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Ultrafast processes in metal clusters

L. WÖSTE

Freie Universität Berlin, Institut für Experimentalphysik, Arnimallee 14, 1000 Berlin 33, Germany

Metal clusters exhibit extraordinary chemical and catalytical properties. These features arise from the fact that with the transition from the atom to the bulk the electronic and dynamic structure drastically changes. This causes significant alterations of the energetic behaviour of the clusters, going from single electronic transition lines to more and more complex features, finally leading to band structures. The phenomenon is easily observable by means of laser spectroscopy. The spectroscopy of metal clusters on the other hand also exhibits the origin of the dynamical properties. In the case of alkalines for example, resonant two photon ionization is the best fitted method to obtain mass selective excitation spectra up to the trimer. Beyond that, however, no spectra were observed by this method. In the process of laser excitation larger particles tend to fragmentize. This is due to an increased amount of energetically very similar electronic states (bunching), and vibronic coupling. Only with the help of depletion spectroscopy spectra of these particles have become observable.

In quantum chemistry structure and electronic properties are commonly regarded as independent features. The symmetry of a system, however, has a drastic influence on the density of states. As a consequence the speed of fragmentation should increase with cluster size; except for cases, whenever a species of higher symmetry is examined. The observation of the phenomenon requires time-resolved spectroscopy. We applied transient two-photon-ionization spectroscopy in the pico- and femtosecond regime to alkaline clusters with a tunable delay between excitation and ionization (pump & probe). As expected a wavepacket introduced into a dimer oscillates for ever ($< 10^{-8}$ sec). The trimer already exhibits an attenuated oscillation about 5psec. With growing cluster size ($< \text{Na}_9$) this time continuously decreases leading to ultrafast unimolecular decay. The data provide an insight into vibronic coupling, which constantly increases with state density. This phenomenon is of highest relevance for photochemical and catalytic applications.