

Extending the Observation Time of Charged Helium Droplets to the Minute Timescale

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We report on the successful trapping of micrometer-sized, multiply-charged helium droplets over periods of several seconds up to a minute—the first observation of trapped charged helium droplets. Within a meter-long multireflection mass spectrometer, the droplets complete thousands of revolutions, opening up new possibilities for precision measurements of these unique systems. Their confinement is limited by collisions with residual gas, which gradually reduce their kinetic energy through evaporation of helium atoms from the droplet and prevent extraction from the trap below a critical value. The rate of energy loss is strongly influenced not only by the residual gas density, but also by the type of species picked up by the droplets. In particular, infrared absorption by water cluster ions inside the droplets enhances energy transfer to the helium matrix, leading to accelerated evaporation of helium atoms.

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Trapping of ions has become a standard method in many different fields of physics and chemistry, including quantum physics [1], high-precision spectroscopy [2], molecular and chemical physics [3], laboratory astrophysics [4], and nanophysics [5]. For this wide range of research topics, various instruments have been developed and utilized, including Penning traps [6,7], ion cyclotron resonance traps [8], radio frequency traps [9], electrostatic traps [10,11], and storage rings [12–14]. These devices are often cooled to cryogenic temperatures [12,13,15–17] in order to isolate ions from the environment and achieve state selectivity. The long interrogation times and the possibility of manipulating and controlling ions has revolutionized molecular and chemical physics, providing unparalleled precision for studying molecular spectroscopy [18] and ion-molecule reactions [19].

Action spectroscopy using messenger tagging with rare-gas ions is used for determining the structure of fragile biomolecular ions [20], as it minimizes fragmentation, or to determine absorption spectra of cold molecular ions [17]. Because of its weak interaction with ions, helium tagging produces the smallest matrix shift and is thus the preferential messenger [21]. Helium tagging of cations has been achieved in cryogenic ion traps [22,23] and via electron ionization of doped helium droplets [24]. Pickup into charged helium droplets and extraction of low-mass helium tagged ions has been demonstrated via shrinking [25] and

splashing [26] of helium droplets upon surface collisions, both for cations and anions.

So far helium nanodroplets (HNDs) formed by pulsed or continuous nozzle expansions into vacuum have typical velocities of a few 100 m/s [27,28] which limits the time until they collide with a wall of the vacuum chamber to a few ms. This short time represents a clear disadvantage compared to ion-storage devices and hampers many experiments that require long data acquisition times. Recently, Brown *et al.* reported magnetic trapping of mm sized neutral helium droplets that were formed upon condensation of helium vapor in the levitation point inside a ⁴He bath cryostat for several hours [29]. The combination of evaporative cooling in ultrahigh vacuum, long measurement time, and large droplet size enables novel experiments with superfluid He droplets, which are, however, limited to neutral species. A few years ago, Laimer *et al.* discovered that large HNDs passing an intense electron beam can become highly charged [30] and can be deflected by relatively moderate voltages applied to ion optical elements such as deflector pairs, electrostatic quadrupole benders [25], or sector fields [31].

In this Letter, we demonstrate that HNDs containing between 10⁶ and 10⁷ He atoms and about 8 × 10⁵ He atoms per charge state can be trapped up to minutes in an electrostatic ion beam trap composed of two opposing ion mirrors based on a multireflection time of flight mass spectrometer [11]. By measuring the number of released HNDs as a function of the gas pressure in the ion trap we show that shrinking of the HNDs upon pickup and cluster formation of residual gas in general and of water molecules in particular due to blackbody infrared radiative dissociation (BIRD) [32] are limiting ion detection at longer storage times.

