Spectral Properties of Correlated Materials: Local Vertex and Non-Local Two-Particle Correlations from Combined GW and Dynamical Mean Field Theory

Thomas Ayral, Philipp Werner, and Silke Biermann

1Theoretical Physics, ETH Zurich, 8093 Zürich, Switzerland
2Centre de Physique Théorique, École Polytechnique, CNRS UMR7644, 91128 Palaiseau Cedex, France
3Institut de Physique Théorique (IPhT), CEA, CNRS, URA 2306, 91191 Gif-sur-Yvette, France
4Department of Physics, University of Fribourg, 1700 Fribourg, Switzerland
5Japan Science and Technology Agency, CREST, Kawaguchi 332-0012, Japan

We present a fully self-consistent combined GW and dynamical mean field (GW+DMFT) study of the spectral properties of the extended two-dimensional Hubbard model. The inclusion of the local dynamical vertex stemming from the DMFT self-energy and polarization is shown to cure the problems of self-consistent GW in the description of spectral properties. We calculate the momentum-resolved spectral functions, the two-particle polarization and electron loss spectra, and show that the inclusion of GW in extended DMFT leads to a narrowing of the quasi-particle width and more pronounced Hubbard bands in the metallic regime as one approaches the charge-ordering transition. Finally, the momentum-dependence introduced by GW into the extended DMFT description of collective modes is found to affect their shape, giving rise to dispersive plasmon-like long-wavelength and stripe modes.

Modern spectroscopic techniques are able to measure one- and two-particle spectra of condensed matter systems with remarkable precision, characterizing not only quasi-particle excitations but unveiling also satellite structures. Examples include Hubbard bands in photoemission spectroscopy, stemming from the atomic-like behavior of the electrons in partially filled narrow d- or f-shells [1], or collective excitations such as plasmonic features. Addressing such effects requires an accurate description of one- and two-particle spectral functions within the framework of many-body theory. The quantitative prediction of satellite features has even been used as a quality marker for many-body techniques. The failure of self-consistent perturbation theory in the screened Coulomb interaction, the self-consistent GW approximation, to describe plasmon satellites in the electron gas for example has provided arguments in favor of a non-self-consistent (“one-shot GW”) treatment [2, 3]. For real solids, few fully self-consistent calculations are available [4, 5], and no consensus concerning the virtues of self-consistency has been reached so far. A popular scheme, dubbed quasi-particle self-consistent GW (QPSC-GW) [6] yields reasonable estimates both for total energies and spectra. Yet, most of the calculations within this scheme were applied to semiconductors, and applications to correlated metals only start to appear [7]. The inclusion of an appropriate vertex correction is expected to resolve the ambiguities around the self-consistency question, and it has been in particular proposed that a combined GW and dynamical mean field scheme [8] would enable self-consistent calculations even for spectral properties. Early pioneering calculations on a three-dimensional extended Hubbard model [9, 10] have benchmarked several flavors of combined schemes along these lines. However, the numerical difficulty of solving the DMFT equations with frequency-dependent interactions has until recently prevented the direct investigation of spectral properties.

Implementing the combined GW+DMFT scheme in a fully self-consistent manner for the two-dimensional extended Hubbard model, we here demonstrate that this technique indeed successfully overcomes the deficiencies of GW. The implicit inclusion of a non-perturbative local vertex enables fully self-consistent calculations for spectral properties. In the correlated metal regime, the GW+DMFT self-energy encodes both, band renormalization effects and Hubbard satellite features. As expected from the physical ingredients, the theory also describes the Mott insulating state for strong local Coulomb interaction, which is inaccessible in GW alone, as well as the charge-ordered state driven by intersite interactions, absent from standard DMFT. In addition, dynamical screening effects give rise to plasmonic features in the local spectral function. While the local spectral functions in the intermediate to strong correlation regime are little affected by the non-local self-energy contributions stemming from the GW approximation, a substantial k-dependent modulation of the peak widths is observed in the momentum-resolved spectral functions. We analyze momentum-resolved two-particle spectra and show that the self-consistent combination of GW and EDMFT strongly affects the shape of collective modes, giving rise to dispersive plasmon-like long-wavelength modes and stripe modes.

