Time-dependent Gutzwiller theory for multi-band Hubbard models

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Abstract. We formulate a multi-band generalization of the time-dependent Gutzwiller theory. This approach allows for the calculation of general two-particle response functions, which are crucial for an understanding of various experiments in solid-state physics. As a first application, we study the momentum- and frequency-resolved magnetic susceptibility in a two-band Hubbard model. As in the case of underlying ground-state approaches, we find significant differences between the results of our method and those from a time-dependent Hartree–Fock approximation.

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1. Introduction

The study of materials with medium to strong Coulomb-interaction effects has been a central subject for experimental and theoretical solid-state physics over many years. Despite enormous effort and significant progress in some fields, however, our theoretical toolbox is still far from satisfactory for such systems. For quite some time, theoreticians in many-particle physics have focused on relatively simple model systems, such as the Heisenberg or the single-band Hubbard models. Only in the past 10 years has attention shifted towards the study of more realistic models, e.g. multi-band Hubbard models. A very important impetus in that direction came from the limit of infinite spatial dimensions ($D \to \infty$). The exact solution of Hubbard models in this limit leads to the dynamical mean-field theory (DMFT), in which the original lattice model
is mapped onto an effective single-impurity system that has to be solved numerically [1–5]. Although significant progress has been made in recent years in developing numerical techniques for the solution of the DMFT equations, it is still quite challenging and can be carried out only with limited accuracy. It is particularly difficult for the DMFT to study multi-orbital Hubbard models when the full (local) Coulomb and exchange interaction is included.

An alternative method that also relies on infinite-\(D\) techniques is the Gutzwiller variational approach. It allows for the approximate study of ground-state properties and single-particle excitations with much less numerical effort than within DMFT and has been applied in a number of works in recent years [6–24]. Another approach that leads to the same energy functional for multi-band models is the slave-boson mean-field theory [25–30].

The theoretical interpretation of a number of experiments requires the study of two-particle response functions. For example, in magnetic neutron scattering the frequency- and momentum-resolved magnetic susceptibility is measured. The textbook method for the calculation of such response functions is the random-phase approximation (RPA), which can be interpreted as a time-dependent generalization of the Hartree–Fock (HF) theory in the small-amplitude limit, i.e. where the perturbation is considered to be sufficiently small. For electronic systems with medium or strong correlation effects, however, the ground-state description of an HF theory is well known to be often inaccurate. Therefore, the RPA, as the time-dependent generalization of the HF theory, is also a questionable approach for such systems.

A time-dependent Gutzwiller theory for the calculation of two-particle response functions was developed for single-band Hubbard models by Seibold et al [31, 32]. In recent years this approach has been applied with astonishing success to quite a number of such models and response functions [33–43]. It is the main purpose of this work to generalize the time-dependent Gutzwiller theory for the investigation of multi-band models. A brief introduction to our method has already been given in [44]. All technical details, however, will be first presented here.

Our presentation is organized as follows. In sections 2 and 3 we summarize the main results of the Gutzwiller variational theory for multi-band Hubbard models. In section 4 the reader will be reminded of the derivation, which introduces the RPA as a time-dependent generalization of the HF theory. In a very similar way, the time-dependent Gutzwiller theory (‘Gutzwiller RPA’) is introduced in section 5. The general Gutzwiller RPA equations are used in section 6 for the calculation of response functions for Hubbard-type lattice models. As a first application, we study the magnetic susceptibility in a two-band model in section 7. A summary and conclusions close our presentation in section 8. The more technical parts of our derivation are referred to in four appendices.

2. Multi-band Hubbard models and Gutzwiller wave functions

We study the general class of multi-band Hubbard models

\[
\hat{H} = \sum_{i\neq j; \sigma, \sigma'} t_{i,j}^{\sigma,\sigma'} \hat{c}_{i,\sigma}^{\dagger} \hat{c}_{j,\sigma'} + \sum_{i} \hat{H}_{\text{loc},i} = \hat{H}_0 + \hat{H}_{\text{loc}}. \tag{1}
\]

Here, the first term describes the hopping of electrons between \(N\) spin–orbital states \(\sigma, \sigma'\) on \(L_s\) lattice sites \(i, j\), respectively. The Hamiltonian

\[
\hat{H}_{\text{loc},i} = \frac{1}{2} \sum_{\sigma_1, \sigma_2, \sigma_3, \sigma_4} U_{i}^{\sigma_1, \sigma_2, \sigma_3, \sigma_4} \hat{c}_{i,\sigma_1}^{\dagger} \hat{c}_{i,\sigma_2}^{\dagger} \hat{c}_{i,\sigma_3} \hat{c}_{i,\sigma_4} + \sum_{\sigma_1, \sigma_2} \epsilon_{i}^{\sigma_1, \sigma_2} \hat{c}_{i,\sigma_1}^{\dagger} \hat{c}_{i,\sigma_2} \tag{2}
\]
contains all local terms, i.e. the two-particle Coulomb interactions (∼U_i) and the orbital on-site energies (∼ε_i). We further introduce the eigenstates |Γ⟩_i of \( \hat{H}_{loc,i} \) and the corresponding energies E_{Γ,i}, i.e.

\[
\hat{H}_{loc,i}|Γ⟩_i = E_{Γ,i}|Γ⟩_i.
\]

Within the Gutzwiller theory, the Hamiltonian (1) is investigated by means of the variational wave function

\[
|\Psi_0⟩ = \hat{P}_G|Ψ_0⟩ = \prod_i \hat{P}_i|Ψ_0⟩,
\]

where |Ψ_0⟩ is a normalized single-particle product state and the local Gutzwiller correlator is defined as

\[
\hat{P}_i = \sum_{Γ,Γ'} \lambda_{i;Γ,Γ'}|Γ⟩_i⟨Γ'|.
\]

For example, in the case of the single-band Hubbard model

\[
\hat{H}_{sb} = \sum_{i ≠ j} \sum_{σ = \uparrow, \downarrow} t_{i,j} \hat{c}_{i,σ}^{†} \hat{c}_{j,σ} + \sum_i U|d⟩_i⟨d|,
\]

the local correlation operator reads

\[
\hat{P}_i = \lambda_{i,d}|d⟩_i⟨d| + \lambda_{i,↑}|↑⟩_i⟨↑| + \lambda_{i,↓}|↓⟩_i⟨↓| + \lambda_{i,↑,↓}|↑⟩_i⟨↓| + \lambda_{i,↑,↓}|↓⟩_i⟨↑|,
\]

\[
+ \lambda_{i,↑,↑}|↑⟩_i⟨↑| + \lambda_{i,↓,↓}|↓⟩_i⟨↓|.
\]

Here, we introduced the atomic states |Γ⟩_i for doubly occupied sites |d⟩_i, singly occupied sites |↑⟩_i and |↓⟩_i, and empty sites |∅⟩_i, as well as the abbreviation λ_{i,Γ} for the diagonal variational parameters λ_{i,Γ}. In terms of the fermionic operators \( \hat{c}_{i,σ}^{†} \), the operator (7) has the form

\[
\hat{P}_i = \lambda_{i,d}\hat{n}_{i,↑}\hat{n}_{i,↑} + \lambda_{i,↑}\hat{n}_{i,↑}(1 - \hat{n}_{i,↑}) + \lambda_{i,↓}\hat{n}_{i,↓}(1 - \hat{n}_{i,↓})
\]

\[
+ \lambda_{i,↑,↓}\hat{c}_{i,↑}^{†}\hat{c}_{i,↓} + \lambda_{i,↓,↑}\hat{c}_{i,↓}^{†}\hat{c}_{i,↑} + \lambda_{i,↑,↑}\hat{c}_{i,↑}^{†}\hat{c}_{i,↑} + \lambda_{i,↓,↓}\hat{c}_{i,↓}^{†}\hat{c}_{i,↓},
\]

where \( \hat{n}_{i,σ} \equiv \hat{c}_{i,σ}^{†}\hat{c}_{i,σ} \). The correlation operator (7) is the most general ansatz for single-band models without superconductivity. The latter would require additional terms of the form \( ∼ |d⟩_i⟨∅| \) and \( ∼ |∅⟩_i⟨d| \); cf [45].

3. Variational energy

As shown in [7, 46], the expectation value of the Hamiltonian (1) with respect to the variational wave function (4) can be evaluated in the limit of infinite spatial dimensions. We consider the expectation values of the local Hamiltonian \( \hat{H}_{loc} \) and the one-particle Hamiltonian \( \hat{H}_1 \) separately in sections 3.1 and 3.2. Additional constraints, which arise through the derivation in infinite dimensions, are discussed in section 3.3. In section 3.4, we recall how the standard Gutzwiller energy functional for a single-band model is recovered from our general multi-band results.
3.1. Local energy

The expectation value of the local Hamiltonian (3) in infinite dimensions reads (see footnote 2)

$$\langle \hat{H}_{\text{loc}} \rangle_{\psi_0} = \sum_{\Gamma} E^{\text{loc}}_{\Gamma, \Gamma} \equiv E_{\text{loc}},$$

(9)

where

$$m_{\Gamma, \Gamma'} \equiv \langle (|\Gamma\rangle \langle \Gamma'|) \rangle_{\psi_0} = \langle (\hat{P}^\dagger |\Gamma\rangle \langle \Gamma'| \hat{P}) \rangle_{\psi_0} = \sum_{\Gamma, \Gamma'} \lambda^*_{\Gamma, \Gamma'} \tilde{\lambda}_{\Gamma', \Gamma'} m^0_{\Gamma, \Gamma'},$$

(10)

and

$$m^0_{\Gamma, \Gamma'} \equiv \langle (|\Gamma\rangle \langle \Gamma'|) \rangle_{\psi_0}.$$  

(11)

To further evaluate the expectation value (11), we introduce the basis of Fock states (i.e. ‘Slater determinants’)

$$|I\rangle = \prod_{\sigma \in I} \hat{c}_{\sigma}^\dagger |0\rangle$$

(12)

in which certain spin–orbit states $\sigma \in I$ are occupied. Mathematically, the indices $I = (\sigma_1, \sigma_2, \ldots, \sigma_n)$ are considered as ordered sets of spin–orbit states $\sigma$. Therefore, we can use all standard set operations, such as $I \cup \sigma$ or $I \setminus \sigma$. In addition, we define the number of orbitals in $I$ as $|I|$. The states $|I\rangle$ provide a basis of the local (atomic) Hilbert space. Hence, we can use them for an expansion of the eigenstates $|\Gamma\rangle$,

$$|\Gamma\rangle = \sum_{I} T_{I, \Gamma} |I\rangle,$$

(13)

and write the expectation value (11) as

$$m^0_{I, I'} = \sum_{I, I'} T_{I, \Gamma} T^*_{I', \Gamma'} m^0_{I, I'}.$$  

(14)

Finally, the uncorrelated expectation values of the transfer operators $|I\rangle \langle I'|$,

$$m^I_{I', I'} \equiv \langle (|I\rangle \langle I'|) \rangle_{\psi_0},$$

(15)

can be written as the determinant

$$m^I_{I', I'} = \begin{vmatrix} \Omega^I_{I', I'} & -\Omega^I_{I', I'} \\ \Omega^I_{I', I'} & \tilde{\Omega}^I_{I', I'} \end{vmatrix}. $$

(16)

Here, $\Omega_{I, I'}$ are the matrices

$$\Omega_{I, I'} = \begin{pmatrix} C^0_{\sigma_1, \sigma'_1} & C^0_{\sigma_1, \sigma'_2} & \cdots & C^0_{\sigma_1, \sigma'_I} \\ C^0_{\sigma_2, \sigma'_1} & C^0_{\sigma_2, \sigma'_2} & \cdots & C^0_{\sigma_2, \sigma'_I} \\ \cdots & \cdots & \cdots & \cdots \\ C^0_{\sigma_I, \sigma'_1} & C^0_{\sigma_I, \sigma'_2} & \cdots & C^0_{\sigma_I, \sigma'_I} \end{pmatrix},$$

(17)

in which the entries are elements of the uncorrelated local density matrix

$$C^0_{\sigma, \sigma'} = \langle \hat{c}_{\sigma}^\dagger \hat{c}_{\sigma'} \rangle_{\psi_0}.$$  

(18)
that belong to the configurations \( I = (\sigma_1, \ldots, \sigma_{|I|}) \) and \( I' = (\sigma'_1, \ldots, \sigma'_{|I'|}) \). The matrix \( \tilde{\Omega}^{I,I'} \) in (16) is defined as

\[
\tilde{\Omega}_{J,J} = \begin{pmatrix}
1 - C^{0}_{\sigma_1, \sigma_1} & - C^{0}_{\sigma_1, \sigma_2} & \cdots & - C^{0}_{\sigma_1, \sigma_{|I|}} \\
- C^{0}_{\sigma_2, \sigma_1} & 1 - C^{0}_{\sigma_2, \sigma_2} & \cdots & - C^{0}_{\sigma_2, \sigma_{|I|}} \\
\vdots & \vdots & \ddots & \vdots \\
- C^{0}_{\sigma_{|I|}, \sigma_1} & - C^{0}_{\sigma_{|I|}, \sigma_2} & \cdots & 1 - C^{0}_{\sigma_{|I|}, \sigma_{|I|}}
\end{pmatrix},
\]

with \( \sigma_j \in J \equiv (1, \ldots, N) \setminus (I \cup I') \).

In applications of the Gutzwiller theory to multi-band systems, it would be quite cumbersome to evaluate the determinants (16) if the local density matrix (18) is non-diagonal. Fortunately, we are free to choose the local orbital basis in a way that suits us best. Therefore, we introduce an orbital basis, defined by (local) operators \( \hat{c}_\gamma^{\dagger} \), for which

\[
C^{0}_{\gamma, \gamma'} = \tilde{C}^{0}_{\gamma, \gamma'} \equiv \delta_{\gamma, \gamma'} \langle \hat{h}_\gamma^{\dagger}, \gamma \hat{h}_\gamma, \gamma' \rangle_{\Psi_0} \equiv n^{0}_{\gamma'}.
\]

With such a basis the expectation value (16) has the simple form

\[
m^{0}_{I,I'} = \delta_{I,I'} \prod_{\gamma \in I} n^{0}_{\gamma} \prod_{\gamma \not\in I} (1 - n^{0}_{\gamma}).
\]

Note that, for simplicity, we always use the same variable \( I \) for configuration states of the form (12) irrespective of the underlying orbital basis (e.g. \( \hat{c}_\sigma^{\dagger} \) in (12) or \( \hat{h}_\gamma^{\dagger} \) in (21)).

As will be shown in section 5, the time-dependent Gutzwiller theory requires one to calculate the first and second derivatives of the energy with respect to all elements of the local density matrix, including the non-diagonal terms. Since the local density matrix enters the energy functional solely through matrices of the form (16), we only need to expand these matrices with respect to small perturbations

\[
C^{0}_{\gamma, \gamma'} = \tilde{C}^{0}_{\gamma, \gamma'} + \delta C^{0}_{\gamma, \gamma'}
\]

up to second order in \( \delta C^{0}_{\gamma, \gamma'} \) around the diagonal ground-state matrix (20). This expansion is explicitly carried out in appendix A.

In our derivation of the ground-state energy, all local onsite energies were considered as part of the local Hamiltonian (2). For later use, however, we also need an expression for the expectation value of a general local one-particle Hamiltonian

\[
\hat{H}_{\text{onsite}} = \sum_{\sigma, \sigma'} \epsilon^{\sigma, \sigma'} \hat{c}_\sigma^{\dagger} \hat{c}_{\sigma'}.
\]

This expectation value is given as

\[
\langle \hat{H}_{\text{onsite}} \rangle_{\Psi_0} = \sum_{\sigma, \sigma'} \epsilon^{\sigma, \sigma'} C^{\sigma}_{\sigma, \sigma'},
\]

where

\[
C^{\sigma}_{\sigma, \sigma'} = \sum_{\Gamma_1, \Gamma_2, \Gamma_3, \Gamma_4} \lambda^{\pm}_{\Gamma_2, \Gamma_1} \lambda_{\Gamma_3, \Gamma_4} (\Gamma_2 | \hat{c}_\sigma^{\dagger} \hat{c}_{\sigma'} | \Gamma_3) m^{0}_{\Gamma_1, \Gamma_4}
\]

is the ‘correlated’ local density matrix.
3.2. Kinetic energy

The expectation value of a hopping term in \( \hat{H}_0 \), equation (1), is given as

\[
\langle \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma'} \rangle_{\psi_0} = \sum_{\sigma_1,\sigma_2,\sigma'_1,\sigma'_2} q_{i,\sigma_1}^{\sigma'_1} \left( q_{j,\sigma_2}^{\sigma'_2} \right)^* \langle \hat{c}_{i,\sigma_1}^\dagger \hat{c}_{j,\sigma_2} \rangle_{\psi_0},
\]

(26)

where we have introduced the (local) renormalization matrix \(^2\)

\[
q_{\sigma}^\sigma = \sum_{\Gamma_1, \ldots, \Gamma_4} \lambda_{\Gamma_1, \Gamma_4}^2 \lambda_{\Gamma_3} \Gamma_2 | \hat{c}_{i}^\dagger | \Gamma_3 \rangle \sum_{I_{i,l}} T_{I_{i,l}} T_{I_{j,l}}^* H_{I_{i,l}}^{\sigma}.
\]

(27)

The matrix \( H_{I_{i,l},I_{j,l}}^{\sigma} \) contains three different contributions depending on whether the index \( \sigma' \) is an element of \( I_{1} \cap I_{4} \), \( I_{4} \setminus (I_{1} \cap I_{4}) \), or \( J = (1, \ldots, N) \setminus (I_{1} \cup I_{4}) \). With the abbreviation \( f_{\sigma,1} \equiv \langle I | \hat{c}_{i}^\dagger \hat{c}_{i} | I \rangle \), we can write \( H_{I_{i,l},I_{j,l}}^{\sigma} \) as

\[
H_{I_{i,l},I_{j,l}}^{\sigma} \equiv (1 - f_{\sigma',1}) | I_{1} | \hat{c}_{i}^\dagger | I_{1} \cup \sigma' \rangle m_{I_{i,l},I_{j,l}}^{0,\sigma'},
\]

\[
+ (1 - f_{\sigma',1}) | I_{1} \setminus \sigma' \rangle \hat{c}_{i}^\dagger | I_{1} \rangle \left( f_{\sigma',1} m_{I_{i,l},I_{j,l}}^{0,\sigma'} + (1 - f_{\sigma',1}) m_{I_{i,l},I_{j,l}}^{0,\sigma'} \right).
\]

(28)

The expectation value \( m_{I_{i,l},I_{j,l}}^{0,\sigma'} \) in (28) has the same form as that in (16), except that the index \( J \) has to be replaced by \( J \setminus \sigma' \). In the case of a diagonal local density matrix, one finds

\[
H_{I_{i,l},I_{j,l}}^{\sigma} = \delta_{I_{1},I_{j,l}} \langle I_{1} \setminus \sigma' | \hat{c}_{i}^\dagger | I_{1} \rangle \frac{m_{I_{i,l},I_{j,l}}^{0,\sigma'}}{1 - C_{\sigma,\sigma'}^{0}},
\]

(29)

in agreement with the results derived earlier [7]. Note that, in general, the renormalization matrix is not Hermitian, i.e.

\[
q_{\sigma}^\sigma \neq (q_{\sigma}^\sigma)^*.
\]

