Canonical perturbation theory for inhomogeneous systems of interacting fermions

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We consider a model of interacting fermions on a lattice with a strong on-site term and a weak intersite one. The model is defined in terms of Hubbard operators and takes into account the local lattice defects as well as the magnetic field. Using a canonical perturbation theory we obtain explicit formulas for parameters of the effective Hamiltonian exact up to second order with respect to the intersite term. We show how the method can be generalized recursively to higher orders and exemplify the procedure with an application to a single band Hubbard model with defects and to its two-orbital extension.

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I. INTRODUCTION

A many-body problem of strongly interacting electrons on a lattice presents an important challenge in the condensed matter physics. Analytical solutions are scarce, whereas approximate treatments using Green's functions methods are complex and often difficult to control. Numerically exact solutions for finite systems are increasingly more available with a growth of computer power, but they critically depend on a number of degrees of freedom of a system under study.

One of the most useful approaches which reduce the complexity of the original problem is a perturbation method based on a canonical transformation.[1–9] In recent years the method was frequently applied to the Hubbard model,[10–13] as well as its many band counterparts,[14–18] to study spectral properties or to describe various electron orderings, including the magnetic and superconducting ones.

The canonical perturbation approach can be formulated in a recursive way[8] that is fully consistent with the perturbation theory based on a resolvent expansion.[19,20] Its practical advantages over the resolvent method is that it allows to obtain in a straightforward way the effective Hamiltonians which are valid for all spectrum range of the original model, rather than just for the the lowest energy region.[19] Although this feature of canonical formulation of the perturbation theory is relatively seldom used in studies of bulk systems and in thermodynamic equilibrium, it can give an important advantage for investigations of nanosystems far from equilibrium conditions.

In the original formulation[8] the recursive canonical approach was developed for the model with a single orbital per site on a perfect lattice. It is of interest to see if the method can be extended to the more complicated cases like multiorbital systems or imperfect lattices. The purpose of the present paper is to describe in detail a formulation of the method which can be applied to a fairly general Hamiltonian of strongly interacting particles. Using this approach various special cases considered so far can be analyzed from the common point of view.

The method can be applied to fermion and boson models with correlations, for example, to study many-body physics of interacting photons (or polaritons) on optical lattice[21] and ultracold atomic gases.[22,23] In this paper we focus on fermion Hamiltonians with a single and many orbitals on lattices with defects, and particularly, on mesoscopic systems, like nanocomposites,[24] coherently coupled quantum dots,[25,26] and magnetic molecules.[27–31] We derive an effective many-body Hamiltonian which properly describes both low and high energy charge and magnetic excitations. Our derivations are general; the new Hamiltonian includes various types of interactions and can be applied to search for the ground state and excitations in multiferroics,[32,33] molecular crystals with charge transfer,[34,35] cobaltates[36] and manganites,[37] or charge and spin dynamics in nanostructures in nonequilibrium situations.[29,30,38–40]

In Sec. II we describe the considered system of electrons with strong local interactions on a lattice and construct the general Hamiltonian. Using Hubbard operators we can express many-body terms of the Hamiltonian in a compact form suitable for the canonical transformation method. Next, we perform a derivation of the effective Hamiltonian up to the second order perturbation (for the sake of clarity of the presentation some more technical aspects of the derivation are shifted to Appendixes A and B). A more specific example of both second order and third order calculations is presented in Sec. III, where we consider the Hubbard model with local defects and in the presence of a flux of magnetic field. A derivation of the effective Hamiltonian for an extension of the Hubbard model with two orbitals per site is presented in Sec. IV. We summarize the results and discuss a scope of possible applications of the presented approach in Sec. V.

II. MODEL OF INTERACTING FERMIONS

A Hamiltonian of interacting electrons on a lattice can be written in a concise way using Hubbard operators [41,42]

$$X_i^{\alpha''\alpha'} = |\alpha''\rangle_i \langle\alpha'|_i, \tag{1}$$

defined by means of a set $\{|\alpha\rangle_i\}$ of local states for any lattice site i (or more generally for any ligand). It is assumed that for each individual site we know all these states $|\alpha\rangle_i$ for different charge and spin configurations. Among them one can also find excited many-electron states (e.g., to describe local excitons or charge-transfer[14,15,34]).

The Hubbard operator $X_i^{\alpha''\alpha'}$, by its construction, describes the transition between the local states: from the initial state $|\alpha'\rangle_i$ to the final state $|\alpha''\rangle_i$. To simplify the notation, in most of the paper we use a single vector index $\alpha = (\alpha'',\alpha')$ to uniquely identify the Hubbard operators (i.e., we write X_i^{α} to mean

$X_i^{\alpha''\alpha'}$). According to this convention the first component of the vector index α denotes the final state and is distinguished with a double primed character, whereas the second component corresponding to the initial state is denoted with a single prime.

