Electron Hole Excitations in Transition Metals Investigated via Inelastic X-ray Scattering: An Experimental Perspective

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Introduction

Inelastic x-ray scattering (IXS) provides a unique probe of the electronic response of condensed matter at large wave vectors, the regime for which electronic correlations are most important. The combination of meV-resolution to eV-resolution IXS measurements on third-generation synchrotrons with *ab initio*, all-electron, time-dependent density-functional theory (TDDFT) calculations [1] represents a breakthrough that provides greatly enhanced opportunities for the fundamental investigation of electron dynamics in systems ranging from elemental solids to compounds such as the transition-metal oxides and other strongly-correlated materials.

Methods, Materials, and Results

Figure 1 shows IXS measurements (energy resolution of 1.4 eV) of the dynamical structure factor of Sc and Cr for wave vectors of ~2 $Å^{-1}$. A novel feature of the experiment is that the data have been reduced to absolute units through the use of IXS measurements in Al. Also shown are TDDFT calculations of the loss spectra. The thin lines refer to the random-phase approximation, and the thick lines refer to the so-called time-dependent local density approximation (TDLDA) [1]. Since there are no adjustable parameters in this comparison, the agreement between theory and experiment for energies below the M edge (~34 and 42 eV for Sc and Cr, respectively) is quite remarkable. Note in particular that the respective spectra are quite different below ~10 eV. Clearly, a different response regime involving an extremely localized initial state sets in at the M edge. It is apparent that either or both the local and the adiabatic approximations, which underpin the TDLDA, break down at the edge. This result provides a significant benchmark for more refined studies of dynamical correlations in transition metals.

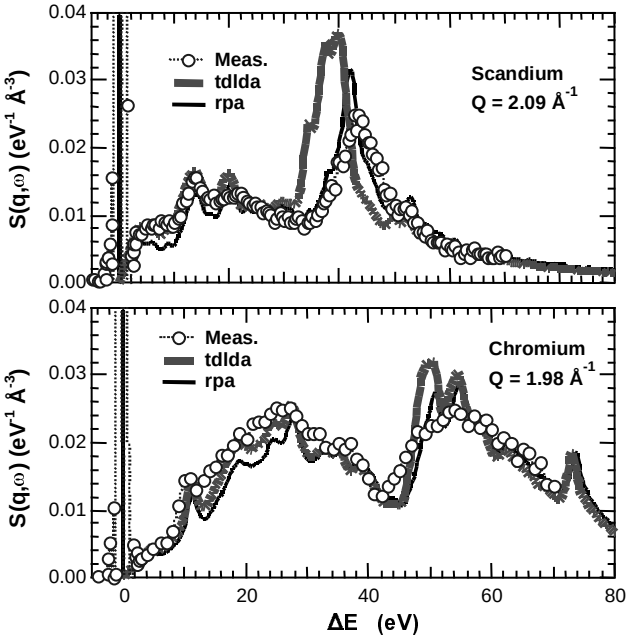

FIG. 1. IXS measurements and TDDFT calculations of the dynamical-structure factor S(q,ω) for Sc and Cr for wave vectors of ~2 $Å^{-1}$.

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