Manifestations of electronic correlation in finite and extended systems

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Abstract. In this work we discuss the influence of electronic correlation in finite and extended systems. In particular we stress the fact that the electronic correlation renormalizes the particle-particle interaction in a characteristic way and interpolate these ideas, well-known for extended systems, to small systems. We also sketch briefly how the thermodynamics and critical phenomena in finite systems may be treated.

INTRODUCTION

Over the past decade there has been an impressive progress in miniaturization techniques that aim ultimately at the fabrication of atomic-size devices whose features are controlled primarily by the quantal behaviour of a finite number of correlated particles [1]. Therefore, it is of interest to develop microscopic theoretical models that connect phenomena akin to few-body quantum systems to those occurring in the thermodynamic limit (large volume V, larger number of particle N and finite particle density $n = N/V$). In this work we explore differences and common features in the behaviour of small and extended systems. Two aspects are emphasized: the cooperative response of a system to an external perturbation and the treatment of critical phenomena in finite systems.

COLLECTIVE RESPONSE AND SHORT-RANGE DYNAMICS

The primary source of knowledge on a given system is provided by its characteristic response to external perturbations. In many cases this response is dependent on the collective behaviour of the constituents of the systems, as in the Faraday effect where the delocalized electrons in a metallic surface re-arrange among them self as to shield an external electric field. These correlated fluctuations of the density are determined by the so-called polarization operator $\Pi(\mathbf{q},\omega)$ which depends on the momentum $\mathbf{q}$ and the frequency ω. On the other hand the polarization of the medium modifies the properties of the particle-particle interaction $U(\mathbf{q},\omega)$. The modified potential U_{eff} is related to U and $\Pi(\mathbf{q},\omega)$ through the integral equation [2, 3]

$$U_{eff} = U + U\Pi U_{eff}. \tag{1}$$

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This relation can be formally written as

$$U_{eff} = \frac{U}{1 - U\Pi}. \tag{2}$$

The *screening* term $\kappa(\mathbf{q}, \omega) := 1/(1 - U\Pi)$ is usually called the generalized dielectric function [3] and plays a central role in a variety of phenomena. E.g. the electrical conductivity $\sigma(\mathbf{q}, \omega)$ of a plasma is obtained from $\kappa(\mathbf{q}, \omega)$ as $\sigma(\mathbf{q}, \omega) = i\omega(1 - \kappa)$. >From Eq.(2) it is clear that the determination of the renormalized interaction U_{eff} and of the dielectric function κ requires the knowledge of the polarization function Π. In essence Π is a two-point Green function that describes the particle-hole excitations. Its lowest order approximation Π_0 is provided by the random phase approximation (RPA) as

$$i\Pi_0(\mathbf{q}, \omega) = \frac{2}{(2\pi)^4} \int d\mathbf{p} d\xi G_0(\mathbf{q} + \mathbf{p}, \omega + \xi) G_0(\mathbf{p}, \xi). \tag{3}$$

Here G_0 is the free, single particle Green function. The evaluation of the integrals (3) can be performed analytically for a homogeneous system [3]. In this work we concentrate on the long wave-length limit (long-range screening) in which case one obtains $\Pi_0 \approx -2N(\mu)$ where $N(\mu)$ is the density of states at the Fermi level μ. If we are dealing with an electronic system, like a metallic cluster, the naked interaction $U(\mathbf{q})$ is given $U(\mathbf{q}) = 4\pi/q^2$ and according to Eq.(2) the screened interaction reads in the long wave-length limit