We consider the half-filled extended Hubbard model...
on a two-dimensional square lattice
\[ H = -t \sum_{i,j,\sigma} \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} + \sum_i (U n_{i\uparrow} n_{i\downarrow} - \mu n_i) + \frac{V}{2} \sum_{i,j} n_i n_j, \]
where \( \hat{c}_{i\sigma} \) and \( \hat{c}_{i\sigma}^\dagger \) denote the annihilation and creation operators of a particle of spin \( \sigma = \uparrow, \downarrow \) at the lattice site \( i \), \( n_{i\sigma} = \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma} \), and \( n_i = n_{i\uparrow} + n_{i\downarrow} \). \( \sum_{i\neq j} \) is the sum over all nearest-neighbor sites, \( t > 0 \) is the hopping amplitude between two neighboring sites, \( \mu \) is the chemical potential, \( U \) the on-site repulsion between electrons of opposite spin and \( V \) the repulsion between two electrons on neighboring sites. The Fourier-transformed bare interaction term thus reads \( v_k = U + 2V (\cos(k_x) + \cos(k_y)) \). All energies are given in units of the half-bandwidth \( D = 4t \). We show results for inverse temperature \( \beta D = 100 \), restricting our study to the paramagnetic phase.

The GW+DMFT approach is derivable from a free energy functional [11]. The Legendre transform of the free energy with respect to the Green’s function \( G \) and the screened interaction \( W \) can be expressed as a sum of the Hartree-Fock part and a Luttinger-Ward-like correlation functional \( \Psi[G, W] \), which sums up all skeleton diagrams built from \( G \) and \( W \) [12]. The GW+DMFT scheme consists in approximating \( \Psi \) as \( \approx \Psi_{\text{EDMFT}}[G_{\text{ii}}, W_{\text{ii}}] + \Psi_{\text{GW, nonloc}}[G_{ij}, W_{ij}] \), where the first term is calculated from a (dynamical) impurity problem as in extended dynamical mean field theory (EDMFT) [13][15] and the second term is the non-local (\( i \neq j \)) part of the GW functional \( \Psi_{\text{GW, nonloc}}[G_{ij}, W_{ij}] \). The GW+DMFT scheme self-consistently constructs the Green’s function \( G \) and the screened interaction \( W \) of the system as a stationary point of the free energy functional. The self-energy \( \Sigma \) and polarization \( P \) are formally obtained by functional differentiation of \( \Psi \) with respect to \( G \) and \( W \), leading to the expressions
\[ \Sigma(k, \omega) = \Sigma_{\text{imp}}(\omega_n) + \Sigma_{\text{GW, nonloc}}(k, \omega) \quad \text{and} \quad P(k, \omega_n) = P_{\text{imp}}(\omega_n) + P_{\text{GW, nonloc}}(k, \omega_n). \]
These, in turn, are used as inputs to a dynamical impurity model, which we solve using a continuous-time Monte Carlo algorithm [16] [18] to obtain updated local self-energies. The whole scheme is iterated until convergence. The calculations have been performed on a 64 \times 64 momentum grid, while the analytical continuation of the imaginary-time data has been performed using the Maximum Entropy method [19]. We monitor the following quantities: (i) the local spectral function \( A_{\text{loc}}(\omega) = \frac{1}{\pi} \text{Im} G_{\text{loc}}(\omega) \), (ii) the local and non-local self-energy, (iii) the local and non-local polarization, (iv) the electron energy-loss spectrum (EELS) \( \text{Im} [-\epsilon(k, \omega)^{-1}] \) where \( \epsilon \) is the dielectric function, \( \epsilon(k, \omega) = 1 - v_k P(k, \omega) \).

