TIME-RESOLVED PHOTOELECTRON IMAGING OF INTRABAND RELAXATION DYNAMICS IN MERCURY CLUSTER ANIONS

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Finite cluster studies permit size-incremental investigation of material properties – both structural and dynamical – evolving with increased aggregation. Clusters of divalent metals (e.g., Hg, Mg) exhibit van der Waals, covalent, and metallic bonding character within various size regimes, and thus offer a set of systems ripe for studying the changes induced through aggregation on the nature and timescale of electronic relaxation processes. We have applied time-resolved photoelectron imaging (TRPEI) to the intraband relaxation dynamics of mercury cluster anions following excitation of the excess electron below the detachment threshold at 1.0 and 1.5 eV. Relatively long timescales (2-30 ps) for relaxation of initially prepared nonthermal distributions are measured for both excitation schemes and for all cluster sizes probed (n=7-18); these observations are in stark contrast to ultrafast relaxation timescales measured in similar experiments on small transition metal cluste rs.[a] Faster relaxation rates are observed for excitation at lower energy, while the longest relaxation times observed with both schemes occur when clusters are excited just below threshold. This latter effect has been investigated on like-sized clusters (n=10-12) excited close to threshold. Furthermore, we observe subtle wavepacket oscillations early in the dynamics for smallest cluster sizes (n=6-8). All observations have been applied towards elucidating the nature of these clusters near the van der Waals-to-covalent bonding-type transition.

[a] N. Pontius, P. S. Bechthold, M. Neeb, and W. Eberhardt, Phys. Rev. Lett. 74, 1132 (2000)