The two-particle propagator and magnetic susceptibility in the Hubbard model

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Abstract
We treat the two-particle Green function in the Hubbard model using the recently developed τ-CPA, a hybrid treatment that applies the coherent potential approximation (CPA) up to a time τ related to the inverse of the bandwidth, after which the system is averaged using the virtual crystal approximation. This model, with suitable approximations, does predict magnetism for a modified Stoner criterion. The evaluation of the two-particle propagator in the τ-CPA requires the solution of the pure CPA, within whose formalism the vertex correction and the weighted Green functions are obtained. The dynamical susceptibility, including the vertex correction and the weighted scattering by the residual interaction, is calculated and shows a spin wave spectrum in the ferromagnetic regime.

1. Introduction

In the previous paper [1], to be referred to as I, we developed an improved treatment of the magnetic state of the simple Hubbard model, called the τ-CPA. This was constructed as intermediate between the mean field Stoner model, equivalent to a virtual crystal approximation (VCA) treatment of the energy bands, and the Hubbard's alloy analogy method, which is equivalent to the CPA (literature on the original and subsequent developments of the CPA is to be found in [2–5]). The single-particle Green functions, and the properties such as the density of states and magnetization which can be developed from them, are given in I and will be used here without further derivations.

Several important magnetic properties in metallic systems, such as the susceptibility, depend on two-particle propagators. The treatment of these is straightforward in VCA-type theories but within the CPA method a number of complications arise. In particular, because of the correlated scattering of the two particles from a single site, a vertex correction has to be included. The theory of such systems in the alloy situation was developed by Velický [6] in the 1960s and applied to the calculation of the electrical conduction. More recently, Schwabe

and Elliott [7, 8] have shown how to derive weighted averages of such propagators which they applied to the theory of electron–hole pairs combined to form excitons.

In the present problem the susceptibility is related to the propagation of a pair of electrons of opposite spin, but because electrons of different types see different scattering centres these theories have to be generalized to the case of a four-component alloy. In addition, the residual interaction between electrons of opposite spin on the same site has to be included separately and is the dominant cause of the appearance of spin waves within the model.

These properties are examined in the pure Hubbard CPA case which, as has been pointed out [9, 10], has only a paramagnetic phase except in special circumstances. The treatment can be carried over into the τ-CPA, again requiring a matching of the propagators, for $t < \tau$ treated in the CPA with their vertex corrections, to the VCA solutions appropriate to longer times. A full treatment of this situation with further approximations to improve the tractability does indeed predict the existence of a broadened spin wave branch in the magnetic response functions.

The equation of motion for the two-particle propagator needed for the susceptibility is developed in section 2. Section 3 treats the two-particle Green function in the pure CPA. In section 4 the results are combined in order to construct the solution in the τ-CPA. The dynamical susceptibility is then calculated in section 5.

2. Two-particle Green functions in the Hubbard model

As was done in I we consider the Hubbard model

$$H = H_0 + U \sum_i c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} \qquad \text{with } H_0 = \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^+ c_{j\sigma} \tag{1}$$

for N spins at temperature $T = 0$. The free part of the Hamiltonian H_0 is evaluated in the Bloch states $H_0 = \sum_k \epsilon_k c_{k\sigma}^+ c_{k\sigma}$ using a tight-binding approximation for the band structure ϵ_k. In the linear response theory the induced moment is given in term of the susceptibility tensor $\chi^{\mu\nu}(\mathbf{x} - \mathbf{x}', t - t')$, $\mu, \nu \in \{x, y, z\}$. It is completely determined by the longitudinal susceptibility χ^{zz} and the transverse susceptibilities χ^{-+} and χ^{+-}. We will concentrate in this paper on the transverse susceptibilities, which are given as

$$\chi^{-\alpha\alpha}(\mathbf{x} - \mathbf{x}', t - t') = \mathrm{i}2\pi\theta(t - t')\langle[\sigma^{-\alpha}(\mathbf{x}, t), \sigma^\alpha(\mathbf{x}', t')]\rangle \tag{2}$$

where $\alpha \in \{+, -\}$ and $\sigma^+(x) = c_{x\uparrow}^\dagger c_{x\downarrow}$, $\sigma^-(x) = c_{x\downarrow}^\dagger c_{x\uparrow}$. The general Green functions we want to evaluate are therefore of the type

$$K_{ijkl}(t) = \langle\langle c_{i\downarrow}^\dagger(t) c_{j\uparrow}(t); c_{k\uparrow}^\dagger c_{l\downarrow}\rangle\rangle \tag{3}$$

with $\chi^{-+}(x, t) = -K_{xx00}(t)$. The equation of motion for K_{ijkl} in the Hubbard Hamiltonian reads

$$E K_{ijkl} = T_0[K] + U\langle\langle(c_{i\downarrow}^\dagger c_{j\uparrow}\hat{n}_{j\downarrow} - \hat{n}_{i\uparrow}c_{i\downarrow}^\dagger c_{j\uparrow}); c_{k\uparrow}^\dagger c_{l\downarrow}\rangle\rangle_E \tag{4}$$

$$T_0[K] := \delta_{jk}\langle c_{i\downarrow}^\dagger c_{l\downarrow}\rangle - \delta_{il}\langle c_{k\uparrow}^\dagger c_{j\uparrow}\rangle + \sum_n t_{jn} K_{inkl} - \sum_n t_{ni} K_{njkl} \tag{5}$$

where $T_0[K]$ is the free part of the equation of motion and $\hat{n}_{i\sigma}$ the particle number operator $c_{i\sigma}^\dagger c_{i\sigma}$. A usual procedure for closing the equation of motion is to do a substitution $\hat{n}_{i\sigma} \to n_{i\sigma}$, where $n_{i\sigma}$ is a scalar. In the simplest approximation $\hat{n}_{i\sigma}$ is replaced by the average number of spins N^σ. This leads to the VCA treatment (or the Stoner model). A more sophisticated approach consists in using the analogy between the Hubbard model and the binary alloy [11] and replacing $\hat{n}_{i\sigma}$ by 0 or 1. This leads to the CPA treatment developed in the next section.

However, replacing the operators by numbers must be done carefully, as for $i = j$ the two interaction terms in (4) are both zero. In order to conserve this feature we write (4) as

$$EK_{ijkl} = T_0[K] + U\langle\langle(n_{j\downarrow} - n_{i\uparrow})c_{i\downarrow}^\dagger c_{j\uparrow}; c_{k\uparrow}^\dagger c_{l\downarrow}\rangle\rangle_E - U\delta_{ij}\langle\langle(n_{j\downarrow} - n_{i\uparrow})c_{i\downarrow}^\dagger c_{j\uparrow}; c_{k\uparrow}^\dagger c_{l\downarrow}\rangle\rangle_E.$$
(6)

The evaluation of the two-particle propagator in the Hubbard model using the τ-CPA methods requires the solution of (6) in the pure CPA and in the pure VCA case. The latter case is straightforward; the former is the subject of the next section.

3. Two-particle Green function in the pure CPA

In this section $n_{i\sigma}$ in (6) takes the value 1 or 0, depending on whether there is a spin σ at i or not. We first solve the equation of motion ignoring the last term in (6), and obtain an expression for the weighted Green functions. The residual interaction is then included by means of a weighted expansion.

