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Excitation Energies from Time-Dependent Density Functional Theory within Modified Linear Response: Inclusion of the Electron-Hole Hartree-Fock Interaction CHUNPING HU, OSAMU SUGINO, Institute for Solid State Physics, University of Tokyo, and also CREST, Japan Science and Technology Agency, YOSHIYUKI MIYAMOTO, Fundamental and Environmental Res. Labs, NEC, Japan — Time-dependent density functional theory (TD-DFT) within linear response (LR) has gained enormous popularity in the calculation of electronic excitations, whereas it is known to give considerably underestimated excitation energies for Rydberg and charge-transfer excitations. Although the incorrect long-range behavior of exchange-correlation (XC) potentials has been blamed for this problem, a different point of view on the LR scheme without any correction of XC potentials is presented here. Analyzing approximate excitation energies from LR within adiabatic local density approximation (ALDA) and the exact exchange (EXX) scheme, we propose a modified LR theory to strictly include the electron-hole Hartree-Fock interaction kernel, and to make excitation energy expression in ALDA explicitly similar to the EXX one. TD-LDA calculations within modified LR on typical diatomic molecules show that excitation energies of both Rydberg and charge-transfer excitations can be greatly improved to the EXX-level accuracy.

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