(30)

Using (26), the expectation value of the one-particle Hamiltonian \( \hat{H}_0 \) can be written as

\[
\langle \hat{H}_0 \rangle_{\psi_0} = \sum_{\sigma_1,\sigma_2,\sigma'_1,\sigma'_2} q_{\sigma_1}^{\sigma'_1} (q_{\sigma_2}^{\sigma'_2})^* E_{\sigma_1,\sigma_2,\sigma'_1,\sigma'_2} \langle \hat{c}_{i,\sigma_1}^\dagger \hat{c}_{i,\sigma_2} \rangle_{\psi_0},
\]

(31)

where we introduced the tensor

\[
E_{\sigma_1,\sigma_2,\sigma'_1,\sigma'_2} \equiv \frac{1}{L^3} \sum_{i \neq j} r_{i,j}^{\sigma_1,\sigma_2} \langle \hat{c}_{i,\sigma_1}^\dagger \hat{c}_{j,\sigma_2} \rangle_{\psi_0}.
\]

(32)

3.3. Physical constraints

As it turns out through the evaluation of expectation values in infinite dimensions, the variational parameters \( \lambda_{\Gamma_{\tau}} \) need to obey certain local constraints. These are

\[
\langle \hat{P}^\dagger \hat{P} \rangle_{\psi_0} = 1,
\]

(33)

\[
\langle \hat{c}_{\sigma}^{\dagger} \hat{P}^\dagger \hat{P} \hat{c}_{\sigma'} \rangle_{\psi_0} = \langle \hat{c}_{\sigma}^{\dagger} \hat{c}_{\sigma} \rangle_{\psi_0} = C_{\sigma,\sigma'}^{0}.
\]

(34)

\(^2\) Note that, in local quantities, we frequently drop lattice site indices.
Note that moving the operator $\hat{P}^\dagger \hat{P}$ relative to $\hat{c}_\uparrow^\dagger$ or $\hat{c}_\alpha'$ in (34) would not alter the whole set of constraints. With the explicit form (5) of the correlation operator $\hat{P}$, the constraints read

$$1 = \sum_{\Gamma_1, \Gamma_2} \lambda_\uparrow^\dagger \Gamma_1 \lambda_\uparrow \Gamma_2 m^0_{\Gamma_1, \Gamma_2},$$

$$C^0_{\sigma, \sigma'} = \sum_{\Gamma_1, \Gamma_2, \Gamma_3} \lambda_\uparrow^\dagger \Gamma_2 \lambda_\uparrow \Gamma_2 \lambda_\uparrow \Gamma_3 \langle \Gamma | \hat{c}_\sigma^\dagger | \Gamma_1 \rangle \times \langle \Gamma | \hat{c}_\sigma' | \Gamma' \rangle m^0_{\Gamma_1, \Gamma'}.$$  

3.4. Recovery of the ‘standard’ single-band energy functional

In the case of a single-band model, the atomic eigenstates $|\Gamma\rangle$ coincide with the configuration states $|I\rangle$. If we assume a translationally invariant ground state and the most general form of a local density matrix (see footnote 2)

$$C^0 = \begin{pmatrix} (\langle \hat{c}_\uparrow^\dagger \hat{c}_\uparrow \rangle \psi_0 - |\langle \hat{c}_\uparrow^\dagger \hat{c}_\uparrow \rangle \psi_0|)^2 & (\langle \hat{c}_\uparrow^\dagger \hat{c}_\downarrow \rangle \psi_0 \langle \hat{c}_\uparrow^\dagger \hat{c}_\downarrow \rangle \psi_0) \\ (\langle \hat{c}_\uparrow^\dagger \hat{c}_\downarrow \rangle \psi_0 \langle \hat{c}_\uparrow^\dagger \hat{c}_\downarrow \rangle \psi_0) & (\langle \hat{c}_\downarrow^\dagger \hat{c}_\downarrow \rangle \psi_0 - |\langle \hat{c}_\downarrow^\dagger \hat{c}_\downarrow \rangle \psi_0|^2) \end{pmatrix},$$

where $(\Delta^0_{\uparrow, \downarrow})^* = \Delta^0_{\uparrow, \downarrow} = \Delta^0$, we find

$$m^0_{\uparrow, \uparrow} = (1 - n^0_{\uparrow})(1 - n^0_{\downarrow}) - |\Delta^0|^2,$$

$$m^0_{\uparrow, \downarrow} = n^0_{\uparrow}(1 - n^0_{\downarrow}) + |\Delta^0|^2,$$

$$m^0_{\downarrow, \downarrow} = \Delta^0_{\downarrow, \downarrow},$$

$$m^0_{\downarrow, \uparrow} = n^0_{\uparrow} n^0_{\downarrow} - |\Delta^0|^2$$

for those of the expectation values (15) that are finite. Here, we used the notation

$$\uparrow = \downarrow \quad \text{and} \quad \downarrow = \uparrow.$$  

As a consequence, the expectation value of the local Coulomb interaction in (6) reads

$$\sum_i U(|d_i\rangle \langle d_i|) \psi_0 = L_s U |\lambda_\uparrow|^2 m^0_{\downarrow, \downarrow}.$$  

For the single-band model with the correlation operator (7) and the local density matrix (37), the elements of the renormalization matrix have the form

$$q^0_\sigma = \lambda_\sigma^\dagger \lambda_\sigma (1 - n^0_\sigma) + \lambda_\sigma^\dagger \lambda_\sigma n^0_\sigma + (\lambda_\sigma^\dagger \lambda_\sigma^\dagger + \lambda_\sigma^\dagger \lambda_\sigma) \Delta^0_{\sigma, \sigma},$$

$$q^0_{\sigma'} = \Delta^0_{\sigma, \sigma'} (\lambda_\sigma^\dagger \lambda_\sigma - \lambda_\sigma^\dagger \lambda_{\sigma'} + \lambda_{\sigma'}^\dagger \lambda_\sigma n^0_\sigma + \lambda_{\sigma'}^\dagger \lambda_{\sigma'} (1 - n^0_\sigma)).$$

Finally, the constraints in this case are given as

$$1 = |\lambda_\uparrow|^2 m^0_{\uparrow, \uparrow} + |\lambda_\downarrow|^2 m^0_{\downarrow, \downarrow} + (|\lambda_\uparrow|^2 + |\lambda_{\downarrow}|^2) m^0_{\uparrow, \downarrow} + (|\lambda_\uparrow|^2 + |\lambda_{\uparrow}|^2) m^0_{\downarrow, \uparrow} + (\lambda_\uparrow^\dagger \lambda_\downarrow + \lambda_\downarrow^\dagger \lambda_\uparrow) \Delta^0_{\uparrow, \downarrow} + (\lambda_{\downarrow}^\dagger \lambda_{\downarrow} + \lambda_{\uparrow}^\dagger \lambda_{\uparrow}) \Delta^0_{\downarrow, \uparrow},$$

$$n^0_\sigma = (|\lambda_\sigma|^2 + |\lambda_{\sigma'}|^2) m^0_{\downarrow, \downarrow} + |\lambda_\sigma|^2 m^0_{\sigma, \sigma},$$

$$\Delta^0_{\sigma, \sigma'} = -(\lambda_\sigma^\dagger \lambda_\sigma + \lambda_{\sigma'}^\dagger \lambda_{\sigma'}) m^0_{\downarrow, \downarrow} + |\lambda_\sigma|^2 \Delta^0_{\sigma, \sigma'}.$$  

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As mentioned before, it is possible to overcome the complications that arise from a non-diagonal local density matrix by a simple transformation

\[ \hat{h}_\gamma^\dagger = \sum_\sigma u_{\sigma,\gamma} \hat{c}_\sigma^\dagger \]  

(49)

to a new orbital basis for which the local density matrix is diagonal by definition,

\[
\begin{pmatrix}
\langle \hat{h}_1^\dagger \hat{h}_1 \rangle_{\psi_0} & \langle \hat{h}_1^\dagger \hat{h}_2 \rangle_{\psi_0} \\
\langle \hat{h}_2^\dagger \hat{h}_1 \rangle_{\psi_0} & \langle \hat{h}_2^\dagger \hat{h}_2 \rangle_{\psi_0}
\end{pmatrix} = 
\begin{pmatrix}
\tilde{n}_1^0 & 0 \\
0 & \tilde{n}_2^0
\end{pmatrix}.
\]  

(50)

In this new basis the constraints have the rather simple form

\[
\begin{align*}
1 &= \bar{\lambda}_\emptyset m_{\emptyset,\emptyset} + \bar{\lambda}_d m_{d,d}, \\
\tilde{n}_\gamma^0 &= \bar{\lambda}_d^2 m_{\gamma,\gamma} + \bar{\lambda}_d^2 m_{d,d},
\end{align*}
\]  

(51, 52)

where the non-diagonal constraints are automatically fulfilled by working with a diagonal correlation operator (\(\bar{\lambda}_{1,2} = 0\)). Equations (51), (52) can be readily solved by introducing the expectation values

\[
\begin{align*}
\tilde{m}_d &\equiv \bar{\lambda}_d^2 m_{d,d}, \\
\tilde{m}_\emptyset &\equiv \bar{\lambda}_d^2 m_{\emptyset,\emptyset} = 1 - \tilde{n}_1^0 - \tilde{n}_2^0 + \tilde{m}_d, \\
\tilde{m}_\gamma &\equiv \bar{\lambda}_d^2 m_{\gamma,\gamma} = \tilde{n}_\gamma^0 - \tilde{m}_d,
\end{align*}
\]  

(53, 54, 55)

which leaves us with only one variational parameter, the expectation value \(\tilde{m}_d\) for double occupancy. The resulting renormalization matrix is then diagonal and its elements have the well-known form [47–49]

\[
q_\gamma \equiv q_\gamma^0 = \bar{\lambda}_d \bar{\lambda}_\gamma (1 - \tilde{n}_\gamma^0) + \bar{\lambda}_d \bar{\lambda}_\gamma \tilde{n}_\gamma^0 \frac{1}{\sqrt{\tilde{n}_\gamma^0 (1 - \tilde{n}_\gamma^0)}} \left( \sqrt{\tilde{m}_\emptyset \tilde{m}_\gamma} + \sqrt{\tilde{m}_d \tilde{m}_\gamma} \right).
\]  

(56)

Hence, the single-particle energy (31) is given as

\[
E_0 = \sum_\gamma q_\gamma^2 \sum_{i \neq j} t_{i,j} \langle \hat{h}_{i,\gamma}^\dagger \hat{h}_{j,\gamma} \rangle_{\psi_0},
\]  

(57)

where we used the orthonormality relation

\[
\sum_\sigma u_{\sigma,\gamma} u_{\sigma,\gamma}^* = \delta_{\gamma,\gamma'}.
\]  

(58)

In order to formally show the equivalence of both approaches, we write the parameters \(\lambda_{\Gamma,\Gamma'}\) in (7) as

\[
\begin{align*}
\lambda_\emptyset &\equiv \bar{\lambda}_\emptyset, \\
\lambda_d &\equiv \bar{\lambda}_d, \\
\lambda_{\sigma,\sigma'} &\equiv \sum_\gamma u_{\sigma,\gamma} u_{\sigma',\gamma} \bar{\lambda}_\gamma.
\end{align*}
\]  

(59, 60, 61)
With these relations it is easy to show that the constraints (46)–(48) are indeed fulfilled. For example, the first constraint (46) can be written as

\[ 1 = \lambda_0^2 m_{\Psi,0}^0 + \lambda_0^2 m_{d,0}^0 + \sum_{\sigma,\sigma',\alpha} \lambda_{\sigma'\sigma}^* \lambda_{\sigma'\sigma} m_{\alpha,\sigma'}^0. \]  

(62)

If we use

\[ m_{\alpha,\sigma'}^0 = \sum_{\gamma} u_{\alpha,\gamma}^* u_{\sigma',\gamma} m_{\gamma}^0 \]

(63)

and the orthonormality relation (58), we readily find that (62) is indeed solved by the parameters (59)–(61). In the same way, one can show the equivalence of the ground-state energy functionals.

4. Time-dependent Hartree–Fock approximation

The approximation most frequently applied to two-particle Green’s functions is the RPA. This approach can be derived in various ways, e.g. by an equation of motion technique or in diagrammatic perturbation theory [50]. In this section, we use a different derivation, that introduces the RPA as a time-dependent generalization of the HF theory; see e.g. [51, 52]. If derived in this way, the approach can be generalized quite naturally to formulate a time-dependent Gutzwiller theory. This will be the subject of section 5.

4.1. Hartree–Fock approximation

In the HF approximation, a single-particle product wave function \( |\Psi_0\rangle \) is used in order to investigate the ground-state properties of a many-particle system. Note that such wave functions are included in the Gutzwiller variational space by setting \( \lambda_{i,j} = \delta_{i,j} \). The expectation value of a many-particle Hamiltonian with respect to an HF wave function is a function of the single-particle density matrix. For example, for the Hamiltonian (1) it reads

\[ E_{HF}(\tilde{\rho}) = \langle \tilde{H} \rangle_{\Psi_0} = \sum_{i \neq j; \sigma, \sigma'} \rho_{(j\sigma'),(i\sigma)} + \sum_{i; \sigma_1, \sigma_2} \epsilon_{i}^{\sigma_1,\sigma_2} \rho_{(i\sigma_2),(i\sigma_1)} + \sum_{i} E_{HF,loc,i}(\tilde{\rho}), \]

(64)

where

\[ \rho_{(j\sigma'),(i\sigma)} \equiv \langle \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma'} \rangle_{\Psi_0} \]

(65)

are the elements of the single-particle density matrix \( \tilde{\rho} \) and

\[ E_{HF,loc,i}^{\tilde{\rho}} = \frac{1}{2} \sum_{\sigma_1,\sigma_2,\sigma_3,\sigma_4} U_{i}^{\sigma_1,\sigma_2,\sigma_3,\sigma_4} \left[ \rho_{(i\sigma_4),(i\sigma_1)} \rho_{(i\sigma_3),(i\sigma_2)} - \rho_{(i\sigma_3),(i\sigma_1)} \rho_{(i\sigma_4),(i\sigma_2)} \right] \]

(66)

is the expectation value of the Coulomb interaction in the Hamiltonian (2). Note that it will be more convenient in both the time-dependent HF and Gutzwiller theory to use a different order of subscripts in the definition (65) of density matrices than, e.g., in section 3 or in previous work on the Gutzwiller theory.
To keep notations simple, we use the abbreviations $\nu \equiv (i, \sigma)$ for local single-particle states and $Y = (\nu, \nu')$ for pairs of these indices. For example, the elements of $\tilde{\rho}$ can then be written as

$$\rho_Y = \rho_{\nu_1, \nu_2} = \rho_{(i_1, \sigma_1), (i_2, \sigma_2)}. \quad (67)$$

With these new notations, the HF energy (64) reads

$$E_{\text{HF}}(\tilde{\rho}) = \sum_{\nu_1, \nu_2} \varepsilon_{\nu_1, \nu_2} \rho_{\nu_1, \nu_2} + \frac{1}{2} \sum_{\nu_3, \nu_4} \rho_{\nu_3, \nu_4} W_{\nu_1, \nu_2; \nu_3, \nu_4} \rho_{\nu_3, \nu_2}$$

$$= \sum_Y \varepsilon_Y \rho_Y + \frac{1}{2} \sum_{Y, Y'} \rho_{Y} W_{Y, Y'} \rho_{Y'}. \quad (68)$$

where

$$\varepsilon_{(\sigma_1), (\sigma_2)} \equiv \delta_{i, j} \varepsilon_{i, j}, \quad \delta_{i, j} \epsilon_{\sigma_1, \sigma_2}$$

and

$$W_{(\nu_1, \nu_2), (\nu_3, \nu_2)} \equiv U_{i}^{\sigma_1, \sigma_2, \sigma_3, \sigma_4} - U_{i}^{\sigma_1, \sigma_2, \sigma_4, \sigma_3} \quad (70)$$

for indices $\nu_k = (i, \sigma_k)$ that belong to the same lattice site $i$. Further, we introduced the ‘inverse’ index $\bar{Y} \equiv (\nu', \nu)$ for $Y = (\nu, \nu')$. Note the symmetries

$$W_{(\nu_1, \nu_4), (\nu_3, \nu_2)} = W_{(\nu_2, \nu_3), (\nu_4, \nu_1)} = -W_{(\nu_1, \nu_3), (\nu_4, \nu_2)}, \quad \bar{Y}'$$

which will be employed in the following section.

The energy functional (68) has to be minimized with respect to all density matrices that belong to a single-particle product state. Such matrices are idempotent, i.e. they obey the matrix equation

$$\tilde{\rho}^2 = \tilde{\rho}. \quad (72)$$

If one imposes this constraint via a Lagrange parameter matrix $\tilde{\eta}$ with elements $\eta_{\nu, \nu'}$, the following equation has to be solved:

$$\frac{\partial}{\partial \rho_{\nu, \nu'}} [E_{\text{HF}}(\tilde{\rho}) - \text{tr}(\tilde{\eta}(\tilde{\rho}^2 - \tilde{\rho}))] = 0. \quad (73)$$

This condition leads to

$$\tilde{h}(\tilde{\rho}) + \tilde{\eta} - \tilde{\rho} \tilde{\eta} - \tilde{\rho} \tilde{\eta} = 0, \quad (74)$$

where we introduced the matrix $\tilde{h}(\tilde{\rho})$ with the elements

$$\tilde{h}_Y(\tilde{\rho}) = \frac{\partial}{\partial \rho_Y} E_{\text{HF}}(\tilde{\rho}) = \varepsilon_Y + \sum_{Y'} W_{Y, Y'} \rho_{Y'}. \quad (75)$$

Equation (74) is solved if $\tilde{\rho}$ satisfies both (72) and

$$[\tilde{h}(\tilde{\rho}), \tilde{\rho}] = 0. \quad (76)$$

Starting with a certain density matrix $\tilde{\rho}$, we can introduce the ‘HF’ basis

$$|\alpha\rangle = \sum_{\nu} u_{\nu, \alpha} |\nu\rangle \quad (77)$$
of states that diagonalize the Hamilton matrix \( \tilde{h}(\tilde{\rho}) \), i.e.

\[
\sum_{\nu'} h_{\nu,\nu'}(\tilde{\rho}) u_{\nu',\alpha} = E_{\alpha} u_{\nu,\alpha}.
\]

Equation (76) is then solved by setting

\[
\rho_{\alpha,\alpha'} = \delta_{\alpha,\alpha'} \Theta(E_F - E_{\alpha}),
\]

where the Fermi energy \( E_F \) is determined by the total number of particles

\[
N = \sum_{\alpha} \Theta(E_F - E_{\alpha}).
\]

The density matrix (79) has to be reinserted into (68), (75) until self-consistency is reached. We denote the solution of these equations as \( \tilde{\rho}^0 \) and introduce the corresponding Hamilton matrix \( \tilde{h}^0 \equiv \tilde{h}(\tilde{\rho}^0) \).