$$U_{eff} = \frac{4\pi}{(q^2 + 8\pi N(\mu))}. \tag{4}$$

The form of this potential in configuration space is obtained via a Fourier transform: $U_{eff} = \frac{e^{-r/\lambda}}{r}$. The advantage of this simplified form of the interaction is that it allows a transparent discussion of the nature of collisions from many-particle systems: In a charged two-particle scattering events with small momentum transfer (far collisions) dominate as deduced from the form factor of the naked potential $U \propto 1/q^2$. In scattering from a polarizable medium these events are cut out due to the finite range of the renormalized scattering potential U_{eff}, i.e. scattering occurs for close collisions where the medium is not able to screen the external field. For a detailed discussion of this point in the case of ionizing electron collisions from C_{60} we refer the interested reader to the work [5] of this volume. Here we would like to emphasize that in scattering processes from many electron systems it is important to account for the cooperative behaviour of the target electrons which results in screening. The reward for resolving the non-trivial task of evaluating the screening effects is that the interactions are then of a short-range and the description of the scattering dynamics can be done using standard methods of scattering theory, such as the first order Born approximation (see the discussion below). Thus, the real obstacle in describing low-energy collisions from many-particle systems is in obtaining an adequate expression for the polarization propagator.

For a uniform dense electron gas one can employ the RPA to obtain useful approximate expression for Π. For a real system such as a metal or semi-conductor surface one should however start with a realistic single particle Green function in Eq.(3) in order to derive the particle-hole excitations. Such a starting Green function can be obtained from

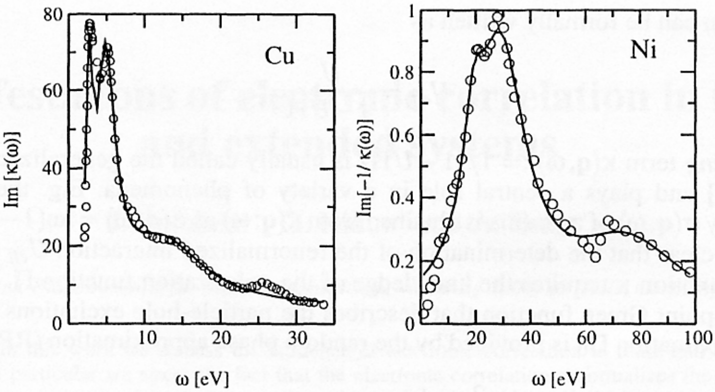

FIGURE 1. The calculated imaginary part of the (bulk) dielectric functions of Cu and the loss function of Ni are compared to the experimental values as deduced from the electron-energy loss spectra [6]. For Ni the experiment is done for a wave vector value of $q = (0.25, 0, 0)\,\frac{2\pi}{a}$ where the lattice constant a is $a = 6.65 a_0$ (a_0 is the Bohr radius).

density-functional theory within the local-density approximation. The strategy adopted here is the so-called *GW* method [4] which is conceptually well-known for a long time but computationally still poses a challenge, in particular in the case of surfaces. According to this method one derives first the dynamical response χ of the system. From this we deduce the screened interaction U_{eff}, as done above which in turn determine the so-called self-energy Σ and, via the Dyson equation, the interacting single-particle Green function. In matrix form this can be written as

$$\begin{aligned} \chi(\mathbf{r},\mathbf{r}',\tau) &= -iG(\mathbf{r},\mathbf{r}',\tau)G(\mathbf{r}',\mathbf{r},\tau) && (5)\\ U_{eff}(\mathbf{r},\mathbf{r}',\tau) &= U(\mathbf{r},\mathbf{r}') + \int d^3x \int d^3x'\, U(\mathbf{r},\mathbf{x})\chi(\mathbf{x},\mathbf{x}',\tau)U_{eff}(\mathbf{x}',\mathbf{r}',\tau) && (6)\\ \Sigma(\mathbf{r},\mathbf{r}',\tau) &= iG(\mathbf{r},\mathbf{r}',\tau)U_{eff}(\mathbf{r},\mathbf{r}',\tau) - \delta(\mathbf{r}-\mathbf{r}')V_{xc}(\mathbf{r}) && (7)\\ G(\mathbf{r},\mathbf{r}') &= G_0(\mathbf{r},\mathbf{r}') + \int d^3x \int d^3x'\, G_0(\mathbf{r},\mathbf{x})\Sigma(\mathbf{x},\mathbf{x}')G(\mathbf{x}',\mathbf{r}'). && (8) \end{aligned}$$

In these equations we operate in the configuration space and in the time domain. In principle, Eqs.(5-8) has to be solved self consistently starting from the Kohn-Sham Green function in Eq.(5) to arrive at the Green function (8) which is then inserted in Eq.(5). In this procedure, as done in Eq.(7), one should subtract the exchange and correlation potential to arrive at the correct self-energy.

