MO 20: Resonant Energy Transfer and Interatomic Coulombic Decay I

Time: Thursday 14:00–15:45 Location: PA 1.150

Invited Talk MO 20.1 Thu 14:00 PA 1.150

Energy and charge transfer processes in helium nanodroplets — •Marcel Mudrich — Department of Physics and Astronomy, Aarhus University, Denmark

Currently the focus is shifting towards ICD and related interatomic decay processes occurring in more complex media such as biomolecules and condensed phase systems. In this respect, pure and doped helium nanodroplets offer interesting perspectives: They have a homogenous, superfluid density, and they are able to embed, aggregate, and cool atoms and molecules, while their constituents are simple two-electron atoms. Thus helium droplets are ideal for tailoring all kinds of unsupported hetero-complexes.

I will report on our recent experiments probing ICD, ETMD, and related correlated decay processes in pure and doped helium nanodroplets initialed by cw or pulsed EUV radiation. In particular, we find that multiple ionization of embedded species can be either facilitated or hampered by the surrounding helium nanodroplets.

MO 20.2 Thu 14:30 PA 1.150

Superexchange Interatomic Coulombic Decay: a bridged assisted mechanism — •Nicolas Sisourat[1], Tsveta Miteva[1], Sévan Kazandjian[1], Petra Votavová[2], and Přemysl Kolorenč[2] — [1]Sorbonne Universités, UPMC Univ Paris 06, UMR 7614, Laboratoire de Chimie Physique Matière et Rayonnement, F-75005 Paris, France — [2]Charles University, Faculty of Mathematics and Physics, Institute of Theoretical Physics, V Holešovičkách 2, 180 00 Prague, Czech Republic

Interatomic Coulombic Decay (ICD) is an ultrafast energy transfer process. Via ICD, an excited atom can transfer its excess energy to a neighboring atom which is thus ionized.

The rates of ICD depend on the distance between the interacting species. At large interatomic distances R, the process can be viewed as an exchange of a virtual photon between the interacting species. In this so-called virtual photon exchange mechanism the decay rates display a $1/R^6$ dependence.

We recently demonstrated that the ICD rates are substantially enhanced in the presence of an ICD inactive atom, i.e. an atom whose ionization potential is greater than the excess energy of the excited species. This enhancement occurs due to coupling of the resonance state to intermediate virtual states of the bridge atom.

During the talk, I will present the Fano-Stieltjes method we employed for computing the ICD rates. Accuracy of the method will be shortly discussed. Finally, some examples illustrating this novel mechanism will be reported.

MO 20.3 Thu 14:45 PA 1.150

Light induced inter-Coulombic decay in quantum dots controled via laser focus, field strength, and polarization — •Anika Haller and Annika Bande — Institute of Methods for Material Development, Helmholtz-Zentrum Berlin für Materialien und Energie, Albert-Einstein-Str. 15, 12489 Berlin, Germany

The inter-Coulombic decay (ICD), an ultrafast energy transfer process induced by long-range electron correlation, has been shown to take place in pairs of atom-like nanostructures, namely quantum dots (QD). We theoretically investigate the electron dynamics for a system of two singly-charged non-coupled GaAs QDs. After resonant excitation by a laser pulse the system efficiently decays from its resonance excited state via ICD: inter-QD energy transfer causes excitation of one electron from one QD into the continuum, while the other relaxes to a lower bound state of the other QD. We show the field strength dependence of the ICD initiation including a changing number of Rabi oscillations during population. In addition, we study the impact of the laser focus on the second, direct ionization channel. Furthermore, we analyze the effect of electric field polarization on the efficiency of ICD, and thus open up the applicability in a further group of material systems. A space-resolved wavefunction ansatz is applied by means of the antisymmetrized multiconfiguration time-dependent Hartree method (MCTDH). A state-resolved representation has been developed as an alternative approach, which lets us conclude on the importance of multi-photon processes.

