Did Millikan observe fractional charges on oil drops?

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We have reanalyzed Millikan's 1913 data on oil drops to examine the evidence for charge quantization and for fractional residual charge. We find strong evidence in favor of charge quantization and no convincing evidence for fractional residual charges on the oil drops.

I. INTRODUCTION

Recent reports[1] of the observation of fractional residual charges on superconducting niobium spheres have aroused interest in re-examining Millikan's original oil drop data. In 1910 Millikan, himself, reported, "I have discarded one uncertain and unduplicated observation apparently on a singly charged drop, which gave a value of the charge on the drop some 30 per cent lower than the final value of e."[2] He explained this by rapid evaporation of the water droplet on which the observation was made.[3] A modern reanalysis[4] of Millikan's 1913 data has shown two anomalous events that are consistent with fractional charges. These are not, however, evidence in favor of free quarks because not only are the total charges on the oil drops integral multiples of the fractional charge, but each change in the charge on the drops is also an integral multiple of that fractional charge. It would indeed be remarkable if fractional changes in charge occurred only on drops that were themselves fractionally charged.

In this paper we reanalyze Millikan's data to examine the evidence for both charge quantization and fractional residual charge on the oil drops. Charge quantization will be shown by looking at the changes in the charge on the oil drops, a point Millikan emphasized.[5] We look for fractional residual charges by two methods: first, we examine the intercept generated by fitting a least-squares straight line to the data in n vs $(1/t_g + 1/t_f)$ for each drop, (see below for details); second, we calculate, for each drop, the average deviation from integral charge obtained by dividing the total charge on the drop, for each individual measurement, by the best modern value for e, $4.803\,24\times 10^{-10}$ esu.

II. CALCULATIONS

The equation of motion of an oil drop moving in an upwards electric field F is $m\ddot{x} = mg - K\dot{x} - QF$, where Q is the drop's charge. According to Stoke's law $K = 6\Pi a\mu$, where a is the drop's radius and μ is the air's viscosity. To take into account the particulate character of air, Millikan replaced K by $K/(1 + b/pa)$, where p is the air pressure and b a parameter fixed by experiment. Since all measurements were made at terminal velocities, $\ddot{x} = 0$, whence

$$Q = \frac{mg}{F}\frac{V_f + V_g}{V_g}; \tag{1}$$

here the subscripts indicate terminal velocities without (V_g) and with (V_f) the field, respectively. Now m, compensated for the bouyant force of air, can be replaced by a using $m = (4/3)\Pi a^3(\sigma - \rho)$, σ and ρ being the densities of oil and air, respectively; a can be done away within favor of μ using Stoke's law; and the ratios of distance d to times of fall and rise, t_g and t_f, can be substituted for the velocities. Altogether

$$Q = ne = 9\Pi d\sqrt{\frac{2}{g}}\,\frac{1}{F}\left(\frac{\mu^3}{(\sigma-\rho)(1+b/pa)^3}\right)^{1/2} \times\left[\sqrt{V_g}\left(\frac{1}{t_g}+\frac{1}{t_f}\right)\right]. \tag{2}$$

The first equality in Eq. (2) expresses the assumption that charge is quantized.

An alternative calculation makes use of successive rise times $t_f, t_{f'}$, in cases where, in the intervening fall, the drop acquired an increment of charge Δn. In this case,

$$\Delta Q = (\Delta n)e = 9\Pi d\sqrt{\frac{2}{g}}\,\frac{1}{F}\left(\frac{\mu^3}{(\sigma-\rho)(1+b/pa)^3}\right)^{1/2} \times\left[\sqrt{V_g}\left(\frac{1}{t_f}-\frac{1}{t_{f'}}\right)\right]. \tag{3}$$

The data used in our recalculations are contained in a microfilm of Millikan's laboratory notebooks obtained from the Millikan Collection at the California Institute of Technology.[6] The notebooks cover the period 28 October 1911 to 16 April 1912. In our recalculation we have used only drops measured after 13 February 1912, the first drop published by Millikan. Prior to that date Millikan had doubts about the proper operation of his apparatus and was, in particular, concerned about convection problems.[7] Of the 107 drops in this period we have excluded 8 for experimental reasons given by Millikan, such as drop flick-

