

## **A Device for Precision Neutralization of Electric Charge of Small Drops Using Ionized Air**

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For use in our Millikan type liquid drop searches for fractional charge elementary particles we have developed a simple ionized air device for neutralizing a narrow stream of small drops. The neutralizer has been used for drops ranging in diameter from 10 to 25  $\mu\text{m}$ . The width of the produced charge distribution is given by the Boltzmann equilibrium charge distribution and the mean of the distribution is set by a bias voltage.

Using the bias voltage, the mean can be set with a precision of better than  $e$ , the electron charge. The use of the neutralizer is illustrated in an application to mineral oil drops produced with charges of the order of  $1000e$ . We also show the interesting case of silicone oil drops that are produced in our drop generator with a charge distribution narrower than the Boltzmann equilibrium charge distribution, the charge distribution being broadened by the neutralizer.

## 1. INTRODUCTION

It has been known for a long time that the electric charge on small drops can be neutralized by passing the drops through an ionized gas [1-4], the ionization being produced by a radioactive source. The method is described in the classic paper of Cooper and Reist [1] and of Liu and Pui [2] and in aerosol textbooks [3,4]. In this paper we describe an ionized air neutralizer that uses a small electric field across the ionizer and makes use of the well known difference between the electrical mobilities of the ions produced in ionized air [5]. This enables us to control the ratio of the positive and negative ion concentrations and allows us to control the center of the drop charge distribution to a fraction of an electron charge. We developed this method to use in our searches for fractional charge elementary particles using a new version of the Millikan oil drop experiment [6,7]. A qualitative model of how the neutralizer works is given in the Appendix.

In our fractional charge search experiments the drops produced by a drop-on-demand method [6,8] have diameters in the range of 10 to 25  $\mu\text{m}$ , but in any particular search the drop diameter is maintained to a fraction of a per cent over the life of the search. It is important in these searches to keep the average drop charge close to zero and to keep the spread of the drop charges within a range of about  $+15e$  to  $-15e$ . Here  $e$  is the magnitude of the electron charge. This spread restriction is important because the error on the measurement of the drop charge is proportional to the square of the charge.

In the past our searches have used Dow Corning 200 silicone oil and as illustrated in Fig. 1, our drop generator with this oil produces a very narrow spread of drop charges close to zero charge. This narrow spread enabled us to set the maximum rms charge error per drop to less than  $0.03e$  [6]. We do not know the reason for this narrow spread. We surmise that it has to do with the lack of ions in this oil [9]. We also do not know the reason for the asymmetric charge distribution.

However, very large charges appear on drops produced by our drop generator using most other liquids such as mineral oils or water or mixtures of water and glycols. These charges range from several  $\pm 100e$  to several  $\pm 1000e$  and we have found no way to control the charge distribution using the generator or surrounding voltages. This, of course is a well known phenomenon in aerosol science experiments and has led to the development of neutralizers [1-4]. In this paper we describe a compact neutralizer that enables precise centering of the charge distribution on zero charge with the rms width of the distribution,  $\sigma_q$ , close to the Boltzmann equilibrium charge distribution [4] of

$$\sigma_q^2 = [4\pi\epsilon_0rkT/e^2] \quad (1)$$

Here in MKS units,  $r$  is the drop radius,  $k$  is the Boltzmann constant,  $T$  is the absolute temperature and  $\epsilon_0$  is the free space permittivity. The physics behind Eq. 1 is that the right side is the ratio of  $kT$  to the electrostatic energy of a drop containing one  $e$  of charge.

## II. DESCRIPTION OF NEUTRALIZER

As shown in Fig. 2 the neutralizer consists of a  $200\ \mu\text{C}\ ^{90}\text{Sr}$  source illuminating an open channel volume that is 3.0 in long in the vertical direction, 0.40 in wide along the direction of the beta rays and 1.60 in wide in the direction perpendicular to the beta rays. The drops being neutralized fall vertically downward through the open channel. The immediate surfaces containing this volume are made of insulating plastic, except on the 1.6 in wide surfaces where thin copper plates allow a voltage,  $V_B$ , to be placed across the 0.40 in width. The copper sheet of the grounded bias plate is 0.003 in thick so that the beta rays can pass through it. The axis perpendicular to the bias plates is called  $x$ , with  $x = 0$  midway between the plates and  $x = \pm w = \pm 0.20$  in at the plates.