### 4.2. Equation of motion for the density matrix

We consider two-particle Green’s functions of the form

\[
G_{(\nu_2,\nu_1), (\nu_3,\nu_4)}(t - t') \equiv \langle \langle \hat{c}^{\dagger}_{\nu_1}(t) \hat{c}_{\nu_2}(t); \hat{c}^{\dagger}_{\nu_3}(t') \hat{c}_{\nu_4}(t') \rangle \rangle
\]

\[
\equiv -i\Theta(t - t') \langle \Phi_0||[\hat{c}^{\dagger}_{\nu_1}(t)\hat{c}_{\nu_2}(t), \hat{c}^{\dagger}_{\nu_3}(t')\hat{c}_{\nu_4}(t')]|\Phi_0 \rangle,
\]

where \( |\Phi_0 \rangle \) is the exact ground state of our multi-band Hubbard Hamiltonian (1), and \( \hat{c}^{(t)}_{\nu}(t) \) is the Heisenberg representation of the operators \( \hat{c}^{(t)}_{\nu} \) with respect to \( \hat{H} \). As shown in most textbooks on many-particle physics, the Green’s functions (82) naturally arise in ‘linear-response theory’ because they describe the time-dependent changes

\[
\delta \langle \hat{c}^{\dagger}_{\nu_1} \hat{c}_{\nu_2} \rangle_t \equiv \langle \hat{c}^{\dagger}_{\nu_1} \hat{c}_{\nu_2} \rangle_t - \langle \hat{c}^{\dagger}_{\nu_1} \hat{c}_{\nu_2} \rangle_{-\infty} \equiv \delta \rho_{\nu_2,\nu_1}(t)
\]

of the density matrix \( \tilde{\rho} \) in the presence of a small time-dependent perturbation

\[
\hat{V}_f(t) = \sum_{\nu,\nu'} f_{\nu,\nu'}(t) \hat{c}^{\dagger}_{\nu} \hat{c}_{\nu'}
\]

added to \( \hat{H} \) [53–55]. After a Fourier transformation and using again the abbreviation \( Y = (\nu, \nu') \), equation (83) reads

\[
\delta \rho_Y(\omega) = \sum_{Y'} G_{Y,Y'}(\omega) f_Y(\omega)
\]

with

\[
G_{Y,Y'}(\omega) \equiv \int_{-\infty}^{\infty} d\tau G_{Y,Y'}(\tau)e^{i\omega\tau},
\]

and \( f_Y(\omega) \) and \( \delta \rho_Y(\omega) \) defined accordingly.

Ideally, we would like to calculate the time dependence of the density matrix

\[
\rho_{\nu',\nu}(t) \equiv \langle \Psi(t)|\hat{c}^{\dagger}_{\nu'} \hat{c}_{\nu}|\Psi(t) \rangle,
\]
where $|\Psi(t)\rangle$ is the exact solution of the time-dependent Schrödinger equation for the Hamiltonian

$$\hat{H}(t) = \hat{H} + \hat{V}_f(t).$$

(88)

The expectation value (87) obeys the Heisenberg equation

$$-i\dot{\hat{\rho}}_{\nu',\nu}(t) = \langle \Psi(t) | [\hat{H}, \hat{c}_{\nu'}\hat{c}_{\nu}] | \Psi(t) \rangle,$$

(89)

which contains the commutator

$$[\hat{H}(t), \hat{c}_{\nu'}\hat{c}_{\nu}] = \sum_{\nu_1} (\epsilon_{\nu_1,\nu} + f_{\nu_1,\nu}(t))\hat{c}_{\nu_1}\hat{c}_{\nu'} - \sum_{\nu_1} (\epsilon_{\nu',\nu_1} + f_{\nu',\nu_1}(t))\hat{c}_{\nu_1}\hat{c}_{\nu'}$$

$$+ \frac{1}{2} \sum_{\nu_1,\nu_2,\nu_3} \left( W_{(\nu_1,\nu_3),(\nu',\nu_2)}\hat{c}_{\nu_1}\hat{c}_{\nu_2}\hat{c}_{\nu_3} + W_{(\nu_1,\nu_2),(\nu_3,\nu')}\hat{c}_{\nu_1}\hat{c}_{\nu_2}\hat{c}_{\nu_3} \right).$$

(90)

In the time-dependent HF approximation, it is assumed that the solution $|\Psi(t)\rangle$ of the Schrödinger equation at any time $t$ is approximately given by a single-particle product wave function. In this case, the expectation value of the commutator (90) can be evaluated by means of Wick’s theorem. This leads to the equation of motion

$$i\dot{\hat{\rho}}(t) = [\hat{h}(\hat{\rho}(t)) + \bar{f}(t), \hat{\rho}(t)]$$

(91)

for $\hat{\rho}(t)$, where the matrix $\hat{h}(\hat{\rho})$ has been introduced in (75). Equations (75) and (91) will also be crucial for our formulation of a time-dependent Gutzwiller theory in section 5.

4.3. Expansion for weak perturbations

We are only interested in cases where

$$\hat{V}_f(t) \rightarrow \delta \hat{V}_f(t) = \sum_{\nu,\nu'} \delta f_{\nu,\nu'}(t)\hat{c}_{\nu'}\hat{c}_{\nu}$$

(92)

is a weak perturbation to the time-independent Hamiltonian $\hat{H}$. In this case, the density matrix $\hat{\rho}(t)$ and the Hamilton matrix $\hat{h}(t)$ are given as

$$\hat{\rho}(t) \approx \hat{\rho}^0 + \delta \hat{\rho}(t),$$

(93)

$$\hat{h}(t) \approx \hat{h}^0 + \delta \hat{h}(t),$$

(94)

where $\delta \hat{\rho}(t)$ describes a ‘small’ time-dependent perturbation around the ground-state density matrix $\hat{\rho}^0$, and

$$\hat{h}_Y^0 = \epsilon_Y + \sum_{Y'} W_{Y,Y'}\rho_Y^0,$$

(95)

$$\delta \hat{h}_Y(t) = \sum_{Y'} W_{Y,Y'}\delta \rho_Y(t).$$

(96)

With the expansion (93)–(94), the equation of motion (91) becomes

$$0 = [\hat{h}^0, \delta \hat{\rho}],$$

(97)

$$i\delta \dot{\hat{\rho}}(t) = [\hat{h}^0, \delta \hat{\rho}(t)] + [\delta \hat{h}(t) + \delta \bar{f}(t), \hat{\rho}^0].$$

(98)
These equations have to be solved for density matrices \( \tilde{\rho}(t) \) that obey the matrix equation (72). After applying the expansion (93), equation (72) reads (to leading order in \( \delta \tilde{\rho}(t) \))

\[
\begin{align*}
\tilde{\rho}^0 &= (\tilde{\rho}^0)^2, \\
\delta \tilde{\rho}(t) &= \tilde{\rho}^0 \delta \tilde{\rho}(t) + \delta \tilde{\rho}(t) \tilde{\rho}^0.
\end{align*}
\]

Note that equations (97), (99) just recover the time-independent Hartree–Fock equations derived in section 4.1.

### 4.4. Random-phase approximation equations

Mathematically, the density matrix is a projector onto ‘hole’ states, \( \tilde{\rho}_h \equiv \tilde{\rho}^0 \). In addition, we define the projector onto ‘particle’ states as

\[
\tilde{\rho}_p \equiv 1 - \tilde{\rho}^0.
\]

With these two operators, we can decompose all matrices into their four components

\[
\begin{align*}
\delta \tilde{\rho}^{vw}(t) &\equiv \tilde{\rho}_v \delta \tilde{\rho}(t) \tilde{\rho}_w, \\
\delta \tilde{f}^{vw}(t) &\equiv \tilde{\rho}_v \delta \tilde{f}(t) \tilde{\rho}_w, \\
\tilde{h}^{0;vw}(t) &\equiv \tilde{\rho}_v \tilde{h}^0 \tilde{\rho}_w,
\end{align*}
\]

where \( v, w \in \{p, h\} \). Note that \( \tilde{h}^{0;vw} \) has the elements

\[
\begin{align*}
\tilde{h}_{u,a}^{0;vw} &= \delta_{v,w} \delta_{a,a'} E_a.
\end{align*}
\]

An evaluation of the condition (100) for the components \( \delta \tilde{\rho}^{vw}(t) \) yields

\[
\begin{align*}
\delta \tilde{\rho}^{vw}(t) &= \delta \tilde{\rho}^{ww}(t) + \delta \tilde{\rho}^{vw}(t) \delta \tilde{\rho}^{vw}(t) + \delta \tilde{\rho}^{vw}(t) \delta \tilde{\rho}^{ww}(t) \\
\delta \tilde{\rho}^{ww}(t) &= 0,
\end{align*}
\]

where \( v \neq w \). Hence, the components \( \delta \tilde{\rho}_{pp}(t) \) and \( \delta \tilde{\rho}_{hh}(t) \) can be neglected in the following compared to the leading fluctuations \( \delta \tilde{\rho}_{ph}(t) \) and \( \delta \tilde{\rho}_{hp}(t) \).

We express the time-dependent quantities \( \delta \tilde{\rho}^{vw}(t) \) and \( \delta \tilde{f}^{vw}(t) \) by their respective Fourier transforms \( \delta \tilde{\rho}^{vw}(\omega) \) and \( \delta \tilde{f}^{vw}(\omega) \). The equation of motion (98) then leads to

\[
(\omega + \delta h^{0;vw}) (\omega) = (E_{a_1} - E_{a_2}) \delta \rho^{vw}_{a_1,a_2} (\omega) \pm (\delta h^{vw}_{a_1,a_2} (\omega) + \delta f^{vw}_{a_1,a_2} (\omega)),
\]

where the plus and minus signs correspond to \( vw = ph \) and \( vw = hp \), respectively. With the abbreviation \( A = (a_1, a_2) \) for pairs of indices \( a \), we find

\[
\delta h^{vw}_A (\omega) = - \sum_{A'} U_{A,A'} (\delta \rho^{vw}_{A'} (\omega) + \delta \rho^{vw}_{A'} (\omega)).
\]

Here, elements of the matrix \( U \) are given as

\[
U_{A,A'} = U_{(a_1,a_2),(a_1',a_2')} \equiv - \sum_{v_1,v_2} u^{v_1,a_1}_{v_1',a_1'} u^{v_2,a_2}_{v_2',a_2'} W_{(v_1,v_2),(v_1',v_2')} u^{v_1,a_1}_{v_1',a_1'} u^{v_2,a_2}_{v_2',a_2'}.
\]
The coefficients $u_{\nu,\alpha}$ in (110) have been introduced in equation (77) and determine the solutions $|\alpha\rangle$ of the HF equations. Equations (108) and (109) then yield

$$\begin{pmatrix}(\omega - \tilde{E}) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \tilde{U} \end{pmatrix} \begin{pmatrix} \delta \tilde{\rho}^{ph}(\omega) \\ \delta \tilde{\rho}^{hp}(\omega) \end{pmatrix} = \begin{pmatrix} \delta \tilde{f}^{ph}(\omega) \\ \delta \tilde{f}^{hp}(\omega) \end{pmatrix},$$

with a matrix $\tilde{E}$ defined as

$$E_{\alpha,\alpha'} = E_{(\alpha_1,\alpha_2),(\alpha'_1,\alpha'_2)} = \delta_{\alpha_1,\alpha'_1} \delta_{\alpha_2,\alpha'_2} (E_{\alpha_1} - E_{\alpha_2}).$$

By comparing equations (111) and (85), we find

$$\tilde{G}^{-1}(\omega) = \begin{pmatrix}(\omega + i\delta - \tilde{E}) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \tilde{U} \end{pmatrix}$$

for the inverse of the two-particle Green’s function

$$G_{\alpha,\alpha'}(\omega) = G_{(\alpha_1,\alpha_2),(\alpha'_1,\alpha'_2)}(\omega) = \sum_{\nu_1,\nu_2,\nu'_1,\nu'_2} u_{\nu_1,\alpha_1} u_{\nu'_2,\alpha_2} G_{(\nu_1,\nu_2),(\nu'_1,\nu'_2)}(\omega) u_{\nu'_1,\alpha'_1} u_{\nu_2,\alpha'_2},$$

Here, we have added an increment $i\delta$ with $\delta = 0^+$ in order to ensure the correct boundary conditions of a retarded Green’s function. For $\tilde{U} = 0$, the inverse Green’s function (113) reads

$$\tilde{\Gamma}^{-1}(\omega) \equiv \pm (\omega + i\delta - \tilde{E}),$$

which leads to

$$\Gamma_{\alpha,\alpha'}(\omega) = \Gamma_{(\alpha_1,\alpha_2),(\alpha'_1,\alpha'_2)}(\omega) = \delta_{\alpha_1,\alpha'_1} \delta_{\alpha_2,\alpha'_2} \rho^{0}_{\alpha_2,\alpha_2} - \rho^{0}_{\alpha_1,\alpha_1} \over \omega - (E_{\alpha_1} - E_{\alpha_2}) + i\delta.$$  

Note that $\tilde{\Gamma}$ is not the exact Green’s function for the single-particle Hamiltonian $\hat{H}_0$ since we just set $\tilde{U} = 0$ in (113) but kept the ‘HF self-energy’ contributions finite,

$$\Sigma_A \equiv \sum_{\alpha'} W_{A,\alpha'} \rho^{0}_{A'},$$

which usually change the ‘eigenvalues’ $E_\alpha$ in (116); cf equations (75) and (78).

With the Green’s function (116), we can write (113) as

$$\tilde{G}(\omega) = \tilde{\Gamma}(\omega) [1 + \tilde{U} \tilde{\Gamma}(\omega)]^{-1} \equiv \tilde{\Gamma}(\omega) + \tilde{\Gamma}(\omega) \tilde{U} \tilde{G}(\omega),$$

where, in the second line, we expanded $[1 + \tilde{U} \tilde{\Gamma}(\omega)]^{-1}$ into a power series with respect to $\tilde{U} \tilde{\Gamma}$.

Both equations (118) and (119) are familiar expressions for the two-particle Green’s function in the RPA.

5. Time-dependent Gutzwiller theory

The time-dependent Gutzwiller approximation was first introduced for single-band Hubbard models by Seibold et al [31, 32]. In this section, we generalize this approach for the investigation of multi-band models. To this end, we set up an effective energy functional of the density matrix in section 5.1, which is used in sections 5.2–5.4 to derive the Gutzwiller RPA equations.
5.1. **Effective energy functional**

As summarized in section 3, the expectation value of the multi-band Hamiltonian (1) in the Gutzwiller theory is a function of the variational parameters $\lambda_{\Gamma,\Gamma'}$ and of the one-particle wave function $|\Psi_0\rangle$. Like in the HF theory, the single-particle wave function $|\Psi_0\rangle$ enters the energy functional solely through the elements (65) of the non-interacting density matrix $\tilde{\rho}$. It is therefore possible to consider the energy

$$ E = E(\vec{\lambda}, \tilde{\rho}) $$

(120)
as a function of the density matrix $\tilde{\rho}$ and of the ‘vector’

$$ \vec{\lambda} = (\{\lambda^s_{\Gamma,\Gamma'}\}, \{\lambda_{\Gamma,\Gamma'}\}) = (\lambda_1, \ldots, \lambda_{n_p}) $$

(121)
of $n_p$ variational parameters $\lambda_{\Gamma,\Gamma'}$ (and $\lambda^s_{\Gamma,\Gamma'}$ for $\Gamma \neq \Gamma'$). The density matrix in the energy functional (120) must be derived from a single-particle wave function and, therefore, it has to obey the condition (72). Note that, in the following considerations, the density matrix will be considered either as a matrix (with respect to its two indices $(i, \sigma)$ and $(j, \sigma')$) or as a vector (with respect to its single index $Y$). To distinguish both cases, we will denote the density matrix $\tilde{\rho}$ in some equations as $\vec{\rho}$ in order to indicate its vector interpretation.

The constraints (35)–(36) are also functions of $\vec{\lambda}$ and $\tilde{\rho}$ and will be denoted as

$$ g_n(\vec{\lambda}, \tilde{\rho}) = 0, \quad 1 \leq n \leq n_c. $$

(122)

Here, $n_c$ is the (maximum) number of independent constraints, which, due to symmetries, is usually smaller than its maximum value $N_{so}^2 + 1$, where $N_{so}$ is the number of spin–orbital states per lattice site. We assume that the functions (122) are real, i.e. in the case of complex equations (33)–(34) their real and imaginary parts are treated separately.

By solving equations (122), we can, at least in principle, express $n_c$ of the variational parameters ($\equiv \lambda^d_X$) through the density matrix $\rho_Y$ and the remaining ‘independent’ parameters ($\equiv \lambda^i_X$),

$$ \lambda^d_X = \lambda^i_X(\vec{\lambda}^i, \tilde{\rho}). $$

(123)

In this way, we obtain an energy functional

$$ E^{GA}(\vec{\lambda}^i, \tilde{\rho}) \equiv E(\vec{\lambda}^d(\vec{\lambda}^i, \tilde{\rho}), \vec{\lambda}^i, \tilde{\rho}), $$

(124)

which has to be minimized without constraints apart from equation (72) and the condition that the total particle number

$$ N = \sum_\upsilon \rho_{\upsilon,\upsilon} $$

(125)
is conserved.

For a fixed density matrix $\tilde{\rho}$, the minimization of (124) with respect to the parameters $\lambda^i_X$,

$$ \frac{\partial}{\partial \lambda^i_X} E^{GA}(\vec{\lambda}^i, \tilde{\rho}) = 0, $$

(126)
determines these parameters

$$ \vec{\lambda}^i = \vec{\lambda}^i(\tilde{\rho}) $$

(127)
as a function of $\tilde{\rho}$. This allows us to define the ‘effective’ energy functional

$$ E^{eff}(\tilde{\rho}) = E^{GA}(\vec{\lambda}^i(\tilde{\rho}), \tilde{\rho}), $$

(128)
which, for a fixed density matrix \( \bar{\rho} \), is given as the minimum of \( E^{GA} \) with respect to \( \bar{\lambda} \). With this effective functional, we will formulate the time-dependent Gutzwiller theory in the following section.