3.1. The four-component alloy

If we leave aside the last term of (6), the model (1) can be reduced to

$$H_{\text{red}} = \sum_{i,j,\sigma} t_{ij}c_{i\sigma}^\dagger c_{j\sigma} + \sum_{l,\sigma} \zeta_{l\sigma}c_{l\sigma}^\dagger c_{l\sigma} \qquad \text{with } \zeta_{j\uparrow} = n_{j\downarrow}U \quad \text{and} \quad \zeta_{i\downarrow} = n_{i\uparrow}U.$$
(7)

The rest of this subsection is dedicated to finding an approximation for K_{ijkl}^0, where K_{ijkl}^0 is a solution of the reduced equation of motion

$$EK_{ijkl}^0 = T_0[K^0] + U(n_{j\downarrow} - n_{i\uparrow})\langle\langle c_{i\downarrow}^\dagger c_{j\uparrow}; c_{k\uparrow}^\dagger c_{l\downarrow}\rangle\rangle_E.$$
(8)

The quadratic Hamiltonian (7) allows us to decompose the two-particle Green function into pairs of one-particle Green functions $G(z)$. Moreover, it is now possible to apply the CPA formalism directly [2]. The kinds of random energies defined in (7) suggest the following extension of the binary alloy analogy to the two-particle case. As seen from (7), an up spin travelling on the lattice is interacting only on the sites where there is a down spin. This can be interpreted in the CPA as a particle, for example a spin up, moving in the lattice and meeting either a 'host' (site with no spin down) or an 'impurity' (site with a spin down). Since a travelling spin up does not see the same 'hosts' and 'impurities' as a spin down, we need to define the lattice as being made of four kinds of sites rather than just two. Let n^+, n^-, $n^\pm$ and $n^\emptyset$ be respectively, for each site, the probability of finding a single electron of spin up, a single electron of spin down, both one spin up and one spin down, and no electron at all. The set $\{n^+, n^-, n^\pm, n^\emptyset\}$ defines the four-component alloy for the system. Thus $N^\uparrow = n^+ + n^\pm$ is the concentration of 'impurities' seen by a down spin and $N^\downarrow = n^- + n^\pm$ the one seen by an up spin. The CPA equation can be written in the form [2]

$$\frac{N^{-\sigma}(U - \Sigma^\sigma)}{1 - (U - \Sigma^\sigma)F^\sigma} - \frac{(1 - N^{-\sigma})\Sigma^\sigma}{1 + \Sigma^\sigma F^\sigma} = 0$$
(9)

where F^σ represents the trace of $\bar{G}^\sigma$, the CPA approximation for the one-particle propagator. F^σ and the self-energy Σ^σ must be solved self-consistently for both $\uparrow$ and $\downarrow$, along with the equilibrium condition on $N^\uparrow$ and $N^\downarrow$.

Within (7) the two-particle Green function decouples as products of a correlation functions and Green functions. K_{ijkl}^0 can therefore be written in terms of the one-particle retarded

propagator $G(z)$ at temperature $T = 0$ in the following manner:

$$K_{ijkl}^0(E) = \frac{i}{2\pi} \int_{-\infty}^{\mu} d\omega \, [G^{\uparrow}(j, k; \omega + E) \, G^{\downarrow}(l, i; \omega) - G^{\uparrow}(j, k; \omega + E) \, G^{*\downarrow}(l, i; \omega)$$
$$+ G^{\uparrow}(j, k; \omega) \, G^{*\downarrow}(l, i; \omega - E) - G^{*\uparrow}(j, k; \omega) \, G^{*\downarrow}(l, i; \omega - E)]. \tag{10}$$

This expression must now be averaged over all possible configurations of spins. This involves the evaluation of quantities such as $\langle G^{\uparrow}(z1)G^{\downarrow}(z2)\rangle$. The up spins do not see the same impurities as the down spins but are nevertheless subject to statistical correlations due mainly to the doubly occupied sites and $\langle G^{\uparrow}(z1)G^{\downarrow}(z2)\rangle \neq \langle G^{\uparrow}(z1)\rangle\langle G^{\downarrow}(z2)\rangle$. The vertex corrections arising from statistical correlations are calculated below.

3.2. Vertex corrections for the four-component alloy

In this part the vertex correction is developed for a four-component alloy that is consistent with the CPA. This is a straightforward generalization of the binary alloy case obtained by Velický [6]. We adopt Velický's procedure and look into evaluating $G^{(2)}(z_1, z_2) := \langle G^{\uparrow}(z_1)CG^{\downarrow}(z_2)\rangle$, where C in our case is a diagonal operator, since for the magnetic susceptibility only quantities of the type $\langle G_{l,m}^{\uparrow}(z_1)G_{m,l}^{\downarrow}(z_2)\rangle$ are needed. The result, after directly following Velický's algebra, can be summarized as follows:

$$G_k^{(2)}(z_1, z_2) = \frac{a_k(z_1, z_2)}{1 - \Lambda(z_1, z_2)a_k(z_1, z_2)} \qquad \text{with } a_k(z_1, z_2) = \frac{1}{N} \sum_q \bar{G}_{q+k}^{\uparrow}(z_1)\bar{G}_q^{\downarrow}(z_2). \tag{11}$$

The vertex correction, as a function of the scattering matrix elements S_n^{σ},

$$\Lambda(z_1, z_2) = \frac{\langle S_n^{\uparrow}(z_1)S_n^{\downarrow}(z_2)\rangle}{1 + F^{\uparrow}(z_1)\langle S_n^{\uparrow}(z_1)S_n^{\downarrow}(z_2)\rangle F^{\downarrow}(z_2)}, \tag{12}$$

gives the correlated scattering of the two particles for the same sites. We recall that the CPA result for the S_ns is $S_n(z) = (\zeta_n - \Sigma(z))[1 - [\zeta_n - \Sigma(z)]F(z)]^{-1}$, where ζ_n is the random potential felt at site n by the travelling particle. The evaluation of Λ for the four-component alloy follows again on the same lines as for the binary alloy in the single-site representation. The average $\langle S_n^{\uparrow}(z_1)S_n^{\downarrow}(z_2)\rangle$ is obtained by summing the contributions from the four species on the lattice. Using the CPA result $F^{\sigma} = (\Sigma^{\sigma} - N^{-\sigma}U)[\Sigma^{\sigma}(U - \Sigma^{\sigma})]^{-1}$, we obtain the following vertex correction:

$$\Lambda^{-1} = \frac{1}{n^{\pm} - N^+N^-} \left(\frac{N^+N^-n^0}{\Sigma^{\uparrow}\Sigma^{\downarrow}} + \frac{n^+N^-(1 - N^+)}{\Sigma^{\uparrow}(U - \Sigma^{\downarrow})} \right.$$
$$\left. + \frac{n^-N^+(1 - N^-)}{\Sigma^{\downarrow}(U - \Sigma^{\uparrow})} + \frac{n^{\pm}(1 - N^+)(1 - N^-)}{(U - \Sigma^{\downarrow})(U - \Sigma^{\uparrow})} \right). \tag{13}$$

In the limit $U \to 0$, $\langle S_n^{\uparrow}S_n^{\downarrow}\rangle = \langle S_n^{\uparrow}\rangle\langle S_n^{\downarrow}\rangle = 0$, so that Λ tends to zero. $n^{\pm}$ takes the same value as in the purely random case where the two particles up and down are not interacting with anything, that is $n^{\pm} \to N^+N^-$. In this respect we note that the vertex correction, with its factor $(n^{\pm} - N^+N^-)$ in the numerator, gives a measure of the deviation from the random value.

In the limit $U \to \infty$, $\Sigma^{\sigma} \to (-N^{-\sigma})[F^{\sigma}]^{-1}$, so that $\Lambda \to (n^{\pm} - N^+N^-)[n^0 F^{\uparrow}F^{\downarrow}]^{-1}$. In this limit the interaction should prevent any double occupation of the same site, so that we expect $n^{\pm} \to 0$. The resulting Λ is proportional to both the concentration of species $\uparrow$ and $\downarrow$, and inversely proportional to the proportion of vacant sites. The large U limit is conveniently obtained in conjunction with the limit $w \to 0$, where w is half the bandwidth. This limit is to be treated with caution when dealing with the susceptibility, since no transitions are possible if

the bands have no width. It is nonetheless possible to get an idea of what will be the respective weights associated with the bands in the split-band limit. In this limit, the behaviour of the Green function in the CPA is given by [7] $\bar{G}^\sigma(E) = (1 - N^{-\sigma})/E + N^{-\sigma}/(E - U)$, so that the two-particle Green function $G_0^{(2)}(E_1, E_2) = G^\uparrow(E_1)G^\downarrow(E_2)$ becomes

$$G_0^{(2)}(E_1, E_2) = \frac{N^\uparrow N^\downarrow}{(E_1 - U)(E_2 - U)} + \frac{N^\uparrow(1 - N^\downarrow)}{E_1(E_2 - U)} + \frac{(1 - N^\uparrow)N^\downarrow}{E_2(E_1 - U)} + \frac{(1 - N^\uparrow)(1 - N^\downarrow)}{E_1 E_2}.$$
(14)

