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Search for Stable Fractionally Charged Particles*

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A search for stable quarks in terrestrial materials has been carried out using ultraviolet spectroscopy and an oil-drop technique of charge measurement. The spectroscopy involved a search for the Lyman series of emission lines from hydrogenic atoms formed with a $+\frac{2}{3}$ quark as a nucleus. The oil-drop method attempted to detect $\frac{1}{3}$ integral charges on oil drops approximately 20 μ in diam. Electric fields were used to enrich the samples in quarks and thereby improve the sensitivity of the experiments. The concentration of quarks in the materials examined was determined to be less than 10^{-18} quarks/nucleon for the spectroscopic experiments and less than 10^{-20} quarks/nucleon for the oil-drop experiments.

I. INTRODUCTION

In 1964 Gell-Mann[1] and Zweig[2] independently proposed that a fundamental explanation of the success of the "eightfold way" in classifying strongly-interacting particles could be found by introducing a unitary triplet from which all other hadrons are constructed. This triplet corresponds to the lowest nontrivial representation of the $SU(3)$ symmetry of the eightfold way. The three particles, called quarks, have remained purely mathematical entities since no physical evidence of their existence has been found. In this paper we shall summarize attempts[3] to detect these particles by means of optical spectroscopy and oil-drop techniques.

II. EXISTENCE OF QUARKS

Experiments have been performed using accelerators in attempts to produce quarks in nucleon collisions.[4–9] These experiments have set lower limits on the mass of a quark or its production cross section, $M_Q \geq 4$ GeV/c^2 or $\sigma_Q \leq 10^{-2}\,\mu$b, respectively. Experiments have also been performed with cosmic-ray showers in an attempt to detect quarks produced in primary cosmic-ray interactions in the upper atmosphere.[10–17] The results of these experiments indicate that the flux of fractionally charged particles in cosmic rays is $\leq 2\times10^{-10}$ cm^{-2} sr^{-1} sec^{-1}.

In addition to experiments which attempt to detect quarks as they are produced either in accelerators or by cosmic rays, experiments have been performed to detect them as new chemical elements existing in nature. This paper describes such a search. Other searches for stable quarks have involved mass-spectroscopic methods,[18] magnetic levitation of graphite particles,[19] and examination of the solar spectrum.[20–22] The mass-spectroscopic measurements indicated that quark concentrations are $<10^{-17}$, 5×10^{-27}, 3×10^{-29} per nucleon in iron meteorites, air, and sea water, respectively, under the assumption that quark-bearing molecules vaporize at a temperature less than a few hundred degrees centigrade. The magnetic levitation sets concentration limits $\leq 10^{-16}$ per nucleon in graphite. The data from the solar spectrum analysis are somewhat ambiguous. In any event, it seems that limits $\leq 10^{-10}$ per nucleon can be set from solar spectra.

One cannot make a definite statement about the stability of quarks under strong decay, except that the lightest must be stable for decay into any of the others. Indeed, it is possible[23] that the stable fractionally charged particle would have charge $|Q| = 4/3, 5/3, 7/3, 8/3$, etc. A quark-antiquark annihilation is virtually the only method for their disappearance. In the presence of atoms, the quark and antiquark would be separated, so that annihilation would be impossible. The positive

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quark would pick up an electron and the negative quark would go into a very low-lying orbit around an atomic nucleus. They would then be shielded from each other by the intervening electrons.

In the present experiment we have searched for naturally occurring stable $+\frac{2}{3}e$ quarks spectroscopically and $\pm\frac{2}{3}e$, $\pm\frac{1}{3}e$ quarks with a modified oil-drop technique. The oil-drop technique will be discussed in Sec. V. The present discussion will center on the spectroscopic search for $+\frac{2}{3}$ quarks.

In the presence of electrons one would expect a $+\frac{2}{3}e$ or a $+\frac{1}{3}e$ quark to form a stable hydrogenic atom with the quark as a nucleus. These atoms would have a net $-\frac{1}{3}e$ or $-\frac{2}{3}e$ charge and their spectra would be similar to that of hydrogen with a factor of either 4/9 or $\frac{1}{9}$ in the rydberg. The atom formed by a $+\frac{2}{3}$ quark would have its Lyman series at 2733, 2306, 2186 Å, etc., with an uncertainty of ~ 1 Å due to the unknown reduced-mass correction for the quark. The atom formed by a $+\frac{1}{3}$ quark and an electron would have its Lyman series beginning with L_α at 1.09 μ. The L_α line of the $+\frac{2}{3}$ quark atom lies in a particularly sensitive region of the optical spectrum where detector efficiencies are high, about 25%, and light losses by transmission and reflection are small. Indeed the 2733 Å line from "quarkium" should be one of the most persistent lines in nature if quarks exist. Searching for the $+\frac{1}{3}$ quark's L_α line at 1.09 μ is not nearly so favorable for optical spectroscopy and this species is probably best sought by other detection schemes.

The present experiment is predicated on a search for the Lyman-α line at 2733 Å. Any promising line was then further tested by improved concentration techniques and a search for the next line, L_β, of the series at 2306 Å whose position could be accurately predicted from the reduced mass of the first line.

Our experiment had its maximum sensitivity for an atom with a $\frac{2}{3}$ charged nucleus, which could result if the $+\frac{2}{3}$ quark were stable or a $-\frac{1}{3}$ quark proton combination were stable. It is interesting to note that an atom with a $+\frac{4}{3}$ charged nucleus would have its second Balmer line at 2733 Å. Therefore, even though the present search was for the L_α line of the $+\frac{2}{3}$ quark at 2733 Å, there was some sensitivity for the $+\frac{4}{3}$ species also.