Within extended DMFT and GW+DMFT, in the absence of intesre repulsion, the Mott transition takes place at \( U_c \approx 2.5 \). This value is slightly modified by intesre repulsions \( V < V_c \). At \( V_c \) a transition to a charge-ordered phase occurs. In the following we study the local spectral properties in the metallic phase with weak \( (U = 0.5, V = 0.1) \) and intermediate \( (U = 1.5, V = 0.4) \) interactions as well as in the Mott insulator at \( U = 3.5 \) and \( V = 3 \). Figure 1 shows the local spectral function \( A_{\text{loc}}(\omega) \) obtained within (i) self-consistent EDMFT, (ii) self-consistent GW+DMFT, (iii) self-consistent GW and (iv) quasi-particle self-consistent GW (QPSG-GW). The latter scheme was implemented by computing the lattice Green’s function from the GW self-energy via \( G(k, \omega) = \omega_n Z_k \omega_n \approx \epsilon(k, \omega) - \epsilon(k, \omega) \approx \epsilon(k, \omega) \approx \epsilon(k, \omega) \approx \epsilon(k, \omega) \), where \( Z_k \approx 1 - \text{Im} \Sigma_{\text{GW}}(k, \omega) \), which is the quasi-particle weight as estimated from the value of the self-energy at the first Matsubara frequency. For small interactions (panel 1a), correlation effects are negligible, and the four schemes result in indistinguishable spectra within the numerical accuracy. As the local interaction \( U \) becomes significant (panel 1b) deviations start to appear, with EDMFT and GW+DMFT exhibiting stronger correlation effects. Upon increasing local interactions (panel 1c), the quasi-particle renormalization becomes stronger, the width of the coherent central peak shrinks, and the corresponding spectral weight is transferred to higher energies. This – physically expected – behavior is realized by the EDMFT and GW+DMFT spectra, which exhibit higher-energy structures at \( \omega \approx \pm U/2 \) already for \( U = 1.5 \). The Hubbard bands gain spectral weight as \( U \) increases further. Interestingly, they are more pronounced within GW+DMFT than within EDMFT. Finally, at \( U = 3.5 \), a Mott gap has opened,
and EDMFT and GW+DMFT spectra are similar. In addition to the two Hubbard bands, the EDMFT and GW+DMFT spectra display two symmetric high-energy satellites, whose spectral weight depends on the intersite interaction \( V \). The QPSC-GW spectra display only a weak renormalization of the bandwidth as \( U \) increases from the weak to the strong coupling limit, and at all correlation levels the spectra remain metallic. The same is true within the self-consistent GW method, although with increasing correlations some spectral weight is shifted to higher frequencies, albeit in a featureless way.

These observations show that both self-consistent GW approaches yield a correct result only in the weak-correlation regime. As correlations increase, GW fails to describe the shift of spectral weight to high-energy incoherent bands, present in DMFT. We note that in the local GW+DMFT spectra the Hubbard bands are enhanced compared to the EDMFT or GW spectra. This effect can be ascribed to the self-consistency, which allows the local quantities to re-adjust to the non-local self-energies \( \Sigma_{GW} \) and \( P_{GW} \).

Another salient characteristic of EDMFT and GW+DMFT spectra is the presence of additional high-energy satellites in the Mott phase. These directly reflect the frequency-dependence of the local interactions \( U(\omega) \) induced by the nearest-neighbor repulsion term \( V \). As demonstrated in Ref. [20], a pole in \( U(\omega) \) (such as a plasmon pole) leads to multiple satellites in the local spectral function. In our case, the local interactions \( U(\omega) \) are characterized by a broad continuum of poles centered at some energy \( \omega_G \), resulting in only two symmetric satellites in the Mott spectra. In the metallic phase, these satellites are present, but they are broad and merged with the Hubbard bands, making them hardly distinguishable.

The failure of both self-consistent GW schemes to capture Hubbard bands or high-energy satellites is consistent with the well-known observation that self-consistency in GW for the homogeneous electron gas results in a smearing out and displacement of high energy satellite features [2]. In light of this observation, most modern GW schemes therefore adopt a “best-G-best-W” strategy, rather than aiming at full self-consistency. Figure 2 illustrates the virtues and limitations of this strategy by displaying the spectra obtained in different one-shot GW schemes: (i) in “\( G_0W \)”, the non-interacting Green’s function \( G_0 \) and the converged GW+DMFT \( W \) are taken as inputs to a one-shot GW calculation, (ii) “\( GW_0 \)” takes the converged GW+DMFT \( G \) and \( W_0 = v(1 - vG_0G_0)^{-1} \) within the random phase approximation as inputs and (iii) “best G, best W” takes the converged GW+DMFT \( G \) and \( W \) as inputs. At all correlation levels (\( U = 0.5 \) to \( U = 3.5 \)), these three GW schemes produce results very similar to self-consistent GW. In particular, they remain metallic. Even the “best G, best W” scheme in the Mott phase (\( U = 3.5 \)) yields a metallic self-energy, despite the Mott-like character of the input \( G \) and \( W \). This phenomenon is due to the lack of Hedin’s three-legged vertex \( \Lambda \) in GW schemes, as shown in the lower-right panel of Figure 2. There, an estimate of the local part of \( \Lambda \) is computed from EDMFT results at \( V = 0 \). Remembering that, schematically, the irreducible vertex function \( \Lambda \) appears in the self-energy as \( \Sigma = GWA \) [21], a rough estimate – neglecting the true frequency structure – is computed as follows: one computes an effectively vertex-corrected screened interaction \( \widetilde{W}(\tau) = \Sigma_{imp}(\tau)/G_{imp}(\tau) \) from EDMFT, then Fourier-transforms it to \( \widetilde{W}(i\nu_n) \); finally, the static vertex estimate is obtained as \( \Lambda(0) \approx \widetilde{W}(i\nu_n)/W_{loc}(i\nu_n) \). Crude as it is (the full vertex depends on two independent frequencies), this estimate nonetheless clearly demonstrates the
role of vertex corrections for the Mott transition: from unity in the weakly correlated regime, it increases with $U$ until it diverges at the Mott transition. This indicates that within the language of Hedin’s equations, the divergence of the local vertex is the driving force of the Mott phenomenon, making any vertex-less approximation unfit to capture it.

