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FOR BASIC SCIENCES
Block JD, Sector III, Salt Lake, Kolkata 700 106

DEPARTMENTAL SEMINAR

Condensed Matter and Materials Physics

15th February,2023 **4.00 PM** **ONLINE/ FERMION**

SPEAKER

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TITLE OF THE TALK

COMPUTATIONAL ELECTRONIC STRUCTURE AND X-RAY SPECTROSCOPY: A SYMBIOTIC FRIENDSHIP

ABSTRACT

With the advent of improved synchrotron-based X-ray sources, core-level spectroscopy has secured its position as one of the most lucrative avenues for probing the electronic structure of materials as well as for fingerprinting various complex chemical phenomena. Evidently, dedicated theory-efforts are needed to complement the experimental research. At the same time, leveraging the rapidly expanding database of experimental spectra, computational core-level spectroscopy can offer valuable insights on the methods and approximations employed in electronic structure theory and thereby inspire the formulation of new theoretical approaches. Following a brief "teaser" report on X-ray absorption to demonstrate the utility of simulations, in this talk I will explore the efficacy and interrelation of two popular first-principles frameworks in the context of simulating core-level emission spectroscopy. For non-resonant X-ray emission spectroscopy (NXES), I will show that a constrained-occupation based optimized-orbital Kohn-Sham treatment is more reliable than the adiabatic linear-response (LR) approach. The inadequacy of the latter is, in fact, rooted in the neglect of dynamical electron-hole screening, which can be associated with the change in polarization resulting from the core-ionization. I will demonstrate that an inexpensive determinant-based formalism reliant only on two self-consistent field calculations is typically sufficient for modeling NXES. Equipped with this understanding, we can develop a flexible framework for simulating the resonant inelastic X-ray scattering (RIXS) process by treating the effects of the core-excitations within the constrained-occupation approach and valence excitations with the LR method. Such a division of treatment not only ensures fast and accurate simulation of the RIXS spectrum, but also helps with its interpretation by automatically assigning the spectral features to the relevant electronic transitions. In a pseudopotential-based implementation, this approach circumvents the need for all-electron calculations. This framework can also be straightforwardly extended to incorporate the effects of additional excitations like vibrations, charge-transfer, etc.

References

● S. Roychoudhury, L. A. Cunha, M. Head-Gordon, and D. Prendergast Changes in polarization dictate necessary approximations for modeling electronic de-excitation inten- sity: an application to x-ray emission ,Physical Review B 106, 075133 (2022).
● S. Roychoudhury and D. Prendergast CleaRIXS: A Fast and Accurate First-Principles Method for Simulation and Analysis of Resonant Inelastic X-ray Scattering, Physical Review B 106, 115115 (2022).
● S. Roychoudhury and D. Prendergast Efficient core-excited state orbital perspective on calculating X-ray absorption transitions in determinant framework, arXiv:2208.11261 (2022).

HOST FACULTY

Prof. Priya Mahadevan, Senior Professor

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