Exploring Resonant Photoemission and Coulomb Interactions in Transition Metal Chalcogenides

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Abstract. We investigate the complex interplay between various decay mechanisms under resonant conditions, focusing on radiation-less Raman Auger and Classical Auger emissions in Transition Metal Chalcogenides (TMCs), including selenides and sulphides. By performing resonant Angle-Resolved Photoemission Spectroscopy (ARPES) measurements and complementary SPR-KKR photoemission calculations, we explore the role of the Coulomb interaction U, a crucial parameter governing electron correlations in these materials. The Coulomb interaction is particularly pronounced in low-dimensional systems, such as quasi-2D TMCs, where it induces a variety of electronic phases, including Charge Density Wave (CDW) order, the coexistence of CDW with superconductivity, and topologically nontrivial phases. We apply the method of Cini and Sawatzky to determine the on-site Coulomb interaction for each element by analysing the energy separation between the main spectral features in the resonant photoemission spectrum and the two-valence-hole (VV) Auger final state. This procedure involves a comparison between the resonant spectrum and the self-convolution of single-hole states derived from non-resonant photoemission, providing a direct measure of the Coulomb energy. On the theoretical front, the SPR-KKR calculations, based on a one-step model, incorporate the full matrix elements of the photoemission process, allowing us to probe the underlying electronic structure with high precision. We further examine the computed ARPES, X-ray Absorption Spectroscopy (XAS), orbital- and element-resolved band structures, and density of states (DOS). The theoretical results show excellent agreement with experimental data, offering valuable insights into the electronic properties of TMCs and their complex many-body interactions.