The transverse susceptibility χ^{-+} consists of transitions from the down spin states to the up spin states. Though (14) suggests that four peaks will appear, the features of χ^{-+} strongly depend on the filling of the bands. In a less than half-filled band, the chemical potential will typically fall within the main sub-band. After the energy convolution on (14), only the last two terms in (14) will remain. The inclusion of the vertex correction (13) has the effect of modifying the weights in the following manner:

$$G^{(2)}(E_1, E_2) = \frac{n^\pm}{(E_1 - U)(E_2 - U)} + \frac{(N^\uparrow - n^\pm)}{E_1(E_2 - U)} + \frac{(N^\downarrow - n^\pm)}{E_2(E_1 - U)} + \frac{1 - N^\uparrow - N^\downarrow + n^\pm}{E_1 E_2}.$$
(15)

Since $n^\pm$ is always smaller than or equal to $N^\uparrow N^\downarrow$, the first and last terms of (15) are reduced compared to (14). In contrast, the two middle terms are enhanced. It is therefore to be expected that the vertex corrections will favour transitions from the main band to the impurity band and reduce transitions between the impurity bands (if they were allowed) or between the main bands. We note that the four weights in (15) are actually the respective concentrations of species.

3.3. Weighted Green functions

The concept of weighted Green functions becomes necessary when different kinds of objects, such as the A and B atoms for the binary alloy, are on the lattice. Weighting in the CPA means that we choose from which type of site the particle is starting the motion, as well as in some cases the type of site where the motion ends. This results in a statistical weight given for each component. The properly weighted Green function then allows us to identify and analyse the effects of each component of the alloy. Recently, Schwabe and Elliott [7] calculated all the necessary weights for the two-particle propagator in the binary alloy. In the subsequent paper [8], they showed how to use the weights to obtain a more refined treatment of the interacting exciton in this context. The two-particle weighted Green functions in the four-component alloy will be required when the residual interaction in (6) is included in the calculations. Moreover, it will be shown that the one-particle weighted Green functions can be used to determine $n^\pm$.

3.3.1. Weighted one-particle Green functions. We consider first the one-particle propagator and assume that a particle propagates between 'impurity' sites i and 'host' sites h (as it does in the four-component alloy as well). The singly weighted Green functions are defined such that

$$G^{io}(l, m) = \begin{cases} G(l, m) & \text{if } l \text{ is an impurity} \\ 0 & \text{otherwise.} \end{cases}$$
(16)

We also define $G^{oi}(l, m)$ as corresponding to the original propagator $G(l, m)$ only if m is an impurity, and zero otherwise, and the doubly weighted Green function $G^{ii}(l, m)$ that is not
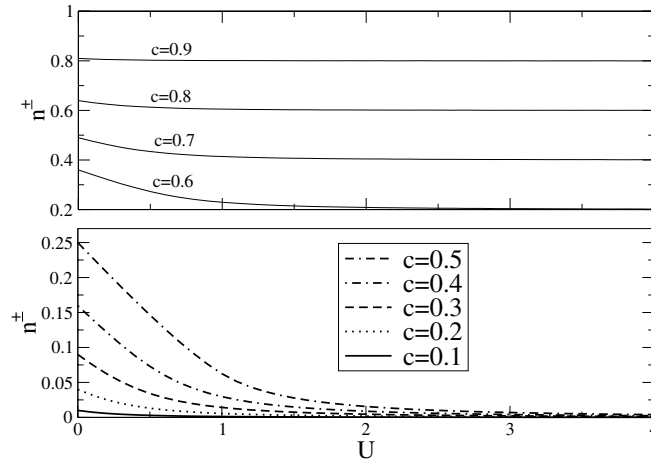
Figure 1. $n^{\pm}$ for various values of the particle concentration c in the CPA treatment of the Hubbard model in the paramagnetic state.

zero only if both l and m are impurities. We define similarly $G^{ho}(l,m)$, $G^{oh}(l,m)$, $G^{ih}(l,m)$, $G^{hi}(l,m)$ and $G^{hh}(l,m)$. The weighted Green function in an alloy of the type $H = H_0 + V$ like (7) can be obtained simply by applying the diagonal interaction operator:

$$G^{io} = \frac{V}{U}G, \qquad G^{oi} = G\frac{V}{U}, \qquad G^{ii} = \frac{1}{U^2}VGV. \qquad (17)$$

An estimation of the averages such as $\bar{G}^{io}$ is needed in order to study how, on average, the processes on the different species of the alloy contribute to the one-particle properties. In the CPA the one-particle single weights are found to be [2, 7]

$$\bar{G}^{io} = \bar{G}^{oi} = \frac{\Sigma}{U}\bar{G} \qquad \bar{G}^{ho} = \bar{G}^{oh} = \left(1 - \frac{\Sigma}{U}\right)\bar{G}. \qquad (18)$$

$\bar{G}^{io}$ offers a method for obtaining the average number of sites that are doubly occupied in the Hubbard model for the paramagnetic case. At zero temperature, the average number of sites per atom with an $\uparrow$ spin is given by $N^{\uparrow} = \langle c_{i\uparrow}^{\dagger} c_{i\uparrow}\rangle = \int_{-\infty}^{\mu} dE\,(-1/\pi)\,\mathrm{Im}\,\bar{G}_{ii}^{\uparrow}(E)$. The Green function $\bar{G}^{\uparrow}(E)$ is evaluated in the CPA. This formulation counts all sites where an $\uparrow$ spin is present, including sites where both up and down spins are found. Recalling the alloy analogy, the $\uparrow$ spin sees the sites where a $\downarrow$ spin is present as defect sites i, where the particle interacts with energy U. The weighted Green function $\bar{G}_{ii}^{\uparrow\,io}$ is thus 0 on sites where there is no $\downarrow$ spin, filtering out the singly occupied sites. Hence the number of sites per atom $n^{\pm}$ where two spins are found is, on average in the CPA approximation and with (18), (9),

$$n^{\pm} = \int_{-\infty}^{\mu} dE\left(-\frac{1}{\pi}\right)\mathrm{Im}\left[F^{\uparrow}(E)\frac{N^{\downarrow}}{1 - (U - \Sigma^{\uparrow}(E))F^{\uparrow}(E)}\right]. \qquad (19)$$

In the limit $U \to 0$, $n^{\pm} \to N^{\uparrow}N^{\downarrow}$, as expected. The results for $n^{\pm}$ in the CPA treatment and for the paramagnetic case, $N^{\uparrow} = N^{\downarrow}$, are given in figure 1. For less than half-filling, that is for $c = N^{\uparrow} = N^{\downarrow} \leqslant 0.5$, $n^{\pm}$ decreases rapidly from the random value c^2 to 0. At $U = 1$, $n^{\pm}$ becomes almost negligible. Beyond half-filling, saturation forces a minimum number of up and down spins to cohabit even at large U, reaching a minimum value of $2c - 1$ at $U \to \infty$.