This neutralizer is designed to act on a *narrow*, vertical stream of drops, the diameter of the stream being about 0.5 mm or less. The effect of the neutralizer on the drops depends upon the densities of the positive and negative ions in the region through which the drops fall, and therefore as discussed in the Appendix the neutralizer effect depends upon  $x$ . In the results described in this paper the drop stream is set at  $x = 0$ , that is, midway between the plates.

The neutralizer has a source shield not shown in Fig. 2 that allows the ionization region to be completely closed off from the source. The shield can be opened by different amounts, thus controlling the ionization. In all the data reported in this paper the shield is either fully

closed or fully open. When the shield is fully open the source produces of the order of  $10^{13}$  ion pairs/m<sup>3</sup>.

The neutralizer is used in connection with our fractional charge search apparatus shown schematically in Fig. 3. The drop generator emits drops into the top of the neutralizer. The drops are at terminal velocity as they fall through the ionized air, for example a 25  $\mu\text{m}$  diameter drop has a terminal velocity of about 1.7 cm/s. The charge measuring part of the search apparatus [6] lies just below the neutralizer and uses a horizontal, alternating electric field to give the drops horizontal, alternating motion, the extent of the motion being proportional to the charge on the drop. The position of the drops as they fall under gravity and move horizontally under the alternating electric field is recorded onto a computer using a lens and CCD system with a stroboscopic LED light source.

### III. NEUTRALIZATION OF HIGHLY CHARGED MINERAL OIL DROPS

To demonstrate the use of the neutralizer we turn to its most recent use with 12.5  $\mu\text{m}$  radius drops of Ultraol 50 NF mineral oil [10] containing a suspension of about 2% powdered meteorite and some surfactant. (We are using these drops to search for fractional charge particles in meteoritic material.) Figure 4a and 4b show two typical initial drop charge distributions for these drops. That is, this is what we measure when the source is closed, so that the neutralizer is not being used. The distribution in Fig. 4a has a mean charge  $q_m = -731e$  and an rms width  $\sigma_q = 362e$ . The respective values for the distribution in

Fig. 4b are  $q_m = -1012e$  and  $\sigma_q = 331e$  for the same type and size drops produced at another time. We have already noted that the charge distribution varies with the operation parameters of the drop generator in ways we do not understand, and even for the same operation parameters, the distribution changes with time.

Figure 5 shows the charge distribution for the same drops when the neutralizer is fully on and the bias voltage,  $V_B$ , is set to 0.25 volts. This bias voltage has been selected to move the mean charge,  $q_m$ , close to zero. We see that the distribution is now much narrower. We find  $\sigma_q$  is  $15.5e$  compared to  $\sigma_q = 14.9e$  given by Eq. 1 for  $r = 12.5 \mu\text{m}$ . Thus the neutralizer can give a charge distribution that obeys the conventional Boltzmann law.

Figure 6 shows in more detail how  $q_m$  and  $\sigma_q$  change with  $V_B$ . The former is quite sensitive to  $V_B$ , but the latter is not. In the use of the neutralizer we set  $V_B$  so that  $q_m$  is within a unit of  $e$  of zero. Over several days of operation there may be a drift in  $q_m$  of about one unit of  $e$ , and we then slightly change  $V_B$ . As shown in Fig. 6b this does not change  $\sigma_q$ .

#### IV. NEUTRALIZER EFFECT ON SILICONE OIL DROPS

As we have already noted and shown in Fig. 1, silicone drops are produced with close to zero charge by our drop generator. For example, the silicone oil charge distribution in Fig. 1 has  $\sigma_q = 3.2e$ , much smaller than the  $\sigma_q = 15e$  predicted by Eq. 1. (The drops have a

radius of 12.3  $\mu\text{m}$ .) In our work we are of course well satisfied with this situation and do not use the neutralizer for searches using silicone oil drops.

