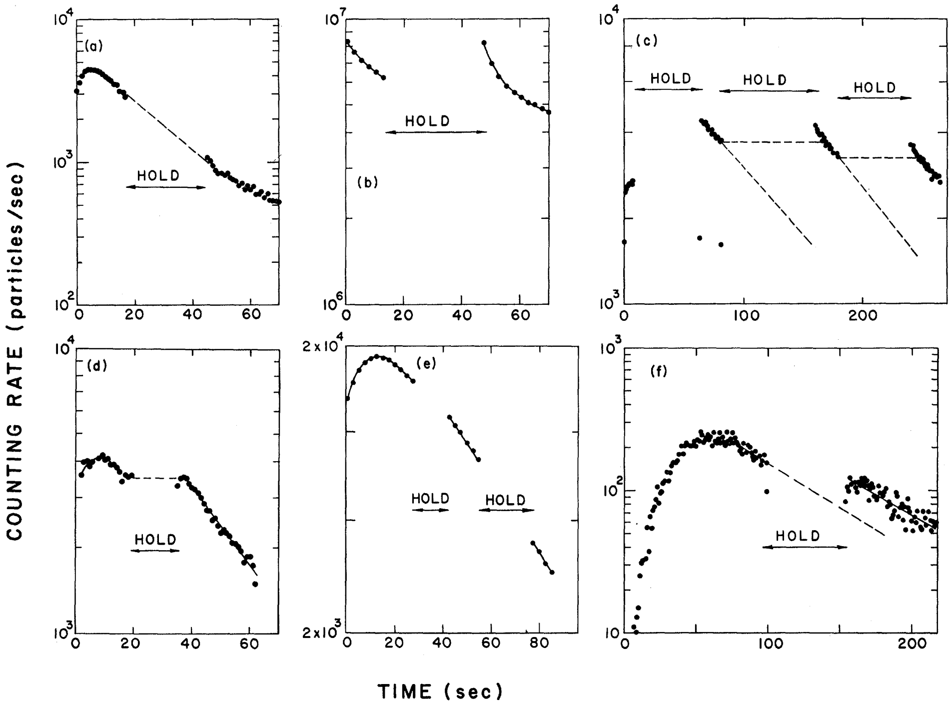

FIG. 1. The evaporation rate of various samples. In each curve, the evaporation is interrupted by introducing a "holding potential" such that the charged species being accelerated could not leave the filament. (a) Evaporation of a normal sample with acceleration of negative ions, in which case the rate is determined by the evaporation of electrically neutral species. (b) The evaporation of Cs^+ ions, with a negligible neutral component, showing the holdup in exponential decay. (c), (d), and (f) show similar behaviors for negative ions from dust, air, and sea water, respectively. The holdup here is unexplained and not expected for normal negative species but would be expected from fractionally charged atoms, or a hitherto unobserved negative species with unique properties. The curve in (e) shows the evaporation of $(FeCl_4)^-$ ions, identified as a large component in collections from air and dust samples. The initial rise in each curve corresponds to the time required for the Pt filament to reach thermal equilibrium.

iment depends on acceleration by an electric field. If this field is reversed the evaporation should proceed undisturbed because, for all known materials that form negative ions, the evaporation is primarily in the form of neutral atoms. Such behavior is shown in Fig. 1(a). This is not the case for all positive ions. For cesium, for instance, atoms come off predominantly in a positively charged state. If the decay is interrupted by reversing the electric field momentarily, the evaporation of positively charged particles is inhibited. When the field is again such as to accelerate the Cs^+ ions, the rate resumes at the level at which it stopped; or a buildup of concentration on the surface may result in an even higher rate. This is shown in Fig. 1(b). Such behavior would not be expected for any known negative ions because negative ions are much more difficult to form, particularly from the surface of a material (such as platinum) with a high work function. On resuming acceleration after a reversal of the electric field, one expects the particles to be desorbing at the rate obtained by extrapolating from the exponential decay occuring previously, because most of the particles will be evaporating as neutral atoms, as in Fig. 1(a). Since any atom with a negative quark attached is permanently charged, one would expect it to show the same behavior as that seen for cesium. In fact, in some of the measurements from the atmosphere, we observed such behavior [Fig. 1(c) and 1 (d)]. However, this behavior occurred only on a few occasions and was not clearly reproducible. In the course of attempts at mass analyses, we have observed an appreciable intensity of ions of the form $(FeCl_4)^-$ and other iron tetrahalides, although such negative ions had not previously been observed in the gaseous state. The evaporation rate of these ions with reversed electric field [Fig. 1(e)] does not exhibit the holding properties of Cs^+ ions. Since relatively little is known about negative ions in general, we cannot reach any firm conclusions on this matter. The particles could have been quarks or an unknown species of negative ion. Because of the lack of reproducibility in our atmospheric and dust measurements, presumably because of uncontrollable meteorological factors, we abandoned these fields of experimentation.

The next medium we studied in some detail was sea water. This has the advantage that it provides a very steady, reproducible source of material; one sample of sea water is very much like any other sample. Also, one can estimate that whatever the chemical property of atoms with quarks attached, they are likely to be present in solution in sea water. The increased solubility of fractionally charged argon atoms has already been noted; one can make similar guesses regarding the solubility of water molecules in which an oxygen atom has a quark attached. Hydrogen atoms with $-\frac{1}{3}e$ quarks attached would be very similar to the $+\frac{2}{3}e$ quarks which, as mentioned above, are expected to remain in sea water and to be best observed as a negative species. It may be, however, that particles with charge $+\frac{2}{3}e$ can be released only by heating the sample to a temperature high enough to volatilize NaCl (~800°C).

