Electronic excitations: ab initio calculations of electron energy loss spectra and applications to zirconia, titania and cuprous oxide

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In this work, we address the question of the theoretical prediction and interpretation of experimental spectra obtained by the excitation of valence electrons, such as electron-energy loss spectra (EELS). We particularly expose the case of the transition metal oxides ZrO$_2$ [1], TiO$_2$ [2] and Cu$_2$O, and compare our theoretical results with experimental data. We show which levels of theory are appropriate, starting with the joint density of states $\sum_{val,cond} \delta(E - E_{val} + E_{cond})$, where the energies of the valence and conduction bands are obtained from ground-state density-functional calculations [3], to the effect of the inclusion of matrix elements of the dipolar operator (random phase approximation RPA) and the inclusion of off-diagonal elements in the response function (RPA with crystal local-field effects). Additional exchange and correlation local fields within the adiabatic time dependent local density approximation, and many body effects in the quasiparticle and in the TDDFT frameworks [4,5] are presented in order to explain the reasons why the RPA turns out to be a level of approximation appropriate for EELS.

References
[6] This research was supported by the joint research program "ISMIR" between CEA and CNRS. Computer time has been granted by CEA/DSM on the cluster of Compaq Alphaserver (project p543) and NEC SX6 (project p93) and by IDRIS on the NEC SX5 (project 544). We acknowledge support from EU's 6th Framework Programme through the NANOQUANTA Network of Excellence (NMP4-CT-2004-500198).