However, it is interesting to explore the effect of the neutralizer on a drop charge distribution that is initially almost neutral and that has a  $\sigma_q$  much smaller than given by Eq. 1. In Fig. 1 the initial charge distribution has  $q_m = -4.2e$  and  $\sigma_q = 3.2e$ . But if we open the neutralizer exposing the silicone oil drops to the ionized air we find that the distribution broadens, given  $\sigma_q = 16.1e$ , close to the Eq. 1 predicted value. Fig.7 is an example. Fig. 8a surveys the effect on that initial charge distribution with the neutralizer being open for various values of  $V_B$ . The mean charge  $q_m$  is shifted by  $V_B$ . The qualitative behavior is the same as that for the mineral oil drops [Fig. 6a].

Turning to Fig. 8b. we see that the distribution is broadened to approximately the  $\sigma_q$  value given by Eq. 1, the broadening being approximately independent of  $V_B$  for small values of  $V_B$ .

We note that for very small silicone oil drop of radius, for example less than 1  $\mu\text{m}$ , Eq. 1 gives  $\sigma_q < 4.2e$ . Hence a narrow charge distribution, such as the  $\sigma_q = 3.2e$  we have here could be maintained while adjusting  $q_m$ .

## **V. CONCLUSION**

We have designed and operated a simple ionized air neutralizer that allows drop charge distributions to be set to the equilibrium Boltzmann charge distribution with the mean charge being set within a unit of  $e$ .

## **ACKNOWLEDGMENTS**

This work was supported by United States Department of Energy contract DE-AC03-76SF00515.



## APPENDIX: QUALITATIVE MODEL OF NEUTRALIZER OPERATION

A full quantitative model for the operation of this neutralizer is beyond the scope of this paper. It would have to take into account the various processes occurring in air in the presence of ionizing radiation and an electric field – creation of ion pairs, recombination of ion pairs, the history of the various ions including the gaseous impurities in the closed air space, and the complicated processes by which drops acquire and lose charge. All this would take place while the ions move due to (a) the force of the non-uniform electric field and (b) diffusion driven by the non-uniform ion concentrations.

Instead we have used a qualitative model to understand the general behavior of the neutralizer. We made the following assumptions:

- a. The model is one-dimensional with the only important axis being the horizontal axis  $x$  of length  $2w$  extending from one  $V_B$  bias plate to the other, Fig. 2. We set  $x=0$  midway between the plates with  $x = w$  at the positive bias plate and  $x = -w$  at the negative bias plate.
- b. There is only one species of negative ion and one species of positive ions with concentrations  $n_-$  and  $n_+$  ions/m<sup>3</sup> and mobilities  $\mu_-$  and  $\mu_+$  m<sup>2</sup>/Vm.

c. The drops fall about midway between the bias plates and we ignore the effects of diffusion using the following argument. The ion current due to diffusion is given by  $J_D = eD (dn/dx)$  where  $D$  is the coefficient of diffusion. The  $(dn/dx)$  is of order of magnitude  $n/w$  where  $n$  is the maximum ion concentration and  $w$  is one half the spacing between the bias plates [Fig. 2]. The ion current due to the electric field of magnitude,  $E$ , is  $J_E = eE\mu n$ . Then  $J_D/J_E = D/(\mu V_B)$ . Since  $D/\mu$  is of the order of  $10^{-2}$  volts and in our neutralizer  $V_B$  is of order 1 volt, the neglecting of the diffusion current in this qualitative model is justified in most of the neutralization channel. The exception is close to the bias plates.

When the ionized gas is in a steady state the following relations hold. From charge conservation

$$dJ_+/dx = -e(A - Rn_+n_-), \quad dJ_-/dx = e(A - Rn_+n_-) \quad (2)$$

$$J_+ = eE\mu_+ n_+ \quad \text{and} \quad J_- = eE\mu_- n_- \quad (3)$$

where  $J_+$  and  $J_-$  are the currents carried by the + and – ions,  $A$  is the number of ion pairs produced per unit volume per second by the beta rays and  $R$  is the recombination constant.

Also from the Poisson equation

$$dE/dx = (e/\epsilon_0)(n_+ - n_-) \quad (4)$$

Finally, since diffusion is ignored the boundary conditions are  $n_+ = 0$  at  $x = w$  and  $n_- = 0$  at  $x = -w$ . Then once  $V_B$ , the integral of  $E$ , is fixed, the dependence on  $x$  of  $E$ ,  $n_+$  and  $n_-$  is determined.

