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Electronic polarons, cumulants and doubly dynamical mean field theory: Theoretical spectroscopy for correlated and less correlated materials



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ABSTRACT

The use of effective local Coulomb interactions that are *dynamical*, that is, frequency-dependent, is an efficient tool to describe the effect of long-range Coulomb interactions and screening thereof in solids. The dynamical character of the interaction introduces the coupling to screening degrees of freedom such as plasmons or particle-hole excitations into the many-body description. We summarize recent progress using these concepts, putting emphasis on dynamical mean field theory (DMFT) calculations with dynamical interactions (“doubly dynamical mean field theory”). We discuss the relation to the combined GW+DMFT method and its simplified version “Screened Exchange DMFT”, as well as the cumulant schemes of many-body perturbation theory. On the example of the simple transition metal SrVO₃, we illustrate the mechanism of the appearance of plasmonic satellite structures in the spectral properties, and discuss implications for the low-energy electronic structure.

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1. Theoretical spectroscopy: from many-body perturbation theory to dynamical Hubbard interactions

Determining the behavior of a single electron in a periodic potential, created for example by the ions in a crystalline solid, is a textbook exercise of quantum mechanics. Determining the wave function of *all* the electrons in the solid, however, is an intractable many-body problem. The Pauli principle imposes full antisymmetry under exchange of any two electrons to this object, and electronic Coulomb interactions prevent it from being a simple Slater determinant.

The good news is that in practice the knowledge of the full many-body wave function of the inhomogeneous electron gas in the solid is barely necessary: the relevant electronic properties are determined by the low-energy response to external perturbations, and the knowledge of these low-energy excitations requires much less information than the full ground-state wave function. In this sense, solid state spectroscopies are a most efficient means for characterizing the properties of a solid state system. An important example are photoemission experiments – angle-resolved or angle-integrated – where information about the electron removal and addition spectra are obtained. Within the simplest possible

model for the photoemission process, the so-called “three-step model”, the photocurrent can be expressed in terms of the one-particle spectral function $A(k, \omega) = -\frac{1}{\pi} \text{Tr} \Im G(k, \omega)$, and computing this quantity from first principles, that is, without adjustable parameters, is one of the central challenges of modern theoretical spectroscopy.

Important progress has been achieved over the last decades within many-body perturbation theory: a first order expansion of the many-body self-energy Σ in the screened Coulomb interaction W [1,2] leads to a conceptually simple approximation $\Sigma = iGW$ which can be calculated within realistic electronic structure codes based on density functional theory (DFT). For reviews of successful applications of the GW approximation and developments based on it, we refer the reader to [3,4]. For more strongly correlated materials, where perturbative techniques reach their limits, the last 15 years have seen the development of a non-perturbative theory, combining dynamical mean field theory (DMFT) [5] with density functional theory. This so-called “DFT+DMFT” approach [6,7] builds on the success of DMFT for the description of lattice models for correlated fermions but extends its scope to the solid by treating a realistic (multi-orbital) Hamiltonian with effective local Coulomb interaction, often parametrized as Hubbard U and Hund’s J .

While DFT+DMFT – at least in its early implementations – can simply be understood as the DMFT solution of a multi-orbital Hubbard model (for reviews see [8–10], for some more modern

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implementations [11,12]) recent efforts have been spent in order to promote DMFT-based techniques to truly first-principles techniques [13]. This implies not only addressing the question of how to relate effective local Hubbard interactions to the full Coulomb interactions in the continuum (while taking care to avoid double-counting of screening), that is the *ab initio* calculation of the infamous effective local “Hubbard U”; since at the DFT level no rigorous distinction between contributions of “correlated degrees of freedom” and “uncorrelated” ones can be made, a truly double-counting free theory can only be achieved by eliminating the reference to the DFT Kohn-Sham Hamiltonian altogether. A successful route is the combination of Hedin’s GW approximation with DMFT, the so-called GW+DMFT method [14–17]. A summary of recent progress along these lines can be found in [13]; for most recent applications both, in the model and realistic electronic structure context we refer the reader to Refs. [18–22]. The common point between the GW method and the combined GW+DMFT scheme is the absence of adjustable interaction parameters. Both theories can be viewed as approximations to a free energy functional [23], where the free energy of the solid is written as a functional of the Green’s function G and the screened Coulomb interaction W . This implies that screening is described within the theory, instead of being introduced into it through an effective parameter. Besides the screened Coulomb interaction W , the GW+DMFT theory introduces an effective local interaction \mathcal{U} used as the bare interaction within an effective local model. The GW+DMFT equations require this interaction to be calculated self-consistently such as to reproduce the local part of the fully screened interaction W when the local model is solved by many-body techniques. This implies that the two interactions are related by a two-particle Dyson (or Bethe–Salpeter) equation $\mathcal{U}^{-1} - W_{loc}^{-1} = P_{loc}$, where P_{loc} is the polarisation function of the local problem. The physical content of this construction can be described as follows: instead of using the full long-range Coulomb interaction within a full continuum description, an effective local interaction \mathcal{U} is used within an effective local problem, but the interaction \mathcal{U} is determined such that the two problems reproduce the same *fully screened local interaction* W . The price to pay is that the effective interaction \mathcal{U} inherits from the fully screened interaction W its dynamical, i.e. frequency-dependent character (even though the bare interaction in the full Hilbert space, the bare Coulomb interaction, is frequency-independent).

