Investigation of radiative cooling of small metal cluster anions by laser-induced electron detachment

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Radiative cooling is a fundamental process that determines the internal temperature of vibrationally excited ions as a function of time, eventually bringing them into thermal equilibrium with their environment. We have investigated the cooling of Cu_n^- (n=4...7) and Co_n^- (n=3, 4) anions. After production with high vibrational excitation in a Cs sputter ion source the clusters were size-selected and stored in the Cryogenic Trap for Fast ion beams (CTF) [1], at a kinetic energy of 6 keV. This electrostatic ion beam trap was kept at a temperature below 15 K by a closed-cycle helium refrigeration system. The extremely low pressure (few 10^{-12} mbar) achieved by cryopumping of residual hydrogen resulted in a very low background of collision-induced charge exchange reactions and thus a long beam lifetime of several minutes.

The stored ion beam was crossed with the beam of a pulsed Nd:YAG laser emitting at 1064 nm. A small fraction of the stored clusters was thus photoexcited above the detachment threshold to study vibrational autodetachment (also called delayed detachment). The rate for this process persisted for several ms after each laser pulse. It approximately follows a power law in time, with a slope that depends on the population of rotational and vibrational levels in the beam just before excitation. It can therefore be used to detect radiative cooling of the stored ions [2].

We have measured the rate of neutral particles emerging from the trap as a function of the time after each laser pulse, with pulses applied every 20 ms for up to 60 s after ion injection into the trap. Events from delayed reactions were distinguished from direct (vertical) photo-detachment by their characteristic power-law dependence. For most ion species the exponent of this power law changed over the first few seconds of storage time, indicating the cooling of the ions' vibrational excitation. However, for Cu_4^- and Cu_5^- no such change in the exponent was observed. Moreover, although their electron affinities [3,4] exceed the photon energy, delayed events were observed for all ions even after 60 s of storage, with the exception of Co_4^- where the signal disappeared after 1 s. As the ions' vibrational energy is expected to have diminished sufficiently after 60 s to prevent electron detachment, the observations suggest that further energetic aspects (e.g. rotational excitation) or decay pathways such as delayed dissociation (which has an experimental signature very similar to detachment) have to be taken into account in the analysis.

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References:
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