Quarks could be produced by high-energy primary cosmic-ray particles colliding with terrestrial nucleons by astronomical sources, and indeed they might even exist as free particles in the universe, condensing along with the other elements to form solid bodies. From the cosmic-ray experiments mentioned earlier, a rough estimate on the upper limit of the concentration of quarks in the earth's crust can be established. If the flux of cosmic-ray primaries has remained constant for 5×10^9 years, the flux limit for quarks mentioned above[16] determines an upper limit for their concentration in the earth's crust. For a flux of quarks $\leq 2\times10^{-10}$ cm^{-2} sec^{-1} sr^{-1}, the total number of quarks collected per cm^2 of the earth's atmosphere is $\leq 2\times10^8$. If these quarks are mixed into the earth's crust and oceans to a depth of the order of a few kilometers, then the concentration of quarks will be $\sim 10^3$/cm^3, which corresponds to a concentration of $\leq$one quark per 2×10^{21} nucleons. Of course the intensity of cosmic rays might not have been constant during this time; hence quarks might exist in higher or lower concentrations than the above data suggest.

The possibility of quarks existing as free particles after the primary expansion of the universe has also been investigated.[24] A range of concentration of quarks from 10^{-9} to 10^{-18}/nucleon was estimated depending on the present density and radiation temperature of the universe. It seemed possible that with the aid of natural concentrating mechanisms or laboratory concentration techniques, quarks could be spectroscopically detectable.

The exact chemical properties of an atom with a $+\frac{2}{3}$-charged nucleus are difficult to predict; however, one can make some rough estimates. From Hylleraas's solution to the problem of the energy of normal helium and heliumlike ions,[25] one can calculate the binding energy of an extra electron to a quarkium atom. Hylleraas found

$$I_2=\frac{R_\infty hc}{1+m/M}\left(Z^2-\frac{5}{4}Z+0.315-\frac{0.0175}{Z}+\cdots\right) \quad (1)$$

for the first ionization energy of a two-electron atom. The substitution $Z=\frac{2}{3}$ in Eq. (1) yields $I_2=-0.12$ eV, indicating that the second electron is repelled by the quarkium atom. The single-electron ionization energy for the $+\frac{2}{3}$ quarkium atom will be 4/9 that of hydrogen; hence $I_1=6.1$ eV. Now Mulliken[26] has shown that the average of I_2, the electron affinity, and I_1, the first ionization energy, is a direct measurement of the electronegativity x of an atom. The relation is[27]

$$(I_1+I_2)/5.4=x. \quad (2)$$

Thus Eq. (2) predicts $x=1.1$ for quarkium. Table I shows a comparison between the predicted properties of quarkium and a few other chemical elements.

From the fact that ions such as Li$^+$, Cl$^-$, etc., have a rare-gas electronic configuration and their solubilities are much higher in polar liquids than those of the rare gases, one can infer that the excess charge on the atom is the major factor in determining its solubility. Hence quarks or quark-containing molecules should be extremely soluble in water. It seems likely that even though a quark atom is structurally quite similar to hydrogen, it will behave more like Li or one of the alkalis, since it will be highly electropositive and not

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TABLE I. Chemical and physical properties of the quark atom.

	Ionization[a] (eV)	A_0[b] Å	Ion radius (Å)	Electron affinity (eV)	Electro-negativity
Q	−6.1	0.79	$0.79^{(-)}$	−0.117	1.05
H	−13.6	0.52	$2.08^{(-)}$	0.72	2.1
Li	−5.4	1.52	$0.60^{(+)}$	0	1.0
Na	−5.1	1.86	$0.95^{(+)}$	0	0.9
K	−4.4	2.31	$1.33^{(+)}$	0	0.8
F	−18.6	0.64	$1.36^{(-)}$	4.25	4.0
Cl	−12.9	0.99	$1.81^{(-)}$	4.05	3.0
Br	−11.8	1.14	$1.95^{(-)}$	3.8	2.8

[a] See Ref. 26.
[b] See Ref. 27.

capable of joining in hydrogenlike pair bonding, because the attractive force of its vacant 1*s* orbital is compensated by its net negative charge.

From considerations involving the ionic character of quarkium and its solubility in water, the sea represents the most logical place in which to search for quarks. The sea acts as a chemical melting pot for the elements and indeed makes up a large fraction of the earth's crust. The fact that all known chemical elements are concentrated by natural or biological processes certainly implies that it would be quite surprising if quarks were completely uniformly distributed over the earth's crust. Indeed, many trace elements were first discovered in the sea by relying on biological concentration. Various animals, sediment, etc., derived from the sea, could conceivably be quark enriched. To this end we have examined an arbitrary selection of organisms from the sea in addition to sea water and sea salt for quarks.

III. SPECTROSCOPIC APPARATUS

The basic instrumentation for the spectroscopic search for quarks was a Spex 1400 double monochromator.[3] In many of the experiments, the high rejection ratio of the monochromator for scattered light allowed the detection of spectral lines to be limited solely by the dark current in the photomultiplier detector.

The detector used in conjunction with the Spex 1400 monochromator was an EMI 9558QA photomultiplier tube which was selected for its high-quantum efficiency and low-dark current. With cooling and magnetic focusing, dark currents corresponding to 10 electrons/sec. were obtained at the cathode. This dark current coupled with the quantum efficiency of the tube (30%) corresponded to an equivalent photon background of 30–40 photons/sec at the exit slit of the monochromator.

