Unoccupied continuum and resonant orbitals in GW quasiparticle calculations

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The first-principles GW method, an interacting many-body Green's function method for *ab initio* calculations of the quasiparticle energies, in its standard formulation[1] involves infinite summations over single-particle orbitals of a mean-field starting Hamiltonian. In practical calculations, these summations are truncated but the number of orbitals required to converge the quasiparticle self-energies increases rapidly with increasing system size. Generating single-particle orbitals within the pseudopotential planewave density functional theory (DFT) method becomes the limiting factor in GW calculations for large systems. In this work[2], we demonstrate that the unoccupied orbitals above a certain energy cutoff (usually not far above the Fermi level) can effectively be replaced with a continuum of appropriately prepared planewave orbitals and discrete resonant states. The continuum orbitals are constructed from symmetrized planewaves and the resonant states are computed with short-range localized basis functions (such as in the SIESTA code). The Gram-Schmidt process is applied to orthogonalize thus constructed unoccupied orbitals with respect to the exact low-energy DFT Kohn-Sham orbitals and among themselves. This approach allows us to carry out accurate GW calculations for large systems using only a small number of unoccupied Kohn-Sham orbitals. For a typical molecular calculation in a $25\times25\times25$ a.u.^{3} supercell, the time required to generate 4,000 unoccupied orbitals decreases from 1,000 CPU hours down to 1 CPU hour, and the subsequent GW calculation takes about 10 CPU hours. This work was supported by National Science Foundation Grant No. DMR10-1006184, the U.S. Department of Energy under Contract No. DE-AC02-05CH11231. Computational resources have been provided by NSF through TeraGrid resources at NICS and DOE at Lawrence Berkeley National Laboratory's NERSC facility.

[1] M.S. Hybertsen and S. G. Louie, Phys. Rev. B 34, 5390 (1986).

[2] G. Samsonidze, M. Jain, J. Deslippe, M. L. Cohen, and S. G. Louie (unpublished).