

A laboratory instrument for the precise, non-destructive determination of the charge and mass of single, well localized and isolated submicron particles under ultra high vacuum conditions is described. A mass resolution of better than  $3 \cdot 10^{-4}$  is reported. Secondary electron emission is monitored online event by event and used to determine the particles absolute charge state. Future applications for the investigation of gas-grain interaction such as determination of sticking coefficients and sublimation temperatures are discussed.

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## Introduction

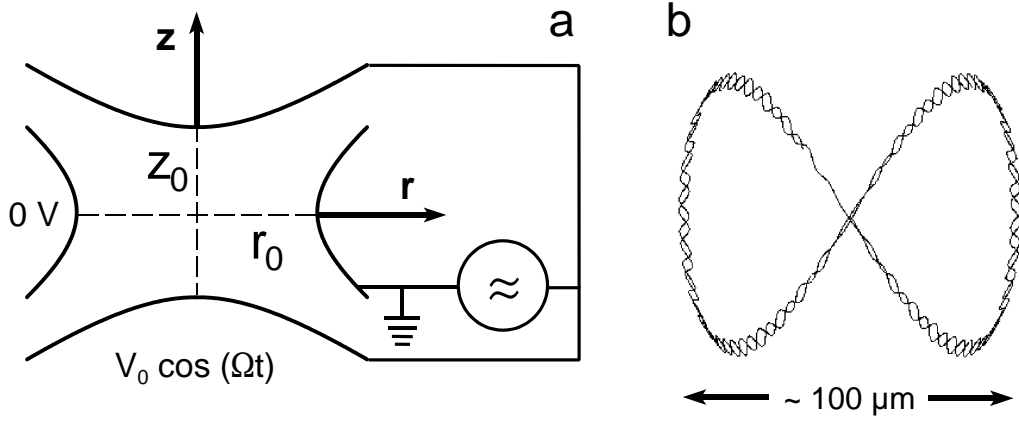
Dust is an important constituent of the interstellar medium, especially in star forming regions (van Dishoek 1998). Its presence has been deduced from the observation of a continuous extinction of starlight from near infrared to far UV. However, a fairly wide range radial size distribution is necessary to account for the observed extinction curve. Besides the extinction bump at 217 nm which is characteristic enough to be attributed to small graphite particles, the nature of the dust material is the matter of a lot of speculation. It has been pointed out by David Williams (Williams 1994) that most interpretations of the simple extinction curve include sufficient parameters that the fitting procedure does not provide serious constraints for any plausible dust model. Many laboratory experiments have been set up in order to simulate the astrophysical observations. For conventional extinction experiments, e.g. FTIR-spectroscopy, large quantities of dust are necessary to obtain a reasonable signal. These dust ensembles usually consist of particles with a fairly large size distribution. Therefore most laboratory work suffers from the same problem as observation, the congestion of spectral information by size and shape variations as well as different possible compositions. A new approach is based on trapping techniques (Schlemmer et al. 1998) which allow to characterize and study single localized objects. In this contribution we describe a trap experiment where a *single, isolated, submicrometer sized particle* can be investigated over periods of days. The particle is very well localized at the center of the trap. Here it is interrogated by the light of a collimated laser beam. Quadrupole trapping and scattered light detection serve as a non-destructive method of accurate mass and charge determination. Details of this method will be described. As a first result we present the detection of individual events of secondary electron emission from a single SiO<sub>2</sub> sphere. The precision of the mass determination and the localization of the trapped particle open up a new class of laboratory experiments on interstellar dust analogous particles which will be discussed in the conclusions.

## Experimental

The principles of electrodynamic trapping have been developed in the 1950's for the use as mass filter for ions. However, quadrupole ion traps have also been used for storage of micrometer sized dust particles (Wuerker et al. 1959) or droplets (Dawson 1976) already from the very beginning. Cooling of the object in a trap with a three-dimensional minimum leads to localization to the center point of the trap and has therefore been most efficiently used for particle trapping with

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\* e-mail: <schlemmer@physik.tu-chemnitz.de>



**Fig. 1:** a) Schematic view of a quadrupole trap. The trapping potential is formed by a high voltage at audio frequency ( $V_0 \cos(\Omega t)$ ) supplied to the cap electrodes. b) Calculated trajectory of a charged particle in the trap (view in  $r$ - $z$  plane).

optical detection. Detection of light absorption of a single atomic ion demonstrated the sensitivity of this method.

