Overcoming Experimental and Intrinsic Broadening in Excited State Spectroscopies using Richardson-Lucy Deconvolution

W.T. ELAM, T.T. FISTER, G.T. SEIDLER, K.P. NAGLE, J.J. KAS, University of Washington, J.O. CROSS, Advanced Photon Source, Argonne National Laboratory — The oscillatory signature of the photoelectron interference phenomenon central to core shell spectroscopies is frequently broadened by experimental or intrinsic (i.e., core-hole lifetime) energy resolutions, limiting the interpretation of the measurement. For example, this problem occurs in x-ray absorption fine structure (XAFS) measurements of heavy elements where the core-hole lifetime is very short ($\hbar/\tau_{\text{core-hole}} \geq 5$ eV), and also in non-resonant x-ray Raman scattering measurements where the instrumental resolution (typically $\sim 1$ eV) can be nearly an order of magnitude larger than the intrinsic energy resolution. Given the small statistical uncertainties in typical XAFS data and in recent XRS measurements using dedicated multielement spectrometers, the question naturally arises as to deconvolving the data with respect to the known instrumental or intrinsic resolutions. Here, we demonstrate that the Richardson-Lucy iterative algorithm provides a robust maximum likelihood method for addressing this issue in both XAFS and XRS. We demonstrate this method on core-hole broadened Ag XAFS data and experimentally broadened diamond and graphite XRS data.