In our setup (see Fig. 1), HNDs are produced via supersonic expansion of precooled helium gas into vacuum

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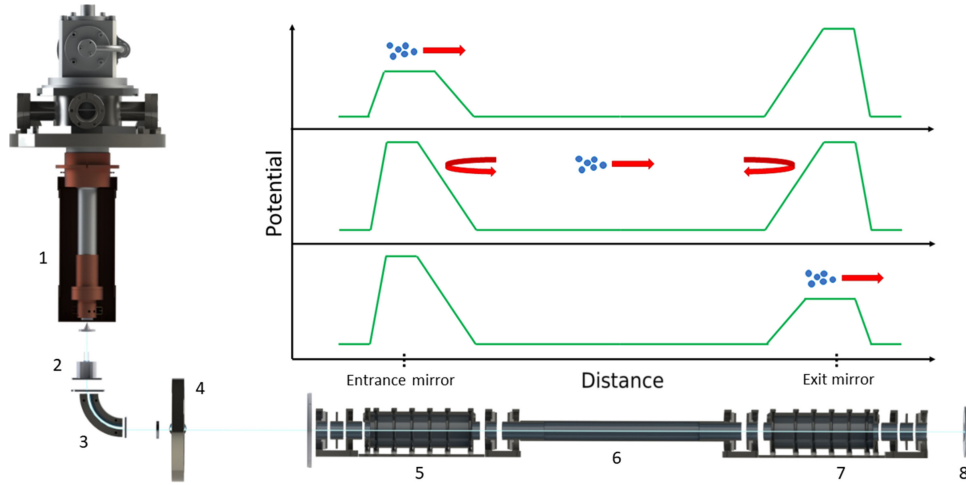


FIG. 1. Cross-sectional view of the setup: (1) closed cycle cryostat, (2) electron impact ion source, (3) electrostatic sector with pickup region, (4) gas barrier for differential pumping, (5) entrance mirror, (6) drift tube, (7) exit mirror, (8) CEM detector.

through a cryogenic nozzle with an approximate diameter of $5\ \mu\text{m}$, under a stagnant pressure of 20 bar. The resulting neutral HNDs are then ionized by electron impact ionization using 90 eV electrons, leading to the formation of multiply-charged cationic droplets. To minimize the energy spread of the charged droplets prior to the injection into the multireflection trap, an electrostatic sector is employed. Once an ensemble with uniform energy per charge state is selected, it is injected into the trap, where it is stored for a defined duration. The length of the stored bunch can be set by the inlet time t_{inlet} , i.e., the duration of potential lowering of the outermost electrode of the entrance ion mirror, discretizing the continuous HND beam to a defined bunch size. After some storage time t_s , the outermost electrode of the exit ion mirror at the far end of the device is switched off, enabling the trapped droplets with sufficient kinetic energy to leave the trap and to be detected by a channel electron multiplier (CEM). Since only the outermost mirror electrode is switched, the remaining potential barrier serves as a discriminator for lower energies. In this Letter, the effective potential as seen by the droplets on the optical axis of the device is lowered from 490 V in the on-state to 350 V in the off-state. This high-pass energy filter allows the monitoring as a function of time of the number of HNDs that have not yet lost the ability to overcome the remaining barrier height—a crucial feature for the experiments described below. In the vacuum chamber housing the electrostatic sector, gas inlets can be used to dope the droplets with molecules of interest, giving rise to a number of novel experiments.

The multireflection time of flight component, i.e., the section from the entrance to the exit ion mirror connected by a drift tube, was developed and produced by a collaboration between the Technical University of Darmstadt and the University of Greifswald [33]. A total of eight devices were distributed among different research groups with a very diverse research interest, highlighting the versatility of this device. Among those, we are the only

one working with clusters in the nano- to micrometer size range. To obtain suitable mirror potentials, first a Monte Carlo simulation was performed, randomly probing the six-dimensional potential space for a configuration with a high transmittance of the device. The best potentials were then further refined using a Nelder-Mead gradient descent algorithm. These potentials served as a starting point for the mirror optimization at the experimental setup. It should be noted that optimizations were only made toward high transmittance at longer storage times and not toward high mass resolution, as mass spectrometry was completely disregarded in the present Letter.