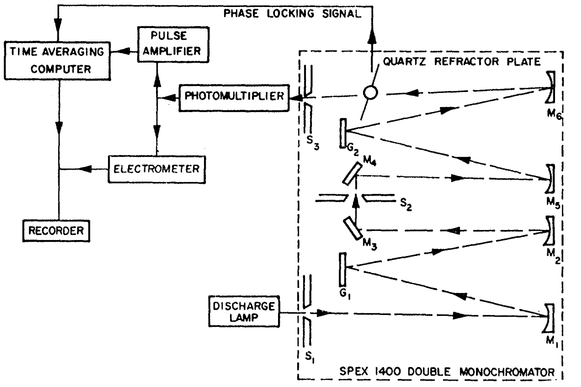

FIG. 1. Block diagram of the spectrometer.

The monochromator could be operated in two quite different modes. It was fitted with a quartz rotating refractor plate which could scan a 2 Å wavelength interval repetitively 20 times/sec. The monochromator could also be scanned in the conventional fashion by rotating the gratings with a motor. In both modes of operation we used slit widths of 10–30 μ which gave a resolution of 0.1–0.3 Å, respectively. The rotating refractor plate was an essential feature of the experiment since it permitted a very rapid scan of the regions where quark lines should exist. Impurities in discharge lamps present in small quantities have a tendency to be driven into the walls or electrodes of the lamp where they are removed from the discharge. The time constant for this process can vary from seconds to minutes. Since most discharges tend to "clean up," we expected the lines from quarks in the lamp to be transitory. For lines that persist for less than a few seconds, the optical scanning of the refractor plate proves to be the only reliable method of detection. For lines which last longer than a few seconds, mechanical scanning of the instrument is more satisfactory. (See Fig. 1.)

Wavelength calibration was obtained from an iron hollow cathode discharge lamp. An additional check of the calibration was provided by a helium hollow cathode discharge. In the spectrum of He^+ there is a series of lines for a quark mass equal to that of an α particle. The existence of these lines can be derived from the Bohr formula for a one-electron atom:

$$\nu = RZ^2(1/n_2^2 - 1/n_1^2). \tag{3}$$

Hence transitions between states in He^+ with principal quantum numbers n which are multiples of three, 3, 6, 9, 12, ⋯ will have identical wavelengths to the Lyman transitions in quarkium. Thus $n=6 \rightarrow n=3$ corresponds to the L_α line of quarkium.

The sensitivity of the instrumentation to quarkium was estimated from measurements on the Li resonance line at 6708 Å. Lithium was selected for these measurements because of its strong resonance line and its expected chemical similarity to quarkium. Solutions of LiCl were prepared in various concentrations as samples and distilled water was used as a control. Sea water was also used as a source of Li in some of the tests. The experiments involved inserting a known amount of Li solution into a discharge, and measuring the signal-to-noise ratio of the Li resonance line. We found that a few times 10^{10} Li atoms were detectable at 6708 Å.[3] 6700 Å does not lie in the region of optimum sensitivity for our instrument. The gratings are blazed at 3000 Å and photomultiplier quantum efficiencies fall off considerably in the red. Therefore, the loss in sensitivity has been measured from 3000 to 6700 Å by means of a comparison of the blackbody spectrum from a pyrometer lamp operating at 2200°K with the output of the spectrometer over the same region 3500 Å → 6700 Å. From these measurements it was found that the sensitivity of the instrument was approximately 30 times better in the near ultraviolet, ~3000 Å, than at 6700 Å in terms of the number of photons required at the entrance slit to produce a given signal at the output of the photomultiplier. From these considerations we concluded that on the order of 10^9 quarkium atoms in the discharge lamp would be detectable by our instrument.

IV. SPECTROSCOPIC PROCEDURE

All spectroscopic experiments relied on a single basic concentration technique. This consisted of vaporizing the sample and collecting any ions released during vaporization by an electric field. After collection the ions were placed in a discharge tube and their spectra were recorded. The vaporization temperatures ranged from the boiling point of water to ~1700°C. The substances examined were sea water, lake water, seaweed, oysters, and plankton. Typical mobilities for ions in air are ~3 cm^2/V sec., and therefore quarks should have mobilities ~ 1 cm^2/V sec. Typical collecting fields used in these experiments were ~500 V/cm.

The processing of sea water was accomplished with two slightly different arrangements. In the first, diagrammed in Fig. 2(a), sea water was distilled through two aluminum electrodes which composed the lower half of a Geissler discharge tube. The lower part of the still was composed of a 2-liter flask which could be filled with water through a side tube. Twenty-liter samples of sea water were evaporated through this device and any ions in the stream were collected on the aluminum electrodes by an electric field of 500 V/cm. One electrode was held at a positive potential with respect to the water; the other electrode was held at a corresponding negative potential. During the distillation procedure, the Geissler tube composing the top half of the still was wrapped in heater tape and held at a temperature above 100°C so that no vapor could condense there. Care was also taken to prevent salt solution from splashing up into the discharge tube. These precautions were taken to keep the discharge tube as clean as possible in order to reduce the background spectrum of the lamp. In general we were successful in that most of our lamps were essentially limited by dark current. After evaporization of the water was complete, the sea salt remaining in the 2-liter flask was thoroughly dried at a temperature of 150°C. The Geissler tube was then separated from the flask with the collecting voltage maintained across the electrodes, and cautiously attached to a vacuum system. After the lamp cooled to room temperature, the voltage across the aluminum electrodes was reduced to ~50 V in preparation for evacuation. This was done to prevent a discharge between the electrodes during evacuation of the lamp. We thought that this voltage was more than sufficient to keep the quarks on the electrodes. After evacuation the lamp was baked out and filled with argon or neon gas. The discharge spectrum of the lamp was then measured in the region of the expected quarkium lines. Since there is no *a priori* way to determine the optimum conditions for excitation of quarkium in the discharge, a number of separate experiments were performed varying the gas pressure from ~0.1 mm to ~5 mm Hg and with both ac and dc excitation. The strength of the discharge was also varied from weak-discharge currents up to currents capable of melting the aluminum electrodes of the lamp.