Using a Lagrange-parameter matrix \( \tilde{\eta} \) as in section 4.1, we find

\[
\frac{\partial}{\partial \rho_{\mu,\nu}} [E_{\text{eff}}(\tilde{\rho}) - \text{tr}(\tilde{\eta}(\rho^2 - \tilde{\rho}))]_{\tilde{\rho} = \tilde{\rho}} = 0,
\]

which leads to

\[
0 = [\tilde{h}(\rho), \tilde{\rho}].
\]

Here we introduced the matrix \( \tilde{h}(\rho) \) with the elements

\[
h_{\bar{Y}}(\rho) = \frac{\partial E_{\text{eff}}(\tilde{\rho})}{\partial \rho_{\bar{Y}}},
\]

and used again the notation \( \bar{Y} \equiv (j, \sigma' ; i, \sigma) \) for \( Y = (i, \sigma ; j, \sigma') \). The self-consistent solution of equations (130)-(131) then yields the ground-state density matrix \( \tilde{\rho}_0 \), the matrix \( \tilde{h}_0 \equiv \tilde{h}(\tilde{\rho}_0) \) and the corresponding single-particle ‘Gutzwiller–Hamiltonian’

\[
\hat{h}_0 \equiv \sum_{i,j;\sigma,\sigma'} h_{0,i;\sigma ; j,\sigma'}(\tilde{\rho}_0) \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma'}.
\]

5.2. Gutzwiller random-phase approximation equations

The derivation of RPA-type equations within the time-dependent Gutzwiller theory goes along the same lines as discussed in section 4 for the time-dependent HF theory. We add a small time-dependent field

\[
\delta \tilde{V}_f(t) = \sum_{i,j;\sigma,\sigma'} \delta f_{i,\sigma ; j,\sigma'}^0(t) \hat{c}_{j,\sigma}^\dagger \hat{c}_{i,\sigma} + \text{h.c.}
\]

to our multi-band Hamiltonian (1). With the particular time dependence

\[
\delta f_{i,\sigma ; j,\sigma'}^0(t) = \delta f_{i,\sigma ; j,\sigma'}^0(\omega) e^{-i\omega t},
\]

the expectation value of \( \delta \tilde{V}(t) \) reads

\[
\langle \delta \tilde{V}_f(t) \rangle = \sum_{i,j;\sigma,\sigma'} \delta \tilde{f}_{i,\sigma ; j,\sigma'}(\omega) e^{-i\omega t} \rho_{j,\sigma';i,\sigma} + \text{c.c.,}
\]

where

\[
\delta \tilde{f}_{i,\sigma_1; j,\sigma_2}(\omega) = \delta_{i,j} \tilde{f}_{i,\sigma_1; i,\sigma_2}^0(\omega) \frac{C_{\sigma_1;\sigma_2}}{\rho_{\sigma_2;\sigma_1}} + (1 - \delta_{i,j}) \sum_{\sigma_1',\sigma_2'} \delta \tilde{f}_{i,\sigma_1'; j,\sigma_2'}^0(\omega) q_{\sigma_1'}^{\sigma_1}(q_{\sigma_2'}^{\sigma_2})^*.
\]
Our main assumption is now that $\delta \tilde{\rho}(t)$ obeys the same equation of motion,

$$i\hbar \frac{\partial}{\partial t} \tilde{\rho}(t) = \left[ \tilde{h}_0, \delta \tilde{\rho}(t) \right] + \left[ \delta \tilde{h}(t) + \delta \tilde{f}(t), \tilde{\rho}_0 \right],$$

as the density matrix in the time-dependent HF theory; see equation (98). Here, however, the Hamilton matrix

$$\tilde{h}(t) \approx \tilde{h}_0(t) + \delta \tilde{h}(t)$$

is not derived from the Hartree–Fock functional (68), but from the effective energy functional (128),

$$h_Y(t) = \frac{\partial}{\partial \rho} E_{\text{eff}}(\tilde{\rho}) \approx h^0_Y + \sum_{Y'} K_{Y,Y'} \delta \rho_Y(t) \equiv h^0_Y + \delta h_Y(t),$$

where the matrix $\tilde{K}$ is given as

$$\tilde{K}_{Y,Y'} \equiv \frac{\partial^2 E_{\text{eff}}}{\partial \rho_Y \partial \rho_{Y'}} \bigg|_{\tilde{\rho}=\tilde{\rho}_0}. \quad (141)$$

The diagonalization of $\tilde{h}_0$ (or equivalently of the Gutzwiller Hamiltonian $\hat{h}_0$) yields a basis $|\alpha\rangle$ with

$$h^0_{a,a'} = h^0_{a'} = \delta_{a,a'} E_a \quad (142)$$

and a ground-state density matrix that is given as

$$\tilde{\rho}^0_{a,a'} = \rho^0_{a'} = \delta_{a,a'} \Theta(E_F - E_a). \quad (143)$$

With the projectors $\tilde{\rho}_h \equiv \tilde{\rho}^0$ and $\tilde{\rho}_p \equiv 1 - \tilde{\rho}^0$, we define the particle and hole components of all matrices, as we did in equations (102)–(104). The components $\delta \tilde{\rho}^{vu}(t)$ of the density matrix fluctuations obey equations (106)–(107), i.e. to leading order we can neglect $\delta \tilde{\rho}^{hh}(t)$ and $\delta \tilde{\rho}^{pp}(t)$. Hence, after a Fourier transformation we end up with the same form of RPA equations,

$$\begin{bmatrix}
(\omega - \tilde{E}) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \tilde{K}
\end{bmatrix}
\begin{pmatrix}
\delta \tilde{\rho}^{ph}(\omega) \\
\delta \tilde{\rho}^{hp}(\omega)
\end{pmatrix}
= \begin{pmatrix}
\delta \tilde{f}^{ph}(\omega) \\
\delta \tilde{f}^{hp}(\omega)
\end{pmatrix}, \quad (144)$$

as in equation (111). Here, however, the bare matrix of Coulomb parameters $\tilde{U}$ is replaced by the matrix $\tilde{K}$, defined in (141), and the energies $E_a$ in the matrix $\tilde{E}$, equation (112), are the eigenvalues of the Gutzwiller Hamiltonian (132). Comparison with (85) leads to the final result

$$\tilde{G}(\omega) \equiv \begin{pmatrix}
(\omega + i\delta - \tilde{E}) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \tilde{K}
\end{pmatrix}^{-1} \quad (145)$$

for the two-particle Green’s function matrix within the time-dependent Gutzwiller approximation.

One should keep in mind that the external ‘fields’ $\delta \tilde{f}(\omega)$ in (144) are ‘renormalized’, i.e. they are not the bare fields as they appear in (85); see equation (136). On the other hand, on the lhs of (85) appears the ‘correlated’ expectation value of the density matrix, while in (144) we work with the fluctuations of the uncorrelated density matrix. Therefore, the ‘true’ Green’s function seen in experiments may, in fact, be given as

$$\tilde{G}_{Y,Y'}(\omega) \equiv c_{Y,Y'} G_{Y,Y'}(\omega), \quad (146)$$
with certain frequency-independent factors $c_{Y,Y'}$. These factors, however, are of minor importance since they only affect the overall spectral weight and not the frequency dependence of the Green's function matrix $\tilde{G}(\omega)$. We can calculate them with the assumption that correlated and uncorrelated density-matrix fluctuations are related through the same renormalization factors as the corresponding ground-state density matrices. For the one-band model, it has been checked that this prescription is in fact the correct procedure for which the correlation functions fulfil the standard sum rules $[34, 37, 43]$.

5.3. Second-order expansion of the energy functional

For an evaluation of the Gutzwiller RPA equations (144), we need to determine the matrix $\tilde{K}$ which is given by the second derivatives (141) of the effective energy functional (128). This end, we expand $E^{GA}$ up to second order around the ground-state values $\tilde{\rho}^0$ and $\tilde{\lambda}^{i0} = \tilde{\lambda}^i(\tilde{\rho}^0)$,

$$ E^{GA}(\tilde{\lambda}^i, \tilde{\rho}) = E_0 + \text{tr}(\tilde{h}^0 \delta \tilde{\rho}) + \frac{1}{2} \sum_{Y,Y'} \delta \rho_Y M^\rho_{Y,Y'} \delta \rho_{Y'} + \sum_{Z,Z'} \delta \lambda^i_Z M^{\lambda^i}_{Z,Z'} \delta \lambda^{i'}_Z $$

$$ + \sum_{Z,Y} \left( \delta \lambda^i_Z M^{\lambda^i}_{Z,Y} \delta \rho_Y + \delta \rho_{Y} M^{\rho^i}_{Y,Z} \delta \lambda^{i'}_Z \right) $$

$$ \equiv E_0 + \text{tr}(\tilde{h}^0 \delta \tilde{\rho}) + \delta E^{(2)}. $$

(147)

Here, we introduced the matrices $\tilde{M}^\rho$, $\tilde{M}^{\lambda^i}$, and $\tilde{M}^{\rho^i}$ with the elements

$$ M^\rho_{Y,Y'} = \frac{\partial^2 E^{GA}}{\partial \rho_Y \partial \rho_{Y'}}, $$

(148)

$$ M^{\lambda^i}_{Z,Y} = \frac{\partial^2 E^{GA}}{\partial \lambda^i_Z \partial \rho_Y} = M^{\rho^i}_{Y,Z}, $$

(149)

$$ M^{\lambda^i}_{Z,Z'} = \frac{\partial^2 E^{GA}}{\partial \lambda^i_Z \partial \lambda^{i'}_{Z'}}, $$

(150)

where the second derivatives on the rhs are evaluated for $\tilde{\rho} = \tilde{\rho}^0$ and $\tilde{\lambda}^i = \tilde{\lambda}^{i0}$. Note that there is no linear term $\sim \lambda^i_{Z'}$ in (147) because of the minimization condition (126). For our further evaluation, it is useful to write the second-order terms in equation (147) in a more compact form by means of matrix–vector products,

$$ \delta E^{(2)} = \frac{1}{2} [(\delta \tilde{\rho})^T \tilde{M}^\rho \delta \tilde{\rho} + 2(\delta \tilde{\lambda}^i)^T \tilde{M}^{\rho^i} \delta \tilde{\rho} + (\delta \tilde{\lambda}^{i'})^T \tilde{M}^{\lambda^i} \delta \tilde{\rho}]. $$

(151)

Here, we used the symmetry

$$ \tilde{M}^{\rho^i} = [\tilde{M}^{\rho^i}]^T. $$

(152)

In the effective energy functional (128), the parameters $\tilde{\lambda}^i$ are determined by the minimization condition (126). Applied to our second-order expansion (151) this condition yields

$$ \frac{\partial}{\partial \delta \lambda^i_Z} \delta E^{(2)}(\delta \tilde{\lambda}^i, \delta \tilde{\rho}) = 0, $$

(153)
which gives us the multiplet amplitudes
\[ \delta \tilde{\lambda}_i = -[\tilde{M}^{\lambda\lambda}]^{-1}\tilde{M}^{\lambda\rho}\delta \tilde{\rho} \]  \hspace{1cm} (154)

as a linear function of the densities \( \delta \tilde{\rho} \). This result leads to the quadratic expansion
\[ E^{\text{eff}}(\tilde{\rho}^0 + \delta \tilde{\rho}) = E_0 + \text{tr}(\tilde{h}^0 \delta \tilde{\rho}) + \frac{1}{2}(\delta \tilde{\rho})^T \tilde{K} \delta \tilde{\rho}, \]  \hspace{1cm} (155)
\[ \tilde{K} = \tilde{M}^{\rho\rho} - \tilde{M}^{\rho\lambda}[\tilde{M}^{\lambda\lambda}]^{-1}\tilde{M}^{\lambda\rho}, \]  \hspace{1cm} (156)

of the effective energy as a function of the density fluctuations \( \delta \tilde{\rho} \). In earlier work on the time-dependent Gutzwiller theory, equations (153) and (154) have been denoted as the ‘antiadiabaticity assumption’. In fact, these equations have the physical meaning that the local multiplet dynamics, described by fluctuations \( \delta \tilde{\lambda}_z(t) \), are fast compared to those of the density-matrix fluctuations \( \delta \tilde{\rho}_Y(t) \). We will use the phrase ‘antiadiabaticity assumption’ in this work too, although, strictly speaking, in our derivation it does not constitute an additional approximation.

With the functional (155), we could now proceed with our evaluation of the Gutzwiller RPA equations (144). For practical applications, however, it is more convenient to determine the ‘interaction kernel’ (156) in a way that avoids the explicit solution of the constraint equations (122). This alternative procedure is the subject of the following section.

5.4. Lagrange-functional expansion

In the second-order expansion, described in section 5.3, we implemented the constraints (122) by explicitly eliminating a certain set of \( n_c \) variational parameters. Although such a procedure can, at least in principle, always be applied, for the numerical implementation it is more convenient to impose the constraints by means of Lagrange parameters. To this end, we define the ‘Lagrange functional’
\[ L(\tilde{\lambda}, \tilde{\rho}, \tilde{\Lambda}) = E(\tilde{\lambda}, \tilde{\rho}) + \sum_{n=1}^{n_c} \Lambda_n g_n(\tilde{\lambda}, \tilde{\rho}), \]  \hspace{1cm} (157)

that depends on all variational parameters \( \tilde{\lambda} \), the density matrix \( \tilde{\rho}(\tilde{\Lambda}) \) and the \( n_c \) Lagrange parameters \( \Lambda_n \). The optimum variational parameters \( \tilde{\lambda}_0 \), density-matrix elements \( \tilde{\rho}_0 \) and Lagrange parameters \( \Lambda_0 \) are then determined by the equations
\[ \frac{\partial L}{\partial \lambda_z} \bigg|_{\tilde{\lambda} = \tilde{\lambda}_0, \tilde{\rho} = \tilde{\rho}_0, \tilde{\Lambda} = \Lambda_0} = \frac{\partial L}{\partial \Lambda_n} = \frac{\partial L}{\partial \rho_Y} = 0, \]  \hspace{1cm} (158)

which have to be solved simultaneously.

We expand the Lagrange functional to leading order with respect to parameter \( (\delta \lambda, \delta \Lambda_n) \) and density fluctuations \( (\delta \rho_Y) \). The second-order contribution has the form
\[ \delta L^{(2)} = \frac{1}{2} \sum_{Y,Y'} \delta \rho_Y L_{Y,Y'}^{\rho\rho} \delta \rho_{Y'} + \sum_{Z,Y} \delta \lambda_Z L_{Z,Y}^{\rho\rho} \delta \rho_Y + \frac{1}{2} \sum_{Z,Z'} \delta \lambda_Z L_{Z,Z'}^{\lambda\lambda} \delta \lambda_{Z'} \]
\[ + \sum_n \delta \Lambda_n \left\{ \sum_Z \frac{\partial g_n}{\partial \lambda_z} \delta \lambda_z + \sum_Y \frac{\partial g_n}{\partial \rho_Y} \delta \rho_Y \right\}, \]  \hspace{1cm} (159)

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with matrices \( \tilde{L}^{\rho\rho}, \tilde{L}^{\lambda\rho}, \tilde{L}^{\lambda\lambda} \) defined as in equations (148)–(150) only with \( E^{GA} \) replaced by \( L \).

The antiadiabaticity conditions
\[
\frac{\partial}{\partial \delta \lambda Z} \delta L^{(2)} = 0 \tag{160}
\]
\[
\frac{\partial}{\partial \delta \Lambda_n} \delta L^{(2)} = 0 \tag{161}
\]
yield the \( n_c \) equations
\[
\sum_{Z} \frac{\partial g_n}{\partial \lambda Z} \delta \lambda Z + \sum_{Y} \frac{\partial g_n}{\partial \rho Y} \delta \rho Y = 0, \tag{162}
\]
and the \( n_p \) equations
\[
\sum_{Z} \tilde{L}^{\lambda\lambda}_{Z,Z'} \delta \lambda Z' + \sum_{Y} \tilde{L}^{\rho\rho}_{Z,Y} \delta \rho Y + \sum_{n} \frac{\partial g_n}{\partial \lambda Z} \delta \Lambda_n = 0. \tag{163}
\]
Together, these equations allow us to express the \( n_p + n_c \) parameter fluctuations \( \delta \Lambda_n, \delta \lambda Z \) in terms of the density fluctuations \( \delta \rho Y \). These can be reinserted into (159) to obtain the desired quadratic functional solely of the density fluctuations,
\[
\delta L^{(2)} = \frac{1}{2} \sum_{Y,Y'} \delta \rho Y \bar{K}_{Y,Y'} \delta \rho Y'. \tag{164}
\]
In appendix B.1, we prove that the interaction matrix \( \bar{K}_{Y,Y'} \) in (164) is, in fact, identical to \( K_{Y,Y'} \) in equations (155)–(156).

6. Two-particle response functions for lattice models

In the previous section, we have developed the general formalism of the time-dependent Gutzwiller theory for the calculation of two-particle Green’s functions. We will be more specific in this section and explain in detail how the response functions that are of interest in solid-state physics can be calculated within our approach.

6.1. Two-particle response functions

In solid-state physics, one is usually not interested in the full two-particle Greens function \( \tilde{G} \) as it has been defined in (82). The properties relevant for experiments are certain linear combinations of elements of \( \tilde{G} \). For our translationally invariant model Hamiltonians (1), these are in particular the two-particle response functions
\[
G_{(\sigma_2,\sigma_1), (\sigma_3,\sigma_4)}(\vec{R}_i - \vec{R}_j, t - t') \equiv \langle \langle \hat{c}^\dagger_{i,\sigma_1}(t) \hat{c}_{i,\sigma_2}(t); \hat{c}^\dagger_{j,\sigma_3}(t'); \hat{c}_{j,\sigma_4}(t') \rangle \rangle \tag{165}
\]
or, more importantly, their Fourier transforms
\[
G_{(\sigma_2,\sigma_1), (\sigma_3,\sigma_4)}(\vec{q}, \omega) = \frac{1}{L^3} \int_{-\infty}^{\infty} dt \ e^{i\omega t} \sum_{i,j} e^{i(\vec{R}_i - \vec{R}_j)} \tilde{q} G_{(\sigma_2,\sigma_1), (\sigma_3,\sigma_4)}(\vec{R}_i - \vec{R}_j, \tau)
\]
\[
= \frac{1}{L^3} \sum_{k,k'} \langle \langle \hat{c}^\dagger_{k,\sigma_1} \hat{c}_{k+q,\sigma_2}; \hat{c}^\dagger_{k',\sigma_3} \hat{c}_{k'+q,\sigma_4} \rangle \rangle \omega. \tag{166}
\]
Here, we introduced the fermionic operators
\[ \hat{c}^{(\dagger)}_{k,\sigma} = \frac{1}{\sqrt{L_s}} \sum_{i} e^{\pm i \mathbf{k}_i \cdot \mathbf{R}_i} \hat{c}^{(\dagger)}_{i,\sigma} \] (167)
and the usual notation
\[ \langle \langle \hat{O}; \hat{O}' \rangle \rangle_{\omega} = \int_{-\infty}^{\infty} d\tau \langle \langle \hat{O}(\tau); \hat{O}'(0) \rangle \rangle \text{e}^{i\omega \tau} \] (168)
for the Fourier transform of a Green’s function with arbitrary operators \( \hat{O}, \hat{O}' \). With the abbreviation \( v = (\sigma, \sigma') \) for spin–orbit indices and the operators
\[ \hat{A}^q_v \equiv \hat{A}^q_{\sigma_2,\sigma_1} \equiv \frac{1}{\sqrt{L_s}} \sum_{k} \hat{c}^{(\dagger)}_{k,\sigma_1} \hat{c}^q_{k,\sigma_2}, \] (169)
we can write (166) as
\[ G_{v,v'}(\mathbf{q}, \omega) = \sum_{v,v'} \kappa_v G_{v,v'}(\mathbf{q}, \omega) \kappa_{v'}. \] (170)
The Green’s functions (166) are still quite general since they include all possible channels of local coupling \( \sigma_1 \leftrightarrow \sigma_2, \sigma_3 \leftrightarrow \sigma_4 \). In experiments, one usually measures response functions that are certain linear combinations,
\[ \chi(\mathbf{q}, \omega) = \frac{1}{L_s} \langle \langle \hat{S}^q_+; \hat{S}^q_- \rangle \rangle_{\omega}, \] (172)
where
\[ \hat{S}^q_+ = \sum_{i} e^{-i \mathbf{q} \cdot \mathbf{R}_i} \hat{S}^+_i = \sum_{k} \sum_{b} \hat{c}^{(\dagger)}_{k, (b\uparrow)} \hat{c}_{k+q, (b\downarrow)}^q, \] (173)
\[ \hat{S}^q_- = \sum_{i} e^{i \mathbf{q} \cdot \mathbf{R}_i} \hat{S}^-_i = \sum_{k} \sum_{b} \hat{c}^{(\dagger)}_{k+q, (b\downarrow)} \hat{c}_{k, (b\uparrow)}^q \equiv \left( \hat{S}^q_+ \right)^{\dagger}, \] (174)
\[ \hat{S}^+_i = \sum_{b} \hat{c}^{(\dagger)}_{i, (b\uparrow)} \hat{c}_{i, (b\downarrow)}, \hat{S}^-_i = \sum_{b} \hat{c}^{(\dagger)}_{i, (b\downarrow)} \hat{c}_{i, (b\uparrow)} \] (175)
are the usual spin-flip operators and \( b \) is an index for the orbitals at each lattice site \( i \). The spin susceptibility of a two-band Hubbard model will be investigated in section 7.

