Monitoring of charging events on particles in a nonpolar liquid

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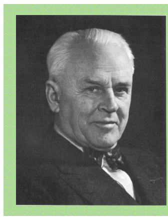
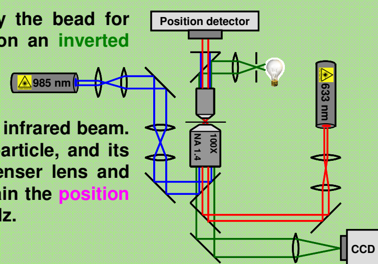


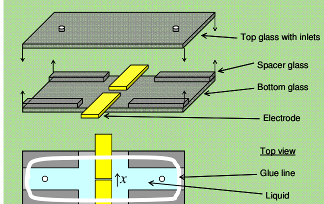
Almost a hundred years ago, Robert Millikan performed his famous experiment on oil-drops in air. He demonstrated the discrete nature of electric charge, and was the first to measure the value of the elementary charge. We performed the same type of experiment on colloidal PMMA particles in a nonpolar liquid. Measuring the elementary charge in a liquid is challenging compared to Millikan's experiment in air, because of the higher viscosity and more rapid fluctuations of the charge.

While Millikan was mainly interested in the value of the electric charge, we want to go further and investigate the statistics of the elementary events, when the particle charge changes with one electron charge. These events are the direct manifestation of the charging and discharging reactions on the surface of the particle, and provide detailed and straightforward information. This information can be used to model the mechanisms responsible for the charge on colloidal particles in a nonpolar liquid.

Experimental setup: optical trapping electrophoresis

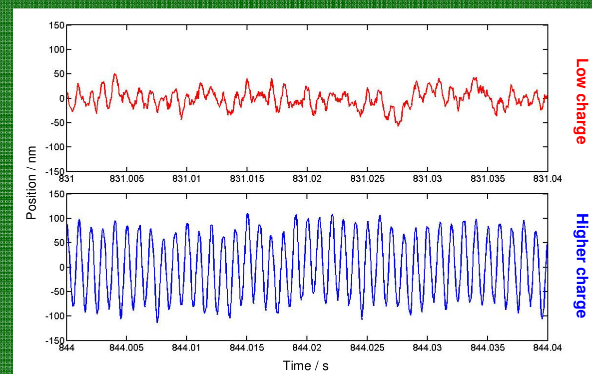
We use optical tweezers to be able to study the bead for thousands of seconds. The setup is based on an inverted microscope with a 100x, 1.4 NA objective lens.
An infrared laser beam is expanded so that it overfills the entrance aperture of the objective.
The bead is attracted towards the focus of the infrared beam.
A second, red laser is also focused on the particle, and its forward scattered light, collected by a condenser lens and imaged on a position detector, is used to obtain the position of the particle at a sampling frequency of 20 kHz.





A mixture of dodecane and 1 µm PMMA beads is inserted into a fluid chamber with two electrodes separated by a distance of 300 µm. One bead is trapped in the middle between these electrodes. We apply a sinusoidally varying voltage with amplitude 1 kV and frequency 1 kHz between the electrodes. The particle then feels a high electric force (necessary for a good resolution), at a high frequency (so that the particle is not pulled out of the trap).

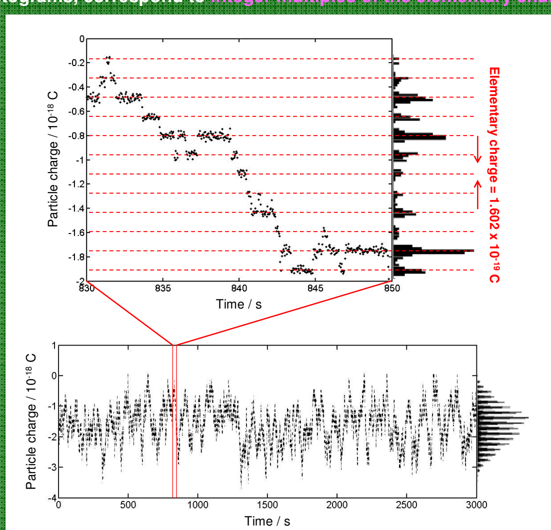
Electrophoretic and Brownian particle motion



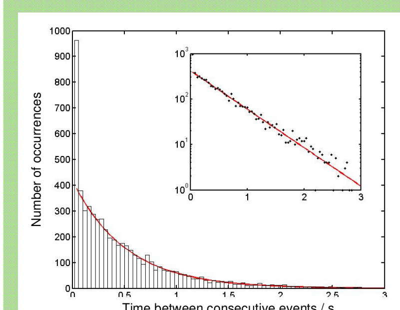
The particle motion nonsists of a sinusoidal electrophoretic contribution (the 'signal' from which we get the charge) and a Brownian contribution (the unavoidable 'noise').

Discrete fluctuations of the particle charge

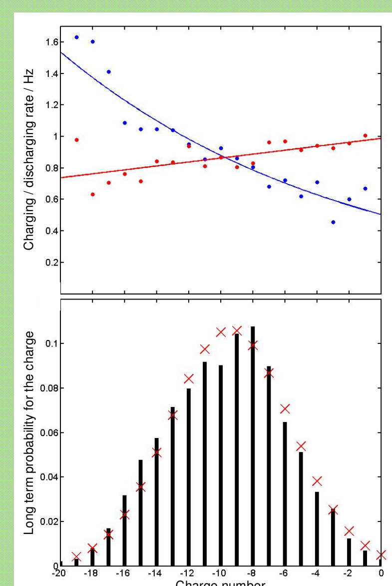
From the amplitude of the motion in the electric field we calculate the charge on the bead, for every 40 ms window. The charge measurements are accurate enough to resolve the elementary charge, fast enough to see the jumps corresponding to a change of one electron charge, and can be performed continuously for 3000 s. The discrete levels in the charge vs. time plots, and the peaks in the histograms, correspond to integer multiples of the elementary charge.



Charging dynamics investigated by elementary event monitoring



We are able to observe the discrete events when the particle charge increases or decreases with one elementary charge, and monitor them during a long enough time to perform a statistical analysis.
As an example, we have plotted the distribution of the times between consecutive events. It can be fitted to an exponential, indicating a Poisson process, in which the events are uncorrelated.



In a more detailed analysis, we investigate the probability (or rate) with which the particle charge increases or decreases with one elementary charge, as a function of the charge number. The results can be compared with a phenomenological model, which explains both the reaction rates and the long term probability distribution for the particle charge.

In conclusion, we have demonstrated the feasability and use of elementary event monitoring to investigate charging processes on solid-liquid interfaces. We were able to measure the charge on a PMMA particle in dodecane, accurately enough to resolve the elementary charge, fast enough to observe jumps from one discrete charge number to another, and during a sufficiently long time to obtain statistically relevant information.

Relevant citations:

1) F. Strubbe et al, 'Detection of elementary Charges on Colloidal Particles', Physical Review Letters 100 (2008) 218301.
2) G. S. Roberts et al, 'Direct measurement of the effective charge in nonpolar suspensions by optical tracking of single particles', Journal of Chemical Physics 126 (2007) 194503.
3) T. A. Wood et al, 'Characterization of microparticles with driven optical tweezers', Faraday Discussions 137 (2008) 319.

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