The action of the Hubbard operator on the given site state produces a the final state with a definite number of fermions $n_{\alpha''}$, which, in general, can be different from the one of the initial state, $n_{\alpha'}$. When the change in a number of fermions on the given site is an odd number, we call the Hubbard operator a fermion-like and substitute letter X by F in its denotation. When $n_{\alpha''} - n_{\alpha'}$ is an even nonzero number we call the Hubbard operator a boson-like and denote it by letter B. Finally, when the number of fermions remains unchanged (i.e., $n_{\alpha''} = n_{\alpha'}$), the Hubbard operator can be identified with one of standard basis (SB) operators often used in the theory of magnetism,[43] and it is denoted by letter L. We point out that the usual fermion operators of the second quantization formalism defined in terms of local one-particle states, can be written as linear combinations of the fermion-like Hubbard operators for the given site, as well as all the Hubbard operators can be represented by linear combinations of products of the one particle fermion operators.

A. Hamiltonian of the model

In our analysis we restrict ourselves to models with strong on-site electron interactions and neglect interactions between electrons from different sites (in Appendix B we discuss an extension of the method to include intersite interactions as well). In the result, the on-site part of the Hamiltonian can be written as

$$W = \sum_{i\alpha} E_i^{\alpha} L_i^{\alpha}. \tag{2}$$

Here we assume that the local problem is already solved and all local interactions are taken into account (also those describing defects) whereas $E_i^{\alpha} \equiv \delta_{\alpha'\alpha''} E_i^{\alpha'}$ is an energy eigenvalue corresponding to the site state $|\alpha'\rangle_i$.

In what follows we assume that the eigenspace of W can be partitioned into two or more subspaces C_q well separated from each other by sizable energy gaps. The energy gaps can be due to strong intrasite Coulomb interactions, Hund's coupling or crystal fields. These type of interactions can be also found in nanoscopic systems, where electron correlations are strong and relevant.[38–40] Additionally, one has to take into account geometrical constrains of nanosystems (e.g., larger level separation), or different local symmetry for hole and electron states in a quantum well.

The lowest eigenspace C_0 corresponds to the ground state and for the case of transition metal compounds it includes degenerate many-electron states with various charge and spin spatial configurations governed by the Hund's rules. To the higher subspaces $C_{q\geqslant 1}$ belong high excited many-body states, which positions depend on interplay of the local interactions. For example, according to the Zaanen-Sawatzky-Allen model[14] an electronic structure in transition metal oxides depends on relation of the intrasite Coulomb interaction energy U and the charge transfer energy Δ_c (see also Ref. 44).

Electron hopping between sites is described by the intersite part of the Hamiltonian, which can be expressed by means of the Hubbard operators as

$$T = \sum_{\substack{q\geqslant 0, p \\ q+p\geqslant 0}} P_{q+p} T P_q = \sum_p T_p = \sum_{\substack{p,(ij) \\ \alpha,\beta}} v_{ij,p}^{\alpha\beta} \overset{+}{F}{}_i^{\alpha} F_j^{\beta}, \tag{3}$$

where (ij) in the last sum above means summation over different (nearest neighbor, NN) sites i, j, and P_q is the projection operator to the subspace C_q. Here and in what follows the Hubbard fermion-like operator that adds a fermion to a site is distinguished by the plus sign over the letter F, whereas the bare letter F denotes the fermion-like operator that removes a fermion from a site (a similar convention is applied below to the boson-like operators). In Eq. (3), a (correlated) hopping parameter $v_{ij,p}^{\alpha\beta}$ is indexed with an integer p to indicate that it corresponds to a term transferring a state from a subspace C_q to C_{q+p}, for all q. In the present considerations we restrict ourselves to single electron hopping, taking into account also correlated hopping between many-electron states with different charge and spin configurations. These terms can include hopping accompanied by spin-flip processes and can be used to describe the spin-orbit interactions.[45]

The total Hamiltonian of the system is defined as

$$H = W + \tau T, \tag{4}$$

where τ is a small parameter used to classify various contributions in the perturbation expansion, and at the end of the calculation we put $\tau = 1$. The total Hamiltonian of the system, Eq. (4), comprises two types of contributions: diagonal and nondiagonal ones. The diagonal terms, the on-site Hamiltonian W and the contribution T_0 to the hopping part, do not mix states from the different subspaces C_q. The nondiagonal part of the Hamiltonian, which includes all the other contributions T_q to the intersite part, transfers states between the different subspaces.

Note that the resolution of the hopping part into the diagonal and the nondiagonal contributions, implied by Eqs. (3), is written in the form of the sum of the two-site terms. This is possible because the division into various subspaces C_q is made here with respect to the spectrum of the Hamiltonian W, which is the sum of the single site terms (see also Appendix B).

B. Derivation of the effective Hamiltonian

We derive now explicitly the effective Hamiltonian up to the second order perturbation for the model of strongly interacting fermions, introduced in the previous chapter, see Eq. (4). For this purpose we employ an extension of the canonical perturbation method developed by MacDonald *et al.*,[8] that recursively eliminates the nondiagonal terms from the unitary transformed Hamiltonian. For completeness, in Appendix A we summarize the derivation of the method in a form which is able to deal with the multiorbital systems.