In the sea-water measurements, 20-liter samples were boiled off through an electric field, and charges were collected much as in the atmospheric experiment. The vapor was diluted with argon gas in order to produce a condition of undersaturation and eliminate the possibility of formation of large clusters. The electric fields employed were sufficiently strong ($>10^3$ V/cm) to collect easily ions with a single shell of water molecules attached. The residual salts from the sea water were heated to 400°C and flushed with argon gas. The number of particles collected was of the order of 10^3 to 10^4 and showed the properties illustrated in Fig. 1(f). The peculiar properties seen in some atmospheric samples are apparent, though less pronounced. We then proceeded to take samples of sea water and mass-analyze them. We have not yet been successful in identifying the mass responsible for the peculiar behavior seen; it will take considerable time and effort to reach conclusive results.

Our results on all measurements are summarized in Table I. The molar concentrations given correspond to the numbers of particles that were observed but not identified with impurities of known masses. The associated quark masses are the ones one would calculate with the stated chemical assumptions, a production cross section[10] assumed to be given by $\sigma_q = \pi(\hbar/M_q c)^2$ for all energies above twice the threshold energy, and the known flux[11] of cosmic rays. It was assumed that the fraction of primaries producing quarks was σ_q/σ_T, where σ_T is the total nucleon-nucleon cross section (which was taken as 30 mb).

Some experiments were also performed with a Millikan oil-drop apparatus with small polyethylene spheres 5 μ in diameter and uniform in size to within 2 %. The particles were allowed to pass through the region in which atoms collected from the atmosphere or sea water were retained by an electric field. It was hoped that if some of these atoms did have fractional charge, then occasionally they would attach themselves to the spheres and be observed as such in the measurement. Approximately 1000 spheres were measured both for air samples and for sea-water samples. All of these exhibited integral charges.

The conclusions to be drawn from all these measurements are rather unsatisfactory. One cannot with any degree of certainty set limits on the existence of fractionally charged atoms. One can only state that under circumstances in which reasonable assumptions might lead one to expect to see quarks, we have not seen fractionally charged atoms. We still need to identify the source of peculiar behavior in the few charged atoms that were observed. This is likely to take a great deal of effort and time. In the meantime, the upper limits given in Table I are the best we can set on the possible existence of such particles. The values for sea water are the ones setting the tightest limits for the quark mass, if one accepts our assumptions as reasonable. The corresponding quark-mass limits from the two estimates obtained in Ref. 10 are 50 and 7 BeV/c^2. It should be remarked that the limits that Ref. 2 sets from direct cosmic-ray measurements correspond to the larger of the two cross-section estimates.

The possibility that negative quarks may decay to $+\frac{2}{3}e$ in less than the time required for capture into a Bohr orbit was not discussed; this would merely mean that positive quarks should be searched for more intensively, though our results are almost as valid for $+\frac{2}{3}e$ quarks as they are for the $-\frac{1}{3}e$; they should both appear as negative species. We have assumed that any quarks would have been produced by cosmic rays during the earth's history. There is no obvious cosmological reason for an extremely low abundance; a recent estimate[12] gives 10^{-9}-10^{-18} per nucleon as a reasonable range after the usual cosmological evolution.

For further measurements it is easy to suggest many natural circumstances (chemical, geological, or biological) under which fractional charges may tend to concentrate. There is

Table I. Summary of limiting concentrations.

Material	Number of molecules	Limit on number of fractionally charged particles	Concentration (per nucleon)	Particles (per liter)	M_q (BeV/c^2)
Meteorites	10^{19}	10^4	$\sim 10^{-17}$		5[a]
Air, 25°C	3×10^{33}	10^2	10^{-33}	3×10^{-9}	
Air, 200°C	3×10^{33}	10^5	10^{-30}	3×10^{-6}	10[b]
Dust, 200°C	10^{32} [c]	10^6	3×10^{-27}	3×10^{-3} per liter of air	
Sea water, 25°C	7×10^{26}	10^2	5×10^{-27}	5	
Sea water, 200°C	7×10^{26}	5×10^4	3×10^{-24}	2500	50[d] (7)[e]

[a] Any larger masses would probably not be stopped in the meteorites.

[b] This is the mass corresponding to the concentration given. This calculation made use of the estimated cross section mentioned in the text and assumed an equilibrium state in which fractionally charged particles accumulated in the air for 10 days. Only capture in Ar was taken into account.

[c] This is the number of air molecules in the volume of air from which the dust was extracted.

[d] This is the mass corresponding to the concentration given. This calculation made use of the estimated cross section given in the text and assumed 10^9 years accumulation. Only capture in oxygen was taken into account.

[e] This is the value corresponding to the cross-section estimate of G. Domokos and T. Fulton, Phys. Letters 20, 546 (1966).

considerable guess work involved in most of these suggestions and we have not discussed them in detail. For the time being, the ocean seems to be the most promising medium. When parts of the lunar surface become available for close examination, these samples promise the experimental advantages of meteorites (cosmic-ray exposure determinable from induced activities and isotope ratios) but without the limitation in size.

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