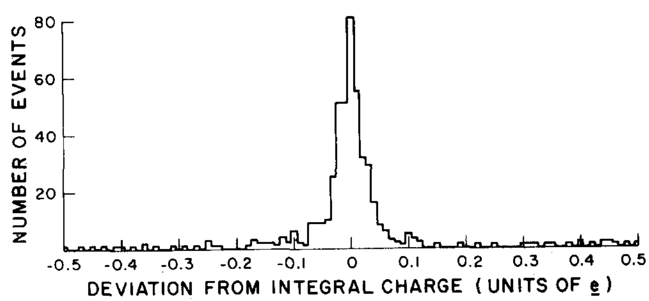

Fig. 1. Deviation from integral charge computed from the change in the charge, ΔQ, of the oil drops.

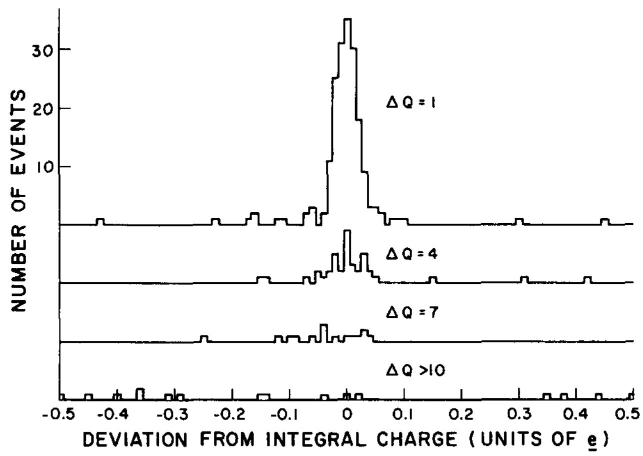

Fig. 2. Deviation from integral charge for different values of the change in the charge, ΔQ, on the drop.

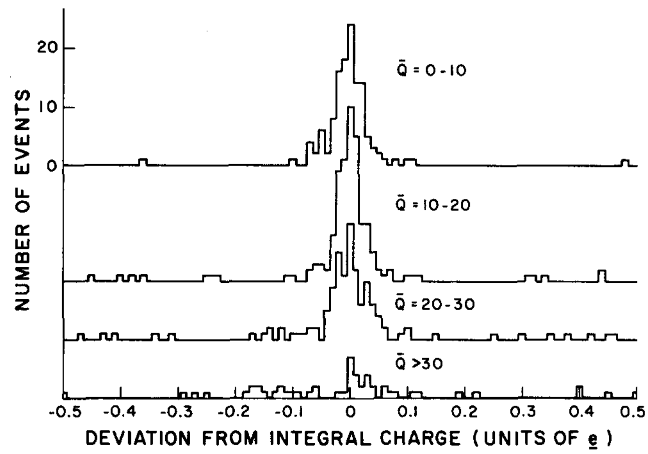

Fig. 3. Deviation from integral charge for different average values ($\bar{Q}$) of the charge on the drop.

ering, irregular voltage, temperature changing rapidly, or inability to obtain a pressure or temperature reading.[8] Fifteen others were excluded because they had values of pressure and drop radius that require a second-order correction to Stoke's law.[9] In our calculation of the change in the charge on the oil drop, four other events were excluded because they had only one value of the total charge, and thus could not give a value for the change in charge. A total of 80 events remain. We then calculate the value for ΔQ for each pair measurements of t_f and $t_{f'}$ using Eq. (3) given above. The results of our calculations are given in Figs. 1–3 and will be discussed below.

We examine the evidence for fractional residual charge on the individual oil drops by two methods. As can be seen from Eq. (2), n should be a linear function[10] of $(1/t_g + 1/t_f)$.[11] We fit a least-square straight line to n as a function of $(1/t_g + 1/t_f)$ of the form

$$n = a(1/t_g + 1/t_f) + b.$$

The intercept b at

$$(1/t_f + 1/t_g) = 0$$

gives the fractional residual charge on the drop. For these calculations, in addition to the 27 drops excluded above, we further exclude 19 drops that did not have at least four unique values of n.[12] This was to insure a reasonable error

