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## Search for Fractional Charges in Water

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Results are presented from a search for fractional charges in water from a variety of natural sources. About 30 000 water drops have been measured, comprising 51  $\mu\text{g}$  of water and dissolved materials. No evidence for fractional charge was seen.

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Recent reports<sup>1</sup> of third-integral charges on superconducting niobium spheres levitated in a magnetic field have heightened interest in the search for quarks in stable matter. Other magnetic-levitometer experiments using steel samples<sup>2,3</sup> have given negative results. These experiments all suffer from the need to use samples of refined or processed materials. Since quarked atoms would appear as impurities in these samples, processing may have depleted the samples of quarked matter. Other experiments have searched smaller samples, but claim greater sensitivity due to enrichment schemes based on specific assumptions about the chemistry of quarked matter.<sup>4</sup> Some of these assumptions are rather uncertain, as the quark mass, the sign of its charge, and the nature of the long-range color force, if any, might affect the chemical behavior of quarked atoms dramatically.<sup>5</sup> One simple property of quarked atoms which is rather generally agreed on is that they would not evaporate easily from solution in a polar liquid. This could lead to concentration of quarks in the ocean.

In the experiment reported here we measured natural samples—ocean water, water from landlocked salt lakes, natural mineral waters, and others—in unrefined form, although to relieve saturation, some distilled water was occasionally added. Some of these samples have probably benefited from concentration of quarks by evaporation. In a 51- $\mu\text{g}$  sample, we find no evidence for fractional charge.

The San Francisco State quark-search apparatus is a modification of the Millikan oil-drop experiment in which small drops are introduced one at a time into a measuring chamber by a piezoelectric drop ejector. The drop's drift ve-

locity in an electric field is measured by timing the passage of an image of the illuminated drop over a series of slits. The measurement is controlled and data are collected by an on-line computer. By switching the field polarity in mid-measurement, the mass and charge of each drop is determined. The measurement is highly redundant, allowing reliable rejection of badly measured drops. Drops which change charge during the measurement are identified and rejected. Drops can be measured at a rate of about 1  $\text{sec}^{-1}$ , and we can measure about 1  $\mu\text{g}$  of water in an hour of good running. The charge on a 15- $\mu\text{m}$ -diam water drop is measured to an accuracy of about 3.5% of  $e$ .

To keep the charge on drops to be measured near zero, a bias wire is inserted into the water in the drop ejector. The potential of this wire (a few volts) is adjusted by the computer. We accepted drops with charge between  $-12e$  and  $+12e$ . Rejection of charges outside this range accounts for most of the roughly 50% rejection rate of single measurements. Further details of the apparatus and a full discussion of criteria for rejection of data have been published previously.<sup>6</sup>

Figure 1 shows the measured velocity of a drop as a function of position. Each value of the velocity is determined from the crossing times for two adjacent slits. The field switches from positive to negative at slit 39. The change in velocity seen at this point corresponds to a charge on the drop of  $-8e$ . The field returns to its original polarity at slit 74, and the velocity returns to nearly its original value. A change in charge during the measurement can be detected by comparing the final velocity with the original one. This is complicated, however, in that we cannot com-

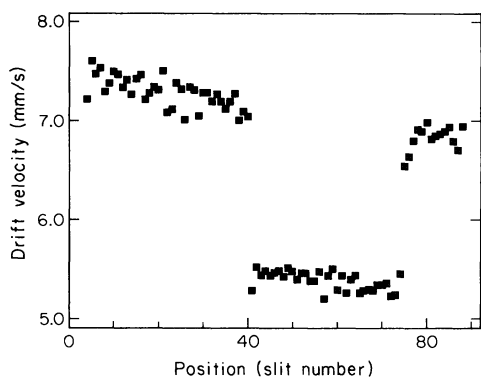


FIG. 1. Drift velocity as a function of position for a typical water drop. Reversals of the electric field occur at slits 39 and 74.

pletely eliminate evaporation of the water drops. The steady mass loss due to evaporation gives rise to a general decrease in drift velocity over the time of the measurement, which can also be seen in Fig. 1. During a run, the drop size and the rate of evaporation are sufficiently constant that an on-line correction can be applied, and a "charge change"  $\Delta Q$  calculated for each drop. Only drops with  $\Delta Q < 0.5e$  are kept in the data sample. The rate of occurrence of values outside this limit is small, less than one per thousand drops measured.

