Simulating Electron Energy Loss Spectroscopy in Large Systems

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The EELS and IXS cross sections in extended systems are proportional to the imaginary part of the diagonal of the inverse dielectric matrix, which can be computed using Time-Dependent (TD) Density Functional Theory (DFT). Current TDDFT-based approaches to dynamical screening involve the computation of a large number of single-particle unoccupied states and the manipulation (multiplication, inversion) of large matrices, two tasks that make them unfit to address systems larger than a handful of atoms. We present a new method, based on TDDFT linear response, that avoids these difficulties by adopting a Lanczos recursion scheme and a representation of the response orbitals borrowed from density-functional perturbation theory [1]. The resulting algorithm allows to compute the EELS and IXS cross sections for a same transferred mometum and in an entire, wide, frequency range with a numerical workload comparable to that of a single ground-state DFT calculation. We have implemented our method in the QUANTUM ESPRESSO distribution of computer codes [2], and successfully benchmarked it on the prototypical examples of bulk silicon and aluminum. The EELS/IXS angle-resolved cross sections in bismuth have been calculated for the first time. Details of the work being presented can be found in Ref. [3].

[1] D. Rocca, R. Gebauer, Y. Saad, and S. Baroni, J. Chem. Phys. 128, 154105. http://dx.doi.org/10.1063/1.2899649

[2] P. Giannozzi, S. Baroni et al., J. Phys.: Cond. Matt. 21, 395502 (2009). http://dx.doi.org/10.1088/0953-8984/21/39/395502

[3] I. Timrov, N. Vast, R. Gebauer, and S. Baroni, arXiv:1305.6233 [cond-mat.mtrl-sci] http://arxiv.org/abs/1305.6233