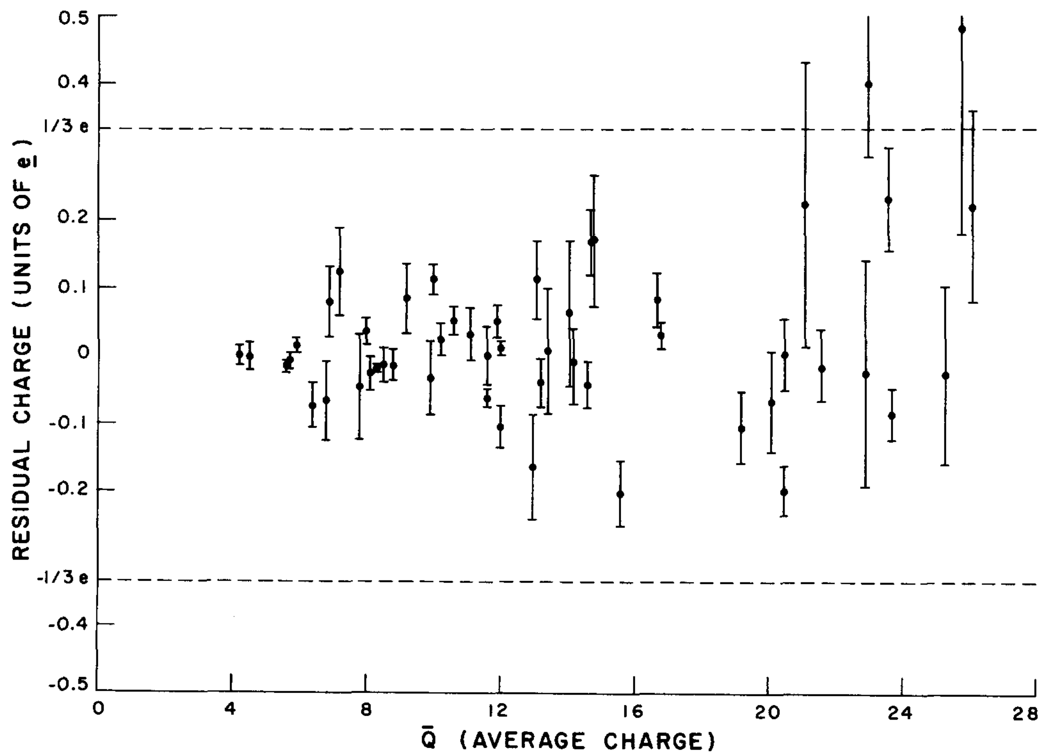

Fig. 4. Residual charge on the oil drop as a function of the average charge $\bar{Q}$ on the drop computed by the least-squares method.