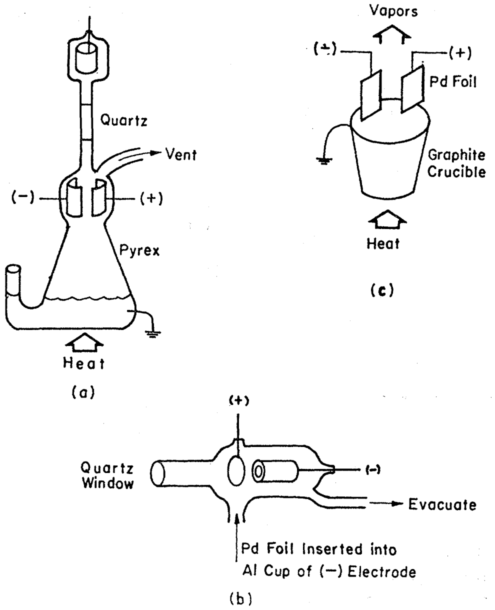

FIG. 2. Distillation apparatus and discharge lamps.

In the second distillation method for sea water, the Geissler tube at the top of the boiling flask was replaced with two palladium or gold-foil collecting electrodes. These foil electrodes were removed from the still after distillation was complete (with the collecting voltage on) and dropped into the cup of a hollow-cathode discharge lamp, shown in Fig. 2(b). The electrodes of the hollow cathode were also held at a 50-V potential difference of the appropriate polarity to accept the foil. Again various gas pressures and excitation methods were tried. The hollow-cathode lamps used in all the experiments had the same basic design, shown in Fig. 2(b). The cathode-cup electrodes had various hole diameters ranging from 3 to 0.5 cm.

The residue remaining in the flask after the water had evaporated was vaporized in a graphite crucible which was heated in a furnace and the vapors were passed between two metal-foil collecting electrodes. The arrangement is shown in Fig. 2(c) and palladium or gold foils were used as the collecting electrodes. The foil was then placed in a discharge lamp and the lamp was baked out, filled, and its spectrum examined as described above.

We found that the foil electrodes inevitably collected vaporized material from the crucible in addition to ions. This extraneous material, mostly salts, produced a spectral background which varied from 10 to 100 times the dark-current noise background. In order to eliminate this background and thereby improve sensitivity, experiments were also performed which involved a second distillation in the presence of an electric field and a new set of foil electrodes. The second distillation was performed in an open flame. The first set of foil electrodes was heated to incandescence and in some cases actually completely melted with a second pair of collecting electrodes positioned above the flame to collect any gaseous ions in the stream. The second set of electrodes had to be placed a few centimeters above the cone of the flame to keep them from striking an arc with the thermal ions inside the flame itself. This procedure reduced the background in most cases to the point where it was comparable to dark current.

The procedure used with lake water was identical with that used to process sea water; of course, the boiling residue represented a considerably smaller fraction of the total volume of the sample. The maximum sample size for lake water was also 20 liters. The samples of oysters, seaweed, and plankton were treated like the residue from the boiling samples and placed directly into the crucible where the water was driven off and solid material was vaporized in the same experiment. The sample sizes used for these materials varied from ~1 to ~2 kg.

With each material experiments were first performed in search of the L_α line at 2733 Å since this line should be the most intense. If a promising L_α line were detected, the corresponding L_β line was searched for. Since small quantities of quarkium would clean up in the discharge, separate experiments were performed in which we first looked for the L_α line and secondly for the L_β line. The sample volume was then increased and the L_β line was searched for, then the L_α line, etc. Since the energy of the two excited states giving rise to these lines is roughly the same in quarkium, the ratio of the intensity of L_α to L_β should be primarily determined by the difference in excitation energy of the two levels and the branching ratios for the various decay modes. The energy difference between L_α and L_β is not large for quarkium; hence the branching ratios will be the major contributing factor. From a consideration of the branching ratios in hydrogen[28,29], we expected L_α:L_β to be ~10:1.