We start here from a formula for the second order canonically transformed Hamiltonian

$$\tilde{H}^{[2]} = W + h^{[1]} + h^{[2]}, \tag{5}$$

where $h^{[1]} = T_0$ and

$$h^{[2]} = \frac{1}{2}\sum_p \left[iS^{[1]}_{-p}, T_p\right]. \tag{6}$$

Above, $h^{[N]}$ and $iS^{[N]}$ denote Nth order contributions to the expansion of the effective Hamiltonian $\tilde{H}$ and the generator of the canonical transformation iS. For the first order effective Hamiltonian to be diagonal between the subspaces C_q the contributions of the generator iS needs to obey the recurrent equations

$$\left[iS^{[1]}_p, W\right] = -T_p, \quad \text{for } p \neq 0, \tag{7}$$

for the first order component of iS. Operator solutions of the recurrence equations can be found in this case if one takes into account the commutation properties of the SB operator L^α_i from Eq. (2), with an arbitrary Hubbard operator X^β_j

$$\left[L^\alpha_i, X^\beta_j\right] = \delta_{i,j}\left(\delta_{\alpha',\beta''} - \delta_{\alpha',\beta'}\right) X^\beta_j, \tag{8}$$

for $\alpha' = \alpha''$. This suggests searching for the solutions in forms of the linear combinations of the operators that appear in the right-hand sides of the recurrent equations, like Eqs. (A7) and (A9). In particular, on the basis of Eq. (3) the solution of Eq. (7) can be anticipated in the form

$$iS^{[1]}_p = \sum_{\substack{(\ell m)\\ \lambda\mu}} s^{\lambda\mu[1]}_{\ell m,p} \overset{+}{F}{}^\lambda_\ell F^\mu_m, \tag{9}$$

for $p \neq 0$. Inserting the above expression into Eq. (7) we obtain

$$\sum_{\substack{(\ell m)\lambda\mu\\ j\alpha}} s^{\lambda\mu[1]}_{\ell m,p} E^\gamma_n \left[\overset{+}{F}{}^\lambda_\ell F^\mu_m, L^\gamma_n\right] = -\sum_{\substack{(ij)\\ \alpha,\beta}} v^{\alpha\beta}_{ij,p} \overset{+}{F}{}^\alpha_i F^\beta_j. \tag{10}$$

Computing the commutators on the left-hand side of Eq. (10) and comparing the results with the right-hand side we obtain

$$s^{\alpha\beta[1]}_{ij,p} = -\frac{v^{\alpha\beta}_{ij,p}}{\Delta^{\alpha\beta}_{ij}}, \tag{11}$$

where $\Delta^{\alpha\beta}_{ij} = E^{\alpha'}_i - E^{\alpha''}_i + E^{\beta'}_j - E^{\beta''}_j$ is an excitation energy related to a fermion transfer between sites from the initial states α', β' and to the final states α'', β'', respectively. Note that $s^{\alpha\beta[1]}_{ij,p}$ is the small parameter because $v \to 0$ and $\Delta^{\alpha\beta}_{ij}$ is large as the initial states and the final ones belong to the different energy subspaces C_q and C_{q+p} for $p \neq 0$.

The procedure of elimination of the nondiagonal terms from the transformed Hamiltonian of model (4), applied above for $N = 1$, can be extended by induction to an arbitrary order N of the perturbation calculation. The recurrent equation to determine $iS^{[N]}$ is obtained by applying the condition that all nondiagonal contributions in the component $h^{[N]}$ of $\tilde{H}$ vanish. Using $h^{[N]}$ given by Eq. (A5) the recurrent equation for $iS^{[N]}$ can be written as

$$\left[iS^{[N]}_p, W\right] = \underbrace{\sum_{\substack{(ij)\\ \alpha\beta}} \mathcal{J}^{\alpha\beta[N]}_{ij,p} X^\alpha_i X^\beta_j + \sum_{\substack{(ijk)\\ \alpha\beta\gamma}} \mathcal{J}^{\alpha\beta\gamma[N]}_{ijk,p} X^\alpha_i X^\beta_j X^\gamma_k + \cdots}_{N \text{ sums}}. \tag{12}$$

The parameters $\mathcal{J}^{\alpha\beta[N]}_{ij,p}$, $\mathcal{J}^{\alpha\beta\gamma[N]}_{ijk,p}$, and so on from Eq. (12) can be explicitly obtained by first doing all the commutators on the right-hand side of Eq. (A5) and next ordering the resulting sums according to the number of different sites involved in the products of the Hubbard operators. Using the commutation properties of the Hubbard operators, Eq. (8), the solution for $iS^{[N]}_p$ can be obtained in the form of the right-hand side of Eq. (12), with each parameter of n-site term $\mathcal{J}^{\alpha_1,\ldots,\alpha_n[N]}_{1,\ldots,n,p}$ (for all n: $2 \leqslant n \leqslant N+1$), replaced by

$$s^{\alpha_1,\ldots,\alpha_n[N]}_{1,\ldots,n,p} = \mathcal{J}^{\alpha_1,\ldots,\alpha_n[N]}_{1,\ldots,n,p} \Big/ \Delta^{\alpha_1,\ldots,\alpha_n}_{1,\ldots,n}, \tag{13}$$

and the denominator of the n-site term given by

$$\Delta^{\alpha_1,\ldots,\alpha_n}_{1,\ldots,n} = \sum_{i=1}^{n}\left(E^{\alpha'_i}_i - E^{\alpha''_i}_i\right). \tag{14}$$

As follows from the above derivations, to obtain the explicit form of the effective Hamiltonian to the order N it is sufficient to know the exact form of the generator iS up to order $N-1$. The Nth order contribution to iS is necessary to eliminate to nondiagonal terms in the transformed Hamiltonian and does not need to be explicitly known.

