Spontaneous decay of hot Ag_n^- clusters in a cryogenic environment

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Synopsis The Double ElectroStatic Ion Ring ExpEriment (DESIREE) has been used to observe the rate of spontaneous neutral particles produced from internally hot small silver cluster anions, $Ag_n^-, n = 2-7$. DESIREE's operational conditions provide long storage times into the hour regime, enabling measurements of the spontaneous decay of anions over much longer time scales than possible in room temperature experiments [1, 2].

The Double ElectroStatic Ion Ring Experiment (DESIREE) provides excellent conditions for the observation of processes occurring on very long time scales [3]. A cryogenic operational temperature of 13 K and residual gas pressure of $\sim 10^{-14}$ mbar provide storage times of tens of minutes. Neutral particle emission rates from small silver clusters were measured using the symmetric ion storage ring in DESIREE [1]. A caesium sputter ion source was used to produce a beam of Ag_n^- ions with a broad distribution of excitation energies. The ions are accelerated to 10 keV, mass selected, injected into the storage ring and stored. The measured storage lifetime for Ag^- was ~ 1600 s. The number of neutral particles produced as a function of time was measured by an MCP detector assembly located in-line with one of the straight sections of the ring, see Fig. 1.

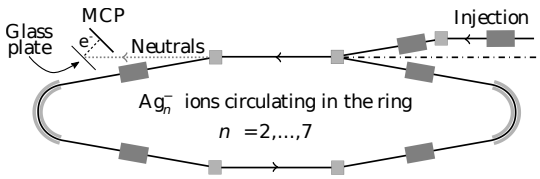

Figure 1. Schematic of the ion storage ring.

The stored, and sufficiently excited, cluster ions can decay via several pathways, including autodetachment ($Ag_n^- \rightarrow Ag_n + e^-$) and fragmentation yielding one neutral (or more) and one charged fragment ($Ag_n^- \rightarrow Ag_{n-m}^- + Ag_m$, $m = 1,\ldots,n-1$). These neutral products are detected as they leave the ring. For an ensemble of anionic clusters with a broad internal energy distribution we expect the neutral particle emission rate to follow a power-law, t^{-p} where $p \approx 1$, when other types of relaxation processes are ignored. However, the ions may also undergo radiative cooling where a photon is emitted due to electronic, vibrational and rotational transitions. For sufficiently large photon-emission energies, these ions may no longer undergo fragmentation or autodetachment. This results in quenching of the neutral particle decay rate, which is then better described by, $t^{-p}e^{-t/\tau}$ where τ is the characteristic photon emission time.

Room temperature studies of small silver clusters ($Ag_n^-, n = 4-9$) have been made using the storage ring ELISA. Short characteristic times for photon emission of less than 20 ms were reported for the n =4,6 and 8 clusters [4]. For Ag_5^- no radiative cooling was observed for storage times up to 50 ms; observation of the decay at later times was not possible due to collisions with the residual gas in ELISA. The cryogenic operation of DESIREE allows us to observe the decay of the Ag_n^- clusters to much longer times. In Fig 2. we show the neutral particle emission rate of Ag_5^-. At early times the neutral particle emission follows a power law, the characteristic photon emission time was found to be 350 ms, and at ~ 1.5 s the signal reaches the collisional residual gas background.

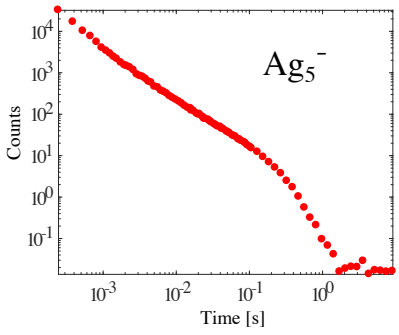

Figure 2. Rate of neutral particle emission from a stored Ag_5^- beam as a function of time after injection.

References

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