V. SPECTROSCOPIC RESULTS

In analyzing the sample spectra for possible quarkium lines, we anticipated the following: (1) the line would

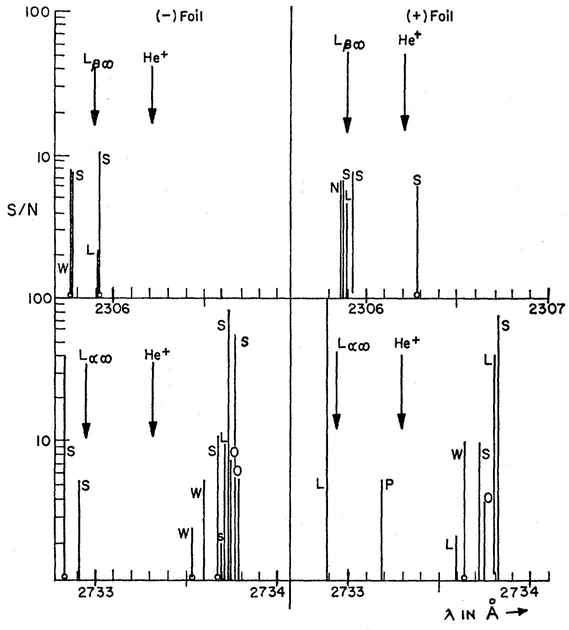

FIG. 3. Results of the spectroscopic experiments. S = sea water, L = lake water, W = seaweed, O = oysters, and P = plankton. The arrows indicate the position of the L_α or L_α line of quarkium for a quark mass equal to a He atom and infinity, respectively. The probable error in the position of a line is about 0.1 Å.

be most intense on the spectrum from either the (+) or (−) collecting plate or foil; (2) the line would clean up from the discharge; (3) an increase in the amount of material processed would increase the intensity of the quarkium lines and make more lines in the series detectable; (4) the lines should fall accurately into a Lyman series for some quark mass ≥a few proton masses. Figure 3 shows the positions and approximate strength of all lines which have been observed excluding a Pt line and a weak argon line at 2733.06 Å which were identified in some of the experiments, and resulted from the use of these materials in the spectroscopic procedure. There is no apparent tendency for the spectral lines to fall into a Lyman series. There are few lines in the region corresponding to a quark mass ≥four protons and there is a cluster of observations at 2733.8 Å from both the (+) and (−) collecting plates. These lines are most likely the same spectral line and the scatter in their wavelength is due to the difficulty of determining the center of a line with low signal to noise. This line appeared in some samples from most sources and in some cases it was relativley intense; we therefore expected a corresponding L_β line to appear in the larger samples if it were truly due to quarkium. This was not the case and we therefore rejected it as evidence for the existence of quarkium in these samples.

To set limits on the concentration of quarks in the various materials which we examined, we can state that we did not detect the L_α line of quarkium with a signal-to-noise ratio of 10:1. Hence if we assume 10^9 quarks detectable as indicated by the experiments on

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[29] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, Cambridge, England, 1963), p. 131.

Li, limits may be set for the concentration of $+\frac{2}{3}$ fractionally charged particles in the examined substances. Of course, such limits are also dependent on the assumptions involved in our collection procedure. We concluded that for the 20-liter samples of sea water and lake water the concentration of $+\frac{2}{3}$ quarks is less than 10^{-18} quarks/nucleon and for the 1-kg samples of oysters, seaweed, and plankton, the concentration is less than 10^{-17} quarks/nucleon.

VI. DESCRIPTION OF OIL-DROP METHOD

In addition to the spectroscopic search for $+\frac{2}{3}$ fractional charges described above, a modified Millikan oil-drop experiment was performed on various oils. This technique seemed promising for fractional charges since one electronic charge could be easily resolved on a drop of radius 10 μ which contains $\sim 2\times 10^{15}$ nucleons. This gives a sensitivity $\sim 1:10^{15}$ before concentration techniques are applied. With fairly simple concentration techniques, it seemed that sensitivities in excess of $1:10^{19}$ in oil could be developed. It was also apparent that the previous determinations of e by the oil-drop method did not rule out the possibility of fractional charges existing in these concentrations. Small drops were used and, of course, an enrichment was attempted. In fact, most of the authors reported a few drops which had fractional charges outside their expected errors and were rejected for a determination of e. These anomalous drops were ascribed to density variations due to impurities in the oil and other experimental difficulties. Schiffer[18] has described an experiment which used enrichment on 2.5-μ polyethylene spheres. The size of the oil drops used previously[30–35] was 1–5 μ in radius, with a few by Hopper on the order of 10 μ (see Table II).

Since large drops were to be examined, a horizontal-deflection technique similar to that described by Hopper and Laby[34] was used. The horizontal-deflection technique consisted of two parallel plates producing an electric field perpendicular to the vertical. The charged oil drops were allowed to fall between these plates in air and their deflections were measured. This method allows the use of large drops which could not be supported against the force of gravity in the conventional Millikan experiment with realizable laboratory electric field strengths. The drops varied in radius from 7 to 13 μ and from 0 to 20 electrons in charge.

One of the difficulties with the horizontal-deflection method is that its precision is limited by the uncertainties of the wall corrections to be applied to the motion of the drop. These corrections have been worked out carefully for the conventional Millikan experiment where the drop moves perpendicular to the plates, but they have not been completely determined for a drop making an arbitrary angle with the condenser plates. By suitable selection of charge, radius, and wall distance, this uncertainty was minimized.

TABLE II. Drop radius used in previous experiments.

a in μ	Observer
0.5–5.8	Millikan, 1913
1.6–2.3	Millikan, 1917
0.8–1.8	Bäcklin and Flenberg, 1936
0.7–4.3	Ishida, 1937
3.3–9.6	Hopper and Laby, 1941
3.3–5.8	Hopper, 1948

The principal advantages of the oil-drop method for the detection of quarks are the following: (1) It measures charge directly (the only well-defined physical characteristic of these particles); (2) the fractional charge of the drop will be preserved regardless of the chemical and/or nuclear reactions which the quarks have undergone; (3) the experiment has a basic simplicity in the sense that a positive result ($\pm\frac{1}{3}$, $\pm\frac{2}{3}e$ charges) is readily identifiable; (4) it is equally sensitive to all six quarks ($\mathcal{P}$, $\mathcal{N}$, λ, $\bar{\mathcal{P}}$, $\bar{\mathcal{N}}$, $\bar{\lambda}$).