In Fig. 2 we show the results from all of our water samples. The horizontal axis is residual charge, the nonintegral part of the measured charge value. A drop containing a quark would

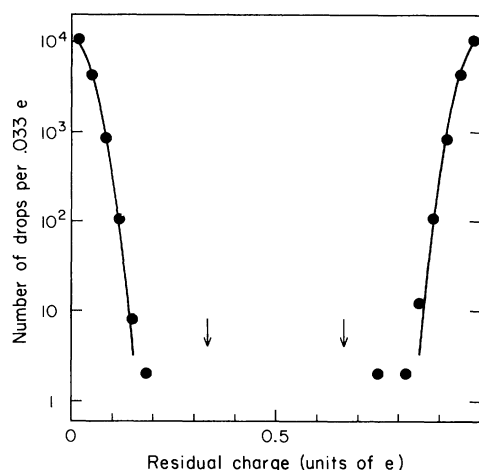


FIG. 2. Distribution of residual charge for all of our water drops. The curve is a fitted Gaussian with a standard deviation  $0.037e$ .

appear near  $\frac{1}{3}e$  or  $\frac{2}{3}e$ , indicated by arrows. A Gaussian curve fitted to the data ( $\sigma = 0.037e$ ), centered at zero residual charge, is drawn on the plot. Most of the measurements roughly follow this single Gaussian. Two measurements with residual charge of about  $0.74e$  seem quite inconsistent with the distribution for integer charges. However, they are not very close to the charge of a quark; with a measuring error of  $0.042e$ , typical of runs at the time these data were taken, they fall about  $1.75\sigma$  from  $\frac{2}{3}e$ . We feel that these two measurements are due to disturbances in the chamber caused by the electric field. Irregular

TABLE I. Sources of water samples giving the mass of each type measured and the amount of the major dissolved materials.

Water source	Description	Mass measured ( $\mu\text{g}$ )	Major solutes ( $\mu\text{g}$ )
Pacific Ocean	Off Ocean Beach, San Francisco	13.4	Na(0.17), Cl(0.29), Mg(0.016) SO <sub>4</sub> (0.014), Ca(0.008) <sup>a</sup>
Lake Louise	Glacial Lake, Canada	15.5	
Calistoga water	Bottled from the Geysers, California	7.3	
Perrier water	From the Perrier Source, France	2.3	
Bumpass' Hell	Fumarole, Lassen Volcanic National Park	4.1	SO <sub>4</sub> (0.001), SiO <sub>2</sub> (0.001) <sup>b</sup>
Mono Lake	Land-locked saline lake, California	0.8	Na(0.03), Cl(0.02), CO <sub>3</sub> (0.02), SO <sub>4</sub> (0.01), K(0.001) <sup>c</sup>
Distilled water		7.5	

<sup>a</sup>See Ref. 7.

<sup>b</sup>Patrick Muffler, U.S. Geological Survey, Menlo Park, Cal., private communication.

<sup>c</sup>B. F. Jones, U.S. Geological Survey Prof. Paper 502-A, 1965 (unpublished). Figures from these listings have been increased by 50%, since diversion of the tributaries to Mono Lake by the Los Angeles water department has increased the lake's salinity since 1965 by about 50%.

TABLE II. Comparisons of the type and quantity of material analyzed by recent quark searches. Only the experiment of LaRue, Phillips, and Fairbank has evidence for quarks. The stated values in the quark/nucleon column for the other null experiments are upper limits.

