Conserving approximations in nonequilibrium green function theory
Stan, Adrian

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
2009

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

Copyright
Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

Take-down policy
If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Downloaded from the University of Groningen/UMCG research database (Pure): http://www.rug.nl/research/portal. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.
Green function methods have been used with great success to calculate a wide variety of properties of electronic systems, ranging from atoms and molecules to solids. Within the Green function approach, these properties are completely determined by the self-energy operator \( \Sigma \), which incorporates all the effects of exchange and correlation in a many-particle system. One of the most successful and widespread methods has been the \( GW \) approximation. In the \( GWA \), the self-energy operator has the simple form \( \Sigma = -GW \), where \( G \) is the Green function that describes the propagation of particles and holes in the system, and \( W \) is the dynamically screened interaction. This quantity describes how the bare interaction \( v \) between electrons is modified due to the presence of the other electrons and appears as a renormalized interaction in terms of Feynman diagrams. In extended systems the screened interaction is much weaker than the bare interaction, and therefore it is much more natural to expand the self-energy in terms of the screened interaction than in terms of the bare interaction. The \( GWA \) has produced excellent results for band gaps and spectral properties of solids but so far has not been explored much for atoms and molecules. However, \( GW \) calculations have been – to the present time – rarely carried out in a self-consistent manner, and the effect of self-consistency is, for this reason, still a topic of considerable debate.

At the core of this thesis lay the self-consistent all-electron \( GW \) calculations for atoms and diatomic molecules. We solve the Dyson equation for atoms and diatomic molecules within the \( GW \) approximation, in order to elucidate the effects of self-consistency and the importance of the electron correlation term of the self-energy in a self-consistent approach. In this respect, we investigated the performance of the \( GW \) at different levels of self-consistency for the case of atoms and diatomic molecules. Our main motivation for studying fully self-consistent \( \Phi \)-derivable schemes was that they provide unambiguous results for different observables and the fact that they satisfy important conservation laws that are important in future nonequilibrium applications of the theory. We addressed the question to what extent partially self-consistent schemes can reproduce the results of a fully self-consistent \( GW \) calculation. In the Chapter 4 of this thesis, we proposed a new partially self-consistent scheme, called \( GW_{fc} \), and we found that, together with the well known \( GW_0 \) method, it yields results in very close agreement with fully self-consistent \( GW \) calculations. A major advantage of the proposed scheme, \textit{i.e.}, \( GW_{fc} \), is that it produces results that are close to the fully self-consistent \( GW \) results at a much lower computational cost (comparable with the widely used \( G_0W_0 \) scheme). It will therefore be very valuable to test this method on solid state systems for which self-consistent \( GW \) calculations are difficult to perform due to the large computational effort. In this way it will be possible to get further insight into the performance of self-consistent \( GW \) for a large class of extended systems. The nonself-consistent \( G_0W_0 \) and the partially self-consistent \( GW_{fc} \) approximation, both violate the number conservation laws but, due to the partial self-consistency in \( GW_{fc} \), the errors are much reduced in this scheme.

After a thorough investigation of the ground state properties of small atoms and diatomic molecules,
within the $GW$ approximation, we have developed a time-propagation scheme for the Kadanoff-Baym equations for general inhomogeneous systems. These equations describe the time evolution of the nonequilibrium Green function for interacting many-body systems in the presence of time-dependent external fields. Our main motivation for developing such a scheme was the recent advancements in the field of molecular electronics which have emphasized the need for further development of theoretical methods that allow for a systematic study of dynamical processes like relaxation and decoherence at the nanoscale. Understanding these processes is of utmost importance for making progress in molecular electronics. We applied this time propagation scheme for the Kadanoff-Baym equations to the study of a double quantum dot and we analyzed in detail the ultrafast dynamics of transients and density distributions, relating them to the level structure displayed in the spectral functions. This study enables a clear interpretation of the transport spectroscopy experiments. Moreover, we showed that the initial correlations and the memory terms present in the Kadanoff-Baym equations, have large effects on time-dependent currents.

The time-propagation scheme for the Kadanoff-Baym equations can be used to further study electron transport through correlated systems considering the effects of correlated leads (two or three dimensional). It is also worthwhile to explore, for example, the dynamics of excitons in polymer chains like polyacetylene or the effect of electron-electron scattering on (non)equilibrium properties of a few electron quantum dots and quantum rings.