The principal disadvantage of the oil-drop method is that it uses a very selected type of material and there is the possibility that quarks may not be present in oil for a variety of reasons related to its chemical formation or manufacture. In light of this we have used natural oils from various sources which have not undergone the extensive refinement of typical low-vapor-pressure oils normally used in this type of experiment.

VII. CONCENTRATION TECHNIQUE IN OIL

In order to improve the sensitivity of the oil-drop experiment, an effort was made to enrich the oil in quarks. Since the charge of a quark is fractional, molecules containing a single quark will have a net charge. It is possible that a molecule containing more than one quark could be neutral if the sum of the quark charges were zero or an integer. However, from a statistical point of view, it is extremely unlikely that a single molecule would contain more than one quark. If quarks exist in oil at all, they would probably be present in chemical combination with other atoms of the oil forming a molecule with a net charge. In the case of positively charged quarks, the quark itself would probably form the nucleus of an atom which could then combine with other atoms to form a molecule. The negative quarks would most likely fall into low-lying mesoniclike orbits of normal atoms. In either case, oil drops containing a quark should exhibit charges of $(n\pm\frac{1}{3})e$, where n is an integer and e is the electronic charge.

The concentration technique consisted of applying a strong electric field to the oil by means of metallic electrodes. Quark-containing molecules would then be attracted to the appropriate electrode since they have a

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net charge. In this way it was hoped that the layer of oil in contact with the electrode would be enriched in quarks. It was in this oil that we hoped to detect $\frac{1}{3}$ fractional charges. Of course, it is possible that the quark-bearing molecules could combine with the electrode and be removed from the liquid. It seems probable that the quark-bearing molecule would not stick to the electrode. A rough estimate of the binding energy at the electrode of a molecule containing a quark can be obtained from its image charge in the metal surface. The electrostatic energy W of a charge q at a distance d from a metallic plane in a medium of dielectric constant ϵ is

$$W=q^2/2\epsilon d. \tag{4}$$

For a molecule containing a quark, q will probably be $\frac{1}{3}e$. At a distance ~ 10 Å such a molecule will have an electrostatic energy equal to the thermal energy at room temperature. A typical dimension for an oil molecule is of the order of 10 Å and the surface layer on the metal formed by oil molecules, normal ions, oxides, etc., would certainly be >10 Å thick; hence it seems probable that an appreciable fraction of the quark-bearing molecules would not become bound tightly to the metal surface.

In the apparatus for the concentration of quarks in oil, the pole of the electromagnet used to produce the oil drops served as one electrode and aluminum foil served as the other. The magnet was held in the center of a glass beaker so that it just touched the surface of the oil. From 5 to 12 kV were applied between the electromagnet and the aluminum foil which covered the inside of the beaker. Three sizes of beakers (1, 4, and 20 liters) were used with collection times 18–48 h, and both polarities of the power supply were tried. The experimental procedure consisted of setting up the beaker and the electromagnet, turning on the voltage between the collecting electrodes, and adjusting a magnetic stirrer to give a slow circulation of the oil. After a number of hours had elapsed, the electromagnet was slowly withdrawn from the oil while the high voltage was still on. This left $\sim\frac{1}{10}$ cm^3 of concentrated oil on the electromagnet. The electric field was then reversed for a few minutes to allow any charges collected in the boundary layer next to the metal a chance to disperse through the oil. The electromagnet was then placed on the apparatus and the charge on oil drops produced from the concentrated oil was measured.

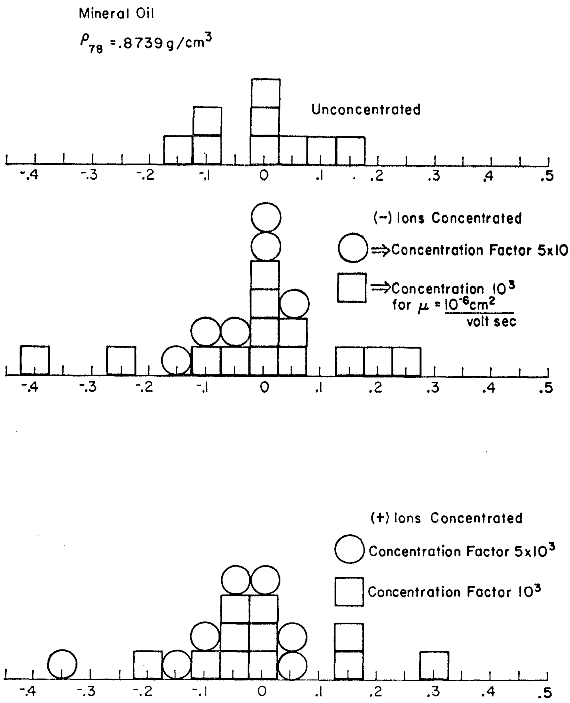

FIG. 4. Histogram of the measurements of the charge on mineral oil drops. The horizontal scale is the deviation from an integral electronic charge on the drop in tenths of an electronic charge.