To write the second order effective Hamiltonian $\tilde{H}^{[2]}$ in a compact form, we need to compute the two-site commutators of the products of fermion-like Hubbard operators that appear in Eq. (6), with T_p and $S^{[1]}_p$ given by Eqs. (3) and (9). We have for $i \neq j$

$$\left[\overset{+}{F}{}^\mu_i F^\nu_j, \overset{+}{F}{}^\alpha_i F^\beta_j\right] = \sum_{\kappa\lambda} f^{\kappa\lambda}_{\alpha\mu,\beta\nu} \overset{+}{B}{}^\kappa_i B^\lambda_j, \tag{15}$$

and

$$\left[\overset{+}{F}{}^\mu_j F^\nu_i, \overset{+}{F}{}^\alpha_i F^\beta_j\right] = -\sum_{\kappa\lambda} f^{\kappa\lambda}_{\alpha\nu,\beta\mu} L^\kappa_i L^\lambda_j, \tag{16}$$

where the factor f is defined by

$$f^{\eta\xi}_{\mu\kappa,\beta\lambda} = \delta^\eta_{\mu\kappa}\delta^\xi_{\beta\lambda} - \delta^\eta_{\kappa\mu}\delta^\xi_{\lambda\beta}, \tag{17}$$

and $\delta^\alpha_{\beta\gamma} = \delta_{\alpha'\gamma'}\delta_{\alpha''\beta''}\delta_{\beta'\gamma''}$. One needs also to know three-site commutators, for example,

$$\left[\overset{+}{F}{}^\mu_i F^\nu_n, \overset{+}{F}{}^\alpha_i F^\beta_j\right] = \sum_\lambda g^\lambda_{\mu\alpha} \overset{+}{B}{}^\lambda_i F^\beta_j F^\nu_n, \tag{18}$$

where $g^{\lambda}_{\mu\alpha} = \delta^{\lambda}_{\mu\alpha} + \delta^{\lambda}_{\alpha\mu}$. Inserting the computed commutators into Eq. (6) we finally obtain the effective second order Hamiltonian in the form

$$\begin{aligned}\tilde{H}^{[2]} = W + T_0 &- \sum_{\substack{(ij)\\ \kappa\lambda}} J^{\kappa\lambda}_{ij} L^{\kappa}_i L^{\lambda}_j + \sum_{\substack{(ij)\\ \kappa\lambda}} \tilde{J}^{\kappa\lambda}_{ij} \overset{+}{B}{}^{\kappa}_i B^{\lambda}_j \\ &+ \sum_{\substack{(jim)\\ \lambda\beta\nu}} \left(I^{\lambda\beta\nu}_{ijm} \overset{+}{B}{}^{\lambda}_i F^{\beta}_j F^{\nu}_m + \text{H.c.} \right) \\ &- \sum_{\substack{(ijm)\\ \lambda\alpha\nu}} \left(K^{\lambda\alpha\nu}_{jim} L^{\lambda}_j \overset{+}{F}{}^{\alpha}_i F^{\nu}_m + \text{H.c.} \right). \end{aligned} \tag{19}$$

The parameters of $\tilde{H}^{[2]}$ read

$$\begin{aligned} J^{\kappa\lambda}_{ij} &= -\sum_{\substack{p,\alpha\beta\\ \mu\nu}} \frac{v^{\alpha\beta}_{ij,-p} v^{\mu\nu}_{ji,p}}{2\Delta^{\mu\nu}_{ji}} f^{\kappa\lambda}_{\alpha\nu,\beta\mu}, \\ \tilde{J}^{\kappa\lambda}_{ij} &= -\sum_{\substack{p,\alpha\beta\\ \mu\nu}} \frac{v^{\alpha\beta}_{ij,-p} v^{\mu\nu}_{ij,p}}{2\Delta^{\mu\nu}_{ij}} f^{\kappa\lambda}_{\alpha\mu,\beta\nu}, \\ I^{\lambda\beta\nu}_{ijm} &= -\sum_{p,\alpha\mu} \frac{v^{\alpha\beta}_{ij,-p} v^{\mu\nu}_{im,p}}{2\Delta^{\mu\nu}_{im}} g^{\lambda}_{\mu\alpha}, \\ K^{\lambda\alpha\nu}_{jim} &= -\sum_{p,\beta\mu} \frac{v^{\alpha\beta}_{ij,-p} v^{\mu\nu}_{jm,p}}{2\Delta^{\mu\nu}_{jm}} g^{\lambda}_{\mu\beta}. \end{aligned} \tag{20}$$

The transformed Hamiltonian $\tilde{H}^{[2]}$, Eq. (19), is a rather general one and may be applied for various specific problems. For instance, for a system of single orbital sites with a strong on-site repulsion the ground state subspace C_0 does not include states with double occupied sites (for the case of less than half-filled band). When we restrict ourselves to the subspace C_0 we can drop from Eq. (19) all the terms that include boson-like operators. The remaining part of the Hamiltonian reduces then to the $t-J$ model.[6] In this form it includes the local term W, the direct hopping term (T_0), the spin-spin Heisenberg interaction (the third term in $\tilde{H}^{[2]}$), and the three-site term of the correlated hopping (the last term in $\tilde{H}^{[2]}$).