3.3.2. Weighted two-particle Green functions. They are defined identically to the one-particle Green functions. We estimate $\langle G_{ij}^{\uparrow n_1 n_2} G_{ji}^{\downarrow n_2 n_1} \rangle$, where n_1 and n_2 denote respectively the type of site i and j can be. The result will be naturally linked to the vertex correction. Schwabe and Elliott [7] presented the calculations used to obtain singly weighted and doubly weighted two-particle Green functions for a binary alloy with clearly defined and fixed impurities i and hosts h. For the susceptibility the following weighted Green functions will be required: $\langle G_{ij}^{\uparrow n^+} G_{ji}^{\downarrow n^+} \rangle$, $\langle G_{ij}^{\uparrow n^-} G_{ji}^{\downarrow n^-} \rangle$, $\langle G_{ij}^{\uparrow n^+ n^+} G_{ji}^{\downarrow n^+ n^+} \rangle$, $\langle G_{ij}^{\uparrow n^- n^-} G_{ji}^{\downarrow n^- n^-} \rangle$. The calculations, though not totally identical, can be made on the very same lines as in the work of Schwabe and Elliott's [7]. The determination of $\langle G^{\uparrow n^{\pm} o} C G^{\downarrow o n^{\pm}} \rangle$ corresponds for example to calculating $\langle G^{\uparrow io} C G^{\downarrow oi} \rangle$, as $n^{\pm}$ is acting as an impurity for both the up spin and the down spin. In the case of single weights we find thus for the susceptibility

$$\langle G^{\uparrow no} C G^{\downarrow on} \rangle = \langle G^{\uparrow on} C G^{\downarrow no} \rangle = \langle G^{\uparrow} C G^{\downarrow} \rangle \xi^n \tag{20}$$

where $n \in \{n^{\pm}, n^+, n^-, n^{\varnothing}\}$ and

$$
\xi^{n^{\pm}} := \frac{\Lambda}{(U - \Sigma^{\uparrow})(U - \Sigma^{\downarrow})} \frac{n^{\pm}(1 - N^{\uparrow})(1 - N^{\downarrow})}{n^{\pm} - N^{\uparrow} N^{\downarrow}} \qquad \xi^{n^{\varnothing}} := \frac{\Lambda}{\Sigma^{\uparrow} \Sigma^{\downarrow}} \frac{n^{\varnothing} N^{\uparrow} N^{\downarrow}}{n^{\pm} - N^{\uparrow} N^{\downarrow}}
$$
$$
\xi^{n^-} := \frac{\Lambda}{(U - \Sigma^{\uparrow})\Sigma^{\downarrow}} \frac{n^- N^{\uparrow}(1 - N^{\downarrow})}{n^{\pm} - N^{\uparrow} N^{\downarrow}} \qquad \xi^{n^+} := \frac{\Lambda}{\Sigma^{\uparrow}(U - \Sigma^{\downarrow})} \frac{n^+(1 - N^{\uparrow}) N^{\downarrow}}{n^{\pm} - N^{\uparrow} N^{\downarrow}}. \tag{21}
$$

The doubly weighted Green functions involve some lengthy algebra. Schwabe and Elliott [7] found, after suitable approximations, that when C is diagonal the total weight is the product of the single weights. Again, the calculations for our four-component alloy can be carried through in the same fashion and we also get

$$\langle G^{\uparrow n_1 n_2} C G^{\downarrow n_2 n_1} \rangle = \langle G^{\uparrow} C G^{\downarrow} \rangle \xi^{n_1} \xi^{n_2} \tag{22}$$

where $n_1, n_2 \in \{n^{\pm}, n^+, n^-, n^{\varnothing}\}$.

We note that the four terms of Λ^{-1} in (13) correspond to the expression for the four weights. In the small U limit, $\Sigma^{\sigma} \to N^{-\sigma} U$ and $\Lambda \to U^2(n^{\pm} - N^{\uparrow} N^{\downarrow})$, so that each weight is simply the corresponding concentration: $\xi^n \to n$, $n \in \{n^{\pm}, n^+, n^-, n^{\varnothing}\}$. In the large U limit the weights retain their energy dependence but it is easy to see that the weights of the $n^{\pm}$, n^+ and n^- sites go to zero, while the one associated with $n^{\varnothing}$ tends to 1. Contributions from occupied sites are eliminated as the repulsive interaction grows, preventing encounters of the particles on those sites when $N_e < 1$.

3.4. The residual interaction

We turn now our attention to the last term in (6). So far we have found the solution K^0_{ijkl} of the reduced equation of motion. The simplest way of treating the term $-U \delta_{ij} \langle \langle (n_{j\downarrow} - n_{i\uparrow}) c_{i\downarrow}^{\dagger} c_{j\uparrow}; c_{k\uparrow}^{\dagger} c_{l\downarrow} \rangle \rangle_E$ is to average the $n_{j\sigma}$s to N^{σ}s, so that it becomes $-U m \delta_{ij} \langle \langle c_{i\downarrow}^{\dagger} c_{j\uparrow}; c_{k\uparrow}^{\dagger} c_{l\downarrow} \rangle \rangle_E$, with $m = N^{\downarrow} - N^{\uparrow}$. This is the VCA treatment of this term. Calling K_{ijkl} the solution of the whole equation of motion (6), it can be checked that $K_{xxx'x'}$ can be expressed in terms of the uncorrected solution by

$$K_{xxx'x'}(t) = K^0_{xxx'x'}(t) - U \sum_{\bar{n}} \int_{-\infty}^{\infty} \mathrm{d}t' \, K^0_{xx\bar{n}\bar{n}}(t - t') K_{\bar{n}\bar{n}x'x'}(t'). \tag{23}$$

Fourier transforming both in time and space, we get

$$K(q, E) = \frac{K^0(q, E)}{1 + U K^0(q, E)}. \tag{24}$$

This result, however, is not consistent with the one-particle Green functions obtained with the CPA, as (24) predicts an instability in the ground state at $U = -1/K^0(0,0)$. Since the CPA does not allow for ferromagnetic solutions, a treatment of the extra term is required that is consistent with the original method.

A more refined treatment can be obtained when $N^\uparrow \neq N^\downarrow$ if the extra term is treated using the weighted Green functions developed in the CPA. Instead of taking their averages, the operators $n_{i\sigma}$ are again replaced by 0 or 1 at random. The integral equation including the extra term now reads

$$K_{xx'}(t) = K_{xx'}^0(t) - \frac{U}{m} \sum_{\bar{n}} \left(n_{\bar{n}\downarrow} - n_{\bar{n}\uparrow} \right) \int_{-\infty}^{\infty} dt' \, K_{x\bar{n}}^0(t-t') K_{\bar{n}x'}(t') \tag{25}$$

where, to simplify the notation, we have written $K_{xx'}^0$ instead of $K_{xxx'x'}^0$. Transformed in the energy space, it can be developed into

$$K_{xx'}(E) = K_{xx'}^0(E) - \frac{U}{m} \sum_{\bar{n}} K_{x\bar{n}}^0(E) \left(n_{\bar{n}\downarrow} - n_{\bar{n}\uparrow} \right) K_{\bar{n}x'}^0(E)$$

$$+ \left(\frac{U}{m} \right)^2 \sum_{\bar{n}_1, \bar{n}_2} K_{x\bar{n}_1}^0(E) \left(n_{\bar{n}_1\downarrow} - n_{\bar{n}_1\uparrow} \right) K_{\bar{n}_1\bar{n}_2}^0(E)$$

$$\times \left(n_{\bar{n}_2\downarrow} - n_{\bar{n}_2\uparrow} \right) K_{\bar{n}_2x'}^0(E) + \cdots. \tag{26}$$

The terms $(n_{\bar{n}\downarrow} - n_{\bar{n}\uparrow})$ can take three possible values, 0, 1 or -1, depending on whether the site n is an empty or doubly occupied site, a single $\downarrow$ site, or a single $\uparrow$ site, respectively. The summations over $\bar{n}$, $\bar{n}_1$, $\bar{n}_2$ etc in (26) can thus be restricted to singly occupied sites. This is just in fact the summation over all sites of the *weighted* Green functions. They have however to be normalized by the impurity concentration, as the averaged weighted Green function is the total counted on all impurity sites. Writing $v_\uparrow \equiv U/(m\,n^+)$ and $v_\downarrow \equiv -U/(m\,n^-)$, the expansion (26) becomes, after Fourier transforming,

$$K(q, E) = K^0(q, E) + \sum_{s \in \{\uparrow, \downarrow\}} v_s K^{0,os}(q, E) K^{0,so}(q, E)$$

$$+ \sum_{s_1, s_2 \in \{\uparrow, \downarrow\}} v_{s_1} v_{s_2} K^{0,os_1}(q, E) K^{0,s_1s_2}(q, E) K^{0,s_2o}(q, E) + \cdots. \tag{27}$$