The determination of the dependence on  $x$  of  $E$ ,  $n_+$  and  $n_-$  is an old problem in the conduction of ions through an ionized gas. For example it is discussed in detail by J. J. Thomson and G. P. Thomson in their 1928 book *Conduction of Electricity Through Gases* [11]. The physics has been studied and discussed extensively because it is the basis of the operation of the continuous plane parallel ionization chamber. But there is no analytic solution and it must be solved numerically. The classic book of Rossi and Staub [12] explain various approximations. In our studies we used the formulation of the problem by Armstrong and Tate [13], a pair of dimensionless equations very convenient for computer calculations.

The treatment of Armstrong and Tate [13] ignores the ion diffusion current. Therefore in our studies we checked that the diffusion current can be neglected under the conditions that we operate the neutralizer.

From Eq. 3 one can see that if  $\mu_+$  is not equal to  $\mu_-$  the distribution of  $n_+$  and  $n_-$  will be asymmetric about  $x=0$  and hence  $n_+$  and  $n_-$  will not be equal at  $x=0$ . Hence it is reasonable

based on Eq. 4 that by adjusting  $V_B$  one can adjust  $n_+$  and  $n_-$  at  $x=0$ , and hence neutralize the drops.

A simple demonstration of this reasoning is obtained by setting the recombination coefficient  $R=0$  in Eq. 2. Then  $J_+$  and  $J_-$  are linear in  $x$  and

$$n_+ = (A/\mu_+ E)(-x + w), \quad n_- = (A/\mu_- E)(+x + w) \quad (5)$$

Note that  $n_+$  and  $-n_-$  are not equal at  $x=0$  if  $\mu_+ \neq \mu_-$ .

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## Figure Captions

Fig. 1. Typical initial charge distribution of Dow Corning 200 silicone oil drops, 24.6  $\mu\text{m}$  in diameter, as produced by our drop generator [6]. The rms width distribution is  $3.2e$ .

Fig. 2. Main features of neutralizer. A  $200\ \mu\text{C}\ ^{90}\text{Sr}$  source emits beta rays into an open air channel through which the drops fall. The main body of the neutralizer is steel. The axis perpendicular to the bias plates is called  $x$ , with  $x = 0$  midway between the plates and  $x = \pm w = \pm 0.20$  in at the plates.

Fig. 3. Schematic showing how we use the neutralizer with our fractional charge search apparatus. The drop generator emits drops into the top of the neutralizer. The drops are at terminal velocity as they fall through the ionized air. The charge measuring part of the search apparatus [6] lies just below the neutralizer and uses a horizontal, alternating electric field to give the drops horizontal, alternating motion, the extent of the motion being proportional to the charge on the drop.

Fig. 4. Two typical *initial* drop charge distributions for 12.5  $\mu\text{m}$  radius drops of Ultraol mineral oil [10] containing a suspension of about 2% powdered meteorite and some surfactant. The distribution in Fig. 4a has a mean charge  $q_m = -731e$  and an rms width  $\sigma_q = 362e$ . The respective values for the distribution in Fig. 4b are  $q_m = -1012e$  and  $\sigma_q = 331e$ .

for the same type and size drops produced at another time. The charge distribution varies with the operation parameters of the drop generator in ways we do not understand, and even for the same operation parameters, the distribution changes slowly with time.

Fig. 5. An example of the altered charge distribution for Ultraol drops with the same size and composition as Fig. 4 when the neutralizer is fully on and the bias voltage,  $V_B$ , is set to 0.25 volts. This bias voltage has been selected to move the mean charge,  $q_m$ , close to zero. The distribution is now much narrower. We find  $\sigma_q$  is  $15.5e$  compared to  $\sigma_q = 14.9e$  given by Eq. 1 for  $r = 12.5 \mu\text{m}$ .