Fig.1 shows the imaginary part of the dielectric function of Cu and the loss function of Ni as obtained from the above *GW* scheme [6]. As indicated by the results of Fig.1 the *GW* approach provides in some cases a reasonable description of the response of a many-body system.

The simple analytical form (3) of the polarization is based on the RPA expression for a uniform medium. The range of validity of such a treatment is estimated from its physical meaning: The interaction creates virtual electron-hole pairs. Within the RPA one considers these events to be incoherent, i.e. the phase of the electron-hole pair is lost right after it's creation and does not affect the creation of the next pair. This assumption is reasonable for systems with a large density of particles, for it is more probable to scatter from different particles consecutively than to undergo multiple scattering from the same scatterer. For a dilute system the RPA is no longer valid and one has to resort to other methods such as the ladder approximation for the polarization propagator [3] in which case the virtual electron-hole pair interacts repeatedly before it disappears. A similar situation of multiple coherent scattering is encountered for systems with few-interacting particles, say three or four electrons. The basic ideas concerning the renormalization of particle-particle interactions are still however valid. This can be seen from the following argument: The fundamental quantity that describes the dynamic of the system is the N-particle Green function $G^{(N)}$. Formally it satisfies the algebraic (Lippmann-Schwinger) relation

$$G^{(N)} = G_0 + G_0 U G^{(N)} = G_0 + G_0 U G_0 + G_0 U G_0 U G_0 + \cdots \tag{9}$$

where G_0 is a reference (solvable) system and the interaction U is given by $U = G_0^{-1} - (G^{(N)})^{-1}$. Usually, one aims at evaluating a limited number of certain terms of the perturbation series (9) involving the naked interaction U. However, Eq.(9) can also be written formally as

$$G^{(N)} = G_0 + G_0 \frac{U}{1 - G_0 U} G_0 = G_0 + G_0 U_{eff} G_0. \tag{10}$$

The operator $1/(1 - G_0 U)$ plays the role of the dielectric function in the RPA (cf. Eq.(2)). The essence of its effect is that it renormalizes the interaction U. As evident from Eq.(9) this renormalization procedure amounts to a sub-sum of all terms in the perturbation series up to the first order (Born) term. In practice G_0 is a diagonal many-body matrix, which is appropriately chosen. Analogous to the many-body case where one has to evaluate the polarization propagator, one needs to invert the matrix $1 - G_0 U$. Having done that the particle-particle interactions can be renormalized and a first-order (Born) treatment of the scattering dynamics is then sufficient.

THERMODYNAMICS PROPERTIES AND PHASE TRANSITIONS IN FINITE SYSTEMS

As stated above the properties of a system are encompassed in the Green function. In this section we point out that thermodynamic properties and critical phenomena and the cross over from the thermodynamic limit to confined small systems can also be described with the help Green function techniques. Strictly speaking, finite systems do not expose phase transitions [7]. However, one expect to observe the onset of a critical behaviour when the system approaches the thermodynamic limit. The traditional theory concerned with these questions is finite-size scaling theory [9]. Here we take another route, originally due to Yang and Lee[7] and Grossmann *et al.* [8] developed to treat critical phenomena in macroscopic systems.

The argument of Yang and Lee[7] is the following: Thermodynamical quantities, such as the specific heat C_V are obtained as a derivative with respect to the inverse temperature

β of the logarithm of the canonical partition function $Z(\beta)$;

$$C_V = \beta^2 \partial_\beta^2 \ln Z(\beta) = f(\beta, Z(\beta))/Z(\beta).$$

Here f is some analytical function and for the Boltzmann constant we assume $k = 1$. Therefore, divergences in the thermodynamic quantities, which signify phase transitions are connected to the zero points of $Z(\beta)$. These zero points are generally complex valued.