To characterize the storage dynamics of the system, the temporal and spatial distribution of droplets was studied at different storage times by lowering the potential of the exit mirror and measuring the HND signal on the CEM. Monitoring these signals as a function of storage time leads to a temporally resolved picture of the droplets over the whole storage duration. Figure 2 shows several of such series of measurements taken at different pressures inside the multireflection trap. Each peak corresponds to a single revolution of the droplet bunch inside the device, with peaks at longer storage times exhibiting a significant broadening due to the finite width of both the initial energy and velocity distributions of the droplet bunch. At the ion mirrors, the droplets are subject to large electrostatic forces acting only onto the charge centers, which may, in principle, “stir up” the droplets and thus lead to additional energy deposition and subsequent mass loss and signal decay. However, the measurements indicate that currently the background pressure in the vacuum is the dominating factor limiting the storage period: Fig. 2 shows a very clear correlation between an increase in background pressure and a faster decay of droplet intensities. Simple calculations show that the mean-free path $\lambda = k_B T / (\sqrt{2} \pi r_{\text{HND}}^2 p)$ of HNDs even in ultrahigh vacuum is in the range of only a few tens of cm ($\lambda = 40\ \text{cm}$ for a droplet with

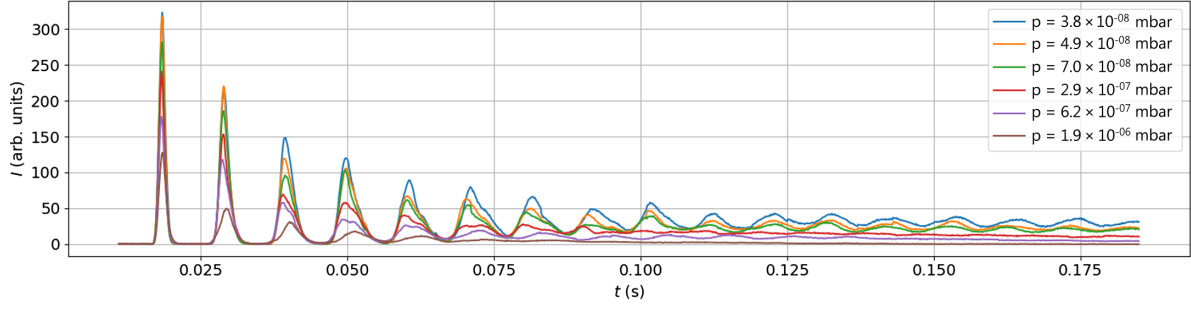


FIG. 2. Storage characteristics of the device for different background pressures. The graph shows the temporal distribution of the droplets inside the trap over the course of several hundred ms.

$N_{\text{He}} = 5 \times 10^6$ helium atoms and a radius of $r_{\text{HND}} \approx 40$ nm at a pressure of $p = 6 \times 10^{-9}$ hPa and ambient temperature T). In the context of our application, this corresponds to tens of thousands of collisions during a storage time of a few seconds or an equivalent distance of a few km traveled by the droplets inside the device. While there is almost no loss of kinetic energy due to momentum transfer in the process of collisions with the background He atoms (a head-on collision leads to a change in velocity of $\Delta v_{\text{HND}} \approx v_{\text{He}}/N_{\text{He}} \approx 10^{-4}$ m/s), each of these collisions leads to evaporation of helium from the droplet and a concomitant loss of kinetic energy due to the loss of mass in that process. Since only the outermost mirror of the reflection mirror stack is switched, droplets with insufficient energy to overcome the remaining barrier cannot be detected.

To extend the storage, i.e., detection time, an effort was made to improve the vacuum conditions by implementation of multiple differentially pumped stages. This improved the vacuum significantly, to a point where our cold-cathode pressure gauges are no longer able to provide a reliable reading. This pressure reduction led to a substantial extension of the signal decay time to about 13 s, as shown in Fig. 3, with individual droplets surviving for more than a minute.

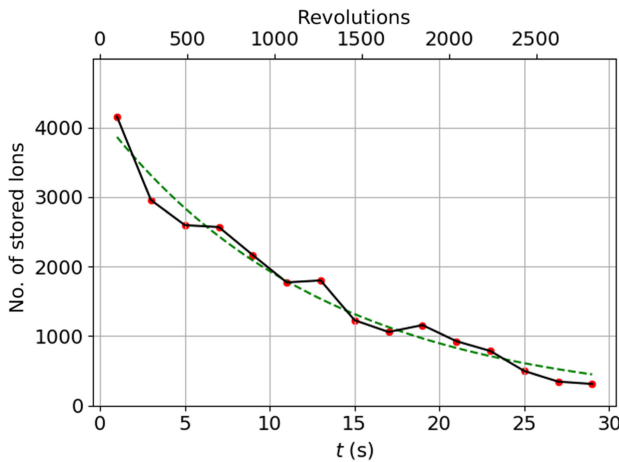


FIG. 3. Long-time storage scan of the droplet signal. The exponential fit (green line) yields a signal decay time of 13(1) s.