Fig. 6. (a) The dependence of  $q_m$  on  $V_B$  for the same  $12.5 \mu\text{m}$  radius drops of Ultraol mineral oil containing a suspension of about 2% powdered meteorite and some surfactant. (b) The dependence of  $\sigma_q$  on  $V_B$  for the same drops. The former is quite sensitive to  $V_B$ , but the latter is not. In the use of the neutralizer we set  $V_B$  so that  $q_m$  is within a unit of  $e$  of zero. Over several days of operation there may be a drift in  $q_m$  of about one unit of  $e$ , and we then slightly change  $V_B$ . As shown in Fig. 6b this does not change  $\sigma_q$ .

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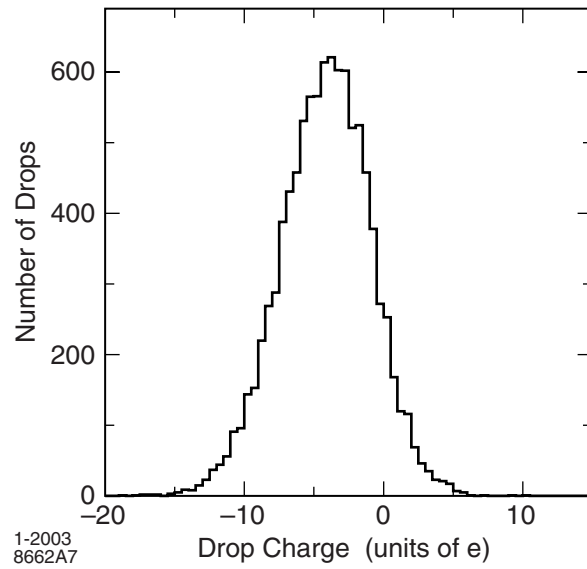


Fig. 1. Typical initial charge distribution of Dow Corning 200 silicone oil drops,  $24.6 \mu\text{m}$  in diameter, as produced by our drop generator [6]. The rms width distribution is  $3.2e$ .

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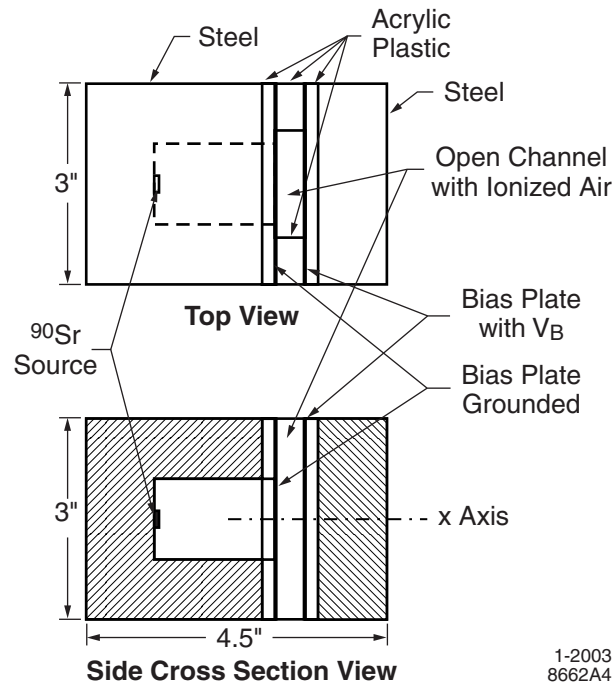


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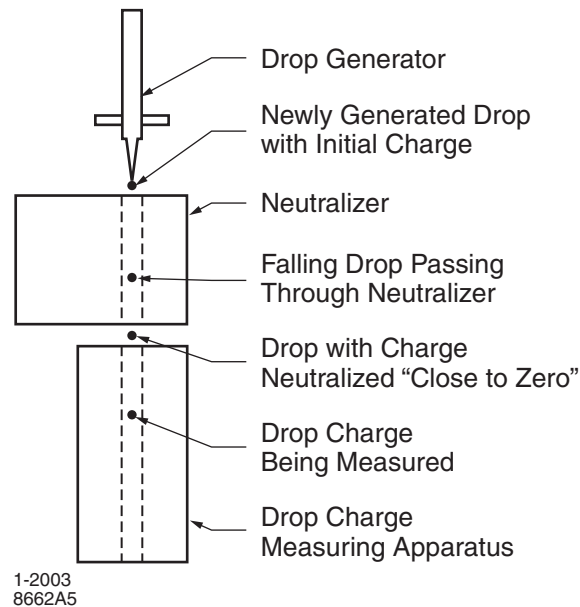