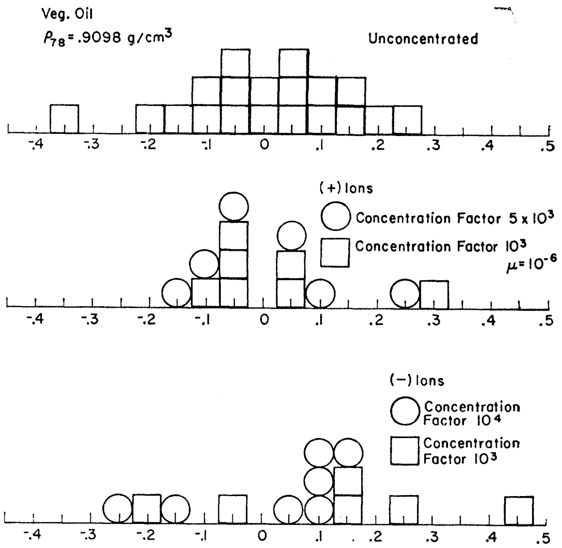

FIG. 5. Histogram of the measurements of the charge on a soyabean oil drop. The horizontal scale is the deviation from an integral electronic charge on the drop in tenths of an electronic charge.

In order to make an estimate of the concentration achieved by this process, the mobility of ions in oil must be determined. The mobility μ can be found from the Einstein relation.[36] For an ion of charge q with a diffusion coefficient D, the mobility is

$$\mu=q\mathrm{D}/kT, \tag{5}$$

where kT is the Boltzmann factor. For a light mineral oil typical of the type of oil used in this experiment, $D=0.03$ cm^2/day[37] for a tridecanoic acid molecule (mol wt 215) of radius ~ 4 Å. Thus from Eq. (5), $\mu=4\times10^{-6}$ cm^2/V sec. In general it appears that the mobility of quark-bearing molecules in oil will be 10^{-5}–10^{-6} cm^2/V sec depending on the size and shape of the particular ion.

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[36] K. Huang, *Statistical Mechanics* (John Wiley & Sons, Inc., New York, 1963).

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The electric field around the pole of the electromagnet falls off roughly as r^{-2}, which means that most of the electric field strength is within a distance equal to a few times the radius of the pole face. The maximum concentration was obtained from 20-liter oil samples for which the electromagnet was held at a potential of 12 kV with respect to the aluminum foil for 48 h. The pole of the electromagnet had a radius of $\frac{1}{2}$ cm and the hemispherical volume of oil enclosed within 1 cm of the electrode will contain approximately 5 cm^3. The time required to sweep out the ions in this volume will be $\approx 1\ \text{cm}/\mu E$, where E is the average electric field in the region and μ is the mobility of the ions. With adequate stirring of the oil, ions will be supplied to this volume as fast as they can be removed. Hence the total number of cubic centimeters which are processed in time t is

$$V = 5t/\mu E. \qquad (6)$$

The largest concentrations were obtained for $t = 1.6\times10^5$ sec (2 days) and an electric field E in the oil of 3×10^3 V/cm. Substituting these numbers in Eq. (6) yields $V = 10^3$ cm^3 for $\mu = 10^{-6}$ cm^2/V sec and $V = 10^4$ cm^3 for $\mu = 10^{-5}$ cm^2/V sec. Since the final volume of oil used in the experiment was 10^{-1} cm^3, the total volume concentration in this case was 10^4–10^5, depending on the mobility of the ions.

VIII. RESULTS OF OIL-DROP EXPERIMENT

Four different natural oils were examined. These were light mineral, soya-bean, peanut, and cod-liver oil. It was not practical to examine all available oils and we thought that these represented a reasonable selection of animal, vegetable, and mineral oils. The oils differed markedly in their adaptability to this experiment. On the average, mineral and peanut oil produced drops of lower charge for a radius $>10\ \mu$ than did soya-bean or cod-liver oil. This made the measurements for these two oils considerably easier.

Experimental errors produced a distribution of drops centered at zero with a standard deviation $\sim(-0.15$ to $+0.15)e$. The standard deviation of the distribution could have been reduced by using smaller drops since their displacement in the apparatus would be larger for a given value of charge. This would be a valuable improvement if a measurment of the electronic charge were the aim of the experiment. However, we wanted to use the drops containing as many atoms as possible since our goal was to search for quarks in the oil. Also, a reduction in the standard deviation of the distribution would not eliminate the anomalous drops of fractional charge observed by other investigators which could be caused by impurity particles affecting the density of the oil, a spurious air current in the apparatus, etc. This was a particularly important consideration since it was not desirable to use particularly pure or clean oil for these experiments. We decided that since a positive indication of the existence of quarks for this experiment would require many drops to have fractional charges of $\frac{1}{3}e$ on them, the standard deviation of the distribution of drops need only be small enough to permit the resolution of a $\frac{1}{3}e$ charge. Such a restriction on the accuracy of the experiment greatly simplified the data taking and construction of the apparatus.

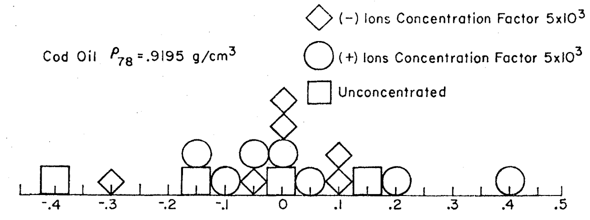

Fig. 6. Histogram of the measurements of the charge on cod-liver oil drops. The horizontal scale is the deviation from an integral electronic charge on the drop in tenths of an electronic charge.