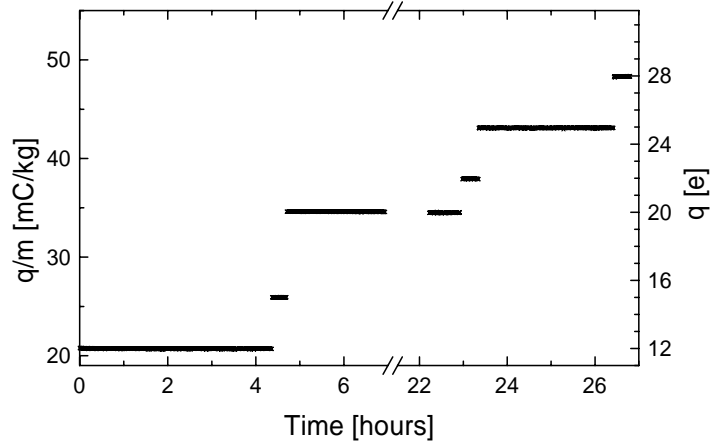
The trap used in the present experiment has been described elsewhere (Schlemmer et al. 1998). Fig. 1a shows a schematic of a quadrupole trap (Paul trap design). It consists of two cap electrodes (top and bottom) and a ring electrode. An alternating voltage ( $V_0 \cos(\Omega t)$ ) applied to the cap electrodes gives rise to the trapping field. Our present trap consists of two cones with axial bored holes. They are opposing each other at a distance of 6.6 mm and act as the top and bottom electrode of the quadrupole trap. The cones are surrounded by eight rods which form a cage like structure. This open design has been chosen in order to obtain a large solid angle for light detection as well as to detect the light at several angles (angular distribution). Moreover the trap volume can be accessed through several ports with additional tools such as the particle source, the laser beam and an electron gun.

The trap is located in a vacuum chamber which is evacuated to UHV pressures prior to trapping. A small oscillating membrane is filled with monodisperse 500 nm diameter  $\text{SiO}_2$  spheres and serves as a particle source. For injection, the membrane vibrates and ejects particles into the trap. Friction due to buffer gas (typically  $10^{-3}$  mbar) is sufficient to dissipate kinetic energy of the particle to be caught in the trapping potential.

Fig. 1b shows a projection of a charged particles trajectory in the  $r$ - $z$  plane of the trap. The particles motion in the trapping potential consists of low frequency oscillations (secular motion,  $\omega_r$  and  $\omega_z$ ) and a high frequency oscillation ( $\Omega$ ) due to the driving voltage. Light from a collimated laser diode (typically  $2.5 \text{ mW/mm}^2$  at 670 nm) is directed near the center of the trap. The residual motion of the trapped particle leads to a modulation of the scattered light. This light is collected by a lens, transferred outside the vacuum in a multimode fibre and transmitted to an avalanche photo diode (APD). A power of up to 4 pW is collected from a detection angle of 0.013 sr for a 500 nm  $\text{SiO}_2$  sphere. The APD-signal is further amplified, filtered and then recorded by an A/D converter. The eigenfrequencies of the secular motion in  $z$ - and  $r$ -direction of the trap are determined from a Fourier Transform of the detected signal. They are proportional to the particles  $q/m$  ratio. For the  $z$ -direction it is given by

$$\omega_z = \frac{\beta_z}{2} \Omega = \frac{\sqrt{2}}{2} \frac{q}{m} \frac{V_0}{\Omega^2 z_0^2} \Omega . \quad (1)$$

Note that  $2z_0$  is the distance of the cap electrodes only for the ideal quadrupole design. In practice



**Fig. 2:** Charging of a single trapped particle using electrons.  $q/m$  has been determined from the eigenfrequency,  $\omega_z/2\pi$ , using Eq. 2. Five steps in  $q/m$  are due to charging in steps of +2 to +5 elementary charges (right axis).

it is a measure of the gradient of the trapping potential. For our trap  $z_0$  has been determined numerically using an ion optics simulation program, (SIMION). The relevant frequency peaks for the motion in  $z$ - or  $r$ -direction of the trap,  $\omega_z/2\pi$  or  $\omega_r/2\pi$ , are recorded every few ten seconds over a period of hours or even days. Due to the inherent precision of the frequency measurement the eigenfrequencies are very sensitive to slight changes in  $q/m$ .