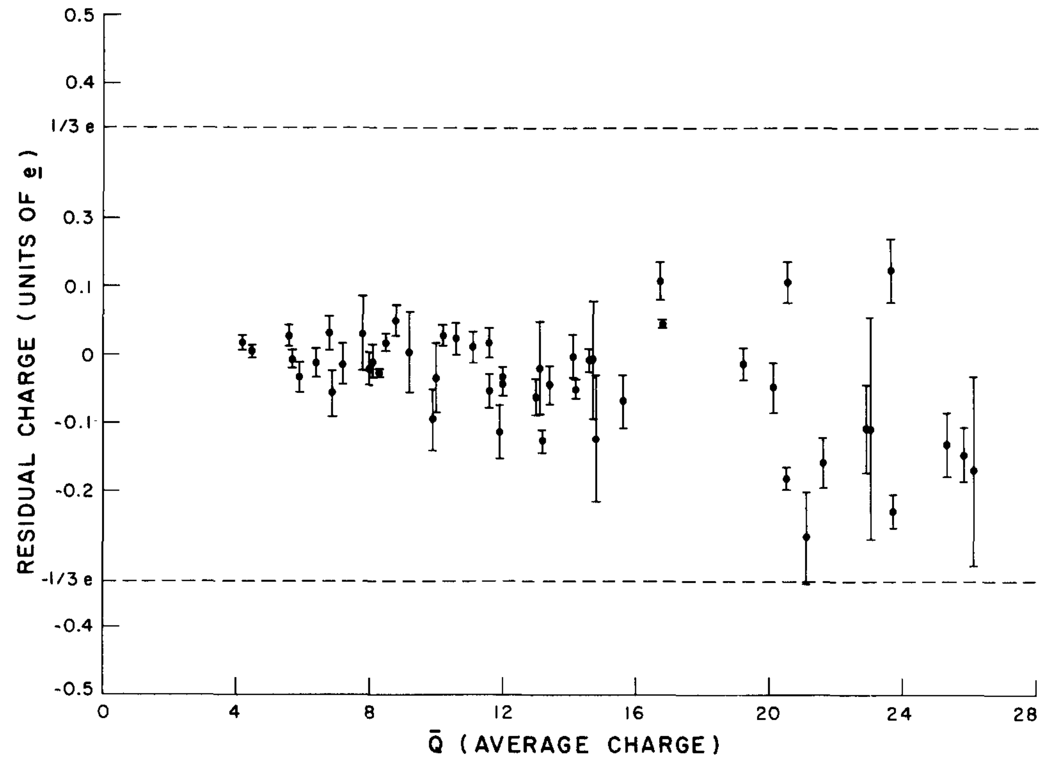

Fig. 5. Residual charge on the oil drop as a function of the average charge $\bar{Q}$ on the drop computed from the average deviation of the total charge on the drop from integral values.

in our least-squares fit. A total of 61 drops remain. In addition we have exluded 13 individual measurements of t_f, out of a total of 804.[13] As seen from Figs. 1–3, the determinaiton of n becomes unreliable for values of $n \gg 20$. Seven of the excluded points are on drops with large charges and for which we cannot uniquely assign a value of n. Six other points have $n < 20$, but have large deviations from integral charge (closer to half-integral n), an unlikely occurrence, and are considered bad observations.[14]

A second method of calculating residual charge is by calculating Q using Eq. (2) and dividing by the best modern value of e, $4.803\,24 \times 10^{-10}$ esu. We then calculate the average deviation from integral charge for each drop using the same 61 drops as above.

The two methods each have different advantages. The least-squares fit is less sensitive to absolute errors in voltage, temperature, pressure, and the viscosity of air, as well as to the Stoke's law correction than is the average deviation method. The latter, however, is less sensitive to a single bad data point, to slow drifts in voltage, and to evaporation.[15] Thus we have used both methods.[16]

III. RESULTS AND DISCUSSION

The results of the deviations from integral values of charge obtained from the calculation of ΔQ, the changes in charge are given in Fig. 1. The distribution has a large peak at zero, as expected and there is no evidence for fractional changes in charge, which would be indicated by a peak at $\pm 1/3$. There is also no evidence for any smaller unit of charge. As Millikan, himself, noted,[17] the changes in charge provide strong evidence for charge quantization. Figures 2 and 3 show the same results for various values of ΔQ, the change in charge, and $\bar{Q}$, the average charge on the drop, respectively. Both graphs show more deviation from integers as both the average charge and the change in charge increase, indicating the increasing unreliability of the method for drops with large total charge.

The results obtained for residual charge on the drops from both the least-squares fit and the average deviation are given in Figs. 4 and 5, respectively. We have plotted only those values for $\bar{Q} < 28$, because of the unreliability of our methods for large values of the total charge. The larger error in our values for the residual charge as $\bar{Q}$ increases is shown in Table I which gives these values for all 61 events. We see no convincing evidence of fractional residual charge for any of these drops. If we adopt as plausible criteria for fractional charge that both methods, least-squares fit (LSF) and average deviation (AD), give results closer to $\pm (1/3)e$ than 0, and that both methods give results at least two standard deviations from 0 we find two candidates. These are the events of 2 March 1912 (second observation) ($\bar{Q} = 21.1$ in Table I) and 29 March 1912 (second observation) ($\bar{Q} = 20.5$ in Table I). The values obtained for the fractional residual charge for these events are $(-0.778 \pm 0.209)e$(LSF), $(-0.272 \pm 0.070)e$ (AD); and $(-0.199 \pm 0.037)e$ (LSF), $(-0.186 \pm 0.016)e$ (AD), respectively. In addition we find that the values for e obtained by using $(n - 1/3)$ rather than n give a better fit to the modern value of e. For the March 2 event we calculate $e = (4.818 \pm 0.015) \times 10^{-10}$ esu (using $n - 1/3$) and $e = (4.742 \pm 0.020) \times 10^{-10}$ esu (n); for March 29, $e = (4.838 \pm 0.007 \times 10^{-10}$ esu $(n - 1/3)$ and $e = (4.759 \pm 0.008) \times 10^{-10}$ esu (n). Although these two events are somewhat more consistent with an interpretation of fractional charge rather than with integral charge the evidence is not strong. Both events also have a reasonably large average charge, $\bar{Q} \simeq 21.1$ and 20.5, respectively,

Table I. Residual charge on the oil drops computed by the least-squares fit and average deviation methods.

