The role of angular momentum in the spontaneous decay of small copper cluster anions measured on long timescales at DESIREE

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Synopsis The spontaneous decay of stored beams of hot copper cluster anions has a complex time structure due to the presence of multiple distinct populations in the beam with very different angular momenta.

We have measured spontaneous decay rates for small copper cluster anions (Cu_n^-, n=3−6) stored in one of the DESIREE (Double ElectroStatic Ion Ring ExpEriments) storage rings [1]. The Cu_n^- ions are produced in a cesium sputter ion source and accelerated to 10 keV/q; a bending magnet is used to select the desired mass per charge before injection into the ring. Neutral particles produced (by electron detachment, unimolecular dissociation or background gas collisions) in one of the straight sections leave the ring and are counted as a function of time t after production. The excellent vacuum conditions in DESIREE permit this spontaneous decay to be observed over long timescales.

As the clusters are produced with a broad range of excitation energies, the measured decay rate follows a power law t^{-p} where p is close to unity [2]. Radiative cooling of the clusters quenches the spontaneous decay after a characteristic time T. Thus the total decay rate R(t) is proportional to $t^{-p}e^{-t/T}$.

For n=4−6, we find that the decay rates are well-described by sums of two such quenched power laws (see Figure 1 and Table 1). Similar results were obtained independently by another group using a cryogenic ion beam trap [3]. Here, we relate these two-component behaviors to the presence of stored ions with higher and lower angular momenta J. We have shown through Density Functional Theory calculations that the ground state geometries of the clusters differ in the high- and low-J regimes. For example, the lowest-energy structure of Cu_4^- is linear at high J and rhomboid at low J. As angular momentum is a conserved quantity, there is no interconversion between these populations despite rather high internal excitation energies.

A single-component decay curve was observed for Cu_3^-, but with an unusual shape that could not be fully reproduced by a direct numerical calculation of the decay rates.

Table 1. Parameters describing decay rates of Cu_n^-

n	p	T (ms)	p	T (ms)
4	1.1	2.6	0.75	830
5	1.2	7.5	1.15	180
6	1.28	5.9	0.3	330

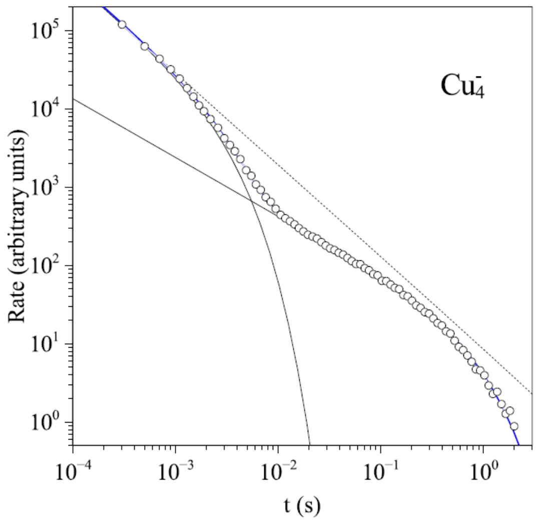

Figure 1. Measured decay rate for Cu_4^- showing two-component behavior. The full black lines are individual fits to $t^{-p}e^{-t/T}$ with values of p and T from Table 1. The dashed line is an extrapolation of the short-time power-law decay rate.

References

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