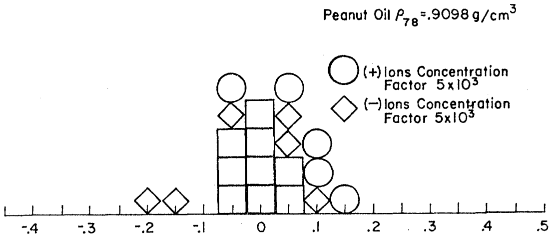

Fig. 7. Histogram of the measurements of the charge on peanut oil drops. The horizontal scale is the deviation from an integral electronic charge on the drop in tenths of an electronic charge.

From an examination of the data presented in Figs. 4–7, we conclude that there is no obvious indication of the presence of quarks at ±0.3. Indeed, if the concentration of quarks in the oil samples were such that on the average there was a 50% chance of a drop containing a quark, the number of data between -0.15 to $+0.15$ would be equal to the number outside that region. In fact, the distribution of the enriched oil samples is quite close to that of the unenriched oil. Note that there is a difference between the distribution for the enriched oil samples and the unenriched ones. There are a few more data points out on the wings of the distribution of the enriched drops. This was a general feature of the data for all the oils examined. We rejected this fact as evidence for the existence of quarks because in none of the experiments were we able to increase appreciably the numbers of these points by increasing the concentration factor of the sample. The concentration factor was increased by increasing the original volume of oil in the beaker, increasing the electric field, and increasing the collection time with no appreciable effect on the distribution. The most probable explanation of the difference in the distributions lies in the fact that there was a very limited amount of enriched oil for each run. Thus the number of enriched drops which can be produced is limited. One therefore must measure the charge on all of the drops which are in a suitable range of charge and radius in order to obtain sufficient statistics for a given sample. In the case of the unconcentrated drops, there was a virtually unlimited supply of oil and our aim was to test the accuracy of the apparatus. We therefore selected only those drops which were in very sharp focus on the film.

In conclusion, we found that a concentration of 10^{-19} quarks/nucleon in these oils would have been easily detected. If quarks existed in a concentration of $1:10,^{19}$ the distribution of data for the enriched oil drops would have been drastically altered. Since we did not observe this effect, we concluded that the concentration of quarks in the four oils examined was less than a few quarks in 10^{20} nucleons for an ionic mobility of 10^{-6} cm^2/V sec. If the mobility were as high as 10^{-5} cm^2/V sec, as it would be for small ions, then the concentration of quarks is less than a few quarks in 10^{21} nucleons.

ACKNOWLEDGMENTS

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Search for the Contribution of the Longitudinal Virtual Photon in Inelastic Electron-Proton Scattering

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We have made a systematic investigation of the inelastic scattering cross section for electrons on hydrogen, keeping constant the two relativistic parameters: four-momentum transfer q^2 and equivalent photon energy K. In the one-photon-exchange approximation, it is then possible to separate the contributions due to the longitudinal and transverse virtual photons. The new feature of our experiment was the use of a thin (12-mm) flat hydrogen target. For our choice of the parameters [$q^2=0.2$, 0.3, and 0.4 (BeV/c)^2; $K=340$ MeV] we did not find any discernible contribution of the longitudinal virtual photon.

I. INTRODUCTION

NUMEROUS authors have studied theoretically and experimentally the inelastic scattering of electrons on hydrogen targets. Among the experimenters, we may mention Panofsky and Allton,[1] Hand,[2] and more recently Lynch, Allaby, and Ritson.[3] These last authors have made a very extensive study of the first inelastic peak, due to excitation of the $N_{\frac{3}{2}}^*$ resonance, taking data at all the possible values of the three parameters available with the facilities at the 1-BeV Stanford Linear Accelerator. The three parameters are E_0 (the energy of the incident electron), and E_1 and θ (the energy and angle of the scattered electron). They used a 112-cm, 90° single-focusing spectrometer and a thick (30 cm long) liquid-hydrogen target.

To present their data, they used the convenient formalism proposed by Hand[2]: Let us denote by $q^2=2E_0E_1(1-\cos\theta)$ the four-momentum transfer, and by $K=E_0-E_1-q^2/2M$ the equivalent photon energy ($M=$mass of the proton); then, in the one-photon-exchange approximation, one may express the cross section as

$$\frac{d^2\sigma}{d\Omega dE_1}(E_0,E_1,\theta)=\Gamma_t(E_0,E_1,\theta)\sigma_t(K,q^2)+\Gamma_s(E_0,E_1,\theta)\sigma_s(K,q^2),$$

with

$$\Gamma_{t,s}=\frac{\alpha}{4\pi^2}\frac{K}{q^2}\frac{E_1}{E_0}\left[a_{t,s}+\frac{q^2\cot^2(\frac{1}{2}\theta)}{q^2+(E_0-E_1)^2}\right],\quad a_t=2,\quad a_s=0.$$

σ_t and σ_s contain the inelastic form factors of the nucleon, and can be interpreted as (and will be called) the total absorption cross sections for transversely and longitudinally polarized virtual photons, respectively; they depend only on the invariants K and q^2.

It is therefore tempting—and this is what has been done by Lynch—to choose the parameters in such a way that K and q^2 remain constant. The kinematic factors Γ_s and Γ_t are easy to calculate, and if one uses the "Rosenbluth plot"—that is, $(1/\Gamma_t)(d^2\sigma/d\Omega dE)$ as a function of Γ_s/Γ_t—one should obtain a straight line whose ordinate at the origin is equal to the transverse cross section, and whose slope is equal to the longitudinal one.

One of the main results of Lynch's investigation is that, in some cases, the longitudinal cross section is not zero. We thought that this result deserved another investigation, and we tried to check it using a com-

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