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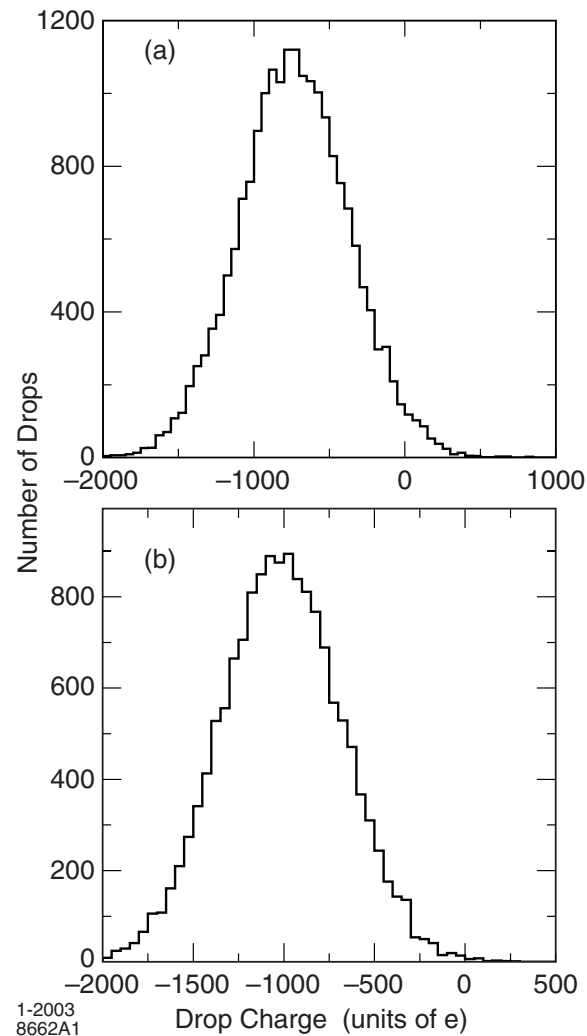


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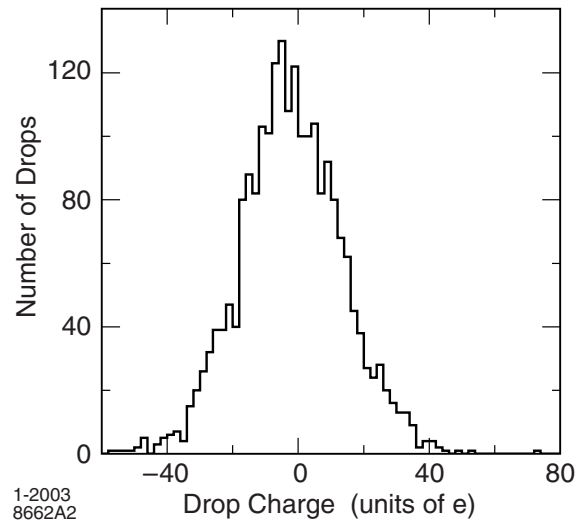


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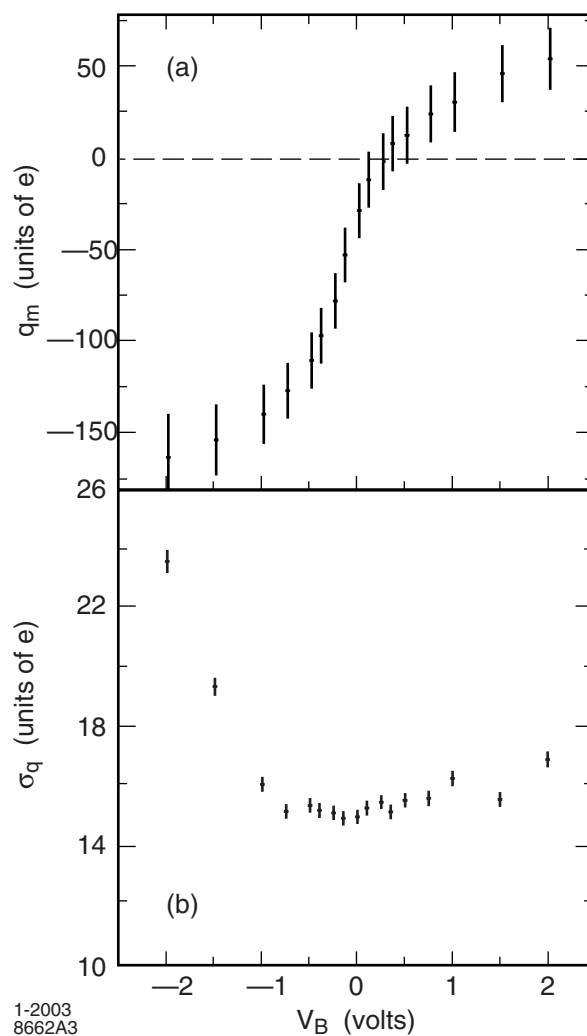