$\bar{Q}$ (Average charge)	Residual charge (Least-squares fit)	(Average deviation)
4.2	0.000 ± 0.014	0.015 ± 0.011
4.5	-0.002 ± 0.019	-0.003 ± 0.008
5.6	-0.016 ± 0.008	0.024 ± 0.016
5.7	-0.008 ± 0.013	-0.009 ± 0.013
5.9	0.014 ± 0.011	-0.035 ± 0.024
6.4	-0.074 ± 0.034	-0.015 ± 0.022
6.8	-0.067 ± 0.059	0.029 ± 0.026
6.9	0.079 ± 0.053	-0.058 ± 0.036
7.2	0.123 ± 0.065	-0.016 ± 0.032
7.8	-0.046 ± 0.078	0.029 ± 0.054
8.0	0.036 ± 0.021	-0.024 ± 0.026
8.1	-0.026 ± 0.025	-0.012 ± 0.024
8.3	-0.019 ± 0.005	-0.029 ± 0.005
8.5	-0.014 ± 0.026	0.015 ± 0.013
8.8	-0.014 ± 0.024	0.047 ± 0.022
9.2	0.084 ± 0.052	0.000 ± 0.060
9.9	-0.033 ± 0.054	-0.097 ± 0.045
10.0	0.112 ± 0.023	-0.037 ± 0.054
10.2	0.024 ± 0.024	0.025 ± 0.015
10.6	0.052 ± 0.021	0.020 ± 0.025
11.1	0.032 ± 0.039	0.008 ± 0.023
11.6	-0.062 ± 0.014	0.014 ± 0.022
11.6	0.000 ± 0.044	-0.056 ± 0.027
11.9	0.050 ± 0.025	-0.115 ± 0.041
12.0	0.011 ± 0.010	-0.035 ± 0.014
12.0	-0.105 ± 0.032	-0.048 ± 0.016
13.0	-0.164 ± 0.078	-0.064 ± 0.028
13.1	0.112 ± 0.058	-0.023 ± 0.070
13.2	-0.040 ± 0.037	-0.129 ± 0.016
13.4	0.007 ± 0.093	-0.047 ± 0.027
14.1	0.063 ± 0.107	-0.007 ± 0.034
14.2	-0.010 ± 0.051	-0.055 ± 0.013
14.6	-0.043 ± 0.034	-0.012 ± 0.018
14.7	0.167 ± 0.049	-0.011 ± 0.089
14.8	0.169 ± 0.097	-0.128 ± 0.095
15.6	-0.203 ± 0.048	-0.073 ± 0.039
16.7	0.082 ± 0.041	0.105 ± 0.028
16.8	0.030 ± 0.020	0.041 ± 0.003
19.2	-0.105 ± 0.054	-0.018 ± 0.023
20.1	-0.067 ± 0.075	-0.053 ± 0.037
20.5	-0.199 ± 0.037	-0.186 ± 0.016
20.5	0.002 ± 0.053	0.102 ± 0.031
21.1	-0.778 ± 0.209	-0.272 ± 0.070
21.6	-0.018 ± 0.059	-0.162 ± 0.038
22.9	-0.025 ± 0.167	-0.112 ± 0.065
23.0	-0.600 ± 0.108	-0.113 ± 0.162
23.6	0.231 ± 0.077	$0.120 + 0.048$
23.7	-0.085 ± 0.038	-0.236 ± 0.026
25.3	-0.027 ± 0.131	-0.136 ± 0.048
25.8	-0.481 ± 0.300	-0.152 ± 0.040
26.1	0.219 ± 0.138	-0.172 ± 0.141
28.6	-0.706 ± 0.364	-0.436 ± 0.103
28.8	0.047 ± 0.064	$-0.014 + 0.028$
29.3	-0.191 ± 0.170	$-0.012 + 0.132$
34.1	-0.247 ± 0.203	$-0.182 + 0.077$
34.4	0.304 ± 0.214	0.125 ± 0.067
35.6	-0.974 ± 0.133	-0.072 ± 0.182
38.8	0.022 ± 0.140	0.035 ± 0.043
51.7	-0.320 ± 0.360	-0.066 ± 0.161
85.4	1.879 ± 0.916	0.025 ± 0.273
122.8	3.251 ± 2.104	0.213 ± 0.341

where our calculational methods are becoming less reliable.