Grossman *et al.* [8] applied the concept of Yang and Lee to the canonical ensemble. In this case the inverse temperature $\beta = 1/T$ is continued analytically to $\beta = \Re(\beta) + i\Im(\beta)$. The phase transitions are then the crossings of the zero points line of $Z(\beta)$ with the real β axis. The crucial point is that in the thermodynamic limit $N \to \infty$, $V \to \infty$ and $v = V/N < \infty$ (V is the volume, N is the number of particles) the zero points approach to an infinitesimal small distance the real axis. For this reason, the characteristic phase-transition divergences appear in the thermodynamical quantities. For finite systems $Z(\beta)$ has only finite zero points which can not lie necessarily infinitely close the real axis. Therefore, the thermodynamic quantities show smooth peaks rather then divergences. The positions and widths of these peaks can be obtained from the real and imaginary parts of the zero points laying closest to the real axis [9].

To apply this approach to quantum finite systems, such as a Bose-Einstein condensate we consider a system of N interacting particles. The canonical partition function of a correlated system can be expressed in terms of the many-body Green function as

$$Z(\beta) = \int dE\, \Omega(E)\, e^{-\beta E}. \tag{11}$$

Here $\Omega(E)$ is the density of states which is related to the imaginary part of the trace of $G^{(N)}$ via

$$\Omega(E) = -\frac{1}{\pi} \Im\, \mathbf{Tr}\, G^{(N)}(E). \tag{12}$$

Therefore, as in the preceding sections the problem reduces to find appropriate expressions for the many-body Green functions. Recently it has been shown that the N-body Green function satisfies a recursion relation where the strength of interaction is successively reduced [10]:

$$G^{(N)} = \sum_{j=1}^{N} G_j^{(N-1)} - (N-1)G_0. \tag{13}$$

For a brief discussion of the limitations of this relation we refer to Ref.[10]. $G_j^{(N-1)}$ is the Green function of a system in which only $N-1$ particles are interacting while particle j is independent of all other particles. G_0 is a reference Green function of an independent-particle system. With the help of Eq.(13) one can construct flow equations to map the interacting system onto a non-interacting one in $N-1$ steps [11].

>From Eqs.(11,13) we deduce for the partition function the recursion relation

$$Z^{(N)} = \sum_{j=1}^{N} Z_j^{(N-1)} - (N-1)Z_0. \tag{14}$$

Here Z_0 is the partition function of the independent particle system (taken as a reference) while $Z_j^{(N-1)}$ is the canonical partition function of a system in which the interaction strength is diluted by cutting all interaction lines that connect to particle j.

Eq(14) allows to study the thermodynamic properties of finite systems on a microscopic level as well as to investigate the inter-relation between the thermodynamics and the strength of correlations. Critical phenomena can be studied using the idea put forward by Yang and Lee. For example if we are interested in the onset of condensation in a quantum Bose gas one should look at the ground-state occupation number $\eta_0(N,\beta)$. This is given by

$$\begin{aligned} \eta_0(N,\beta) &= -\frac{1}{\beta}\frac{\partial_{\varepsilon_0} Z^{(N)}(\beta)}{Z^{(N)}(\beta)} \\ &= -\frac{1}{\beta}\frac{\sum_{j=1}^{N} \partial_{\varepsilon_0} Z_j^{(N-1)} - (N-1)\partial_{\varepsilon_0} Z_0}{Z^{(N)}}. \end{aligned} \tag{15}$$

Here ε_0 is the ground-state energy. By means of this equation one can study systematically the influence of the interaction on the onset of the critical regime or one may chose to find the roots of Eq.(14) in the complex β plane and to identify the zero point that systematically approach the real β axis signifying the transition point in the thermodynamic limit.

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