MO 20.4 Thu 15:00 PA 1.150

Calculating Interatomic Coulombic Decay Rates from Atomic Data: A case study — •Severin Bang[1], Robert Bennett[1], and Stefan Yoshi Buhmann[1,2] — [1]University of Freiburg, Germany — [2]Freiburg Institute for Advanced Studies (FRIAS), Freiburg, Germany

Interatomic Coulombic decay is an ultra-fast decay process by which energy can be transported between molecules. There are two main approaches to the calculation of the ICD rate, namely ab initio quantum chemistry simulations, or in terms of atomic transitions with their associated photon emission and absorption. In this talk, I will discuss the intricacies of the process by which ICD rates can be calculated from atomic line data. The whole ICD process begins with a donor species being in a excited state, followed by a relaxation and the corresponding photon emission, which in turn ionises an acceptor species.

The ultimate aim of this work is to critically evaluate the ability of currently available spectral line data to give reliable predictions for ICD. Using data from the NIST spectral line database, we will show an example calculation for a neon-argon cluster. We will also point out cases where the data necessary for such investigations is incomplete. For some transitions, no dipole moments are available, and the photoionization cross-section data is very sparse. Finally, we compare our asymptotic ICD rates with those from ab initio approaches, noting that the latter experience difficulties when taking the large-separation limit of initially composite systems.

MO 20.5 Thu 15:15 PA 1.150

Ultrafast proton migration in water and ice — •Clemens Richter[1], Clara Saak[2], Melanie Mucke[2], Isaak Unger[2], Minna Patanen[3], Torsten Leitner[4], Ieva Bidermane[4], Caspar Lant[1,4,5], Olle Björneholm[2], and Uwe Hergenhahn[1,6] — [1]Leibniz-Institut für Oberflächenmodifizierung, Leipzig — [2]Uppsala University, Sweden — [3]University of Oulu, Finland — [4]Helmholtz-Zentrum Berlin — [5]New York University, NY USA — [6]Max-Planck-Institut für Plasmaphysik, Greifswald

Inner-shell ionization of water leads not only to a K-shell decay, but also to a substantial nuclear rearrangement. A spectral signature for ultrafast proton transfer has been seen in Auger and X-Ray spectra of liquid water. Detailed theoretical modelling has revealed the role of Intermolecular Coulombic Decay (ICD) and Electron Transfer Mediated Decay (ETMD) in these spectra. An analogous feature we now have observed in KVV Auger spectra of amorphous ice. To minimize charging and radiation damage, we have employed an angle-resolving time-of-flight spectrometer offering a large collection angle for electrons together with good energy resolution at Auger kinetic energies.

In a second set of experiments, we intended to use the proton transfer signature ('proton transfer mediated charge separation', PTMCS) as a monitor for hydrogen bond strength in a variety of systems. Towards this end, we have recorded the K-shell autoionization spectra of water clusters in comparison with methanol clusters, which are known to have a less strong hydrogen network. Several cluster sizes were probed. Differences between the two species will be discussed.

MO 20.6 Thu 15:30 PA 1.150

Recent Progress in the Theoretical Description of the Inter-Particle Coulombic Decay in Paired Metal Nanoparticle - Quantum Dot Systems — •Matthias Berg[1], Aliezer Martínez-Mesa[2], Llinersy Uranga-Piña[2], and Annika Bande[1] — [1]Helmholtz-Zentrum Berlin für Materialien und Energie, Hahn-Meitner Platz 1, 14109 Berlin, Germany — [2]DynAMoS (Dynamical processes in Atomic and Molecular Systems), Faculty of Physics, University of Havana, Cuba

Metal nanoparticles (MNPs) and semiconductor quantum dots (QDs) are two of the main building blocks of nano-scale systems with a wide range of applications in electronics and photonics. In view of the ubiquitous nature of the interparticle Coulombic decay process (ICD), which was also studied for paired QDs, we investigate ICD in the context of MNP-QD dimers. In this proposed scenario, the MNP is excited to a plasmonic state, which drives the emission of an QD electron. The energy transfer between the systems is mediated by the long-range Coulomb interaction. I will report on new developments towards a time-dependent description of the process, based upon the established electron dynamics methodology for paired QDs.