Such an expansion is very similar to what is done in the case of the excitonic absorption in a binary alloy [8]. In the paper, Schwabe and Elliott calculate the optical absorption of an exciton, taking into account the interaction between the hole and the electron. This is done by writing a weighted scattering expansion comparable to (27). Similarly to in [8], (27) can be expressed in a compact matrix form. The following matrices are defined, where all weighted Green functions are understood to depend on q and E:

$$\vec{K}^0 = \begin{pmatrix} K^{0n^+} \\ K^{0n^-} \end{pmatrix} \qquad \hat{U} = \begin{pmatrix} v_\uparrow = \frac{U}{mn^+} & 0 \\ 0 & v_\downarrow = -\frac{U}{mn^-} \end{pmatrix} \qquad \hat{K}^0 = \begin{pmatrix} K^{0n^+n^+} & K^{0n^+n^-} \\ K^{0n^+n^-} & K^{0n^-n^-} \end{pmatrix}. \tag{28}$$

Since the disorder average has been taken, the following simplification can be made: $K^{0n^+n^-} \equiv K^{0n^-n^+}$, and $K^{0n} \equiv K^{0on} \equiv K^{0no}$ for any n. The scattering expansion (27) becomes

$$K(q, E) = K^0(q, E) + \vec{K}^{0^T} \hat{U} \vec{K}^0 + \vec{K}^{0^T} \hat{U} \hat{K}^0 \hat{U} \vec{K}^0 + \cdots$$

$$= K^0(q, E) + \vec{K}^{0^T} \hat{U} \sum_{n=0}^{\infty} [\hat{K}^0 \hat{U}]^n \vec{K}^0. \tag{29}$$

As (29) represents a geometric series, the equation goes over to

$$K(q, E) = K^0(q, E) + \vec{K^0}^T \hat{U}[\mathbb{I} - \hat{K}^0 \hat{U}]^{-1} \vec{K^0}. \tag{30}$$

This expression can be greatly simplified if the small U limit is taken. In that case, the U^2-terms are neglected, and the weights are energy independent. Using the property (22) for the double weights, (30) becomes

$$K(q, E) \rightarrow \frac{K^0(q, E)}{1 + \frac{U}{m} K^0(q, E)\left[\frac{(\xi^{n^-})^2}{n^-} - \frac{(\xi^{n^+})^2}{n^+}\right]} \tag{31}$$

so that the standard result (24) is retrieved.

3.5. The paramagnetic case

As the CPA does not allow for ferromagnetic ground states, the paramagnetic case where $n^+ = n^-$, $N^\uparrow = N^\downarrow$ and $m = 0$ must be examined separately. In this case, the averaged weighted Green functions with respect to n^+ and n^- are expected to be identical. However, the condition that $K^{0\,on^+}$ is equivalent to $K^{0\,on^-}$ is not fulfilled by the expressions for the weights as they are given by (21). This is due to the fact that the denominator of $G^{2,n^-\,oon^-}(z_1, z_2)$ contains $\left(U - \Sigma^\uparrow(z_1)\right) \Sigma^\downarrow(z_2)$, whereas $G^{2,n^+\,oon^+}(z_1, z_2)$ contains $\Sigma^\uparrow(z_1)\left(U - \Sigma^\downarrow(z_2)\right)$. The energy convolution therefore produces two different results for K^{0n^+} and K^{0n^-}, in spite of the physical symmetry in the problem. The symmetry is however restored if K^0_{-+} is considered together with K^0_{+-}. In the paramagnetic regime we have $K^{0n^+}_{-+} = K^{0n^-}_{+-}$, with similar equivalences for the doubly weighted Green functions. It is thus possible in principle to obtain the limit of $K_{xx}(q, E) := K_{-+}(q, E) + K_{+-}(q, E)$ when $m \rightarrow 0$. To simplify the notation, we abandon the wavevector and energy dependences. The first term of the expansion of K_{xx} from (27), K_1, is given by

$$K_1 = \frac{U}{m}\left(\frac{(K^{n^+}_{-+})^2}{n^+} - \frac{(K^{n^-}_{-+})^2}{n^-} + \frac{(K^{n^+}_{+-})^2}{n^+} - \frac{(K^{n^-}_{+-})^2}{n^-}\right). \tag{32}$$

The two terms in the middle and the two remaining terms on the right-hand side of (32) become identical in the paramagnetic limit and can be paired off. We need only consider one pair, for example $\lim_{m\rightarrow 0} = U/m\left((K^{n^+}_{-+})^2/n^+ - (K^{n^-}_{+-})^2/n^-\right)$. We define $\tilde{N}$ so that $N^\uparrow = \tilde{N} - m/2$ and $N^\downarrow = \tilde{N} + m/2$. Similarly, $n^+ = \tilde{n} - m/2$ and $n^- = \tilde{n} + m/2$. In the paramagnetic limit $K^{n^+}_{-+} = K^{n^-}_{+-} \rightarrow K^{\tilde{n}}$. Both $K^{n^+}_{-+}$ and $K^{n^-}_{+-}$ can be developed in powers of m so that

$$\frac{(K^{n^+})^2}{n^+} = \frac{(K^{\tilde{n}})^2}{\tilde{n}} + m\frac{\partial}{\partial\tilde{n}}\left(\frac{(K^{\tilde{n}})^2}{\tilde{n}}\right)\frac{\partial\tilde{n}}{\partial m} + \mathcal{O}(m^2). \tag{33}$$

The two terms together give

$$\frac{(K^{n^+})^2}{n^+} - \frac{(K^{n^-})^2}{n^-} = -m\left(-\frac{(K^{\tilde{n}})^2}{\tilde{n}^2} + \frac{2}{\tilde{n}}K^{\tilde{n}}\frac{\partial K^{\tilde{n}}}{\partial\tilde{n}}\right). \tag{34}$$

Due to the self-consistent nature of the CPA, the quantity $\partial K^{\tilde{n}}/\partial\tilde{n}$ is very difficult to obtain. The self-energy varies in a complex way with the concentration of impurities, as the background itself is affected by a small modification in the number of impurities. The weights themselves are also dependent on the self-energy and the concentration of species in a complex manner. In two cases, however, the quantity $\partial K^{\tilde{n}}/\partial\tilde{n}$ can be established. In the limit $U \rightarrow 0$ it is easy to see that $\partial K^{\tilde{n}}/\partial\tilde{n} \rightarrow K^{\tilde{n}}/\tilde{n}$. The same result is obtained in the split-band limit, since in this
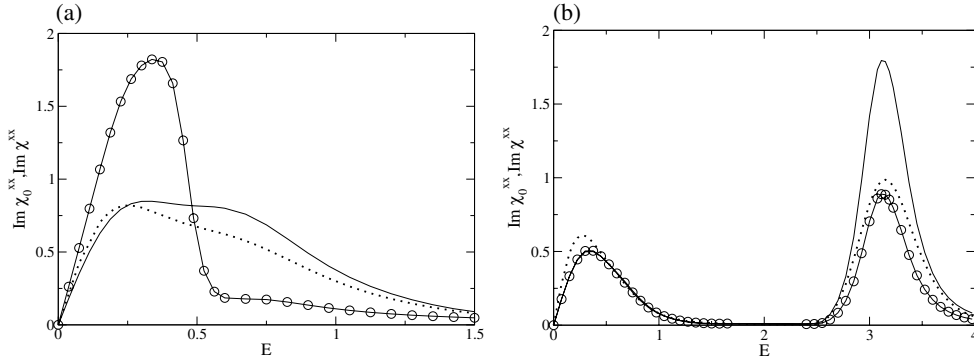
Figure 2. Imaginary part of $\chi_0^{xx}(q, E)$ and χ^{0xx} at $q = 0$ and for $N_e = 0.7$. Dotted line: χ_0^{xx} without the inclusion of the vertex correction; solid line: χ_0^{xx} with the inclusion of the vertex correction; circles: χ^{xx} including both the vertex correction and the residual interaction. (a) $U = 0.5$, $n^{\pm} = 0.04$ and $\mu = -0.05$; (b) $U = 3.0$, $n^{\pm} = 0.0$ and $\mu = 0.02$.

case $K^{\tilde{n}}(z_1, z_2)$ is given simply by $\tilde{n}/[z_1(z_2 - U)]$. In the limit of small and large U, the first term becomes

$$K_1 \rightarrow -\frac{U}{n^+ n^-}\left(K_{-+}^{n^+}\right)^2 - \frac{U}{n^+ n^-}\left(K_{-+}^{n^-}\right)^2. \tag{35}$$

It is easy to see that the subsequent terms in U can all be paired in the same way. Extrapolating the result (35) to the higher order terms, we find

$$K_{xx}(q, E) \rightarrow 2K^0(q, E) - \frac{U\frac{(K^{0n^+})^2}{n^+ n^-}}{1 + U\frac{K^{0n^+n^+}}{n^+ n^-}} - \frac{U\frac{(K^{0n^-})^2}{n^+ n^-}}{1 + U\frac{K^{0n^-n^-}}{n^+ n^-}}. \tag{36}$$

Numerical results are given in the next subsection.