Reference	Technique	Sample material	Mass	Quark/Nucleon
LaRue, Phillips, and Fairbank (Ref. 1)	Levitometer	Niobium	1100 $\mu\text{g}$	$2.1 \times 10^{-20}$
Marinelli and Morpurgo (Ref. 2)	Levitometer	Steel	3700 $\mu\text{g}$	$< 1.3 \times 10^{-21}$
Liebowitz, Binder, and Ziock (Ref. 3).	Levitometer	Steel	720 $\mu\text{g}$	$< 6.9 \times 10^{-21}$
Hodges <i>et al.</i> (Ref. 6)	Millikan	Native mercury	175 $\mu\text{g}$	$< 2.8 \times 10^{-20}$
Ogorodnikov, Samoilo, and Solntsev (Ref. 10)	Desorption	Sea water and ocean sediments	134 kg <sup>a</sup>	$< 4.0 \times 10^{-28}$
Boyd <i>et al.</i> (Ref. 11)	Van de Graaff	Helium	20 $\mu\text{g}$	$< 6.4 \times 10^{-16}$
This experiment	Millikan	Natural waters	51 $\mu\text{g}$	$< 9.8 \times 10^{-20}$

<sup>a</sup>The experiment of Ogorodnikov, Samoilo, and Solntsev is the only experiment listed here which relies on a quark concentration scheme. The scheme involves two assumptions: (a) that the vapor from the heated sample will contain the quarks; and (2) that quarks collected on filter electrodes can be transported to collector electrodes. With an applied electric field, the rate of desorption from the heated collector electrode then sets the limit on quark concentrations.

air currents due to discharges in the chamber are sometimes observed. We have also occasionally observed tiny charged bits of matter flying up or down in the chamber which sometimes affect the motion of the drop being measured. One source of these charged particles is the water drops themselves, which can evaporate to leave a small salt crystal behind.

The origins of our water samples and some details about them are given in Table I. These samples have complicated and varied geologic histories. The spring waters and the fumarole runoff have passed through subterranean rock formations, two of them at high temperatures. Mono Lake is the oldest free-standing body of water in North America (over one million years old). The oceans have collected solutes over the last three billion years. Our samples contain a wide range of dissolved ions; in addition to those listed, they contain trace quantities ( $> 10^6$  atoms) of more than forty elements, including Ge, In, Mo, V, T, U, W, and Nb.<sup>7</sup>

The special role of sea water in quark searches is due to the fact that a single quark will always have an electric monopole moment. The high solubility of polar solutes in water is well known, and is due to hydration (hydrogen-bonding) interactions between solute and water.<sup>8</sup> Thus, quarks of any sign should remain dissolved in water. Perhaps only the unfortunate binding of a quark to its antiquark would allow their removal from water (by precipitation or annihilation).

The observation by Millikan in 1910 of "a value of the charge on the drop some 30% lower than

the final value of  $e$ " was a measurement made on water drops condensed from vapor.<sup>9</sup> His sample in total was about 0.0007  $\mu\text{g}$ , some 100 000 times smaller than ours. Millikan's explanation was that the measurement of that drop was in error as a result of rapid evaporation. Thus the interpretation of this uncertain measurement as evidence for fractional charge should be discounted.

A comparison between our result and the other published stable-matter quark searches of the last four years is shown in Table II. The only positive quark search is the experiment of LaRue, Phillips, and Fairbank<sup>1</sup> which has observed fourteen charges of  $\mp \frac{1}{3}e$ . Compared to their total sample, the water analyzed in our experiment is 20 times less in mass. For the null experiments the quark/nucleon column gives the upper limit on the concentration of quarks. The limits are calculated at the 95% confidence level except for the analysis of Boyd *et al.* which is at 67% and of Ogorodnikov, Samoilo, and Solntsev where the confidence level is not available.

In conclusion, we have measured the largest direct water sample ever, and the only one since Millikan's work, with no evidence for quarks. We find less than one quark per  $1.0 \times 10^{19}$  nucleons of natural water with 95% confidence.

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<sup>3</sup>D. Liebowitz, M. Binder, and K. O. H. Ziock, *Phys. Rev. Lett.* **50**, 1640 (1983).

<sup>4</sup>See the review by L. W. Jones, *Rev. Mod. Phys.* **49**, 717 (1977).

<sup>5</sup>The properties of atoms with quarked nuclei have been discussed by K. S. Lackner and G. Zweig, *Lett. Nuovo Cimento* **33**, 65 (1982).

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<sup>7</sup>P. K. Park, in *Water and Aqueous Solutions*, edited by R. A. Horne (Wiley, New York, 1971), pp. 245-248.

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