### 6.2. Response functions in the time-dependent Gutzwiller approximation

In order to apply the time-dependent Gutzwiller approximation, as developed in section 5, we have to expand the Lagrange functional (157) up to second order with respect to density matrix \( (\delta \tilde{\rho}) \) and variational-parameter fluctuations \( (\delta \lambda_{\Gamma, \Gamma'}) \). This means that we need an expansion of the constraints (35)–(36), of the local energies (9)–(11) and (24), and of the kinetic energy (31)–(32). The second-order expansion of the kinetic energy is more involved than that of the local energies and of the constraints. In the latter there are only contributions from
fluctuations at the same lattice sites while in the kinetic energy local and non-local fluctuations (such as \( \delta(\hat{c}_{j,\sigma}^\dagger \hat{c}_{j,\sigma})_\psi \)) couple. Nevertheless, the calculation of the second-order Lagrange functional is tedious but otherwise straightforward. We therefore refer to appendix \( C \) where details of this derivation are presented. As shown in that appendix, it is useful to introduce the operators

\[
\hat{g}^q_{\omega} \equiv \hat{g}^q_{\alpha_1,\sigma_1,\sigma'_1,\alpha'_2} = \frac{1}{\sqrt{L_k}} \sum_k \epsilon^{\sigma_2,\sigma_1}_{k,\sigma_1} \hat{c}_{k,\sigma_1}^\dagger \hat{c}_{k+q,\sigma'_1},
\]

(176)

\[
\hat{g}^q_{\omega} \equiv \hat{g}^q_{\alpha_1,\sigma_1,\sigma'_1,\alpha'_2} = \frac{1}{\sqrt{L_k}} \sum_k \epsilon^{\sigma_2,\sigma_1}_{k,\sigma_1} \hat{c}_{k,\sigma_1}^\dagger \hat{c}_{k+q,\sigma'_1},
\]

(177)

and to define the auxiliary Green’s function matrix \( \tilde{\Pi}(\tilde{q}, \omega) \) with the elements

\[
\Pi_{\omega, \omega'}(\tilde{q}, \omega) \equiv \begin{pmatrix}
\langle \langle \hat{A}_q^\dagger ; \hat{A}_q^\dagger \rangle \rangle_{\omega} & \langle \langle \hat{A}_q^\dagger ; \hat{B}_q^\dagger \rangle \rangle_{\omega} & \langle \langle \hat{B}_q^\dagger ; \hat{B}_q^\dagger \rangle \rangle_{\omega} \\
\langle \langle \hat{B}_q^\dagger ; \hat{A}_q^\dagger \rangle \rangle_{\omega} & \langle \langle \hat{B}_q^\dagger ; \hat{B}_q^\dagger \rangle \rangle_{\omega} & \langle \langle \hat{B}_q^\dagger ; \hat{B}_q^\dagger \rangle \rangle_{\omega} \\
\langle \langle \hat{B}_q^\dagger ; \hat{B}_q^\dagger \rangle \rangle_{\omega} & \langle \langle \hat{B}_q^\dagger ; \hat{B}_q^\dagger \rangle \rangle_{\omega} & \langle \langle \hat{B}_q^\dagger ; \hat{B}_q^\dagger \rangle \rangle_{\omega}
\end{pmatrix}
\]

(178)

We are actually interested only in the first ‘element’ of this matrix, i.e. the Green’s functions (170), since they allow us to determine any response function of the form (171). As shown in appendix \( D \), however, the time-dependent Gutzwiller approximation leads to the following equation for the entire matrix (178) from which (170) can be extracted,

\[
\tilde{\Pi}(\tilde{q}, \omega) = (1 + \tilde{\Pi}_0(\tilde{q}, \omega) \tilde{V}^q)^{-1} \tilde{\Pi}_0(\tilde{q}, \omega).
\]

(179)

Here, \( \tilde{V}^q \) is the effective second-order interaction matrix, introduced in (C.40), and \( \tilde{\Pi}_0(\tilde{q}, \omega) \) is the Green’s function matrix (178) evaluated for the single-particle Hamiltonian (132). As shown in [10, 46], this Gutzwiller Hamiltonian \( \tilde{H}_0 \equiv \tilde{H}_0^{\text{eff}} \) for our lattice Hamiltonian (1) has the form

\[
\tilde{H}_0^{\text{eff}} = \sum_k \sum_{\sigma_1,\sigma_2} (\tilde{e}^{\sigma_1,\sigma_2}_k + \eta_{\sigma_1,\sigma_2}) \hat{c}_{k,\sigma_1}^\dagger \hat{c}_{k,\sigma_2} = \sum_k \sum_{\alpha} E_{k,\alpha} \hat{h}_{k,\alpha}^\dagger \hat{h}_{k,\alpha},
\]

(180)

where the Lagrange parameters \( \eta_{\sigma_1,\sigma_2} \) are determined by the minimization of the variational ground-state energy and \( \tilde{e}^{\sigma_1,\sigma_2}_k \) is defined as

\[
\tilde{e}^{\sigma_1,\sigma_2}_k = \sum_{\sigma_1',\sigma_2'} q^{\sigma_1,\sigma_2}_{\sigma_1',\sigma_2'} \epsilon^{\sigma_1',\sigma_2'}_k.
\]

(181)

The creation and annihilation operators \( \hat{h}_{k,\alpha}^{(\pm)} \) of the effective single-particle Hamiltonian (180) can be written as

\[
\hat{h}_{k,\alpha}^+ = \sum_{\sigma} u_{\sigma,\alpha} \hat{c}_{k,\sigma}^+, \quad \hat{h}_{k,\alpha}^- = \sum_{\sigma} (u_{\sigma,\alpha})^* \hat{c}_{k,\sigma}^+.
\]

(182)

(183)

where the coefficients \( u_{\sigma,\alpha} \) are determined by a diagonalization of (180). With these eigenstates, the calculation of \( \tilde{\Pi}_0(\tilde{q}, \omega) \) is now a simple task. For example, the first element \( \langle \langle \hat{A}_q^\dagger ; \hat{A}_q^\dagger \rangle \rangle_{\omega}^0 \)
is given as
\[
\langle \langle \hat{A}_{\sigma_1,\sigma_2}^q \rangle \rangle_\omega = \frac{1}{L_s} \sum_{k, q} \sum_{\alpha_1, \alpha_2} \langle \langle \hat{h}_{k,\alpha_1} \hat{h}_{k+q,\alpha_2} \hat{h}_{k,\alpha_1} \rangle \rangle_\omega (u_{\sigma_1,\sigma_2}^k)^* u_{\sigma_1,\sigma_2}^{k+q} u_{\sigma_1,\sigma_2}^k \hat{A}_{\sigma_1,\sigma_2}^q (u_{\sigma_1,\sigma_2}^k)^* u_{\sigma_1,\sigma_2}^k - u_{\sigma_1,\sigma_2}^{k+q} u_{\sigma_1,\sigma_2}^k),
\]
where
\[
n_{k,\alpha}^0 = \Theta(E_F - E_{k,\alpha})
\]
is the ground-state distribution function (143). In the same way, we can calculate all the other elements of \( \hat{P}^0(\alpha, \omega) \). The result is always the same as in (184) only with additional factors \( \sim \epsilon_{k,\sigma'} \) or \( \sim \epsilon_{k+q,\sigma'} \) due to the definition of the operators (176)–(177). For example, the second element in (178) leads to
\[
\langle \langle \hat{A}_{\sigma_1,\sigma_2}^q \rangle \rangle_\omega = \frac{1}{L_s} \sum_{k, q} \sum_{\alpha_1, \alpha_2} \langle \langle \hat{h}_{k,\alpha_1} \hat{h}_{k+q,\alpha_2} \hat{h}_{k,\alpha_1} \rangle \rangle_\omega (u_{\sigma_1,\sigma_2}^k)^* u_{\sigma_1,\sigma_2}^{k+q} u_{\sigma_1,\sigma_2}^k (n_{k,\alpha_1}^0 - n_{k+q,\alpha_1}^0).
\]
(186)

To summarize, with equations (179), (184), (186) and the interaction matrix (C.40), we are now in a position to investigate any two-particle response function for our general class of multi-band models (1). As a first example, we study the magnetic susceptibility for a two-band model in the following section.

7. Magnetic susceptibility of a two-band Hubbard model

In this section, we investigate the magnetic susceptibility of a two-band Hubbard model in three spatial dimensions. The model Hamiltonian and the Gutzwiller wave functions that we use for its investigation are introduced in section 7.1. In section 7.2, we discuss the Green’s function matrices that we need to study in order to calculate the magnetic susceptibilities within the RPA and the Gutzwiller-RPA schemes. The numerical results for the two-band model are presented in section 7.3.

7.1. Model and variational ground state

We investigate a Hubbard model with two degenerate \( e_g \) orbitals per site on a cubic lattice. The local Hamiltonian (2) for this system can be written as
\[
\hat{H}_{1b} = U \sum_e n_{e,\uparrow} n_{e,\downarrow} + U' \sum_{s, s'} n_{1,s} n_{2,s'} - J \sum_s \hat{n}_{1,s} \hat{n}_{2,s} + J \sum_s \hat{c}_{1,s}^\dagger \hat{c}_{2,s} + J \sum_s \hat{c}_{2,s}^\dagger \hat{c}_{1,s}.
\]
(187)

Here, \( e = 1, 2 \) labels the \( e_g \) orbitals, \( s = \uparrow, \downarrow \) is the spin index and we use the convention \( \hat{\uparrow} \equiv \downarrow, \downarrow \equiv \uparrow \). Due to the cubic symmetry, the Coulomb parameters \( U, U' \) and the exchange parameter \( J \) are related to each other through
\[
U' = U - 2J.
\]
(188)
Table 1. Two-particle eigenstates with symmetry specifications and energies.

| No. | Atomic eigenstate | Symmetry | Energy $E_{\Gamma}$ |
|-----|------------------|----------|-------------------|
| 1   | $|\uparrow, \uparrow\rangle$ | $^3A_2$ | $U' - J$ |
| 2   | $(|\uparrow, \downarrow\rangle + |\downarrow, \uparrow\rangle)/\sqrt{2}$ | $^3A_2$ | $U' - J$ |
| 3   | $|\downarrow, \downarrow\rangle$ | $^3A_2$ | $U' - J$ |
| 4   | $(|\uparrow, \downarrow\rangle - |\downarrow, \uparrow\rangle)/\sqrt{2}$ | $^{1}E$ | $U' + J$ |
| 5   | $(|\uparrow, 0\rangle - |0, \uparrow\rangle)/\sqrt{2}$ | $^{1}E$ | $U - J$ |
| 6   | $(|\uparrow, 0\rangle + |0, \uparrow\rangle)/\sqrt{2}$ | $^{1}A_1$ | $U + J$ |

Hence, only two of these three parameters can be chosen independently.

There are four spin–orbital states $\sigma = (e, s)$ per atom, leading to a $2^4 = 16$-dimensional atomic Hilbert space. All eigenstates $|\Gamma\rangle$ of $\hat{H}^{2b}$ with particle numbers $N \neq 2$ are simple Slater determinants of spin–orbital states $|\sigma\rangle$, and their energies are

$$E_{\Gamma} = 0 \quad (N = 0, 1),$$
$$E_{\Gamma} = U + 2U' - J \quad (N = 3),$$
$$E_{\Gamma} = 2U + 4U' - 2J \quad (N = 4).$$

(189)

The two-particle eigenstates are slightly more complicated because some of them are linear combinations of Slater determinants. We introduce the basis

$$|s, s\prime\rangle \equiv \hat{c}_{1,s}^\dagger \hat{c}_{2,s\prime}^\dagger |0\rangle,$$

(190)
$$|\uparrow \downarrow, 0\rangle \equiv \hat{c}_{1,\uparrow}^\dagger \hat{c}_{1,\downarrow}^\dagger |0\rangle,$$

(191)
$$|0, \uparrow \downarrow\rangle \equiv \hat{c}_{2,\downarrow}^\dagger \hat{c}_{2,\uparrow}^\dagger |0\rangle,$$

(192)
of two-particle states, which are used to set up the eigenstates of $\hat{H}_{\text{loc}}$, see table 1. The states of lowest energy are the three triplet states with spin $S = 1$, which belong to the representation $A_2$ of the cubic point-symmetry group. Finding a high-spin ground state is a simple consequence of Hund’s first rule. The two degenerate singlet states of symmetry $E$ and the non-degenerate singlet state of symmetry $A_1$ are higher in energy.

For the variational ground state, we can work with a wave function (4) that contains only diagonal parameters $\lambda_{\Gamma, \Gamma}$. Non-diagonal parameters could only arise if we break the cubic symmetry or want to study states with magnetic orders not collinear to the chosen spin-quantization axis. Note, however, that for the study of spin excitations we must allow for non-diagonal variational parameters; see below.

In our numerical analysis of this two-band model we will consider a tight-binding Hamiltonian $\hat{H}_0$ with generic hopping parameters that were already used in previous works and led to the density of states (DOS) at the Fermi energy shown in figure 1 (left). Due to the maximum in the DOS at approx. $n_\sigma = 0.29$, in that range of band fillings there is the strongest tendency for a ferromagnetic state to be lower in energy than the paramagnet. This has already been demonstrated in [7]. Another important finding in that work is the huge importance of the exchange interaction $J$ for the appearance of ferromagnetic order. This can be seen from the Gutzwiller phase diagram for our model in figure 1 (right). In contrast, the HF phase diagrams
Figure 1. Left: model DOS at the Fermi energy as a function of orbital filling \( n_{\sigma} \). Right: ground-state phase diagram for both HF and GW. The lines mark the instability for a transition from the paramagnetic (PM) to the ferromagnetic (FM) state. The orbital filling is \( n_{\sigma} \approx 0.29 \) and \( t \equiv |t_{dd\sigma}| \) (cf [56]).

show almost no dependence on the size of \( J \); see also [44] where similar results have been reported for a two-band model in infinite dimensions.

7.2. Magnetic susceptibility

For calculation of the spin susceptibility (172), we need to determine a Green’s function matrix of the form (178) in which the operators \( \hat{A}_q \), \( \hat{B}_q \) and \( \hat{\bar{B}}_q \) are given as

\[
\hat{A}_q^{b_1,b_2} = \frac{1}{\sqrt{L_s}} \sum_k \hat{c}_{k,(b_2\uparrow)}^\dagger \hat{c}_{k+q,(b_1\downarrow)}^\dagger,
\]

\[
\hat{B}_q^{b_1,b_2,b_1',b_2'} = \frac{1}{\sqrt{L_s}} \sum_k \epsilon_{k,b_1} \hat{c}_{k,(b_2\uparrow)}^\dagger \hat{c}_{k+q,(b_1\downarrow)}^\dagger,
\]

\[
\hat{\bar{B}}_q^{b_1,b_2,b_1',b_2'} = \frac{1}{\sqrt{L_s}} \sum_k \epsilon_{k+q,b_2} \hat{c}_{k,(b_2\uparrow)}^\dagger \hat{c}_{k+q,(b_1\downarrow)}^\dagger.
\]

The matrix (178) that results from these operators is \( 4 + 16 + 16 = 36 \) dimensional. Due to symmetries, this dimension can be reduced to 20 for a general wave vector \( \vec{q} \). Along symmetry lines the symmetry reduction could even go further. In our numerical calculations, however, we did not exploit such symmetry considerations since the numerical efforts for a two-band model are still moderate, even in three dimensions.

Note that there is a difference between HF and Gutzwiller-RPA calculations concerning the elements of \( \tilde{\Pi}^0(\vec{q}, \omega) \) that have to be taken into account in our calculation of the susceptibility

\[
\chi(\vec{q}, \omega) = \sum_{b,b'} \langle \langle \hat{A}_{q,b,b'}^\dagger (\hat{A}_{q,b',b'}^\dagger) \rangle \rangle_\omega.
\]

In HF-RPA, due to the locality of the interaction terms in Hubbard models and the symmetries of \( e_g \) orbitals, the only elements of \( \tilde{\Pi}^0(\vec{q}, \omega) \) that contribute are \( \langle \langle \hat{A}_{q,b,b'}^\dagger (\hat{A}_{q,b',b'}^\dagger) \rangle \rangle_\omega \), i.e. those
that are diagonal with respect to the local orbital indices. This is different from the Gutzwiller-RPA equations in which all Green’s functions defined by the operators \((193) – (195)\) have to be taken into account. In particular, Green’s functions \(\langle \hat{A}_{q_1}^{b_1} \hat{A}_{q_2}^{b_2} \rangle_0 \) with \(b_1 \neq b_2\) or \(b_3 \neq b_4\) cannot be discarded. The reason for this difference is the non-locality of the interaction matrix \(\tilde{V}\) in the time-dependent Gutzwiller theory.

7.3. Results

We prepare a ferromagnetic ground state in both HF and Gutzwiller approximations at band filling \(n_\sigma \approx 0.2987\) in order to be close to the maximum of the DOS. In general, the two schemes will give different magnetizations for the same set of interaction parameters. Therefore, one could either perform the comparison for fixed parameters or fixed magnetization (cf also \([57]\)). To avoid this inconsistency, we present results for interaction parameters that lead to a fully polarized ferromagnetic ground state in both approximations, i.e. \(m = 2n_\sigma\). Note that for numerical reasons we have to stay slightly below this value in the case of the Gutzwiller approximation. The corresponding interaction parameters are specified in the captions to figures 2 and 3, which display the magnetic excitations obtained within both approximations.

These spectra are composed of a low-energy magnon part due to the breaking of spin-rotational invariance and a high-energy Stoner continuum, that reflects the particle–hole spin-flip excitations of the ‘bare’ system, i.e. \(\Pi^0(q^2, \omega)\), cf equation \((179)\). For both methods we show the excitations along the \((100)\) and \((111)\) directions. The difference in these directions mainly arises due to the orientation dependence of the particle–hole dispersion, which is significantly stronger along the diagonals.