3.6. Numerical illustrations

The numerical applications of the formalism developed for the CPA susceptibility are presented here for a three-dimensional, simple cubic band at $T = 0$. We evaluate $\chi_0^{xx}(q, E)$ (residual interaction neglected) and compare it with $\chi^{xx}(q, E)$ that includes the residual interaction.

The susceptibility is given by $\chi_0^{xx}(q, E) = \chi_0^{-+}(q, E) + \chi_0^{+-}(q, E)$ which, in the paramagnetic ground state of the CPA, becomes simply $\chi_0^{xx}(q, E) = 2\chi_0^{-+}(q, E)$. $\chi_0^{-+}(q, E)$ is obtained as $\chi_0^{-+}(q, E) = -K^0(q, E)$, where $K^0(q, E)$ is calculated from the combination of Green functions (10). Each of these Green functions is obtained from the CPA solution (11) with the vertex correction (13). The quantity $n^{\pm}$ is determined from the weighted Green functions (19). The effects of the vertex correction are depicted in figure 2 (solid line) for small and large U and compared to the non-corrected case (dotted line). They correspond to the predictions of (15) and can be understood from the perspective of the spin up and down propagating on the four-component alloy. As the spin up interacts with the down spins and the spin down with the up spins, it is to be expected that the $n^{\pm}$ sites will play a major role in correlating the particle motions. For both the propagating spins, up and down, the $n^{\pm}$ sites are included in the impurity band (B band), hence enhancing transitions from the main band (A band) to the B band. Similarly, the n^o sites, with which no spin interacts, are included in the A band of each species. Compared to the free particle cases, it is thus expected that the inclusions of correlations will reduce the transitions from one main sub-band to the other.

The result of the approximation (36) for $\chi^{xx}(E)$ at $q = 0$ can be seen (circles in figure 2) in its range of validity. At small U the A–A transitions are enhanced compared to the value including only the vertex correction. At large U the A–A transitions have come back to their vertex correction value. In both cases though, the A–B transitions are strongly reduced.

Even though we could not interpolate (36) for intermediate U, it can be predicted that the peak at the energy of the maximum of the A–B transition will be reduced from its value obtained from the vertex correction. This is an expected behaviour considering the way we split the equation of motion. In the first part of the problem, when considering the propagation of two particles on a four-component alloy, the inclusion of the vertex correction led to the enhancement of the interband transitions. Due to the nature of the problem, it is indeed expected for the particles to become correlated when they meet on an $n^{\pm}$ site, the type of site that represents an impurity for both of them. However, the Pauli principle forbids the $\uparrow$ and $\downarrow$ spin to flip on such a site. When considering one-particle Green functions, the Pauli principle is taken into account through the Fermi statistics. The vertex correction is obtained from the point of view of two particles moving in a lattice of potentials, rather than an up and down spins moving among other up and down spins. It is thus not clear whether the inclusion of the vertex correction conserves the Pauli principle or not. The inclusion of the last term in the equation of motion in the form of a weighted scattering correction appears to adjust the results of the vertex correction so that they comply to the Pauli principle.

4. Two-particle Green functions in the τ-CPA

The τ-CPA treatment developed in I for the one-particle Green functions is extended to the two-particle case, by using the two-particle CPA propagator up to a time τ and thereafter the VCA propagator. The resulting Green function is thus made of a CPA part that includes the vertex correction and of a VCA part, both weighted by factors depending on τ.

The two-particle Green functions in the τ-CPA can be built in a straightforward manner from the one-particle case. The consistent way to build the two-particle Green function $\langle G^{(2)}(t)\rangle$ is to break it into a VCA part and CPA part, as for the one-particle case:

$$\langle G^{(2)}(t)\rangle = \begin{cases} \langle G^{(2)\mathrm{CPA}}(t)\rangle & \text{if } t \leqslant \tau \\ \langle G^{(2)\mathrm{VCA}}(t)\rangle & \text{if } t > \tau. \end{cases} \tag{37}$$

For any time t, $\langle G^{(2)}(t)\rangle = \langle G^{\uparrow}(t)G^{\downarrow}(t)\rangle$. To obtain the two-particle function of the type $\langle G^{(2)}(E)\rangle = \int_{-\infty}^{\infty} d\omega\, \langle G^{(2)}(\omega, E - \omega)\rangle$, where $G^{(2)}(\omega, E - \omega) = G^{\uparrow}(\omega)G^{\downarrow}(E - \omega)$, one has to evaluate

$$\langle G^{(2)}(E)\rangle = \int_{-\infty}^{\infty} dt\, \mathrm{e}^{\mathrm{i}Et}\langle G^{(2)}(t)\rangle = \underbrace{\int_{-\infty}^{\tau} dt\, \mathrm{e}^{\mathrm{i}Et}\langle G^{(2)\mathrm{CPA}}(t)\rangle}_{=:\langle \widetilde{G}^{(2)\mathrm{CPA}}(E)\rangle} + \underbrace{\int_{\tau}^{\infty} dt\, \mathrm{e}^{\mathrm{i}Et}\langle G^{(2)\mathrm{VCA}}(t)\rangle}_{=:\langle \widetilde{G}^{(2)\mathrm{VCA}}(E)\rangle}. \tag{38}$$

The VCA part of the two-particle Green function is easily obtained. Using the definition from I:

$$\mathcal{G}_k^{\sigma\,\mathrm{VCA}}(E) = \frac{1}{2\pi} \int_{\tau}^{\infty} dt\, \mathrm{e}^{\mathrm{i}Et} G_k^{\sigma\,\mathrm{VCA}}(t) = \frac{\mathrm{e}^{\mathrm{i}(E - \epsilon_k - N^{-\sigma}U)\tau}}{E - \epsilon_k - N^{-\sigma}U} \tag{39}$$

the VCA part can be expressed as the convolution of the one-particle functions:

$$\langle \mathcal{G}_k^{(2)\mathrm{VCA}}(E)\rangle = \int_{-\infty}^{\infty} d\epsilon\, \frac{1}{N} \sum_q \mathcal{G}_{k+q}^{\uparrow\,\mathrm{VCA}}(\epsilon) \mathcal{G}_q^{\downarrow\,\mathrm{VCA}}(E - \epsilon) \tag{40}$$

and $\mathcal{G}^{(2)\text{VCA}}(t) = \mathcal{G}^{\uparrow(\text{VCA})}(t)\mathcal{G}^{\downarrow\text{VCA}}(t)$. The factor for matching the two Green functions at time τ is included as usual in the VCA part, so that we have

$$\langle \widetilde{G}_k^{(2)\text{VCA}}(\epsilon_1, \epsilon_2)\rangle = \frac{1}{N}\sum_q \widetilde{G}_{k+q}^{\uparrow\text{VCA}}(\epsilon_1)\widetilde{G}_q^{\downarrow\text{VCA}}(\epsilon_2) \tag{41}$$

with, from I, the one-particle VCA part that includes the normalization β_k is

$$\widetilde{G}_k^{\sigma\,\text{VCA}}(E) := \frac{1}{\beta_k}\left(\frac{e^{\text{Im}\,\Sigma^\sigma(E)\tau}}{E - \epsilon_k - N^{-\sigma}U}\right). \tag{42}$$

