Hunting double excitations

D. Sangalli(1,3), P. Romaniello(2,3), G. Onida(1,3)

(1) Consorzio Nazionale Interuniversitario per le Scienze Fisiche della Materia (CNISM) and Dipartimento di Fisica dell'Università degli studi di Milano (Italy)
(2) Laboratoire Des Solides Irradiés, Ecole Polytechnique, Palaiseau (France)
(3) European Theoretical Spectroscopy Facilities (ETSF)

email: davide.sangalli@gmail.com

Abstract

The spectra of isolated systems is known to be described quite well by Casida's approach to Time-Dependent Density-Functional-Theory (TDDFT) for many closed shell systems, within the adiabatic approximation. On the other hand the same framework is known to perform quite poorly for open shell systems. The reason of this deficiency can be found in the failure of the adiabatic approximation in describing double excitations, that leads, therefore, to the break of the spin symmetries of the system. One would encounter analogous deficiencies using the Bethe Salpeter (BSE) approach with a static kernel W(0,0). A solution to the problem is the use of dynamical kernels. This work aims to scrutinize the dynamical structure that the BSE kernel and/or the exchange correlation kernel of TDDFT should have in order to describe double excitations. Following the same line of previous works on the study of removal/addition of electrons [2] and of excited states of nuclei [3,4], we find out that second order Feynman diagrams do the right job. We show some preliminary tests on model systems.

Conclusions:
- The result (TDA): only diagram K1 here:
  \[ K^{(2)}(\omega) = \sum_{n,m,r,s} \text{det}(X(n,m)) \text{det}(Y(r,s)) \rho^{\text{exc}}(n,m) \rho^{\text{exc}}(r,s) \text{det}(X')^{(1)}(n,m) \text{det}(Y')^{(1)}(r,s) \text{det}(X^{(1)}(n,m)) \text{det}(Y^{(1)}(r,s)) \text{det}(K^{(0)}(n,m)) \text{det}(K^{(0)}(r,s)) \]
- We pointed out:
  - which is the right mathematical structure of a kernel containing double excitations;
  - through Feynman diagrams, which physics should be included;
  - applying the method to a model system, how over-counting disappears;
  - this preliminary result could be the starting point for the research of dynamical kernel which includes more physics than 2nd order one (maybe the RPA kernel).

References:

I'm especially thanks to prof. Gianluca Colò for many very useful discussions on the topic.

No over-counting!!!

How to evaluate all 8 second order Feynman diagrams knowing only one of them...

...ask the author!

"Work in progress":

"No over-counting!!!"