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Multi-electron processes in atoms and molecules

Experimental investigations by coincidence spectroscopy

Jonas Andersson
Institutionen för fysik
Naturvetenskapliga fakulteten

Akademisk avhandling för filosofie doktorsexamen i fysik, som med tillstånd från Naturvetenskapliga fakulteten kommer att offentligt försvaras fredagen den 7 juni 2019, kl. 13:15 i PJ-salen, institutionen för fysik, Fysikgården 2, Göteborg.

ISBN: 978-91-7833-484-1, tryckt version
ISBN: 978-91-7833-485-8, pdf nätet
Tillgänglig via http://hdl.handle.net/2077/59899

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Jonas Andersson
Department of Physics
University of Gothenburg, Sweden

Abstract:

This thesis presents studies on multi-electron processes in atoms and molecules initiated by single-photon absorption. The experimental techniques used for these studies rely on synchrotron radiation offered at large scale user facilities, and a magnetic bottle spectrometer. The magnetic bottle spectrometer is a versatile time-of-flight instrument that collects charged particles emitted from an ionization event using a characteristic magnetic field. The experiments were carried out in a coincidence detection mode, which allows selective analysis of correlated ionization processes.

The work in this thesis includes detailed analyses of Auger decay processes leading to triply ionized final states in atomic Cd and Hg. The experimental data were compared with numerical calculations to identify the triply ionized final states and the Auger cascades leading to these states. The Auger cascade analyses identified important intermediate inner-states involved in the formation of triply ionized final states, and demonstrated the strong influence of Coster-Kronig transitions when energetically allowed. The studies on Cd also demonstrated the involvement of shake-up transitions in reaching the triply ionized ground state from photoionization using 200 eV photons.

A new instrument for multi-electron and multi-ion coincidence studies was developed and used in experimental studies on Auger cascades in atomic Xe and on Coulomb explosion of molecular ICN. We studied the final charge state distributions from photoionization of different subshells in Xe, by measuring the ion mass spectra recorded in coincidence with specific photoelectrons. These results were compared with experimental results on Coulomb explosion of ICN from photoionization of similar subshells in I. The results suggest that the overall degree of ionization in Coulomb explosion of ICN is similar to the charge state distributions from photoionization of the related subshells in Xe.

Furthermore, experimental results on energy sharing distributions of the two emitted electrons from single-photon direct double photoionization of He are presented. Energy sharing distributions were measured by recording the kinetic energies of both electrons in coincidence for excess energies ranging from 11-221 eV. An empirical model was introduced to parametrize the shapes of the distributions and to form benchmarks for future studies on other direct double ionization processes. The experimental distributions were used to extract indirect information on the knock-out mechanism, thought to be partly responsible for the direct double photoionization process. Theoretical shake-off distributions and the experimentally estimated knock-out distributions were parametrized using the same empirical model, and the results are found to be in agreement with numerical simulations.

Keywords: Atomic physics, Molecular physics, Auger cascade, Coulomb explosion, Coster-Kronig decay, Direct double photoionization, Magnetic bottle spectrometer, Coincidence spectroscopy