The same approximation will be assumed to hold also for the CPA part. In that case, the procedure must be adapted to treat the vertex corrections correctly. The τ-CPA says that the interaction is treated in the CPA up to a time τ, from when it is treated in the VCA. In both cases, the interaction is taken in full. Therefore the vertex correction term must involve the full CPA Green functions and not only the CPA equivalent of (42),

$$\widetilde{G}_k^{\sigma\,\text{CPA}}(E) := \frac{1}{\beta_k}\left(\frac{1 - e^{\text{Im}\,\Sigma^\sigma(E)\tau}}{E - \epsilon_k - \Sigma^\sigma(E)}\right). \tag{43}$$

The pure CPA two-particle Green function is given by (11). A treatment similar to that for the one-particle Green functions, where the energy dependence of Σ was ignored temporarily while the time integration was carried over, is applied here as well. The vertex correction to the free particle case, the denominator of (11), is kept unchanged in the pure CPA form while the numerator becomes, as for the VCA case, a convolution on the τ-CPA one-particle Green functions. The following approximation will thus be used for the CPA part of the two-particle Green function in the τ-CPA:

$$\langle \widetilde{G}_k^{(2)\text{CPA}}(\epsilon_1, \epsilon_2)\rangle = \frac{\frac{1}{N}\sum_q \widetilde{G}_{k+q}^{\uparrow\text{CPA}}(\epsilon_1)\widetilde{G}_q^{\downarrow\text{CPA}}(\epsilon_2)}{1 - \Lambda(\epsilon_1, \epsilon_2)\frac{1}{N}\sum_q G_{k+q}^{\uparrow\text{CPA}}(\epsilon_1)G_q^{\downarrow\text{CPA}}(\epsilon_2)} \tag{44}$$

$\Lambda(\epsilon_1, \epsilon_2)$ is calculated using the self-energy obtained from the full CPA equation.

5. Dynamical susceptibility in the τ-CPA

In this section we put together the elements developed in the prior sections. In order to use the formalism developed above to calculate the susceptibility, the residual interaction between the two spins must first be calculated.

The transverse susceptibility, without the residual term, is given by the two-particle Green function $K_k^0(E)$, built from the disorder average of (10). So far we have $K_k^{0\tau\text{CPA}}(E) = \widetilde{K}_k^{0\text{VCA}}(E) + \widetilde{K}_k^{0\text{CPA}}(E)$, where $\widetilde{K}_k^{0\text{VCA}}(E)$ and $\widetilde{K}_k^{0\text{CPA}}(E)$ are given by the convolutions (10) of the two-particle Green functions (41) and (44). The correction for a pure VCA system is given by (24) whereas the pure CPA requires the weighted result (30). Once again, a straightforward treatment would be to Fourier transform (24) and (30), then to integrate the CPA part from 0 to τ and the VCA part from τ to ∞. This would affect both the numerator and the denominator of (24) and (30). The denominators contain the correction due to the interaction. It was argued in the context of the vertex correction for $\langle \widetilde{G}^{(2)\text{CPA}}(E)\rangle$ that the correction is kept unchanged, since the interaction must be included at all time. An identical procedure is applied here, so that the time integration is carried only on the numerators:

$$K_k^{\tau\text{CPA}}(E) \approx \frac{\widetilde{K}_k^{0\text{VCA}}(E)}{1 + UK_k^{0\text{VCA}}(E)} + \frac{\widetilde{K}_{2k}^{0\text{CPA}}(E)}{1 + K_{1k}^{0\text{CPA}}(E)}. \tag{45}$$

$K_k^{0\text{VCA}}(E)$ is given by the convolutions (10) of the complete two-particle Green functions $G_k^{(2)\text{VCA}}(\epsilon_1, \epsilon_2) = N^{-1}\sum_q G_{k+q}^{\uparrow\text{VCA}}(\epsilon_1)G_q^{\downarrow\text{VCA}}(\epsilon_2)$. $K_{1k}^{0\text{CPA}}(E)$ and $\widetilde{K}_{2k}^{0\text{CPA}}(E)$ are defined

below. In order to simplify the notation, the index CPA will be abandoned for the weighted Green function, and n^+ becomes just $+$ (with identical definitions for the other species of the alloy). All functions are evaluated at the energy E.

$$K_{1k}^{0CPA} = \frac{U}{m}\left(\frac{K_k^{0--}}{n^-} - \frac{K_k^{0++}}{n^+}\right) - \frac{U^2}{m^2 n^+ n^-}\left[K_k^{0++}K_k^{0--} - \left(K_k^{0+-}\right)^2\right] \tag{46}$$

$$\widetilde{K}_{1k}^{0CPA} = \frac{U}{m}\left(\frac{\widetilde{K}_k^{0--}}{n^-} - \frac{\widetilde{K}_k^{0++}}{n^+}\right) - \frac{U^2}{m^2 n^+ n^-}\left[\widetilde{K}_k^{0++}\widetilde{K}_k^{0--} - \left(\widetilde{K}_k^{0+-}\right)^2\right] \tag{47}$$

$$\widetilde{K}_{2k}^{0CPA} = \widetilde{K}_k^{0CPA}\left[1 + \widetilde{K}_{1k}^{0CPA}\right] - \frac{U}{m}\left[\frac{\left(\widetilde{K}_k^{0-}\right)^2}{n^-} - \frac{\left(\widetilde{K}_k^{0+}\right)^2}{n^+}\right]$$

$$+ \frac{U^2}{m^2 n^+ n^-} \times \left[\left(\widetilde{K}_k^{0+}\right)^2\widetilde{K}_k^{0--} + \left(\widetilde{K}_k^{0-}\right)^2\widetilde{K}_k^{0++} - 2\widetilde{K}_k^{0+}\widetilde{K}_k^{0-}\widetilde{K}_k^{0+-}\right]. \tag{48}$$

This formulation is not completely consistent with the rest of the picture. The denominators of both VCA and CPA parts do not yet contain the full interaction. We recall that (10) is an integral from 0 to the chemical potential μ. μ is obtained consistently from the one-particle τ-CPA Green function $G^{\tau CPA}$ (see I) and not from the pure CPA or the pure VCA. The problem can be retraced back to the development (25) relating K to K^0. We note that the expression (25) is also valid in the case of the pure VCA, since in this case the factor $\left(n_{\bar{n}\downarrow} - n_{\bar{n}\uparrow}\right)$ is simply averaged to m, consistently with the VCA. Going back to the expansion (25), it is clear that the term m in $U/m\sum_{\bar{n}}\left(n_{\bar{n}\downarrow} - n_{\bar{n}\uparrow}\right)$ comes from the inhomogeneous term $2\pi\delta(t)(\delta_{jk}\langle c_{i\downarrow}^\dagger c_{l\downarrow}\rangle - \delta_{il}\langle c_{k\uparrow}^\dagger c_{j\uparrow}\rangle)$ in the time version of the equation of motion (4). The two correlation functions involved are thus to be calculated at the time $t = 0$. If a pure VCA (CPA) system is considered, the m generated by the inhomogeneous term will be called m^{VCA} (m^{CPA}):

$$m^{VCA} = N^{\downarrow VCA} - N^{\uparrow VCA} = \int_{-\infty}^{\mu} d\epsilon\left(-\frac{1}{\pi}\right)\frac{1}{N}\sum_q \text{Im}[G_q^{\uparrow VCA}(\epsilon) - G_q^{\downarrow VCA}(\epsilon)] \tag{49}$$

$$m^{CPA} = N^{\downarrow CPA} - N^{\uparrow CPA} = \int_{-\infty}^{\mu} d\epsilon\left(-\frac{1}{\pi}\right)\frac{1}{N}\sum_q \text{Im}[G_q^{\uparrow CPA}(\epsilon) - G_q^{\downarrow CPA}(\epsilon)]. \tag{50}$$

