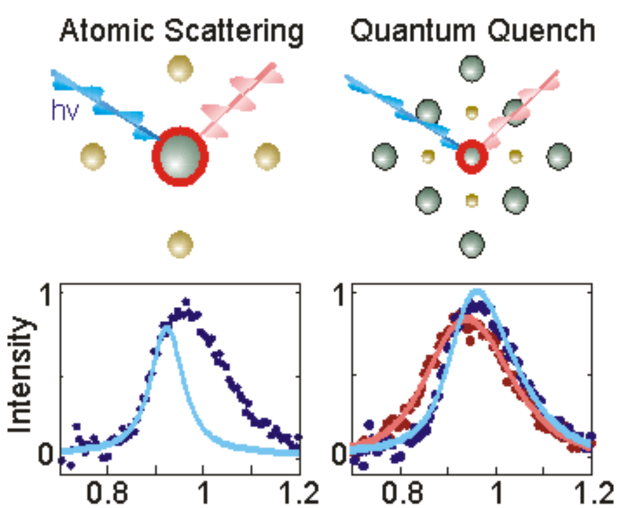
Postdoctoral Fellowship Highlight

L. Andrew Wray, Ph.D., Physics

Years of Fellowship: 2010-2013
Collaborating Institution: Princeton University, Princeton, NJ
Project: BL4.0.3 (MERLIN)

As an ALS postdoctoral fellow, Andrew helped commission Beamline4.0.3 and developed essential experimental protocols for tasks such as data analysis, minimization of scattering artifacts, and optimal operation of x-ray emission spectrograph. His measurements and analysis of data from the beamline established the scientific cases for high resolution resonant inelastic x-ray scattering spectroscopy (RIXS) in strongly correlated electron systems.

A new measurement regime: The MERLIN beamline provides access to numerous low energy resonances in extreme UV (EUV) regime, and demonstrates groundbreaking energy resolution for the RIXS technique in daily operations. The RIXS technique addresses a critical gap in current research capabilities to observe and interact with nanoscale quantum states, in which strong electronic correlations manifest intriguing material properties such as high temperature superconductivity, multiferroicity, insulator-metal transition and colossal magnetoresistance. Understanding how the nanoscale interplay of electronic correlations will advance the development of next generation technologies which exploit the new functionalities from the collective behaviors of many electrons, thus the relevant research lies at the heart of current condensed matter physics.



Photons make a splash: A well resolved charge excitation in CoO is modeled with (left) a standard CoO_6 cluster and (right) an extended $Co_{19}O_{46}$ model that considers how changes in the electronic state on the scattering site create a quantum quench, triggering dynamics on near-neighbor Co atoms [2]. Boltzmann weighting states of the poly-atomic many-body system gives good correspondence with (blue) T=90K and (red) T=320K data.

With resolution better than thermal energy, Andrew demonstrated for the first time that thermally populated in-gap states can create anti-

Stokes-like tails of intensity attached to features in RIXS spectra. The observation of this feature is currently only possible at MERLIN, as no other RIXS spectrometer has achieved energy resolution comparable to the scale of room temperature thermal fluctuations (~26meV). In addition, Andrew has shown that different kind of extended tail on the opposite side (energy loss side) of charge excitations bears rich information about many-body quantum states and quantum quench phenomena. Through comparison between extensive modeling and high resolution RIXS data, he developed atomic-multiplet-based calculations that incorporate lifetime effects and the quantum entanglement between neighboring electronic states to satisfactorily explain the observed spectra of model Mott insulators CoO and NiO [1-3]. These analyses serendipitously revealed an unexpected coherent component to scattering emission, which gives information about the spatial extent of magnetic domains and may in the future enable a new form of X-ray microscopy. The experimental and theoretical achievements by Andrew have laid the strong foundation for high resolution RIXS studies at MERLIN. The acquired knowledge and techniques developed over his postdoctoral research have benefited researchers using X-ray spectroscopies (X-ray absorption, emission and RIXS) at the ALS.

Tools for the future: The recent development of free electron laser (FEL) light sources has encouraged innovative thinking about X-ray spectroscopies. By delving into the physics of the unique photon energy range available at MERLIN, Andrew has identified several promising X-ray scattering techniques for future development at FEL and synchrotron facilities (e.g. NGLS).

The coherent elastic component of EUV scattering identified by Andrew will be a valuable tool for resolving domain boundaries through observing coherent scattering phase interference between them. However, it is not the only way to access phase information within a RIXS experiment. Strong quantum interference between the EUV M_2 and M_3 resonance states in cuprates has enabled the first measurement of phases in inelastic emission [4]. Coupling phase information with scattering intensities allows RIXS excitation processes to be transformed into the time domain, granting insight into the dynamical processes that may be interacted with in pump-probe RIXS experiments at FEL light sources. Andrew has shown that the diverse scattering time scales observed in high resolution studies give a way to probe how charge excitations are "dressed" by many-body modes (e.g. spin) [1,2,4], and might one-day answer key fundamental questions about high-Tc superconductivity by revealing the femtosecond scale response dynamics of a superfluid to different types of strong local perturbations.

The anti-Stokes-like energy gain feature identified in these studies reveals the occupation of low energy states, which is a valuable quantity to measure in pump-probe experiments as it will reveal the non-thermal distribution of energy introduced by a pumping pulse into low energy degrees of freedom. These results provide strong motivation for the development of future time-resolved spectrometers.

Engaging the experimental community: Andrew has publicized the beamline in talks at Stanford, SLAC, Rice, Texas A&M, LBL, SPring-8, Kyoto University, Keio University, SPINTECH6, M2S 2012 and the APS March Meeting. His analysis of resonant scattering in the EUV has helped to lower the barrier for scientists proficient in other spectroscopies becoming involved in measurements at the beamline. He frequently collaborates with users, and has engaged theoretical and sample growth collaborators to focus on the unique opportunities and challenges that the MERLIN RIXS capabilities represent.

1. L. A. Wray *et al.*, *Phys. Rev. B* **86**, 195130 (2012).
2. L. A. Wray *et al.*, CoO results in journal review (Feb, 2013).
3. Y.-D. Chuang, L. A. Wray, J. Denlinger and Z. Hussain, Synch. Rad. News **25**, 23 (2012).
4. L. A. Wray *et al.*, Preprint at http://arxiv.org/abs/1203.2397 (2012).