**Figure 2.** HF excitations for \(U = 10.0t, J/U = 0.30\) resulting in \(m = 0.5975\) (fully polarized) and \(\lambda^- = E^+_{\uparrow} - E^+_{\downarrow} \approx 7.77t\). The magnon dispersion is fitted by \(\varepsilon_{\text{magnon}}(\vec{q})/t = D|\vec{q}|^2(1 + \beta|\vec{q}|^2)\) with \(D^{100} \approx D^{111} = 100 \times 10^{-3}\). Scaling: \(t \equiv |t_{d\sigma}^{(1)}|\) (cf \([56]\)) and \(q_{x,y,z} \in (-\pi, \pi)\).
One first important difference between the HF and Gutzwiller approximations concerns the difference in the magnetic band splitting $\lambda^- = E_F^\uparrow - E_F^\downarrow \approx 2.128t$. In the HF theory, this value is just given by $\lambda^-(\text{HF}) = (U + J)m$, and thus for strongly correlated systems produces a large gap $O(U)$ between the low-energy magnon and a high-energy Stoner continuum. On the other hand, we find that $\lambda^-(\text{GA})$ is significantly reduced with respect to its HF counterpart. For the present system, $\lambda^-(\text{GA}) \approx \frac{1}{2} \lambda^-(\text{HF})$. Given the broadening of the Stoner continuum with increasing transferred momentum, the low-energy magnon thus rapidly merges with the continuum in the time-dependent GA, as can be seen from figure 3. As a consequence, the excitation at $\omega \sim J$, corresponding to a respective spin-flip in the two orbitals, is only visible in HF-RPA along the $(100)$ direction, whereas in the time-dependent Gutzwiller approach it is already within the continuum. Note that the overestimation of the Stoner excitation energy within HF-RPA is a longstanding problem in solid-state theory, as discussed in [58].

At $\vec{q} = 0$ all the weight is contained in the zero-frequency Goldstone mode. The existence of this excitation provides an important consistency check of the Gutzwiller-RPA approach similar to the analogous finding in HF-RPA. The positive dispersion of the magnon further demonstrates that the underlying Gutzwiller solution is a stable energy minimum, that is not destroyed by the fluctuations. The spin-wave stiffness, i.e. the quadratic coefficient of the magnon dispersion, is significantly larger in HF-RPA than in time-dependent Gutzwiller theory. Note, however, that to a certain extent this huge difference is caused by an instability of the ferromagnetic ground state with respect to an incommensurate phase that is found for interaction parameters not much smaller than those used in figure 3.
8. Summary

In this paper, we have given a detailed derivation of the time-dependent Gutzwiller approximation for multi-band Hubbard models. The basic assumptions that underlie the method can be summarized as follows. Firstly, it is assumed that the dynamics of the Slater-determinant (upon which the Gutzwiller projector acts in the starting ansatz) is determined by the so-called Gutzwiller Hamiltonian, equation (140), which leads to an equation of motion similar to standard RPA, equation (138). Secondly, the dynamics of the variational parameters is determined from the assumption that at each instant of time the energy is minimized. This leads to a linear relation between variational parameter and density fluctuations, equation (154). Thirdly, as in the standard HF-RPA approach, it is assumed that the external perturbation and thus the density fluctuations are small, equations (106), (107). We have seen that also in the multi-band case these assumptions lead to a consistent theory in the sense that an instability that is signalled within the Gutzwiller-RPA approach corresponds to a (second-order) phase transition, that one would obtain from the bare variational Gutzwiller approximation. We have further demonstrated that for ferromagnetic ground states the Gutzwiller-RPA approach leads to the appearance of the Goldstone mode, as expected for systems that break continuous spin symmetry.

The formalism as developed in its present form can now be straightforwardly applied to the investigation of correlation functions in strongly correlated multi-band systems such as, e.g., pnictides, manganites, cobaltates, etc. On the other hand, a natural application of the theory would also comprise the investigation of, e.g., orbital quenches for which the small-amplitude assumption for the density matrices has to be abandoned. For single-band Hubbard models, such a fully time-dependent formulation of the Gutzwiller approximation has been recently presented by Schiró and Fabrizio [59, 60] where also the second assumption above has been replaced by separate equations of motion for the variational parameters. Future work should thus address the question of whether their approach reduces to the present theory in the small-amplitude limit and how it eventually can be extended to the multi-band case.

Appendix A. Second-order expansion of determinants

According to equation (16), section 3.1, expectation values $m^0_{I,I'}$, can be written as determinants of certain matrices $A$ with elements that are linear functions of the local density matrix $C^0_{\gamma,\gamma'}$. In the variational ground state, $C^0_{\gamma,\gamma'}$ is diagonal, and if we choose a proper order of the orbitals $\gamma$ to 0th order the matrix $A$ is diagonal too,

$$m^0_{I,I'} = |A| = |A^0| = \begin{vmatrix} A^0_{1,1} & 0 & \ldots & 0 \\ 0 & A^0_{2,2} & \ldots & 0 \\ 0 & \ldots & \ldots & \ldots \\ 0 & 0 & 0 & A^0_{N,N} \end{vmatrix}.$$ \hspace{1cm} (A.1)

For $I \neq I'$, at least one of the diagonal elements $A^0_{i,i}$ vanishes and we find

$$m^0_{I,I'} = \delta_{I,I'} \prod_i A_{i,i} = \delta_{I,I'} \prod_{\gamma} n^0_{\gamma} \prod_{\gamma \in \{1\ldots N\} \setminus I} (1 - n^0_{\gamma}),$$ \hspace{1cm} (A.2)
as expected. In order to calculate the first and second derivatives of \( m^0_{i,j} \), we need to expand the determinant

\[
|A| = |A^0 + \delta a|
\]

up to second order with respect to the matrix elements \( \delta a_{i,j} \). For this expansion one readily finds

\[
|A| - |A^0| = |A^0| \sum_i \frac{\delta a_{i,j}}{A^0_{i,i}} + |A^0| \sum_{i,j} \frac{\delta a_{i,i} \delta a_{j,j} + \delta a_{i,j} \delta a_{j,i}}{A^0_{i,i} A^0_{j,j}}.
\]

Note that for \( A^0_{i,i} = 0 \) the rhs is defined by the corresponding limit \( A^0_{i,i} \to 0 \).

### Appendix B. Invariance of second-order expansions

#### B.1. Equivalence of the Lagrange-functional expansion

In this section, we show that the interaction kernel \( \bar{K}^{\rho,\rho}_{Y,Y} \) in (164) is identical to \( K^{\rho,\rho}_{Y,Y} \) in equations (155)–(156). To this end, we choose again some arbitrary independent and dependent variational parameters \( \lambda^Z_i \) and \( \rho^Y_i \), cf equation (123). By construction, the constraints (122) are automatically fulfilled as a function of \( \lambda^Z_i \) and \( \rho^Y_i \), i.e. we have

\[
g_n(\tilde{\lambda}^Z_i(\lambda^Z_i, \rho^Y_i), \tilde{\rho}^Y_i, \rho^Y_i) = 0.
\]

Consequently, all first- or higher-order derivatives of (B.1) with respect to \( \lambda^Z_i \) and \( \rho^Y_i \) vanish. For example, the first-order derivatives lead to

\[
\begin{align*}
\frac{dg_n}{d\lambda^Z_i} & = \frac{\partial g_n}{\partial \lambda^Z_i} + \sum_X \frac{\partial g_n}{\partial \lambda^X_i} \frac{\partial \lambda^Z_i}{\partial \lambda^X_i} = 0, \\
\frac{dg_n}{d\rho^Y_i} & = \frac{\partial g_n}{\partial \rho^Y_i} + \sum_X \frac{\partial g_n}{\partial \lambda^X_i} \frac{\partial \lambda^Z_i}{\partial \rho^Y_i} = 0.
\end{align*}
\]

Using the matrices

\[
G_{n,X} \equiv \frac{\partial g_n}{\partial \lambda^X_i}, \quad R_{X,Z} \equiv \frac{\partial \lambda^Z_i}{\partial \lambda^X_i}, \quad Q_{X,Y} \equiv \frac{\partial \lambda^Y_i}{\partial \rho^Y_i},
\]

we can write equations (B.2)–(B.3) as

\[
\begin{align*}
\frac{\partial g_n}{\partial \lambda^Z_i} & = -[\bar{G} \bar{R}]_{n,Z}, \\
\frac{\partial g_n}{\partial \rho^Y_i} & = -[\bar{G} \bar{Q}]_{n,Y}.
\end{align*}
\]

With the classification of dependent and independent variables, we are in a position to evaluate the antiadiabaticity conditions (162)–(163). First, equation (162) leads to

\[
\sum_X \frac{\partial g_n}{\partial \lambda^X_i} \delta \lambda^X_i + \sum_Z \frac{\partial g_n}{\partial \lambda^Z_i} \delta \lambda^Z_i + \sum_Y \frac{\partial g_n}{\partial \rho^Y_i} \delta \rho^Y_i = 0.
\]
which together with equations (B.5)–(B.6) yields
\[
\bar{G}[\delta \vec{\lambda}^d - \vec{R} \delta \vec{\lambda}^i - \vec{Q} \delta \bar{\rho}] = \vec{0}.
\] (B.8)

Since the square matrix \( \bar{G} \) should be invertible, the bracket in (B.8) must vanish. Hence, we find the relation
\[
\delta \vec{\lambda}^d = \vec{R} \delta \vec{\lambda}^i + \vec{Q} \delta \bar{\rho},
\] (B.9)

which determines the dependent-parameter fluctuations \( \delta \vec{\lambda}^d \) as a function of \( \delta \vec{\lambda}^i \) and \( \delta \bar{\rho} \).

Applying the separation of dependent- and independent-parameter fluctuations to the second set of equations (163) yields
\[
\left( \begin{array}{c} \vec{A}^T \\ \bar{G}^T \end{array} \right) \delta \vec{\Lambda} = - \left( \begin{array}{cc} \vec{L}^i \\ \bar{L}^d \end{array} \right) \left( \begin{array}{c} \delta \vec{\lambda}^i \\ \delta \vec{\lambda}^d \end{array} \right) - \left( \begin{array}{c} \vec{L}^i \bar{Q} \\ \vec{L}^d \bar{Q} \end{array} \right) \delta \bar{\rho}.
\] (B.10)

Here we introduced the six matrices
\[
L^i_{Z,Y} \equiv \frac{\partial^2 L}{\partial \lambda^i_{Z} \partial \lambda^i_{Y}}, \ldots, L^d_{X,Y} \equiv \frac{\partial^2 L}{\partial \lambda^d_{X} \partial \rho_Y}
\] (B.11)
of second derivatives. With (B.9) and the second ‘row’ of equations (B.10), one can write the Lagrange-parameter fluctuations as a function of \( \delta \vec{\lambda}^i \) and \( \delta \bar{\rho} \),
\[
\delta \vec{\Lambda} = -[\bar{G}^T]^{-1}[(\vec{L}^i + \bar{L}^d \vec{R}) \delta \vec{\lambda}^i + (\bar{L}^d \vec{Q} + \vec{L}^i \bar{Q}) \delta \bar{\rho}] \equiv \Lambda_1 \delta \vec{\lambda}^i + \Lambda_2 \delta \bar{\rho}.
\] (B.12)

Inserting this expression into the first row of equations (B.10) and using
\[
\vec{A}^T = -\bar{R}^T \bar{G}^T,
\] (B.13)

we eventually find
\[
\delta \vec{\lambda}^i = -[\vec{L}^i + \bar{L}^d \vec{R} + \vec{R}^T \bar{L}^d \vec{R}]^{-1}[\vec{L}^i \bar{Q} + \bar{L}^d \vec{Q} + \bar{R}^T \bar{L}^d + \bar{R}^T \bar{L}^d \bar{Q}] \delta \bar{\rho}.
\] (B.14)

Equations (B.14), (B.12), (B.9) now enable us to write all fluctuations \( \delta \vec{\lambda}^i \), \( \delta \vec{\lambda}^d \) and \( \delta \vec{\Lambda} \) as functions of the density fluctuations \( \delta \bar{\rho} \). These relations can be inserted into the second-order expansion of the Lagrange functional,
\[
2\delta L^{(2)} = (\delta \bar{\rho})^T \bar{L}^d \delta \bar{\rho} + (\delta \vec{\lambda}^i)^T \vec{L}^i \delta \vec{\lambda}^i + (\delta \vec{\lambda}^d)^T \bar{L}^d \delta \vec{\lambda}^d + [(\delta \bar{\rho})^T \bar{L}^d \delta \vec{\lambda}^d + (\delta \vec{\lambda}^i)^T \vec{L}^i \delta \vec{\lambda}^d] + 2(\delta \vec{\lambda}^i)^T \bar{G} \delta \vec{\lambda}^i - \bar{R} \delta \vec{\lambda}^i - \bar{Q} \delta \bar{\rho},
\] (B.15)
in order to calculate \( \bar{K}^{(2)}_{\phi \phi} \) in equation (164). However, to prove just the identity of \( \bar{K}^{(2)}_{\phi \phi} \) and \( K^{(2)}_{\phi \phi} \) in (155) it is sufficient to apply only equation (B.9) to the expansion (B.15). This leads to
\[
2\delta L^{(2)} = (\delta \bar{\rho})^T (\bar{L}^d + \vec{Q}^T \bar{L}^d) \delta \bar{\rho} + (\delta \vec{\lambda}^i)^T (\vec{L}^i + \bar{L}^d \vec{R} + \bar{R}^T \bar{L}^d) \delta \vec{\lambda}^i + (\delta \vec{\lambda}^d)^T (\vec{L}^d + \bar{L}^i \vec{R} + \bar{R}^T \vec{L}^i) \delta \vec{\lambda}^d + [(\delta \bar{\rho})^T \bar{L}^d \delta \vec{\lambda}^d + (\delta \vec{\lambda}^i)^T \vec{L}^i \delta \vec{\lambda}^d] + 2(\delta \vec{\lambda}^i)^T \bar{G} \delta \vec{\lambda}^i - \bar{R} \delta \vec{\lambda}^i - \bar{Q} \delta \bar{\rho} + \bar{R}^T \bar{L}^d \delta \vec{\lambda}^i + [\bar{L}^i \bar{Q} + \vec{Q}^T \bar{L}^d \bar{Q}] \delta \bar{\rho}.
\] (B.16)

As we will show below, the matrices (148)–(150) that determine the second-order expansion (151) are the same as the corresponding matrices in (B.16). Hence, we have
\[
\delta E^{(2)} = \delta L^{(2)}.
\] (B.17)
Since the antiadiabaticity condition
\[ \frac{\partial \delta E^{(2)}}{\partial \delta \lambda_Z^{(2)}} = \frac{\partial \delta L^{(2)}}{\partial \delta \lambda_Z^{(2)}} = 0 \]
for \( \delta E^{(2)} \) reproduces equation (B.14), the identity of \( \tilde{K}_{Y,Y}^{\rho} \) and \( K_{Y,Y}^{\rho} \) is then finally demonstrated.

It remains to be shown that the matrices (148)–(150) agree with those in (B.16). To this end, we use the explicit form (124) of the energy functional (120) that appears in the definition of the matrices (148)–(150). As an example, we consider the matrix \( \tilde{M}_{Y,Y}^{\rho} \) and show that it is identical to the matrix in the first line of (B.16). With similar derivations one can prove the same for the other matrices (149), (150) and their counterparts in (B.16).

Using (124) and (148), we find
\[ \tilde{M}_{Y,Y}^{\rho} = \left[ \tilde{E}^{\rho} + \tilde{Q}^T \tilde{E}^{\rho} + \tilde{E} \tilde{Q} + \tilde{Q}^T \tilde{E} \tilde{Q} \right]_{Y,Y} + 2 \sum_X \frac{\partial E}{\partial \lambda_X^d} \cdot \frac{\partial^2 \lambda_X^d}{\partial \rho_Y \partial \rho_Y}. \]
Here, the matrices
\[ \tilde{E}^{\rho} = L^{\rho} - \sum_n \Lambda_n \tilde{g}^{\rho}, \ldots, \tilde{E}^{dd} = L^{dd} - \sum_n \Lambda_n \tilde{g}^{dd} \]
and \( \tilde{g}^{\rho}, \ldots, \tilde{g}^{dd} \) are defined as in (B.11) only with \( L \) replaced by \( E \) or \( g_n \), respectively. Obviously, the matrix in the first line of (B.16) is identical to \( \tilde{M}^{\rho} \) if
\[ 2 \sum_X \frac{\partial E}{\partial \lambda_X^d} \cdot \frac{\partial^2 \lambda_X^d}{\partial \rho_Y \partial \rho_Y} = - \sum_n \Lambda_n \left[ \tilde{g}^{\rho} + \tilde{Q}^T \tilde{g}^{\rho} + \tilde{g}^{dd} + \tilde{Q}^T \tilde{g}^{dd} \tilde{Q} \right]_{Y,Y}. \]
To prove (B.21), we use the fact that the second (total) derivatives of (B.1) with respect to the densities \( \rho_Y \) vanish,
\[ \frac{d g_n}{d \rho_Y} = \left[ \tilde{g}^{\rho} + \tilde{Q}^T \tilde{g}^{\rho} + \tilde{g}^{dd} + \tilde{Q}^T \tilde{g}^{dd} \tilde{Q} \right]_{Y,Y} + 2 \sum_X \frac{\partial g_n}{\partial \lambda_X^d} \cdot \frac{\partial^2 \lambda_X^d}{\partial \rho_Y \partial \rho_Y} = 0. \]
Equation (B.21) is therefore fulfilled if
\[ \sum_X \left( \frac{\partial E}{\partial \lambda_X^d} + \sum_n \Lambda_n \frac{\partial g_n}{\partial \lambda_X^d} \right) \frac{\partial^2 \lambda_X^d}{\partial \rho_Y \partial \rho_Y} = 0. \]
This equation, however, holds trivially, since (158) leads to
\[ \frac{\partial L}{\partial \lambda_Z} = \frac{\partial E}{\partial \lambda_Z} + \sum_n \Lambda_n \frac{\partial g_n}{\partial \lambda_Z} = 0 \]
for all parameters \( \lambda \) and in particular for \( \lambda = \lambda_X^d \) as it appears in (B.24).

### B.2. Linear transformations of the density matrix

In investigations of our translationally invariant lattice systems (1), it turns out to be more convenient to work with fluctuations \( \delta \tilde{\rho} \) that are linearly related to the density-matrix fluctuations,
\[ \delta \tilde{\rho} = \tilde{\Sigma} \cdot \delta \tilde{\mu}; \]
cf equations (C.27)--(C.28) and the resulting Green’s functions (178). The effective second-order functional (155)--(156) in terms of the fluctuations \( \delta \bar{\mu} \) is then given as

\[
\delta E^{(2)}(\delta \bar{\mu}) = \frac{1}{2}(\delta \bar{\mu})^T (\hat{\Xi}^T \hat{K}^{\rho \rho} \hat{\Xi}) \delta \bar{\mu},
\]

(B.26)

with \( \hat{K}^{\rho \rho} \) as defined in (156). For numerical calculations, it is important to show that one obtains the same kernel

\[
\hat{K}^{\mu \mu} \equiv \hat{\Xi}^T \hat{K}^{\rho \rho} \hat{\Xi}
\]

(B.27)
as in (B.26) if the transformation (B.25) and the antiadiabaticity condition are applied in reverse order. If we apply (B.25) first to (151), we obtain

\[
\delta E^{(2)} = \frac{1}{2}(\delta \bar{\mu})^T \hat{\Xi}^T \hat{M}_{\rho \rho} \hat{\Xi} \delta \bar{\mu} + 2(\delta \hat{\lambda}_1)^T \hat{M}_{\rho \rho} \hat{\Xi} \delta \bar{\mu} + (\delta \hat{\lambda}_1)^T \hat{M}_{\rho \rho} \hat{\Xi} \delta \bar{\mu} + (\delta \hat{\lambda}_1)^T \hat{M}_{\rho \rho} \hat{\Xi} \delta \bar{\mu}.
\]

(B.28)
The antiadiabaticity condition for \( \delta \bar{\mu} \) then reads

\[
\delta \hat{\lambda}_1 = -[\hat{M}^{\lambda \lambda}]^{-1} \hat{M}^{\rho \rho} \hat{\Xi} \delta \bar{\mu}.
\]

(B.29)

On being inserted into (B.28), this equation yields

\[
\delta E^{(2)}(\delta \bar{\mu}) = E_0 + \frac{1}{2}(\delta \bar{\mu})^T \hat{K}^{\mu \mu} \delta \bar{\mu},
\]

(B.30)
as claimed above.