$G_k^{\sigma VCA}$ is given by the full VCA one-particle result and by the full CPA (as detailed in I). Hence, the expansion (24) for the VCA case should be written as

$$K^{VCA}(q, E) = \frac{K^{0VCA}(q, E)}{1 + U\frac{m}{m^{VCA}}K^{0VCA}(q, E)}. \tag{51}$$

In a pure VCA case m is simply equivalent to m^{VCA}. The CPA case (30) must also be recast in the same way, by making the substitution $m \to m^{CPA}$ in (46)–(48). If the case of a pure CPA is considered, $m \equiv m^{CPA}$. The approximation for the two-particle Green function, involving the CPA vertex corrections and the extra U-term of the equation of motion, is thus

$$K_k^{\tau CPA}(E) = \frac{\widetilde{K}_k^{0VCA}(E)}{1 + U\frac{m}{m^{VCA}}K_k^{0VCA}(E)} + \frac{\widetilde{K}_{2k}^{0CPA}[m^{CPA}](E)}{1 + K_{1k}^{0CPA}[m^{CPA}](E)}. \tag{52}$$

The chemical potential μ, being obtained from the whole $G^{\tau CPA}$, will in general lead to an m^{CPA} and m^{VCA} quite different from the values obtained for pure systems.

It is crucial to keep the full interaction, as it guarantees the appearance of the spin waves at $E = 0$ and $k = 0$. As ferromagnetism sets in, collective spin wave excitations are expected to appear. The magnetization at $k = 0$ can assume any direction in the averaged system and the energy cost E for exciting spin waves must be zero. This is also found in the τ-CPA, as
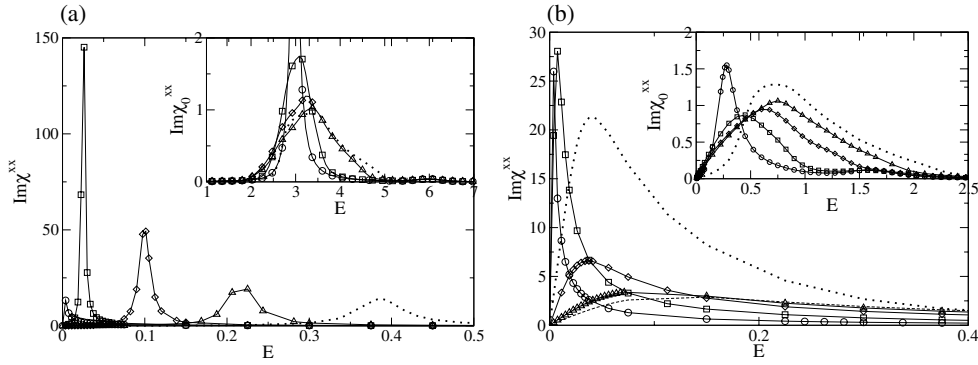
Figure 3. Spin wave part of the spectrum in the ferromagnetic regime of the τ-CPA for $\tau = 1$ and q varying along the $(1, 1, 1)$ direction: $q = 0$ (circles), $q = 1/5Q$ (squares), $q = 2/5Q$ (diamonds), $q = 3/5Q$ (triangles), $q = Q$ (dots). (a) $U = 6.0$, $N^\uparrow = 0.1$, $N^\downarrow = 0.6$; (b) $U = 1.6$, $N^\uparrow = 0.27$, $N^\downarrow = 0.43$.

the spin wave spectrum is generated almost entirely from the VCA part. In a ferromagnetic ground state we find $\lim_{k \to 0} K_k^{0\mathrm{VCA}}(E) \to \frac{m^{\mathrm{VCA}}}{E - Um}$, and $\lim_{k \to 0} K_k^{\mathrm{VCA}}(E) \to \frac{m^{\mathrm{VCA}}}{E}$. For small but finite k, we expect the excitation energy E to increase. As in the Stoner model, the spin waves will finally merge with the continuum. The transverse susceptibility $\chi_k^{-+}(E)$ can now be obtained as $\chi_k^{-+}(E) = -K_k^{\tau\mathrm{CPA}}(E)$. In ferromagnetic ground states, $\chi_k^{+-}(E)$ is also required and can be obtained similarly to $\chi_k^{-+}(E)$.

The paramagnetic limit of (52) is again difficult to obtain, and we will concentrate the application of (52) on ferromagnetic ground states.

Numerical calculations of the susceptibility require the evaluation of $n^\pm$. This value is an equilibrium property of the system along with the chemical potential and cannot be deduced from one-particle properties alone in polarized states. For most cases of interest it is however not necessary to carry through a full self-consistent treatment of $n^\pm$. When the system is close to or beyond the transition from the paramagnetic to the ferromagnetic state, the value of the interaction is fairly large. It is clear from figure 1 that it is then reasonable to put $n^\pm$ to zero under those circumstances, which is what we have done in the numerical examples.

Calculations of the susceptibility for small and large U, using the result (52) for τ-CPA at $\tau = 1$ in a ferromagnetic ground state and for a simple cubic band, are illustrated in figure 3. For both values of U the system is in a state of incomplete ferromagnetism. The insets in figure 3 show $\chi^{0xx}(q, E)$ built from $K_k^{0\tau\mathrm{CPA}}(E) = \widetilde{K}_k^{0\mathrm{VCA}}(E) + \widetilde{K}_k^{0\mathrm{CPA}}(E)$—that is, ignoring the residual expansion. In the case of $U = 6$, figure 3(a), the spectrum of $\chi^{0xx}(q, E)$ remains relatively sharp at all qs, and we observe the progression of the spin waves across the zone. At $U = 1.6$ (figure 3(b)), the spin waves are also seen to have decreased in energy at $q = Q$, but in that case the excitation is very broad and the peak is rather a resonance of $\chi^{0xx}(q, E)$. The τ-CPA causes the general broadening of the spectrum by forcing the VCA Green function to match the CPA Green function at time τ. The tendency of the spin wave to emerge out of the continuum when the wavevector follows the diagonal of the Brillouin zone is however characteristic of an antiferromagnetic instability that is also exhibited in the pure VCA case. It is therefore to be assumed that the region of ferromagnetism would be reduced, as is usually expected in the Hubbard model. While the dynamical mean field theory (DMFT) does not predict ferromagnetism for finite U on the hypercubic lattice, it is expected for the fcc lattice roughly between fillings of 0.2 and 0.9 for $U = 6$ [12]. In two dimensions on the square lattice

the cluster DMFT exhibits a small island of ferromagnetism around a filling of 0.85 [13]. In our case a more thorough study of the antiferromagnetic ground state would have to be done in order to establish the stability of the ferromagnetic ground state.

6. Conclusion

In this paper we have extended our new treatment of the magnetism in the Hubbard model using the τ-CPA developed in paper I to consider the two-particle propagators and the general magnetic susceptibility of the system. The equation of motion suggests finding the solution first within the alloy analogy and then expanding this solution according to the residual interaction. This requires initially the study of the model in the pure CPA (effectively a four-component alloy since electrons of different spins see different scattering centres), the calculation of the vertex corrections and the weight associated with each species of particles.

It is crucial for the full treatment of the magnetic susceptibility to include this residual interaction between pairs of electrons with opposite spins on the same site. It is well known that this gives rise to spin wave excitations even within an RPA treatment of the Stoner (VCA) approximation. Within the τ-CPA this requires the proper use of the weighted two-particle propagators, in part to ensure that the Goldstone mode at $q = E = 0$ is preserved. Some further approximations are necessary to make the calculation tractable, which then provides a satisfactory treatment of the susceptibility. In the paramagnetic case, when treated in the appropriate limit, it shows an enhancement of transitions between sub-bands of different spins and a corresponding reduction of those between same spin bands. In the ferromagnetic case the spin wave spectrum is obtained.

Although the treatment via the τ-CPA reported here is restricted to a single band in the simple cubic lattice with less than half-filling, it is in principle possible to extend the formalism further. It would be desirable in the future to study the model in this new light for other band structures, as well as including in the analysis other forms of magnetic order such as antiferromagnetism.

Acknowledgments

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