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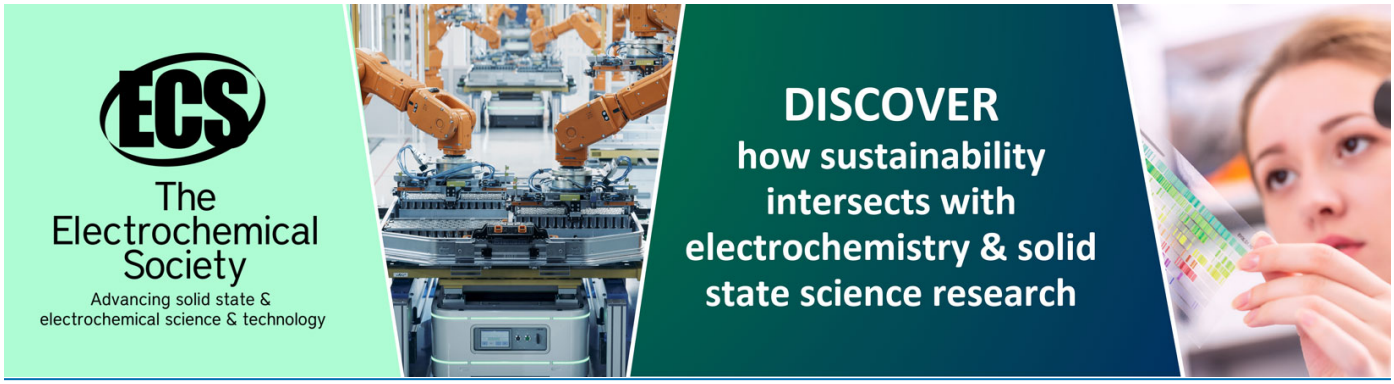

X-ray spectroscopy on ultrafast-decaying core-excited atomic ions

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Synopsis Results from the first soft X-ray user experiment at the European XFEL on nonlinear photon-matter interaction will be presented. Angle-resolved electron time-of-flight spectroscopy employed at the AQS (Atomic-like Quantum Systems) endstation of the SQS (Small Quantum Systems) instrument reveals insight into the character of resonances in highly transient, core ionized neon ions, i.e. Ne^+ $1s^1 2s^2 2p^6$ → Ne^{+*} $1s^0 2s^2 2p^6 np$, together with their respective relaxation dynamics. Enabled by the unique properties of the European XFEL, novel perspectives on efficient nonlinear spectroscopy will be discussed.

Highly intense X-ray pulses from X-ray free-electron lasers (XFELs) enable the study of nonlinear, photo-induced processes in atoms and molecules at short wavelengths. This includes sequential and nonsequential multiphoton ionization and excitation processes. Sequential ionization of core electrons is in strong competition with ultrafast relaxation via Auger emission [1]. At high X-ray irradiation, more than one photon can be absorbed before the Auger decay happens, i.e. typically within few femtoseconds. Consequently, the yield of this sequential photo-absorption by core electrons and accordingly the population of transient, highly excited ions increase significantly. Therefore, an efficient characterization of these short-lived ions becomes spectroscopically feasible. By using the intense pulses from the SASE3 soft X-ray undulators at European XFEL, we could, for the first time, study the core-hole excitation of transient, highly excited atoms by means of resonant Auger electron spectroscopy employing the Small Quantum Systems (SQS) instrument. Here, neon was ionized at the *1s* shell and, depending on the photon energy, either sequentially ionized to Ne^{2+} $1s^0 2s^2 2p^6$ or excited to different Ne^{+*} $1s^0 2s^2 2p^6 np$ states within the same fs X-ray pulse. Thanks to the routinely available tunability of the SASE3 undulator, it was possible to scan the photon energy across the $1s^1 2s^2 2p^6$ → $1s^0 2s^2 2p^6 np$ resonances with photon energy steps of 1 eV (much smaller than the bandwidth). The resulting Auger electron spectra were recorded by electron time-of-flight spectrometers. The results of these measurements are compared to previous studies of single-photon processes populating the same final states [2] as well as with results from calculations based on the Multiconfiguration Hartree-Fock (MCHF) method [3] including relativistic Breit-Pauli corrections (allowing non-orthogonal wave-functions). The results are benchmarking the performance of the SQS endstation at European XFEL and demonstrate the novel possibilities which have been opened up in particular for investigations of nonlinear phenomena and ultrafast decaying transient states in the X-ray domain.

References

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