We conclude that Millikan's original data gives strong evidence for charge quantization and no convincing evidence for fractional residual charge, although two events (out of 61) are consistent with such an interpretation.

[1]George S. LaRue, James D. Phillips, and William M. Fairbank, Phys. Rev. Lett. **46**, 967 (1981); G. S. LaRue, Ph.D. Thesis, Stanford University, 1978 (unpublished); G. S. LaRue, W. M. Fairbank, and A. F. Hebard, Phys. Rev. Lett. **38**, 1011 (1977); G. S. LaRue, W. M. Fairbank, and J. D. Phillips, Phys. Rev. Lett. **42**, 142 (1979); erratum **42**, 1019 (1979); G. S. LaRue, J. D. Phillips, and W. M. Fairbank, Proceedings of the XXth Conference on High Energy Physics, Madison, Wisconsin, 1980 (unpublished).

[2]R. A. Millikan, Philos. Mag. **110**, 209 (1910).

[3]"Such drops, both because of the smallness of their size and the smallness of their charge, are not in equilibrium with multiply charged drops and consequently evaporate so rapidly that their life is relatively short. The single observation mentioned above was probably upon such a drop, but it was evaporating so rapidly that I obtained a poor value of *e*." Reference 2, p. 223.

[4]A. Franklin, Hist. Stud. Phys. Sci. **11** (part 2), 185 (1981).

[5]"The total number of changes which we have observed would be between one and two thousand and *in not one single instance has there been any change which did not represent the advent upon the drop of one definite invariable quantity of electricity or a very small multiple of that quantity* (emphasis in original)." R. A. Millikan, Phys. Rev. **32**, 349 (1911). In his 1913 paper [Phys. Rev. **2**, 109 (1913)], which was primarily devoted to a precise measurement of *e*, rather than to proving charge quantization, Millikan regarded his 1911 work as having definitively shown quantization.

[6]All notebook references are to folders 3.3 and 3.4 in the Robert Millikan Collection and are published by courtesy of the California Institute of Technology Archives. This is the data Millikan published in his 1913 paper (Ref. 5).

[7]See Ref. 4 for details.

[8]In addition half the data on one drop was excluded by both Millikan and ourselves because the voltage was fluctuating.

[9]For large values of $1/pa$, as Millikan himself suspected [Phys. Rev. **2**, 138 (1913)] the first-order correction to Stokes' law is insufficient.

[10]We get a very good estimate of n from the values of Δn, which is, in general, a very small number and easily obtained. "Since n' (our Δn) is always a small number and in some of the changes almost always had the value of 1 or 2, its determination for any change is obviously never a matter of the slightest uncertainty. On the other hand, n is often a large number, but with the aid of the known values of n' it can always be found with absolute certainty as long as it does not exceed say 100 or 150." Millikan (1913), pp. 123–124. As we shall see Millikan was somewhat optimistic. The estimate of n becomes rather unreliable for values greater than 30.

[11]This assumes that V_g = (distance of fall)/t_g has already been calculated.

[12]Thus a drop with successive values of the total charge $n = 4, 3, 4, 5$ would be excluded because there are only 3 unique values of the charge.

[13]By comparison Millikan omitted 12 observations on which he had performed calcuations and 45 on which he did no calculations. For a detailed discussion of Millikan's data analysis and calculations see Ref. 4.

[14]We tried to avoid exclusions except in cases where a measurement was clearly inconsistent with the other values obtained for that drop. Including these measurements actually had no significant effect on our results. We note in passing that four of these excluded measurements come from the same anomalous drop (7 March 1912, fourth observation). See Ref. 4 for details.

[15]We have looked for the effects of evaporation by examining t_g, the time of fall, as a function of time, for each drop. We see no consistent evidence of such evaporation.

[16]The method used by Millikan in 1913 and in the modern reanalysis (Ref. 4), whose primary purpose was to measure e precisely, did not look for the presence of fractional charge.

[17]See Ref. 5.