In the opposite limit of the strong on-site attractive interaction (the negative Hubbard-U), the ground state subspace C_0 does not include states with single occupied sites and we can drop from $\tilde{H}$ all the terms that include fermion-like operators. Then first, third, and fourth terms of $\tilde{H}^{[2]}$ combined make a Hamiltonian of hard-core charged bosons on a lattice. When represented with pseudospins operators this model can be reduced to an isotropic Heisenberg Hamiltonian.[46]

In the systems with dominating on-site electron repulsions the terms including boson-like operators can usually be neglected while considering the lowest energy excitations or low temperature thermodynamics. In some multiorbital systems the stability of boson-like pairs in the low energy regime and the pair hopping (fourth term in $\tilde{H}^{[2]}$) may be supported by the Hund's exchange or by on-site interorbital attraction resulting from an electron-phonon coupling. Otherwise these terms describe the dynamics of high energy excitations which can be created with external electromagnetic fields. They may be also relevant for an analysis of electron transport through nanosystems when a bias voltage exceeds the energy separation of the eigenspaces C_0 and C_1.

Below we consider two particular examples of strongly interacting fermionic systems: the Hubbard model with a single orbital and with two-orbitals per site in a lattice with defects to illustrate the method in detail.

III. HUBBARD MODEL WITH DEFECTS

The parameters of the effective Hamiltonian obtained above are site dependent and may describe situations where both the local orbital energy as well as the fermion repulsion vary across the lattice. This feature allows us to extend the results obtained for the models mentioned in the previous section to a lattice with strong local defects. As a case study we consider here the Hubbard model with the large on-site fermion repulsion. For an arbitrary number of fermions per site this model is often used to describe various strongly correlated metals, as well as systems of quantum dots and molecules. The effective Hamiltonian for a square lattice (and an arbitrary number of fermions) in the presence of defects was studied in the context of high-temperature superconductors.[47] We first reproduce this result in a more complete form including, often omitted, a three site term (Sec. III A). Next we consider a third order perturbation on triangular systems (i.e., triangular lattices or triple quantum dots) for the case of one fermion per site (Sec. III B). We show that unlike the homogeneous system, where the third order correction vanishes in zero magnetic flux, in the presence of local defects the third order correction is nonzero even in absence of the field.

A. Effective second order Hamiltonian

For the Hubbard model the orbital basis for a single site includes four eigenstates of the on-site Hamiltonian: $\{0, \uparrow, \downarrow, 2\}$ (i.e., the empty site, the single occupied site with spin up and down, and the double occupied site). The corresponding local energy eigenvalues depending on the occupation of the site i are $0, \epsilon_i, 2\epsilon_i + U_i$. Let us stress that in the considered model we take into account defects, and the site energy ϵ_i and on-site Coulomb interactions U_i are assumed to be site dependent. On the condition that the differences are not too large the spectrum of W can be partitioned into well separated eigenspaces C_q. The Hubbard operators can be written explicitly in terms of usual fermion operators as

$$\overset{+}{F}{}^{\sigma 0}_i = c^{\dagger}_{i,\sigma}(1 - n_{i,-\sigma}), \quad \overset{+}{F}{}^{2,-\sigma}_i = \sigma\, c^{\dagger}_{i,\sigma} n_{i,-\sigma}, \tag{21}$$

where $\sigma \in \{\uparrow, \downarrow\}$. The diagonal and off-diagonal components of the hopping part of the Hamiltonian read

$$\begin{aligned} T_0 &= \sum_{(ij)\sigma} t_{ij} \left(\overset{+}{F}{}^{\sigma 0}_i F^{0\sigma}_j + \overset{+}{F}{}^{2\sigma}_i F^{\sigma 2}_j \right), \\ T_{+1} &= \sum_{(ij)\sigma} \sigma t_{ij} \overset{+}{F}{}^{2-\sigma}_i F^{0\sigma}_j, \\ T_{-1} &= \sum_{(ij)\sigma} \sigma t_{ij} \overset{+}{F}{}^{\sigma 0}_i F^{-\sigma 2}_j. \end{aligned} \tag{22}$$

Here, the hopping parameters $t_{ij} = |t_{ij}| \exp(i\phi_{ij})$ are, in general, complex numbers, where ϕ_{ij} describes dependence on a magnetic flux. The Aharonov-Bohm effect can be seen in semiconducting nanostructure for magnetic fields of the order of a few Tesla.[48] For a lattice and molecules the phase shift ϕ_{ij} is negligible for realistic magnetic fields, because the enclosed flux in a unit cell is very small. However, in cuprates one expects a flux phase with circulating currents, where the phase ϕ_{ij} is related with an order parameter.[49]

