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Inelastic X-Ray Scattering and Time-Dependent Density-Functional-Theory Investigation of Electronic Excitations and Correlations in Ni

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Inelastic x-ray scattering (IXS) measurements of the energy-loss spectra provide a unique tool for fundamental investigations of electronic correlations in condensed matter. This tool becomes particularly powerful when combined with *ab initio* density-response calculations performed within time-dependent density-functional theory (TDDFT). However, until the advent of 3rd generation synchrotron sources, the strong photo-electric absorption associated with *d*-band materials and related compounds severely restricted the IXS investigation of these important systems.

We have initiated IXS investigations of transition metal Ni along [111] wave-vectors up to 4.5 \AA^{-1} , in conjunction with a new breed of *ab initio* TDDFT density-response calculations. The measurements were performed on the UNICAT undulator beamline using the sagittal and meridial focusing, and the high intensity of the Si (111) high heat load monochromator operated with 1.1 eV resolution at 7.56 KeV. Incident beam intensities of $\sim 4 \times 10^{12}$ photons/sec were obtained, and the beam size on the single crystal Ni sample was 0.5 mm x 0.5 mm. Additional measurements at 0.35 eV resolution were made using a Si (333) channelcut monochromator to narrow the incident energy bandpass. Analysis of the energy loss spectrum was made using a (444) Ge spherically bent (and diced) analyzer configured in near backscattering geometry. To collect IXS spectra, the analyzer remained tuned to 7.56 KeV and the energy of the incident beam was varied.

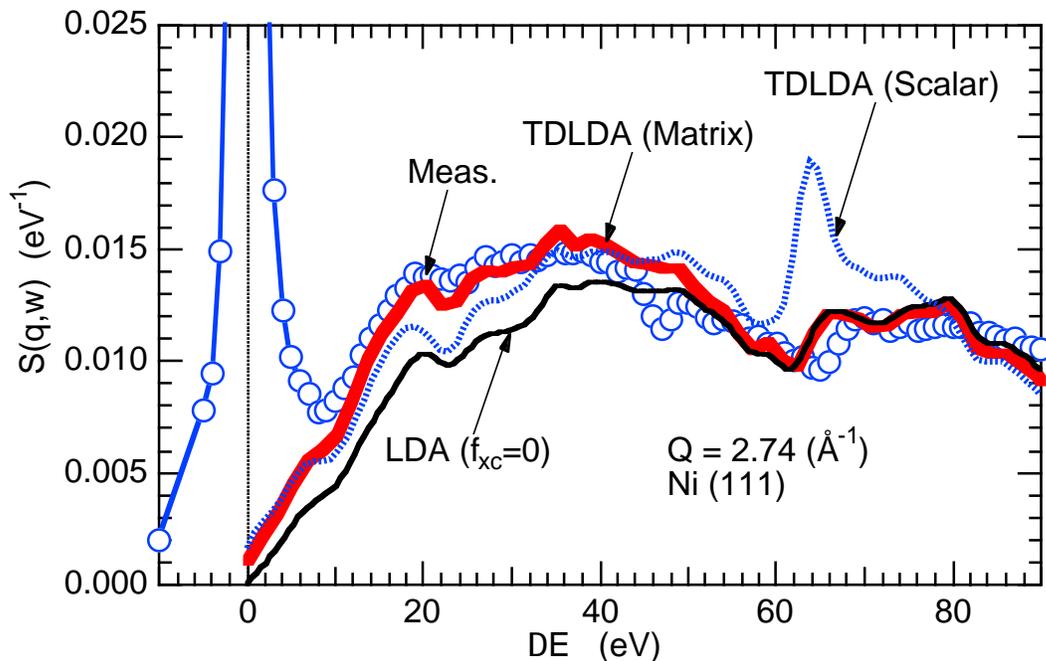


Figure 1 shows an example of the measured inelastic scattering data obtained for a wave-vector of 2.74 \AA^{-1} with 1 eV energy steps over an energy range of 90 eV. Characteristic features are observable in the loss spectra including structure at $\sim 25 \text{ eV}$ and the core-electron edge at $\sim 66 \text{ eV}$. The theoretical loss spectrum labeled TDLDA corresponds to a TDDFT calculation performed with use of the local-density approximation (LDA) in the solution of the ground-state problem and in the evaluation of the so-called many-body kernel $f_{xc}(W)$. The density-response function for Kohn-Sham (KS) electrons was evaluated in terms of LAPW band structure and orbitals; non-diagonal matrix elements of the KS response in reciprocal space bring in the crystal local-field effects. The physical density-response is obtained by solving the integral equation which relates it to the KS response and the many-body kernel.

It is clear that for energies up to $\sim 30 \text{ eV}$, the TDLDA response corresponds extremely well with the measured spectrum. On the other hand, for larger energies a significant discrepancy between theory and experiment is apparent; the same is currently under investigation. The number of reciprocal lattice vectors (G-shells) required for full convergence of the TDLDA spectrum, and the completeness of the LAPW basis functions for these high energies, are likely sources of this problem. It is also possible that the adiabatic approximation for the many-body kernel breaks down for these energies, i.e., the data may be suggesting the need for the development of a non-trivial, frequency-dependent $f_{xc}(W)$. The discrepancy between theory and measurement at the onset of the core excitations provides a signature of the LDA underestimate of the binding energy of the core states; the exaggerated intensity of the edge is likely related to insufficient number of G-shells in the calculation. It is interesting to note that, while the KS band structure has no rigorous meaning in the context of comparison with photoemission, the same enters *rigorously* into the evaluation of the KS response and, therefore, in the comparison with IXS data. Further elaborations of the interplay between theory and experiment illustrated in this Progress Report is expected to lead to new insight into the electronic excitations and correlations in *d*-band materials.

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