Cumulant expansion approaches to excited state electronic structure and specta

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Cumulant expansions for the one electron Green's function have recently had great success in describing many-body excitations in a variety of materials and spectra.[1,2,3,4] In particular, in comparison to the GW method, improved agreement is found for satellite peaks in x-ray photoemission spectra. However, previous treatments based on expansions of the time-ordered Green's function were inconsistent, failing to describe satellites on both sides of the chemical potential, leading to occupation numbers which retain their free electron values, in contrast to QMC calculations and experimental results. Recent theoretical developments give a consistent picture based on an expansion of the retarded one electron Green's function []. Here we describe several methods for obtaining excited state electronic structure and spectra using cumulant expansion based approaches. First, we describe an extended derivation of the retarded cumulant expansion method which allows for the use of quasiparticle self-consistent starting points.[6] The effects of various levels of self-consistency on quantities of interest, such as spectral functions, quasiparticle properties, and occupation numbers are investigated and discussed. Results are shown for a variety of materials, including semi-conductors and oxides.

Second, we present a real-time TDDFT method for calculating the cumulant for the deep-hole Green's functions. This method has been successful in treating charge-transfer excitations in x-ray emission. Here we extend the treatment to describe x-ray absorption as well, and give results for correlated materials such as transition metal oxides and CeO2.

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