With such long storage times achieved, it becomes possible to investigate additional factors that may limit confinement. One such factor is the absorption of black-body radiation by dopants embedded in HNDs. BIRD is a well-known method used in spectroscopy for the probing of the vibrational transitions of molecules or clusters [32,34]. At the presented setup, a series of measurements on HNDs doped with water clusters was conducted, underlining the flexibility of the device for different experimental applications.

To this end, water molecules are picked up by the droplets in the sector chamber (see Fig. 1) and form cationic water clusters at the charge centers of the droplet. The mean water-cluster size of about 25 molecules is set by the appropriate partial pressure of water in the pickup section. Infrared blackbody radiation emitted by the room-temperature vacuum chamber is absorbed by the dopants during the storage time and leads to subsequent evaporation of He atoms and thus a reduction of kinetic energy to the point that the ions can no longer leave the trap due to the remaining Coulomb barrier. Figure 4 shows the effect on

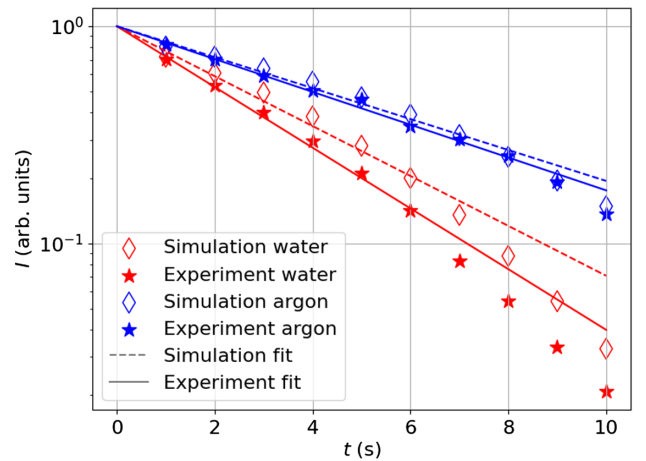


FIG. 4. Comparison of experiment (solid symbols, solid line) and simulation (hollow symbols, dashed line). The data show a significant decrease of the signal decay time for water doped droplets (red) compared to the reference measurement doped with argon (blue).

the detected ion signal as a function of storage time. There is a reduction of the signal decay time by a factor of ~ 1.85 when water clusters are present compared to a reference measurement, where droplets were doped with argon at the same pickup pressure. Since the argon clusters do not absorb blackbody radiation at room temperature, this measurement provides a baseline for the signal decay time under the present conditions.

To model the signal as a function of storage time, the energy loss of each droplet is considered as a function of time, while only droplets with enough energy remaining to overcome the mirror barrier survive and contribute to the signal. For the simulation, several approximations are made. First, the neutral HND sizes are randomly sampled from an exponential size distribution [35]. For the electron-impact ionization, a binomial distribution is assumed with the probability $p_{\text{Ion}} = r_{\text{HND}}^2/r_{\text{Beam}}^2$ of an electron hitting and ionizing the droplet and $N = 2Ir_{\text{Beam}}/(v_{\text{HND}}e)$ the number of electrons with elementary charge e emitted from the electron source while the droplet with radius r_{HND} and velocity v_{HND} is passing the electron beam with radius r_{Beam} , carrying a current of I . The droplet radius can be calculated via $r_{\text{HND}} = 0.22 \sqrt[3]{N_{\text{He}}} \text{ nm}$ [36]. Since N is usually on the order of 10^9 , the distribution can be well approximated by a Gaussian with mean $\mu = Np_{\text{Ion}}$ and $\sigma = \sqrt{Np_{\text{Ion}}(1 - p_{\text{Ion}})}$. The charge state is then discretized by rounding to the nearest integer z_{HND} , while ignoring negative values.