The effect of the non-local GW contributions on EDMFT are illustrated by the momentum-resolved spectral functions, displayed in Figure 3. Generally, the dispersion of the Hubbard bands follows the dispersion of the quasi-particle peak, within both schemes. In the presence of a strong intersite interaction, the non-local self-energy and polarization lead however to an additional $k$-dependent modulation of the linewidth and weights. This is illustrated in the lower panels of Fig. 3 for the $(\pi, \pi)$ point, where a pronounced sharpening of the quasi-particle peak is observed along with enhanced weight of the Hubbard bands.

We now turn to a study of two-particle quantities. Figure 4 shows the momentum-resolved imaginary part of the polarization and the electron energy loss (EELS) spectrum $\text{Im} [\epsilon^{-1}(k, \omega)]$ in the metallic regime. Within EDMFT, the polarization displays a broad mode which reflects the particle-hole excitations of the system. They are centered at $U/2$, reflecting the emergence of the Hubbard bands and the corresponding excitations between Hubbard bands and the quasi-particle peak. In contrast, the polarization spectrum within GW+DMFT is dispersive. While displaying sharper features close to the $\Gamma \equiv (0,0)$ point, it captures particle-hole excitations due to Fermi-surface nesting at wave-vector $(\pi, \pi)$, as well as the zero-sound mode at long wavelengths and low energies. The EELS spectrum contains the particle-hole excitations (poles of the polarization) of the system and its collective modes, which correspond to the solutions of $\text{Re} P(k, \omega) = 1/v_k$. These collective modes are damped out close to particle-hole excitations (when $\text{Im} P(k, \omega)$ is large). This analogue of the free-electron-gas Landau damping occurs at the $(\pi, \pi)$ point in EDMFT and GW+DMFT. It can be directly ascribed to the nearest-neighbor repulsion, which induces scattering along this direction. The energy and lifetime of this collective excitation differs from EDMFT to GW+DMFT. In GW+DMFT it is lower in energy, more dispersive and with a larger lifetime. In GW+DMFT, two modes are visible above the $(\pi, 0)$ point, indicating the existence of two stripe modes at energies $\omega = 1$ and $\omega = 2$ corresponding to stripe-like modulations, where the sign of the density fluctuation varies from row to row in the $x$-direction. For obvious reasons, they are not captured by EDMFT. These two-particle excitations are directly related to the screening in the system as the screened interaction, $W$, is given by $W(k, \omega) = \epsilon^{-1}(k, \omega)v_k$. In particular, they explain the retardation effects at play in the local interactions $U(\omega)$ and the corresponding satellites in the local spectra.

In conclusion, we have demonstrated how the ambiguities on the optimal degree of self-consistency in many-body perturbation theory are resolved by including a non-perturbative local vertex in the calculation. Based on a fully self-consistent implementation of the combined GW+DMFT scheme, we have analyzed one- and two-particle satellite features in correlated materials. While we confirm the well-known “washing out” of satellite features in self-consistent GW calculations, self-consistent GW+DMFT does not suffer from this deficiency. Plasma- and zero-sound-like oscillations involving itinerant carriers as in the electron gas survive only in the regime of small local Coulomb interactions, but are quickly suppressed in the correlated metal. In this regime, excitations related to the creation of doublons become dominant. While local spectral functions are little affected by non-local contributions in a wide range of parameters, the momentum-resolved spectra display a $k$-dependent modulation of the width of the peaks; the momentum-dependence introduced by the GW part becomes truly crucial when assessing dispersions of two-particle spectral properties, differentiating in particular the nature of the collective modes in the $(0, 0)$, $(0, \pi)$, and $(\pi, \pi)$ directions. Our findings have implications for the nature of satellite features in correlated materials. In particular, it becomes obvious that electron-gas-like plasmons in materials stem dominantly from the charge contained in completely filled shells (that is from multi-orbital effects), while partially filled shells give rise to doublon excitations of the kind we describe. The interplay of local correlations and charge ordering phenomena and their intriguing wave-vector dependence call for an extension of our study to realistic two-dimensional systems: recent experimental findings of charge ordering in cuprates [22], cobaltates [23] or in systems of adatoms on.