Interestingly, this concept can be generalized and has proven useful even outside the GW+DMFT scheme. Namely, the full many-body problem can be simplified by eliminating some of the interacting degrees of freedom, at the price of introducing an effective dynamical interaction. The latter is determined from the requirement that when the resulting many-body problem is solved, the fully screened interaction is retrieved. In this work, we describe the different sources of frequency-dependence of effective local Hubbard interaction and investigate their effects on solid state spectroscopies. Section 2 discusses the dynamical character of effective interactions, while Section 3 presents the relation to coupled electron-boson Hamiltonians. Solving such a Hamiltonian within DMFT, that is, by mapping onto a local problem, consists in generalizing the usual DMFT concept to a “doubly dynamical” one, where not only the Weiss mean field is dynamical but also the effective local interactions. We abbreviate this “doubly dynamical mean field theory” in the following as “DDMFT”. Section 4 introduces approximate but very efficient and accurate concepts for solving the dynamical impurity model arising within DDMFT in the anti-adiabatic limit. Section 5 addresses implications for the resulting spectral functions, in particular with respect to satellite structures and spectral weight transfers – concepts that are then applied to the ternary transition metal oxide SrVO₃ in Section 6. A discussion of observable consequences of dynamical screening effects concludes this work.

2. Dynamical interactions: the concept of partial screening

The above equation for the effective local interaction \mathcal{U} can be rewritten as

$$W_{loc} = \frac{\mathcal{U}}{1 - P_{loc} \mathcal{U}} \quad (1)$$

stressing the interpretation of screening of the effective interaction \mathcal{U} by the dielectric function of the effective local problem:

$$\epsilon_{loc}^{-1} = \frac{1}{1 - P_{loc} \mathcal{U}}. \quad (2)$$

Alternatively, one can say that the screened interaction is “unscreened” by P_{loc} to obtain \mathcal{U} :

$$\mathcal{U} = \frac{W_{loc}}{1 + P_{loc} W_{loc}} \quad (3)$$

These observations have motivated generalizations of the concept of partial screening, where a many-body problem is solved in a two-step procedure: first, an effective Hamiltonian (or action) is constructed in a Hilbert space that is a subset of the original space. Finally, this effective many-body problem is solved with some suitable many-body technique. The bare interaction in the subspace is a partially screened interaction in the full space. In order to determine it, one needs some estimates for the fully screened interaction W and the polarization “at the second step”, the polarization $P_{\text{step}-2}$ of the effective many-body problem. Then, the effective interaction is constructed as

$$\mathcal{U} = \frac{W}{1 + P_{\text{step}-2} W}. \quad (4)$$

The most important example of such a “constrained screening approach” (see [25] for a more detailed discussion of the general philosophy) is the so-called “constrained random phase approximation” [24]. The cRPA provides an (approximate) answer to the following question: given the Coulomb Hamiltonian in a large Hilbert space, and a low-energy Hilbert space that is a subspace of the former, what is the effective *bare* interaction to be used in many-body calculations dealing only with the low-energy subspace, in order for physical predictions for the low-energy Hilbert space to be the same for the two descriptions? A general answer to this question not requiring much less than a full solution of the initial many-body problem, the cRPA builds on two approximations: it assumes (i) that the requirement of the same physical predictions be fulfilled as soon as in both cases the same estimate for the fully screened Coulomb interaction W is obtained and (ii) the validity of the random phase approximation to calculate this latter quantity.

The cRPA starts from a decomposition of the polarisation of the solid in high- and low-energy parts, where the latter is defined as given by all screening processes that are confined to the low-energy subspace. The former results from all remaining screening processes:

$$p^{\text{high}} = p - p^{\text{low}}, \quad (5)$$

One then calculates a partially screened interaction

$$W^{\text{partial}} = \epsilon_{\text{partial}}^{-1} \nu. \quad (6)$$

using the *partial* dielectric function

$$\epsilon_{\text{partial}}^{-1} = \frac{1}{1 - p^{\text{high}} \nu}. \quad (7)$$

Screening W^{partial} by processes that live within the low-energy space recovers the fully screened interaction W . This justifies the interpretation of the matrix elements of W^{partial} in a localized Wannier basis as the interaction matrices to be used as bare Hubbard interactions within a low-energy effective Hubbard-like Hamiltonian written in that Wannier basis.

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