Since the thermal dopant molecules in the sector chamber are much faster than the droplets ($\bar{v}_{\text{H}_2\text{O}} \approx 600 \text{ m/s}$, $v_{\text{HND}} \approx 147 \text{ m/s}$), the pickup of dopants can be approximately modeled via the kinetic theory of gases with the HNDs at rest. The particle flux at pressure p and temperature T for a particle of mass m is given by $\Phi = p(2\pi k_B T m)^{-1/2}$. A total number of dopant molecules $N_{\text{dopant}} = 4r_{\text{HND}}^2 \pi \Phi s_{\text{dope}}/v_{\text{HND}}$ is picked up through the surface of the HND along the doping region of length s_{dope} , which is assumed to evenly distribute among the charges of the droplet with charge state z_{HND} . The first water molecule to reach a charge center is ionized by charge transfer, and further neutral water molecules that are picked up attach to these condensation nuclei and form clusters with a mean size of $N_{\text{cluster}} = N_{\text{dopant}}/z_{\text{HND}} \approx 25$. Since the water clusters are singly charged, z_{HND} also denotes the number of clusters inside the droplet. Using the evaporation rate of water clusters at room temperature $k_{\text{evap}} = 0.204 (N_{\text{cluster}} - 1.387) \text{ s}^{-1}$ [34] (this value might change for sub-Kelvin water clusters since for warmer clusters, more energy loss channels are available, leading to a systematic underestimation of the BIRD effect in the simulation) and the binding energy of a water molecule to the cluster $E_{\text{Bind,H}_2\text{O}} \approx 11 \text{ kJ/mol}$ [37] (value taken for neutral water clusters due to a lack of data for cationic water clusters) at the estimated cluster size of ca. 25 molecules,

the rate of energy deposition into the droplet can be expressed as $(dE/dt) = z_{\text{HND}} E_{\text{Bind,H}_2\text{O}} k_{\text{evap}}$.

To model the loss of helium atoms due to the collision of the HNDs with the background gas, which is presumed to consist primarily of He atoms, again the flux of background gas atoms through the surface of the droplet is considered. The energy deposition rate is then calculated via the mean energy of the atoms $(dE/dt) = \Phi \cdot 4r_{\text{HND}}^2 \pi \cdot 2k_B T$. This set of differential equations is then simply propagated using the Euler method.

After each time step, the charge state of the droplet is compared to the critical charge $z_{\text{crit}} = 0.026 r_{\text{HND}}^2 = 1.26 \times 10^{-3} N_{\text{He}}^{2/3}$ [30,36]. If the charge state of the droplet z exceeds z_{crit} , charges are emitted from the droplet until $z < z_{\text{crit}}$. Since our pressure gauges do not provide a reliable reading of the background pressure in the setup, it was approximated to align the simulation to the reference measurement. Such an alignment was also performed for the droplet size because it depends heavily on the nozzle diameter, which is subject to some uncertainty.

Figure 4 shows that despite the simple model, the simulation reproduces the experimental data qualitatively with a decreased signal decay time when BIRD effects are included, while systematically underestimating the effect of BIRD. The simulation suggests a ratio of $\tau_{\text{Ar}}/\tau_{\text{H}_2\text{O}} = 1.60(14)$ compared to the experimentally obtained value of $1.85(10)$. The uncertainties of both the simulation and the experiment are taken directly from the fit of the exponential decay.

The ability to trap multiply-charged HNDs for tens of seconds up to minutes represents a significant advance in the experimental control and manipulation of these cryogenic nanolaboratories. The techniques presented in this Letter open the door to future measurements that can provide unprecedented insights into the dynamics of HNDs and enable entirely new experimental approaches. Because of the modular structure of the trap, further upgrades can be implemented without redesigning the entire setup. A particularly promising next step is the integration of a pickup electrode suitable for charge detection mass spectrometry [38], which would allow simultaneous measurement of mass, charge, and their evolution over time for individual HNDs. Such capabilities would offer novel insights into the temporal evolution of both pristine and doped HNDs, in analogy to recent studies of aqueous nanodrops [39], while simultaneously providing opportunities for nanocalorimetric studies of reactions occurring inside the HNDs. The (almost) linear trajectories of the droplets make this setup an ideal candidate for laser spectroscopy, enabling time resolved measurements like pump-probe or fluorescence investigations of doped helium droplets as well as studies of decay processes [40,41].

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Data availability—The data that support the findings of this article are openly available [42].

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