We start our discussion of the second order effective Hamiltonian for the Hubbard model with the terms including the boson-like operators, $\overset{+}{B}{}_i^{20} = c_{i\uparrow}^\dagger c_{i\downarrow}^\dagger$ and $B_i^{02} = c_{i\downarrow} c_{i\uparrow}$. Using the hopping elements of Eq. (22) in Eq. (20) we obtain the pair hopping term

$$\sum_{\substack{(ij)\\ \kappa\lambda}} \tilde{J}_{ij}^{\kappa\lambda} \overset{+}{B}{}_i^{\kappa} B_j^{\lambda} \to \sum_{(ij)} t_{ij}^2 \left(\frac{1}{\Delta_{ij}} + \frac{1}{\Delta_{ji}} \right) \overset{+}{B}{}_i^{20} B_j^{02}, \tag{23}$$

where $\Delta_{ij} = U_j + \epsilon_j - \epsilon_i$. The three-site term, describing exchange between fermion and boson-like excitations can be represented as

$$\sum_{\substack{(jim)\\ \lambda\beta\nu}} I_{ijm}^{\lambda\beta\nu} \overset{+}{B}{}_i^{\lambda} F_j^{\beta} F_m^{\nu} + \text{H.c.}$$

$$\to \sum_{(jim)\sigma} t_{im} t_{ij} \left(\frac{1}{2\Delta_{im}} + \frac{1}{2\Delta_{ji}} \right) \overset{+}{B}{}_i^{20} F_j^{0\sigma} F_m^{\sigma 2} + \text{H.c.} \tag{24}$$

Equations (23) and (24) confirm that the both terms involving boson-like operators conserve the number of double occupied sites in the system (i.e., they do not mix different subspaces C_q) in accordance with the method of derivation of $\tilde{H}$. These terms give nonzero result only in the eigenspaces of W that include states with double occupied sites, and they can be omitted for considerations of low-energy physics of the Hubbard model with the strong on-site repulsion since the double occupied sites are excluded from C_0. For the latter case we are left with the effective second order Hamiltonian in the form

$$\begin{aligned} \tilde{H}^{[2]} = & \sum_{i\sigma} \epsilon_i L_i^{\sigma\sigma} + \sum_{(ij)\sigma} t_{ij} \overset{+}{F}{}_i^{\sigma 0} F_j^{0\sigma} \\ & + \frac{1}{2} \sum_{\substack{(ij)\\ \sigma\sigma'}} \sigma\sigma' J_{ij}^{(2)} L_i^{-\sigma\sigma'} L_j^{\sigma,-\sigma'} \\ & + \frac{1}{2} \sum_{\substack{(ijk)\\ \sigma\sigma'}} \sigma\sigma' \left(J_{ijk}^{(2)} L_j^{-\sigma\sigma'} \overset{+}{F}{}_i^{\sigma 0} F_k^{0,-\sigma'} + \text{H.c.} \right), \end{aligned} \tag{25}$$

where

$$\begin{aligned} J_{ij}^{(2)} &= 2|t_{ij}|^2 \left(\frac{1}{\Delta_{ji}} + \frac{1}{\Delta_{ij}} \right), \\ J_{ijk}^{(2)} &= 2 t_{ij} t_{jk} \left(\frac{1}{\Delta_{ij}} + \frac{1}{\Delta_{kj}} \right). \end{aligned} \tag{26}$$

The effective Hamiltonian can be rewritten in a more familiar form with a help of the spin and charge operators for sites and bonds, projected to the subspace C_0

$$\begin{aligned} \vec{S}_{jk} &= \frac{1}{2} \sum_{\sigma} (c_{j\sigma}^\dagger c_{k,-\sigma}, \; -i\sigma \, c_{j\sigma}^\dagger c_{k,-\sigma}, \sigma \, c_{j\sigma}^\dagger c_{k\sigma}), \\ \rho_{jk} &= \sum_{\sigma} c_{j\sigma}^\dagger c_{k\sigma}. \end{aligned} \tag{27}$$

The on-site spin and density operators are given by $\vec{S}_j = \vec{S}_{jj}$ and $\rho_j = \rho_{jj}$. Representing the fermion operators (27) by the Hubbard operators, projecting the result to C_0 and using it in Eq. (25) we obtain the effective Hamiltonian in the explicitly spin-rotation invariant form, valid in the subspace C_0

$$\begin{aligned} \tilde{H}^{[2]} = & (W + T_0)|_{C_0} + \sum_{(ij)} J_{ij}^{(2)} \left(\vec{S}_i \cdot \vec{S}_j - \frac{1}{4} \rho_i \rho_j \right) \\ & + 2 \sum_{(ijk)} \text{Re}\left(J_{ijk}^{(2)} \right) \left(\vec{S}_j \cdot \vec{S}_{ik} - \frac{1}{4} \rho_j \rho_{ik} \right). \end{aligned} \tag{28}$$

The resulting Hamiltonian (28) is valid for an arbitrary lattice and it corresponds to the well-known t-J model.[5–7] Its new feature is the fact that two site and three site exchange parameters, $J_{ij}^{(2)}$ and $J_{ijk}^{(2)}$, depend here explicitly on the local values of Coulomb repulsion as well as the local orbital energy through Eq. (26). Note, that the Peierls factors ϕ_{ij} cancel in $J_{ij}^{(2)}$, whereas the three site parameter $J_{ijk}^{(2)}$ depends on the magnetic flux.

