Final Technical Report

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Title: Computational time-resolved and resonant x-ray scattering of strongly correlated materials

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Basic-Energy Sciences of the Department of Energy (BES/DOE) has made large investments in x-ray sources in the U.S. (NSLS-II, LCLS, NGLS, ALS, APS) as powerful enabling tools for opening up unprecedented new opportunities for exploring properties of matter at various length and time scales. The coming online of the pulsed photon source, literally allows us to see and follow the dynamics of processes in materials at their natural timescales. There is an urgent need therefore to develop theoretical methodologies and computational models for understanding how x-rays interact with matter and the related spectroscopies of materials.

The present project addressed aspects of this grand challenge of x-ray science. In particular, our Collaborative Research Team (CRT) focused on developing viable computational schemes for modeling x-ray scattering and photoemission spectra of strongly correlated materials in the time-domain. The vast arsenal of formal/numerical techniques and approaches encompassed by the members of our CRT were brought to bear through appropriate generalizations and extensions to model the pumped state and the dynamics of this non-equilibrium state, and how it can be probed via x-ray absorption (XAS), emission (XES), resonant and non-resonant x-ray scattering, and photoemission processes. We explored the conceptual connections between the time-domain problems and other second-order spectroscopies, such as resonant inelastic x-ray scattering (RIXS) because RIXS may be effectively thought of as a pump-probe experiment in which the incoming photon acts as the pump, and the fluorescent decay is the probe. Alternatively, when the core-valence interactions are strong, one can view K-edge RIXS for example, as the dynamic response of the material to the transient presence of a strong core-hole potential. Unlike an actual pump-probe experiment, here there is no mechanism for adjusting the time-delay between the pump and the probe. However, the core hole predominantly decays via Auger processes, thereby providing an internal time-scale, which limits intermediate-state processes to timescales of a few femtoseconds. Accordingly, a number of activities directed at modeling K-, L- and M-edge RIXS in correlated materials were also pursused by our CRT.

Our research effort supported by this CMCSN grant substantially advanced the understanding of x-ray scattering processes in the time-domain as well as in the more conventional scattering channels, including time-resolved photoemission, and how such processes can be modeled realistically in complex correlated materials more generally. The modeling of relaxation processes involved in time-domain spectroscopies is important also for understanding photoinduced effects such as energy conversion in photosynthesis and solar cell applications, and thus impacts the basic science for energy needs.

The CMCSN funding provided the 'glue' funding without which the collaborative research activities that were undertaken under this award would not have been possible, and the CMCSN funding was thus a critically important enabler in this connection. The collaborative activities started under the CMCSN award led to further collaborative work within the team members even after the CMCSN funding ended.

Details of the research accomplished under this award are presented in the list of publications in open, peer-reviewed literature given below.

In short, we were able to accomplish the goals of our CMCSN project extremely well.

List of CMCSN-supported Publications

1. L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. Hill, and J. van den Brink, "Resonant inelastic X-ray scattering studies of valence excitations", Rev. Mod. Phys. 83, 705 (2011).

2. L. J. P. Ament, M. van Veenendaal, and J. van den Brink, "Determining the electron-phonon coupling strength in correlated electron systems from resonant inelastic x-ray scattering", Europhys Lett. 95, 27008 (2011).

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4. GW+U real-space Green's function calculations of x-ray spectra, Towfiq Ahmed, J.J. Kas and J. J. Rehr, Phys Rev B 85, 165123 (2012).

5. Local time-correlation approach for calculations of x-ray spectra A. J. Lee, F. D. Vila, and J. J. Rehr, Phys. Rev. B 86, 115107 (2012).

6. Y. Ding, J. Fernandez-Rodriguez, J. Kim, F. Lei, D. Casa, M. Upton, T. Gog, H. Mao, and M. van Veenendaal, "Spin ordering mediated orbital hybridization in CoO at high pressure", Phys. Rev. B 86, 094107 (2012).

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11. Joshua Kas, John Rehr, Moniek Tromp, Rowena J. Thomas, and Pieter Glatzel, "Real Space Green's Function Approach to Resonant Inelastic X-Ray Scattering and HERFD XAS", J. Phys.: Conf. Ser. 430 012003 (2013).

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14. B. Moritz, A. F. Kemper, M. Sentef, T. P. Devereaux, and J. K. Freericks, "Electron-Mediated Relaxation Following Ultrafast Pumping of Strongly Correlated Materials: Model Evidence of a Correlation-Tuned Crossover between Thermal and Nonthermal", Phys. Rev. Lett. 111, 077401 (2013).

15. K. Wohlfeld, S. Nishimoto, M. W. Haverkort, and J. van den Brink, "Microscopic origin of spin-orbital separation in Sr2CuO3", Phys. Rev. B.88, 195138 (2013).

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