**Appendix C. Explicit form of second-order expansion**

We calculate the second-order expansion of the Lagrange functional with respect to the variational parameters \( \lambda_{i;\Gamma,\Gamma'} \) and the density matrix (65). For the general consideration in section 5 and appendix B it was convenient to subsume the parameters \( \lambda_{\Gamma,\Gamma'} \) and their conjugates \( \lambda^*_{\Gamma,\Gamma'} \) into a set of \( n_p \) parameters \( \lambda_{Z_i} \); cf equation (121). In this appendix, where we aim to resolve the explicit structure of the second-order expansion, it is better to take the difference between \( \lambda_{\Gamma,\Gamma'} \) and \( \lambda^*_{\Gamma,\Gamma'} \) into account.

The constraints (35)--(36), the local energy (24) and the renormalization matrix (27) are all functions only of \( \lambda^*_{i;\Gamma,\Gamma'} \), \( \lambda_{i;\Gamma,\Gamma'} \) and of the local density matrix \( C^0_{i;\sigma,\sigma'} \). For simplicity, we use the joint variables \( A_i^v \), \( (A_i^v)^* \) for all these local variables, i.e. either

\[
A_i^v = A_i^{v_1}_{\sigma_1,\sigma_2} = (\hat{c}_{i,\sigma_2}^\dagger \hat{c}_{i,\sigma_1}) \quad \text{or} \quad A_i^v = A_i^{v_1}_{\Gamma,\Gamma'} = \lambda_{i;\Gamma,\Gamma'}.
\]

(C.1)

With respect to the parameters \( \lambda^*_{i;\Gamma,\Gamma'} \), \( \lambda_{i;\Gamma,\Gamma'} \), the second derivatives of (35)--(36), (24), (27) are quadratic functions of the form \( \sim (A_i^v)^* A_i^v \). Due to the Hermiticity of the density matrix, the same can be achieved with respect to the local density matrix. Then the only finite second derivatives of the Lagrange functional

\[
L = T + \sum_i E_i, \text{loc}(\{(A_i^v)^*\}, \{A_i^v\}) + \sum_{i,n} \Lambda_{i,n} g_{i,n}(\{(A_i^v)^*\}, \{A_i^v\})
\]

(C.2)

\[
T = \sum_{\text{i # j}} \sum_{\text{\sigma_1,\sigma_2}}^\alpha t_{i,j}^{\sigma_1,\sigma_2} q_{i,\sigma_1}^\sigma \langle \hat{c}_{i,\sigma_1}^\dagger \hat{c}_{j,\sigma_2} \rangle^*
\]

(C.3)
are
\[ \frac{\partial^2 L}{\partial (A_i^v)^* \partial A_i^{v'}} \neq 0 \] (C.4)

whereas
\[ \frac{\partial^2 L}{\partial (A_i^v)^* \partial (A_i^{v'})^*} = \frac{\partial^2 L}{\partial A_i^v \partial A_i^{v'}} = 0. \] (C.5)

The second-order expansion of the constraints and the local energy is straightforward since only local fluctuations \( \delta A_i^v \) couple,
\[ \delta L^{(2)}_{\text{loc}} = \sum_q \sum_{v,v'} (\delta A_q^v)^* K_{v,v'}^{\text{loc}} \delta A_q^{v'}. \] (C.6)

where we introduced
\[ K_{v,v'}^{\text{loc}} = \frac{\partial^2 L_{\text{loc}}}{\partial (A_i^v)^* \partial A_i^{v'}} \] (C.7)

and the Fourier transforms of the local fluctuations
\[ \delta A_i^v = \frac{1}{\sqrt{L_s}} \sum_q e^{-i\mathbf{R}_i \cdot \mathbf{q}} \delta A_q^v. \] (C.8)

All derivatives in this section (e.g. (C.7)) have to be evaluated for the ground-state values of the variational parameters \( \lambda_{i;\Gamma_i} \), the density matrix \( \tilde{\rho} \) and the Lagrange parameters \( \Lambda_{i;n} \). Note that the density-matrix fluctuations \( \delta A_q^{\sigma_2,\sigma_1} \) can be written as
\[ \delta A_q^{\sigma_2,\sigma_1} = \frac{1}{\sqrt{L_s}} \sum_k \delta \{ \hat{c}_{k,\sigma_1} \hat{c}_{k+q,\sigma_2}^{\dagger} \} \] (C.9)
where the operator \( \hat{A}_q^{\sigma} \) has been defined in (169).

In addition to (C.6), we need to take into account the mixed terms \( \sim \delta A_i^v \delta \Lambda_{i;n} \). In real space, their contribution is given as
\[ \delta L_{c}^{(2)} = \sum_{i,n,v} \left( \frac{\partial g_{i,n}}{\partial (A_i^v)^*} \delta (A_i^v)^* + \frac{\partial g_{i,n}}{\partial A_i^v} \delta A_i^v \right) \delta \Lambda_{i;n}. \] (C.10)

If we introduce the Fourier transforms \( \delta \Lambda_n^{\sigma} \) of the fluctuations \( \delta \Lambda_{i;n} \), we can write (C.10) as
\[ \delta L_c^{(2)} = \sum_q \sum_{n,v} (\delta A_q^v)^* K_{c,n}^q \delta \Lambda_n^{\sigma} + (\delta \Lambda_n^{\sigma})^* (K_{c,n}^q)^* \delta A_q^v. \] (C.11)

Here, we used the fact that the constraints \( g_{i,n} \) are assumed to be real and lattice-site independent such that
\[ K_{c,n}^q = \frac{\partial g_{i,n}}{\partial (A_i^v)^*} = \left( \frac{\partial g_{i,n}}{\partial A_i^v} \right)^*. \] (C.12)

More involved than the calculation of (C.6) is the expansion of the kinetic energy. Here we find
\[ \delta T^{(2)} = \delta T_1^{(2)} + \delta T_2^{(2)} \] (C.13)

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with

\[ \delta T_1^{(2)} = \sum_{i \neq j} \sum_{\alpha_1, \alpha_2, \sigma_1, \sigma_2} t^{\sigma_1, \alpha_2}_{ij} \langle \hat{c}^\dagger_{i \sigma_1} \hat{\bar{c}}_{j \sigma_2} \rangle \sum_{v, v'} \left[ \frac{\partial^2 q^{\sigma_1}_{i, \alpha_1}}{\partial (A^v_i)^* \partial A^v_{i'}} \left( q^{\sigma_1}_{j, \alpha_2} \right)^* \left( \delta A^v_{i'} \right)^* \delta A^v_{i'} \right. \\
+ \frac{1}{2} \left( \frac{\partial q^{\sigma_1}_{i, \alpha_1}}{\partial (A^v_i)^*} \frac{\partial \left( q^{\sigma_1}_{j, \alpha_2} \right)^*}{\partial A^v_{i'}} \left( \delta A^v_{i'} \right)^* \delta A^v_{i'} + \frac{\partial q^{\sigma_1}_{i, \alpha_1}}{\partial A^v_{i'}} \frac{\partial \left( q^{\sigma_1}_{j, \alpha_2} \right)^*}{\partial (A^v_i)^*} \left( \delta A^v_{i'} \right)^* \delta A^v_{i'} \right) \right] \] + c.c. \quad (C.14)

and

\[ \delta T_1^{(2)} = \sum_{i \neq j} \sum_{\alpha_1, \alpha_2, \sigma_1, \sigma_2} t^{\sigma_1, \alpha_2}_{ij} \delta \langle \hat{c}^\dagger_{i \sigma_1} \hat{\bar{c}}_{j \sigma_2} \rangle \sum_{v} \left[ \frac{\partial q^{\sigma_1}_{i, \alpha_1}}{\partial (A^v_i)^*} \left( q^{\sigma_1}_{j, \alpha_2} \right)^* \left( \delta A^v_{i} \right)^* + q^{\sigma_1}_{i, \alpha_1} \frac{\partial \left( q^{\sigma_1}_{j, \alpha_2} \right)^*}{\partial (A^v_i)^*} \left( \delta A^v_{i} \right)^* \right] \] + c.c. \quad (C.15)

The fact that the complex conjugates give the terms not explicitly shown in equations (C.14)–(C.15) follows from the relations

\[ \left( \frac{\partial q^{\sigma_1}_{i}}{\partial A^v_{i}} \right)^* = \frac{\partial \left( q^{\sigma_1}_{i} \right)^*}{\partial (A^v_{i})}, \quad (C.16) \]

\[ \left( \frac{\partial^2 q^{\sigma_1}_{i}}{\partial (A^v_{i})^* \partial A^v_{i'}} \right)^* = \frac{\partial^2 \left( q^{\sigma_1}_{i} \right)^*}{\partial (A^v_{i})^* \partial A^v_{i'}}, \quad (C.17) \]

\[ \left( t^{\sigma_1, \sigma_2}_{ij} \right)^* = t^{\sigma_2, \sigma_1}_{ij}. \quad (C.18) \]

For our translationally invariant ground state, it is more convenient to write (C.14)–(C.15) in momentum space. With the Fourier transforms of the local fluctuations, the term (C.14) reads

\[ \delta T_1^{(2)} = \sum_q \sum_{v, v'} (\delta A^q_{v})^* \left[ K^1_{q; v, \nu} + (K^1_{q; v', \nu})^* \right] \delta A^q_{v'}, \quad (C.19) \]

where

\[ K^1_{q; v, \nu} \equiv \sum_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'} \left[ \frac{1}{2} E_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'}(\bar{q}) \left( \frac{\partial q^{\sigma_1}_{\alpha_1}}{\partial (A^v_{i})^*} \frac{\partial \left( q^{\sigma_1}_{\alpha_2} \right)^*}{\partial A^v_{i'}} + \frac{\partial q^{\sigma_1}_{\alpha_1}}{\partial A^v_{i'}} \frac{\partial \left( q^{\sigma_1}_{\alpha_2} \right)^*}{\partial (A^v_{i})^*} \right) \right. \\
+ E_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'}(\bar{q}) \left( \frac{\partial^2 q^{\sigma_1}_{\alpha_1}}{\partial (A^v_{i})^* \partial A^v_{i'}} \left( q^{\sigma_1}_{\alpha_2} \right)^* \right) \left( q^{\sigma_1}_{\alpha_2} \right)^* \right]. \quad (C.20) \]

Here we assumed that the renormalization matrix is lattice-site independent and introduced the tensor

\[ E_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'}(\bar{q}) = \frac{1}{L_s} \sum_{k} \epsilon^{\sigma_1, \sigma_2}_{k+q} \langle \hat{c}^\dagger_{k \sigma_1} \hat{\bar{c}}_{k \sigma_2} \rangle \quad (C.21) \]

with

\[ \epsilon^{\sigma_1, \sigma_2}_{k} = \frac{1}{L_s} \sum_{i \neq j} t^{\sigma_1, \sigma_2}_{ij} e^{i \langle \bar{k}_i - \bar{k}_j \rangle}. \quad (C.22) \]

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Note that for \( \bar{q} = 0 \) the tensor (C.21),
\[
E_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'} = E_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'}(0)
\]  (C.23)
has already been defined in (32). For evaluation of the second (‘transitive’) term (C.15), we write the non-local density-matrix fluctuations as
\[
\delta \langle \hat{c}_{i \sigma_1}^\dagger \hat{c}_{j \sigma_2} \rangle = \frac{1}{L_s} \sum_{k, k'} e^{i(\tilde{K}_k - \tilde{K}_k')} \delta \langle \hat{c}_{k \sigma_1}^\dagger \hat{c}_{k' \sigma_2} \rangle.
\]  (C.24)
Together with (C.8) this yields
\[
\delta T_{t}^{(2)} = \frac{1}{L_s} \sum_{q, k, v, \sigma_1, \sigma_2} (\delta A^q_v)^* \tilde{K}_{k, q; v, \sigma_1, \sigma_2} \delta \langle \hat{c}_{k \sigma_1}^\dagger \hat{c}_{k + q \sigma_2} \rangle + \text{c.c.}
\]  (C.25)
with
\[
\tilde{K}_{k, q; v, \sigma_1, \sigma_2} = \sum_{\sigma_1', \sigma_2'} \left[ \frac{\partial q_{\sigma_1'}^{\sigma_2}}{\partial (A^v_q)^*} (q_{\sigma_2}^{\sigma_1})^* \epsilon_{k \sigma_1, \sigma_2} + q_{\sigma_1'}^{\sigma_2} \frac{\partial (q_{\sigma_2}^{\sigma_1})^*}{\partial (A^v_q)^*} \epsilon_{k' \sigma_2} \right].
\]  (C.26)
In principle, equations (C.25)–(C.26) allow us to calculate all second-order couplings of density-matrix and parameter fluctuations that arise from \( \delta T_{t}^{(2)} \). For numerical calculations, however, these equations are not very useful due to the explicit \( k \) dependence of (C.26). It is much easier to introduce the two auxiliary fluctuations
\[
\delta B_{w}^q \equiv \delta B_{\sigma_2, \sigma_1, \sigma_2', \sigma_1'}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k e_{k \sigma_1, \sigma_2}^\dagger \delta \langle \hat{c}_{k \sigma_1}^\dagger \hat{c}_{k + q \sigma_2} \rangle,
\]  (C.27)
\[
\delta \bar{B}_{w}^q \equiv \delta \bar{B}_{\sigma_2, \sigma_1, \sigma_2', \sigma_1'}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k e_{k + q \sigma_2}^\dagger \delta \langle \hat{c}_{k \sigma_1}^\dagger \hat{c}_{k + q \sigma_2} \rangle,
\]  (C.28)
where \( w \equiv (\sigma_2, \sigma_1, \sigma_2', \sigma_1') \) is an abbreviation for quadruples of indices \( \sigma \). With these definitions we can write (C.25) as
\[
\delta T_{t}^{(2)} = \sum_q \sum_{v, w} \left[ (\delta A^q_v)^* K_{v, w}^{t(1)} \delta B_{w}^q + (\delta A^q_v)^* K_{v, w}^{t(2)} \delta \bar{B}_{w}^q + (\delta B_{w}^q)^* (K_{v, w}^{t(1)})^* \delta A^q_v + (\delta \bar{B}_{w}^q)^* (K_{v, w}^{t(2)})^* \delta A^q_v \right],
\]  (C.29)
where
\[
K_{v, w}^{t(1)} = q_{\sigma_1'}^{\sigma_2}(q_{\sigma_2}^{\sigma_1})^* \frac{\partial q_{\sigma_1'}^{\sigma_2}}{\partial (A^v_q)^*},
\]  (C.30)
\[
K_{v, w}^{t(2)} = q_{\sigma_1'}^{\sigma_2}(q_{\sigma_2}^{\sigma_1})^* \frac{\partial (q_{\sigma_2}^{\sigma_1})^*}{\partial (A^v_q)^*}.
\]  (C.31)
Note that we introduced the two \( \delta B_{w}^q \) fluctuations (C.27), (C.28) only because they allow us to write the second-order expansion in a relatively simple form. In fact, these fluctuations are not independent but related through
\[
\delta \bar{B}_{w}^q_{\sigma_2, \sigma_1, \sigma_2', \sigma_1'} = (\delta B_{\sigma_2, \sigma_1, \sigma_2', \sigma_1'})^*.
\]  (C.22)
Altogether we end up with the following second-order expansion of the Lagrange functional:

\[
\delta L^{(2)} = \frac{1}{L_s} \sum_q \left( \delta \vec{A}^q \ | \ \delta \vec{B}^q \ | \ \delta \vec{z}^q \ | \ \delta \vec{A} \right)^* \tilde{K}^q \left( \begin{array}{c}
\delta \vec{A}^q \\
\delta \vec{B}^q \\
\delta \vec{z}^q \\
\delta \vec{A}
\end{array} \right),
\]

(C.33)

where

\[
\tilde{K}^q = \begin{pmatrix}
\tilde{K}^{(A,A)} & \tilde{K}^{(A,B)} & \tilde{K}^{(A,\tilde{B})} & \tilde{K}^{(A,\tilde{A})} \\
(K^{(A,B)})^\dagger & 0 & 0 & 0 \\
(K^{(A,\tilde{B})})^\dagger & 0 & 0 & 0 \\
(K^{(A,\tilde{A})})^\dagger & 0 & 0 & 0
\end{pmatrix}
\]

(C.34)

and

\[
\tilde{K}^{(A,A)} = \tilde{K}^{\text{loc}} + \tilde{K}^{11} + (\tilde{K}_q^{11})^\dagger,
\]

(C.35)

\[
\tilde{K}^{(A,B)} = \tilde{K}^{(11)}, \quad \tilde{K}^{(A,\tilde{B})} = \tilde{K}^{(12)},
\]

(C.36)

\[
\tilde{K}^{(A,\tilde{A})} = \tilde{K}^c.
\]

(C.37)

As described in section 5.4, the antiadiabaticity condition leads to an effective second-order functional only of the density matrix. This condition can be evaluated directly for the second-order expansion (C.33) since the fluctuations \( \delta \vec{A}^q \), \( \delta \vec{B}^q \) and \( \delta \vec{z}^q \) are some linear functions of the density-matrix fluctuations \( \delta (\tilde{c}_{k,\sigma}, \tilde{c}_{k^\sigma,\sigma}) \); cf appendix B.2. To this end, we distinguish the fluctuations of the local density matrix \( \delta \vec{A}^q \) and of the variational parameters \( \delta \vec{A}^q \) as well as the corresponding blocks in the matrix (C.34),

\[
\tilde{K}^{(A,A)} = \begin{pmatrix}
\tilde{K}^{(A,A)}_{(\lambda,\lambda)} & \tilde{K}^{(A,A)}_{(\lambda,\rho)} \\
(K^{(A,A)})^\dagger_{(\lambda,\rho)} & \tilde{K}^{(A,A)}_{(\rho,\rho)}
\end{pmatrix}, \quad \tilde{K}^{(A,B)} = \begin{pmatrix}
\tilde{K}^{(A,B)}_{(\lambda,\lambda)} \\
(K^{(A,B)})^\dagger_{(\lambda,\rho)} & \tilde{K}^{(A,B)}_{(\rho,\rho)}
\end{pmatrix}, \ldots
\]

(C.38)

The resulting functional is then given as

\[
\delta \tilde{L}^{(2)} = \frac{1}{L_s} \sum_q \left( \delta \vec{A}^q \ | \ \delta \vec{B}^q \ | \ \delta \vec{z}^q \ | \ \delta \vec{A} \right)^* \tilde{V}^q \left( \begin{array}{c}
\delta \vec{A}^q \\
\delta \vec{B}^q \\
\delta \vec{z}^q \\
\delta \vec{A}
\end{array} \right)
\]

(C.39)

with the new kernel

\[
\tilde{V}^q = \begin{pmatrix}
\tilde{V}^{(A,A)} & \tilde{V}^{(A,B)} & \tilde{V}^{(A,\tilde{B})} \\
\tilde{V}^{(B,A)} & \tilde{V}^{(B,B)} & \tilde{V}^{(B,\tilde{B})} \\
\tilde{V}^{(\tilde{B},A)} & \tilde{V}^{(\tilde{B},B)} & \tilde{V}^{(\tilde{B},\tilde{B})}
\end{pmatrix}
\]

(C.40)

\[
= \begin{pmatrix}
\tilde{K}^{(A,A)}_{(\lambda,\lambda)} & \tilde{K}^{(A,B)}_{(\lambda,\lambda)} & \tilde{K}^{(A,\tilde{B})}_{(\lambda,\lambda)} \\
(K^{(A,B)})^\dagger_{(\lambda,\rho)} & 0 & 0 \\
(K^{(A,\tilde{B})})^\dagger_{(\lambda,\rho)} & 0 & 0
\end{pmatrix} - \Delta \tilde{V}^q
\]

(C.41)
where
\[
\Delta \tilde{V}^q \equiv \begin{pmatrix}
\tilde{K}_{\rho,\lambda}^{(A,A)} & \tilde{K}_{\rho,\lambda}^{(A,\Lambda)} \\
(\tilde{K}_{\lambda,\lambda}^{(A,B)})^\dagger & 0 \\
(\tilde{K}_{\lambda,\rho}^{(A,B)})^\dagger & 0
\end{pmatrix}^{-1}
\begin{pmatrix}
\tilde{K}_{\lambda,\rho}^{(A,A)} & \tilde{K}_{\lambda,\rho}^{(A,\Lambda)} \\
(\tilde{K}_{\lambda,\lambda}^{(A,B)})^\dagger & 0 \\
(\tilde{K}_{\lambda,\rho}^{(A,B)})^\dagger & 0
\end{pmatrix}.
\]  
(C.42)

Note that \(\tilde{V}^q\) (unlike \(\tilde{K}^q\)) includes finite couplings also between the fluctuations \(\delta \tilde{B}^q\), \(\delta \tilde{B}^q\). The calculation of \(\tilde{V}^q\) (for fixed \(\vec{q}\)) only involves the handling of finite-dimensional matrices. In contrast, the evaluation of the functional (C.25) (instead of (C.29)) would have led to significantly more complicated equations.