B. Third order effective Hamiltonian for triangular systems

Here we consider a third order contribution to the effective Hamiltonian for the triangular system, restricting ourselves to the ground state subspace C_0 with all sites singly occupied. When the site occupancy is constant in the unperturbed ground state the effective Hamiltonian projected to the subspace C_0 can be fully written in terms of the SB operators because all the terms that include the boson and the fermion-like Hubbard operators disappear from the projection. The third order contribution including only the SB operators takes a form

$$h^{[3]} = \sum_{\substack{(imj)\\ \alpha\beta\gamma}} J_{imj}^{\alpha\beta\gamma} \, L_i^{\alpha} L_m^{\beta} L_j^{\gamma}. \tag{29}$$

Here we assume that the eigenspace of W is divided into two eigenspaces: C_0 and C_1. To get the expressions for the parameters of $h^{[3]}$ we first need to compute the second order contribution $iS^{[2]}$ to the generator iS, which is given for an arbitrary N by Eqs. (12) and (13) and inserting it to Eq. (A11) we arrive after some algebra to the following result:

$$J_{imj}^{\alpha\beta\gamma} = \sum_{\substack{\nu\xi\eta\\ \mu\kappa\lambda\\ p=\pm 1}} \frac{v_{mj,-p}^{\kappa\lambda}}{2\Delta_{ijm}^{\alpha\eta\mu}} \left(\frac{v_{ji,p}^{\eta\nu} v_{im,0}^{\xi\mu}}{\Delta_{ji}^{\eta\nu}} - \frac{v_{ji,0}^{\eta\nu} v_{im,p}^{\xi\mu}}{\Delta_{im}^{\xi\mu}} \right) g_{\nu\xi}^{\alpha} f_{\mu\kappa,\eta\lambda}^{\beta\gamma}. \tag{30}$$

For the present case of the single orbital Hubbard model with one fermion per site the SB operators can be replaced by the spin operators.

To simplify somewhat tedious algebra we first define a matrix

$$\mathcal{L}(S_i) = \begin{bmatrix} \frac{1}{2} + S_i^z & S_i^+ \\ S_i^- & \frac{1}{2} - S_i^z \end{bmatrix}, \tag{31}$$

where $S_i^{\pm}$ are the usual spin lowering and raising operators for the site i. The matrix $\mathcal{L}(S_i)$ facilitates computations of sums over products of SB operators in C_0

$$\begin{aligned} &\sum_{\sigma\sigma'\sigma''} \sigma'\sigma''\, L_j^{-\sigma''-\sigma'} \left[L_m^{\sigma''\sigma}, L_i^{\sigma\sigma'}\right]_s \\ &\quad = \mathrm{tr}\{\, \mathcal{L}(-S_j)\, [\mathcal{L}(S_m), \mathcal{L}(S_i)\,]_s\,\} \\ &\quad = \delta_{s,-1} \left\{ (\vec{S}_j - \vec{S}_m - \vec{S}_i)^2 - \frac{1}{4} \right\} + 2i\, \delta_{s,1}\, \vec{S}_j \cdot (\vec{S}_m \times \vec{S}_i), \end{aligned} \tag{32}$$

where $s = 1$ holds for the commutator and $s = -1$ for the anticommutator of the matrices. Using Eqs. (30), (31), and (32) we obtain the third order contribution to $\tilde{H}$ in a form

$$\begin{aligned} h^{[3]}|_{C_0} = &\sum_{\{ij\}(m)} J_{ij(m)}^{(3)} \left(\vec{S}_i \cdot \vec{S}_j - \frac{1}{4} \right) \\ &+ \sum_{\{imj\}} J_{ijm}^{(3)}\, \vec{S}_i \cdot (\vec{S}_j \times \vec{S}_m). \end{aligned} \tag{33}$$

The summation in the first sum of Eq. (32) goes over two-element sets of sites $\{i,j\}$ and the sites (m) being the nearest neighbors to the both sites of the set. The second summation in Eq. (32) is over three element sets of different NN sites: $\{i,m,j\}$. The exchange parameters in Eq. (33) read