### Charge and Mass Determination

In the following we report the intentional change of  $q$  in steps of elementary charges to determine the absolute net charge of the particle and thus the absolute mass as well. Emission of single photoelectrons upon UV-radiation has been used previously to determine the absolute number of charges of trapped charged particles (Philip et al. 1983). However, the work function for  $\text{SiO}_2$  is too large to use a conventional UV-lamp. Therefore we used an electron gun to charge the particle.

Fig. 2 shows the temporal evolution of a single particles eigenfrequency in  $z$ -direction. Using Eq. 1  $\omega_z$  has been converted to the particles  $q/m$  ratio. The example particle is positively charged, as most of the  $\text{SiO}_2$  particles investigated. This can be checked easily by the polarity of a superimposed DC potential. Upon electron bombardment  $q/m$  raises eventually in steps which can be attributed to an increase in charge in multiple of elementary charges ( $e$ ) due to the emission of secondary electrons. The relative heights of the five observed steps are  $i=3,5,2,3,3$  and correspond to the emission of a charge  $\Delta q = i \cdot Q$  from a particle with an initial net charge of  $+12 Q$ . In further series of measurements with even more charging steps we find values  $i=1-6$ . Also the sticking of an electron,  $\Delta q = -e$ , has been observed, (Schlemmer et al. 1998). Therefore it is safe to assume  $Q=e$  for the experiment shown in Fig. 2 and to use Eq. 1 to determine the particles absolute mass,  $m = 1.8 \cdot 10^{-16}$  kg. This value corresponds to the expected mass of a single 560 nm diameter  $\text{SiO}_2$  sphere. In order to use this instrument for absolute mass determination a calibration with a particle of known mass is necessary. From another series of more than 500 consecutive measurements, in which  $q/m$  was not changed, we determined the precision of the experiment. Details of this measurement are given in (Schlemmer et al. 1998). For a single

13.2 sec measurement a precision of 270 ppm is obtained. This precision is a measure of the reproducibility of the measurement (no change in  $q$  or  $m$ ). Slightly larger systematic deviations of 360 ppm have been determined when  $q$  was increased by 20 %. This is due to deviations from the ideal trapping potential even near the center of the trap. It becomes detectable when lifting the particle upon charging. On the one hand these measurements show that a particles absolute charge state can be determined unambiguously even if the particle carries more than 100 elementary charges. On the other hand a mass determination with a precision of  $\Delta m = 4 \cdot 10^{-20}$  kg and thus submonolayer resolution is now possible.

## Conclusions

The *observation* of spectral features related to gas phase species, dust and ice layers, *modelling* based on the chemical composition and physical environment of gas phase and grain species as well as *laboratory work* make up todays knowledge in astrochemistry. The instrument described in this contribution may be used for various laboratory experiments simulating the gas-grain interaction under astrophysical conditions. First, the charge state  $q$  of grains is of some interest in the interstellar environment. Due to the short penetration depth of electrons in bulk material it is quite clear that the few ten charges on the SiO<sub>2</sub> particles investigated, are localized near the particles surface in clumps of a few positively charged holes. Preliminary results from the current experiment indicate that charge localization as well as charge migration are measurable quantities. Second, using the *non-destructive* mass determination, accretion rates of ambient gases such as CO, H<sub>2</sub>O, etc. can be determined as a function of temperature. Sticking coefficients and sublimation temperatures can be derived from these measurements. Details of the new ISO observations show that depending on the source viewed, gas can be trapped as ice mantles on grains or be present in gasphase, (van Dishoek 1998). However, thermal evaporation (i.e. the sublimation temperature) might depend strongly on the chemical composition of the adsorbate as well as on the kind of binding to the surface, (Sandford & Allamandola 1988). Therefore a quantitative determination of the sublimation temperatures at various chemical conditions is needed. Third, on smaller interstellar dust particles the localization of charges near the surface might put restrictions on the widely accepted basic idea that light species, such as H, H<sub>2</sub>, C, N and O can hop from site to site on a timescale which is much shorter than the evaporation time because atoms and molecules might be trapped at these sites due to the charge induced dipole energy and enhance or reduce reaction rates for the formation of molecules. In summary, the new apparatus allows a large set of experiments on individual, well localized and isolated dust analogue particles.

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