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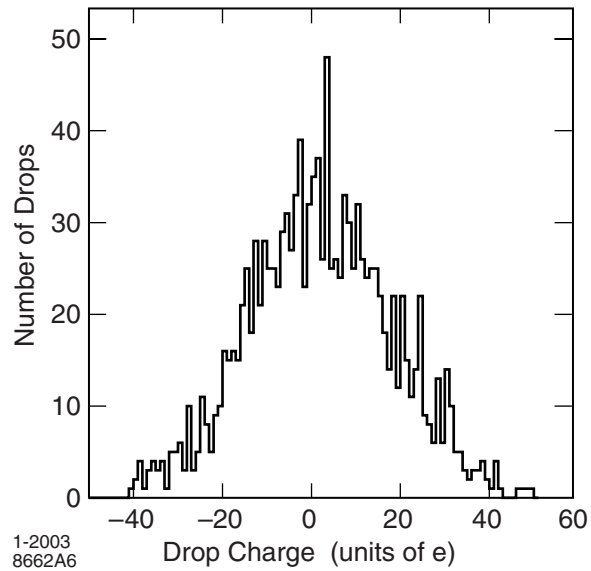


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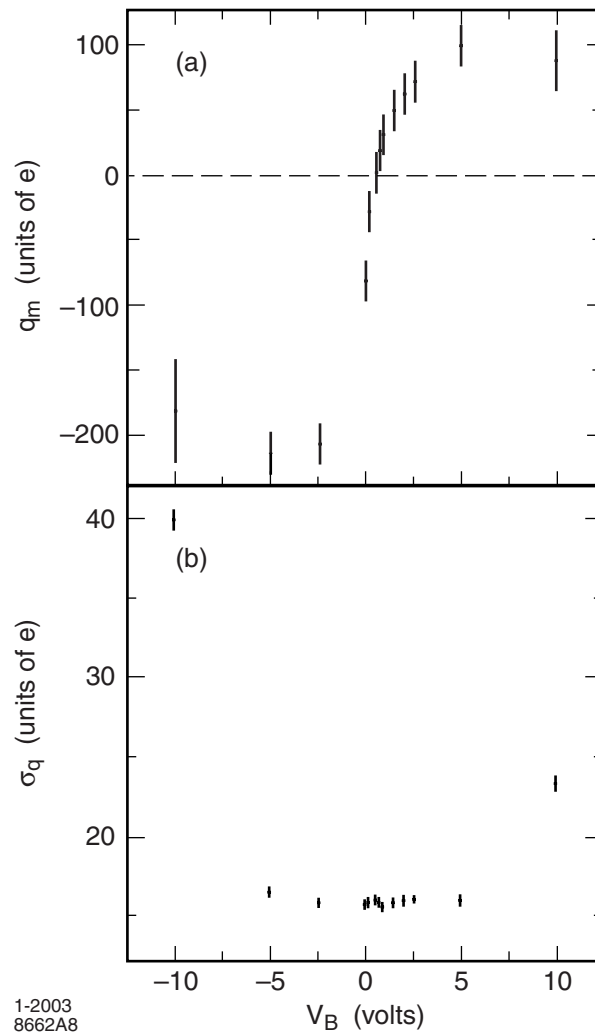


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