Figure 4: $U = 2, V = 0.4$. Upper panels: $\text{Im} P(k, \omega)$ within EDMFT (left) and GW+DMFT (right). Lower panels: $\text{Im} \epsilon^{-1}(k, \omega)$ within EDMFT (left) and GW+DMFT (right).
surfaces [24] are prominent examples.

We acknowledge useful discussions with F. Aryasetiawan, M. Casula, A. Georges, P. Hansmann, M. Imada, M. Katsnelson, A. Millis, and T. Miyake. This work was supported by the French ANR under project SURMOTT, GENCI/IDRIS Orsay under project 1393, by DFG FOR 1346 and by the Swiss National Science Foundation (grant PP0022-118866). Most of the calculations have been performed on the Brutus cluster at ETH Zurich, using a code based on ALPS [25]. We thank L. Boehnke for allowing us to use his MaxEnt code.

[1] M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. 70, 1039 (1998).
[2] U. von Barth and B. Holm, Phys. Rev. B 54, 8411 (1996).
[3] B. Holm and U. von Barth, Phys. Rev. B 57, 2108 (1998).
[4] W. Ku and A. G. Eguiluz, Phys. Rev. Lett. 89, 126401 (2002).
[5] A. Kutepov, K. Haule, S. Y. Savrasov, and G. Kotliar, Phys. Rev. B 82, 045105 (2010).
[6] T. Kotani, M. van Schilfgaarde, and S. V. Faleev, Phys. Rev. B 76, 165106 (2007).
[7] A. Kutepov, K. Haule, S. Y. Savrasov, and G. Kotliar, arXiv (2011), 1112.0214.
[8] S. Biermann, F. Aryasetiawan, and A. Georges, Phys. Rev. Lett. 90, 086402 (2003).
[9] P. Sun and G. Kotliar, Phys. Rev. B 66, 085120 (2002).
[10] P. Sun and G. Kotliar, Phys. Rev. Lett. 92, 196402 (2004).
[11] S. Biermann, F. Aryasetiawan, and A. Georges, Physics of Spin in Solids: Materials, Methods, and Applications (Kluwer Academic Publishers B.V., 2004), pp. 43–65, NATO Science Series II, available electronically as arXiv:0401653.
[12] C. Almbladh, U. von Barth, and R. van Leeuwen, Int. J. Mod. Phys. B 13, 535 (1999).
[13] A. M. Sengupta and A. Georges, Phys. Rev. B 52, 10295 (1995).
[14] Q. Si and J. L. Smith, Phys. Rev. Lett. 77, 3391 (1996).
[15] H. Kajueter, Ph.D. thesis, Rutgers University, New Brunswick (1996).
[16] G. Baym, Phys. Rev. 127, 1391 (1962).
[17] P. Werner and A. J. Millis, Phys. Rev. Lett. 99, 146404 (2007).
[18] P. Werner and A. J. Millis, Phys. Rev. Lett. 104, 146401 (2010).
[19] M. Jarrell and J. Gubernatis, Physics Reports 269, 133 (1996), ISSN 0370-1573.
[20] M. Casula, A. Rubtsov, and S. Biermann, Phys. Rev. B 85, 035115 (2012).
[21] L. Hedin, Phys. Rev. 139, A796 (1965).
[22] T. Wu, H. Mayaffre, S. Kramer, M. Horvatic, W. N. Berthier, Claude and-Hardy, R. Liang, D. A. Bonn, and M.-H. Julien, Nature 477, 191 (2011).
[23] I. R. Mukhamedshin, H. Alloul, G. Collin, and N. Blanchard, Phys. Rev. Lett. 94, 247602 (2005).
[24] P. Hansmann, L. Vaugier, and S. Biermann (2012), submitted to J. Phys. Cond. Matt.
[25] B. Bauer, L. D. Carr, H. G. Evertz, A. Feiguin, J. Freire, S. Fuchs, L. Gamper, J. Guikelerger, E. Gull, S. Guertler, et al., Journal of Statistical Mechanics: Theory and Experiment 2011, P05001 (2011).