**Appendix D. Explicit form of Gutzwiller-random-phase approximation equations**

In this appendix, we prove that the general Gutzwiller-RPA equations (144) lead to the Green’s function matrix (179) if applied to our multi-band Hamiltonian (1). With the abbreviations \(\delta D^q\) and \(\tilde{D}^q\) for the three fluctuations \(\delta A^q\), \(\delta B^q\) and \(\delta \tilde{B}^q\) and the corresponding operators \(\hat{A}^q\), \(\hat{B}^q\) and \(\tilde{B}^q\), we have to show that the Green’s function matrix
\[
\Pi_{\mu,\mu'}(\vec{q}, \omega) = \langle \langle \hat{D}^q_{\mu}; (\hat{D}^q_{\mu})^\dagger \rangle \rangle_\omega,
\]  
(D.1)
as given in (179), obeys the equation
\[
\delta D^q = \sum_{\mu'} \langle \langle \hat{D}^q_{\mu'}; (\hat{D}^q_{\mu'})^\dagger \rangle \rangle_\omega \delta f^q_{\mu'}.
\]  
(D.2)

Using the explicit form (179) of \(\tilde{\Pi}(\vec{q}, \omega)\), this equation can also be written as
\[
\sum_{\mu'} \{1 + \tilde{\Pi}^0(\vec{q}, \omega) \tilde{V}^q\}_{\mu,\mu'} \delta D^q_{\mu'} = \sum_{\mu'} \Pi^0_{\mu,\mu'}(\vec{q}, \omega) \delta f^q_{\mu'}.
\]  
(D.3)

Note that the excitation amplitudes \(\delta f^q_{\mu'}\) enter the problem through the perturbation operator
\[
\delta \hat{V}_f \equiv \sum_{\mu} \delta f^q_{\mu} (\hat{D}^q_{\mu})^\dagger
\]  
\[
= \frac{1}{\sqrt{L_s}} \sum_k \sum_{\sigma_1,\sigma_2,\sigma_1',\sigma_2'} \hat{c}^\dagger_{k+q,\sigma_1'} \hat{c}_{k,\sigma_2'} \left( \delta f^A_{\sigma_1,\sigma_2} \delta \hat{g}_{\sigma_1,\sigma_1'} \delta \hat{g}_{\sigma_2,\sigma_2'} + \delta f^B_{\sigma_1,\sigma_2} \delta \hat{g}_{\sigma_1,\sigma_1'} \delta \hat{g}_{\sigma_2,\sigma_2'} + \delta f^{\tilde{B}}_{\sigma_1,\sigma_2} \delta \hat{g}_{\sigma_1,\sigma_1'} \delta \hat{g}_{\sigma_2,\sigma_2'} \right),
\]  
(D.4)

which is needed to define the general Green’s functions (178).

Before we prove equation (D.3), it is instructive to consider the case \(\tilde{V}^q = 0\) in which the three fluctuations \(\delta \hat{A}^q\), \(\delta \hat{B}^q\) and \(\delta \tilde{B}^q\) are decoupled and we can set \(f^{\tilde{B};q} = f^{B;q} = 0\). We start this derivation in the eigenbasis of the Gutzwiller Hamiltonian (180). It leads to the simplest form of the matrix \(\tilde{E}\) in equation (144), which then reads
\[
(\omega - (E_{k+q,\sigma_1} - E_{k,\sigma_2})) \delta \langle \hat{g}_{k+q,\sigma_1} \hat{g}_{k+q,\sigma_1} \rangle_{ph} = \frac{1}{\sqrt{L_s}} (n_{k,\sigma_2} - n_{k+q,\sigma_1}) \delta f_{(k+q,\sigma_1),(k,\sigma_2)}.
\]  
(D.5)
Here the excitation amplitude is given as

\[ \delta f_{(k+q,a_1),(k,a_2)} = \delta \bar{A}_{\sigma_2}^{k+q} (u_{\sigma_2}^{k+q})^* u_{\sigma_2}^k. \]  

(D.6)

Note that the factor \( n_{k,a_2}^0 - n_{k+q,a_1}^0 = \pm 1 \) in (D.5) represents the particle–hole and the hole–particle channels in equation (144). For simplicity, we will drop the corresponding labels hp/ph in the following.

With the transformations (182), (183), equation (D.5) leads to

\[ \delta A_{\sigma_1,\sigma_2}^q = \frac{1}{L_s} \sum_k \delta \langle \hat{c}_{k,a_1}^{\dagger} \hat{c}_{k+q,a_1} \rangle \]

\[ = \frac{1}{L_s} \sum_k \sum_{a_1,a_2} (u_{\sigma_1,a_2}^k)^* u_{\sigma_2,a_1}^{k+q} \delta \langle \hat{h}_{k,a_2}^{\dagger} \hat{h}_{k+q,a_1} \rangle \]

\[ = \frac{1}{L_s} \sum_k \sum_{a_1,a_2} (u_{\sigma_1,a_2}^k)^* u_{\sigma_2,a_1}^{k+q} (u_{\sigma_1,a_1}^{k+q})^* u_{\sigma_2,a_2}^k (n_{k,a_2}^0 - n_{k+q,a_1}^0) \delta f_{\sigma_1,\sigma_2}^q. \]  

(D.7)

As expected, we therefore find

\[ \delta A_{\sigma_1,\sigma_2}^q = \sum_{\sigma_1,\sigma_2} \langle \langle \hat{A}_{\sigma_1,\sigma_2}^q, (\hat{A}_{\sigma_1,\sigma_2}^q)^\dagger \rangle \rangle_{\omega} \delta f_{\sigma_1,\sigma_2}^q, \]  

(D.8)

with the (‘retarded’) Green’s function

\[ \langle \langle \hat{A}_{\sigma_1,\sigma_2}^q, (\hat{A}_{\sigma_1,\sigma_2}^q)^\dagger \rangle \rangle_{\omega} = \frac{1}{L_s} \sum_k \sum_{a_1,a_2} (u_{\sigma_1,a_2}^k)^* u_{\sigma_1,a_1}^{k+q} (u_{\sigma_2,a_1}^{k+q})^* u_{\sigma_2,a_2}^k (n_{k,a_2}^0 - n_{k+q,a_1}^0) \delta (E_{k+q,a_1} - E_{k,a_2}) + i\delta (n_{k,a_2}^0 - n_{k,a_1}^0) \]

(D.9)

as introduced in (184).

Now we consider the case of a finite interaction matrix \( \tilde{V}^q \). Using our abbreviation \( \delta D_{\mu} \) for the amplitudes \( \delta A_{\nu}, \delta B_{\omega} \) and \( \delta B_{\omega} \), the Lagrange functional \( \delta \bar{L}^{(2)} \) has the form

\[ \delta \bar{L}^{(2)} = \sum_{q,\mu,\nu} \langle \langle \delta D_{\mu}^q, (\delta D_{\nu}^q)^\dagger \rangle \rangle_{\omega} \tilde{V}_{\mu,\nu}. \]  

(D.10)

With this additional interaction term, equation (D.5) reads

\[ (\omega - (E_{k+q,a_1} - E_{k,a_2})) \delta \langle \hat{h}_{k,a_2}^{\dagger} \hat{h}_{k+q,a_1} \rangle + (n_{k,a_2}^0 - n_{k+q,a_1}^0) \sum_{k',a_2} U_{k,a_1,a_2}^{k',a_2} (q) \delta \langle \hat{h}_{k',a_2}^{\dagger} \hat{h}_{k+q,a_1} \rangle \]

\[ = \frac{1}{L_s} (n_{k,a_2}^0 - n_{k+q,a_1}^0) \delta f_{(k+q,a_1),(k,a_2)}, \]  

(D.11)

where

\[ U_{k,a_1,a_2}^{k',a_2} (q) = \frac{\partial}{\partial \delta (\hat{h}_{k+q,a_1}^{\dagger} \hat{h}_{k,a_2})} \frac{\partial}{\partial \delta (\hat{h}_{k',a_2}^{\dagger} \hat{h}_{k+q,a_1})} \delta \bar{L}^{(2)} \]

\[ = \sum_{\mu,\nu} \tilde{V}_{\mu,\nu} \frac{\partial (\delta D_{\mu}^q)^*}{\partial \delta (\hat{h}_{k+q,a_1}^{\dagger} \hat{h}_{k,a_2})} \frac{\partial \delta D_{\nu}^q}{\partial \delta (\hat{h}_{k',a_2}^{\dagger} \hat{h}_{k+q,a_1})} \]  

(D.12)

(D.13)
and
\[
\delta f_{(\kappa, q, \alpha_1), (\kappa, q_2)} = \sum_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} (u^{k+q}_{\alpha_1, \alpha_2})^* u^k_{\sigma'_1, \sigma'_2} \left( \delta f_{A:q}^{\alpha_1, \alpha_2} \delta_{\sigma_1, \sigma'_1} \delta_{\sigma_2, \sigma'_2} + \delta f_{B:q}^{\alpha_1, \alpha_2} e^{\alpha_1, \alpha_2} + \delta f_{A:q}^{\alpha_1, \alpha_2} e^{\alpha_1, \alpha_2} \right).
\]

The derivatives in (D.13) can be further evaluated using the transformation (182), (183).

\[
\frac{\partial \delta D^q_{\mu}}{\partial \delta (\hat{h}^\dagger_{k+q, \alpha_1} \hat{h}_{k+q, \alpha_2})} = \sum_{\sigma_1, \sigma_2} \frac{\partial (\delta D^q_{\mu})^*}{\partial \delta (\hat{c}^\dagger_{k+q, \sigma_1} \hat{c}_{k+q, \sigma_2})} (u^{k+q}_{\alpha_1, \alpha_2})^* u^k_{\sigma_1, \sigma_2},
\]

(D.15)

\[
\frac{\partial \delta D^q_{\mu'}}{\partial \delta (\hat{h}^\dagger_{k', \alpha_1} \hat{h}_{k'+q, \alpha_2})} = \sum_{\sigma_3, \sigma_4} \frac{\partial \delta D^q_{\mu'}}{\partial \delta (\hat{c}^\dagger_{k+q, \sigma_3} \hat{c}_{k+q, \sigma_4})} (u^{k'}_{\alpha_1, \alpha_2})^* u^{k+q}_{\sigma_3, \sigma_4}.
\]

(D.16)

Depending on the particular fluctuations \( \delta D^q_{\mu} \), the remaining derivatives on the rhs of equations (D.15), (D.16) are given as

\[
\delta D^q_{\mu} = \delta A^q_{\mu} : \frac{\partial \delta A^q_{\sigma_1, \sigma_2}}{\partial \delta (\hat{c}^\dagger_{k, \sigma_1} \hat{c}_{k+q, \sigma_2})} = \frac{\partial (\delta A^q_{\sigma_1, \sigma_2})^*}{\partial \delta (\hat{c}^\dagger_{k+q, \sigma_1} \hat{c}_{k+q, \sigma_2})} = \frac{\delta_{\sigma_1, \sigma_1} \delta_{\sigma_2, \sigma_2}}{\sqrt{L_s}},
\]

(D.17)

\[
\delta D^q_{\mu} = \delta B^q_{\mu} : \frac{\partial \delta B^q_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'}}{\partial \delta (\hat{c}^\dagger_{k, \sigma_1} \hat{c}_{k+q, \sigma_2})} = \frac{\partial (\delta B^q_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'})^*}{\partial \delta (\hat{c}^\dagger_{k+q, \sigma_1} \hat{c}_{k+q, \sigma_2})} = \frac{\delta_{\sigma_1, \sigma_1} \delta_{\sigma_2, \sigma_2} \epsilon_{\sigma_2, \sigma_2}}{\sqrt{L_s}},
\]

(D.18)

\[
\delta D^q_{\mu} = \delta \tilde{B}^q_{\mu} : \frac{\partial \delta \tilde{B}^q_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'}}{\partial \delta (\hat{c}^\dagger_{k, \sigma_1} \hat{c}_{k+q, \sigma_2})} = \frac{\partial (\delta \tilde{B}^q_{\sigma_1, \sigma_2, \sigma_1', \sigma_2'})^*}{\partial \delta (\hat{c}^\dagger_{k+q, \sigma_1} \hat{c}_{k+q, \sigma_2})} = \frac{\delta_{\sigma_1, \sigma_1} \delta_{\sigma_2, \sigma_2} \epsilon_{\sigma_2, \sigma_2}}{\sqrt{L_s}}.
\]

(D.19)

With equations (D.12)–(D.19), we are now in a position to evaluate (D.11). To this end, we proceed as in (D.7),

\[
\delta A^q_{\alpha_1, \alpha_2} = \frac{1}{\sqrt{L_s}} \sum_k \sum_{\alpha_1, \alpha_2} (u^{k+q}_{\alpha_1, \alpha_2})^* u^{k+q}_{\alpha_1, \alpha_2} \delta (\hat{h}^\dagger_{k+q, \alpha_1} \hat{h}_{k+q, \alpha_2})
\]

\[
= - \sum_{\mu, \mu'} V^q_{\mu, \mu'} \left[ \left( \frac{1}{\sqrt{L_s}} \sum_k \sum_{\alpha_1, \alpha_2} (u^{k+q}_{\alpha_1, \alpha_2})^* u^{k+q}_{\alpha_1, \alpha_2} (u^{k+q}_{\alpha_1, \alpha_2})^* u^{k+q}_{\alpha_1, \alpha_2} \right) \right]
\]

\[
\times \frac{\partial (\delta D^q_{\mu})^*}{\partial \delta (\hat{c}^\dagger_{k+q, \sigma_1} \hat{c}_{k+q, \sigma_2})} \times \sum_{\sigma_3, \sigma_4} \frac{\partial \delta D^q_{\mu'}}{\partial \delta (\hat{c}^\dagger_{k+q, \sigma_3} \hat{c}_{k+q, \sigma_4})} \delta (\hat{c}^\dagger_{k'+q, \sigma_1} \hat{c}_{k'+q, \sigma_2})
\]

\[
+ \sum_{\mu} \langle \langle \hat{A}_{\alpha_1, \alpha_2}; (\hat{D}^q_{\mu})^\dagger \rangle \rangle^0 \delta f^q_{\mu}.
\]

(D.20)

The sums over \( \mu \) and \( \mu' \) lead to nine contributions that can all be evaluated using equations (D.17)–(D.19). As a result, we find

\[
\delta A^q_{\mu} + \sum_{\mu, \mu'} \langle \langle \hat{A}_{\mu}; (\hat{D}^q_{\mu})^\dagger \rangle \rangle^0 V^q_{\mu, \mu'} \delta D^q_{\mu} = \sum_{\mu} \langle \langle \hat{A}_{\mu}; (\hat{D}^q_{\mu})^\dagger \rangle \rangle^0 \delta f^q_{\mu},
\]

(D.21)
where the ‘non-interacting’ Green’s functions $\langle \langle \hat{A}_q^*; (\hat{D}_q^*)^\dagger \rangle \rangle_\omega^0$ in (D.21) are given as in (D.9), apart from additional factors $e^{\sigma_i \sigma_1 \epsilon_k}$ or $e^{\sigma_i \sigma_2 \epsilon_k}$.

\[
\left( \langle \langle \hat{A}_{q_1}^*; (\hat{B}_{q_1}^*)^\dagger \rangle \rangle \right)_\omega^0 = \frac{1}{L_8} \sum_k \sum_{\alpha_1,\alpha_2} \left( u_{\sigma_1,\alpha_2}^k \right)^* u_{\sigma_1,\alpha_1}^{k+q} \left( u_{\sigma_1,\alpha_1}^{k+q} \right)^* u_{\sigma_1,\alpha_2}^{k+q} \left( e_k^{\sigma_1 \sigma_4} \right)^* \left( e_k^{\sigma_1 \sigma_4} \right) \left( n_{k,\alpha_2}^0 - n_{k,\alpha_1}^0 \right),
\]

(D.22)

With (D.21), we have proved the ‘first’ set of equations (D.3), i.e. those with $\mu = v = (\sigma, \sigma')$. If we replace $\delta A_{\sigma_1,\sigma_2}$ in the first line of equation (D.20) by

\[
\delta B_{\sigma_1,\sigma_2,\sigma_1';\sigma_2'} = \frac{1}{\sqrt{L_8}} \sum_k \sum_{\alpha_1,\alpha_2} \left( u_{\sigma_1,\alpha_2}^k \right)^* u_{\sigma_1,\alpha_1}^{k+q} \delta \langle \hat{h}_{k,\alpha_2}^\dagger \hat{h}_{k,\alpha_1} \rangle e_{\sigma_1'}^{\sigma_1} e_{\sigma_2'}^{\sigma_2}
\]

or by

\[
\delta \bar{B}_{\sigma_1,\sigma_2,\sigma_1';\sigma_2'} = \frac{1}{\sqrt{L_8}} \sum_k \sum_{\alpha_1,\alpha_2} \left( u_{\sigma_1,\alpha_2}^k \right)^* u_{\sigma_1,\alpha_1}^{k+q} \delta \langle \hat{h}_{k,\alpha_2}^\dagger \hat{h}_{k,\alpha_1} \rangle e_{\sigma_1'}^{\sigma_1} e_{\sigma_2'}^{\sigma_2},
\]

(D.24)

the remaining equations (D.3) are derived in the very same way as (D.21). This closes our proof of equation (179).

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