$$\begin{aligned} J_{ij(m)}^{(3)} = &(t_{ji} t_{im} t_{mj} + t_{ji}^* t_{im}^* t_{mj}^*) \\ &\times \left(\Delta_{ij}^{-1}\Delta_{mj}^{-1} + \Delta_{ji}^{-1}\Delta_{mi}^{-1} + \Delta_{mi}^{-1}\Delta_{mj}^{-1}\right. \\ &\left. -\Delta_{ji}^{-1}\Delta_{jm}^{-1} - \Delta_{ij}^{-1}\Delta_{im}^{-1} - \Delta_{im}^{-1}\Delta_{jm}^{-1}\right), \\ J_{ijm}^{(3)} = &-i(t_{ji} t_{im} t_{mj} - t_{ji}^* t_{im}^* t_{mj}^*) \\ &\times \left(\Delta_{ij}^{-1}\Delta_{mj}^{-1} + \Delta_{ji}^{-1}\Delta_{jm}^{-1} + \Delta_{ji}^{-1}\Delta_{mi}^{-1}\right. \\ &\left. +\Delta_{im}^{-1}\Delta_{ij}^{-1} + \Delta_{im}^{-1}\Delta_{jm}^{-1} + \Delta_{mi}^{-1}\Delta_{mj}^{-1}\right). \end{aligned} \tag{34}$$

The parameter $J_{ijm}^{(3)}$ of the three site term is nonzero only in the presence of the magnetic flux. The corresponding (second) term in Eq. (33) describes spin chirality which can be related with circulating persistent currents. Our Hamiltonian is an extension of the results obtained by other authors[10,13,33] to the case of the lattice with defects. For example, one can describe influence of an electric field which breaks the symmetry of the system and changes local electrical polarization as well as spin configuration.

The parameter $J_{ij(m)}^{(3)}$ of the two-site term accounts for modification of the usual nearest neighbor Heisenberg exchange interaction due to defects on sites which are nearest neighbors to both sites of the bond. It vanishes for the uniform case (i.e., when U_i and ϵ_i do not depend on site index). For small ϵ_i (i.e., for $\epsilon_i \ll U$), one gets from Eq. (34) an approximate formula for $J_{ij(m)}^{(3)}$

$$J_{ij(m)}^{(3)} \approx \frac{8\,\mathrm{Re}(t_{ji} t_{im} t_{mj})}{U^3} (2\epsilon_m - \epsilon_i - \epsilon_j). \tag{35}$$

We conclude that for the small site energy modulation the third order term depends linearly on the site energy parameters. This is in contrast to the second order term which depends on the second power of site energy differences [cf. Eq.(26)]. Applying the method to a triangular system in an electric field one can see a linear and quadratic Stark effect in spin-spin correlations.[50]

IV. TWO-ORBITAL MODEL

In this section we exemplify the application of the presented perturbation procedure to a simple model with more than one orbital per site. We start with the Hubbard Hamiltonian extended to the multi-orbital case

$$\begin{aligned} H = &\sum_{\substack{ia,jb \\ (i\neq j)\sigma}} t_{ia,jb}\, c_{ia\sigma}^\dagger\, c_{jb\sigma} + \sum_{ia\sigma} \varepsilon_{ia}\, n_{ia\sigma} \\ &+ \sum_{\substack{ia\sigma \\ jb\sigma'}} \left(1 - \delta_{ij}\delta_{ab}\delta_{\sigma\sigma'}\right) U_{ia,jb}\, n_{ia\sigma}\, n_{jb\sigma'}. \end{aligned} \tag{36}$$

The first term in the above equation represents electron hopping between the sites i,j and the orbitals a,b, the second term accounts for the orbital energy, and the last term describes Coulomb interaction between electrons located on the sites i,j and the orbitals a,b. Depending on the model parameters, such the Hamiltonian can be used as a generic model to describe very different systems from transition metal compounds, like superconducting copper oxides,[51] to systems of multiorbital quantum dots.[52] The model is different from the degenerate Hubbard model[44] in that we did not include Hund's coupling and in our case orbitals are not degenerate.

For simplicity we consider the above Hamiltonian for the special case of two nearly degenerate orbitals per site and one electron per site. Such the model can describe the system of quantum dots recently analyzed in a semiconducting carbon nanotube.[53] We split the model Hamiltonian into the unperturbed part and the perturbation as follows:

$$\begin{aligned} W = &\varepsilon \sum_{ia\sigma} n_{ia\sigma} + \sum_{ia} U_{aa}\, n_{ia\uparrow}\, n_{ia\downarrow} \\ &+ \frac{1}{2} \sum_{\substack{iab(a\neq b) \\ \sigma\sigma'}} U_{ab}\, n_{ia\sigma}\, n_{ib\sigma'}, \\ T = &\sum_{\substack{ija\sigma \\ (i\neq j)}} t_{ia,ja}\, c_{ia\sigma}^\dagger\, c_{ja\sigma} + \sum_{ia\sigma} \Delta\varepsilon_{ia}\, n_{ia\sigma} \\ &+ \frac{1}{2} \sum_{\substack{ijab \\ (i\neq j)\sigma\sigma'}} V_{ia,jb}\, n_{ia\sigma}\, n_{jb\sigma'}. \end{aligned} \tag{37}$$

The unperturbed Hamiltonian W includes the on-site intraorbital and the interorbital Coulomb repulsion parameters, U_{aa} and U_{ab}, respectively, and $a,b \in \{A,B\}$, where A,B denote the orbitals. The perturbation T includes the hopping parameter $t_{ia,ja}$, the energy difference $\Delta\varepsilon_i$ between the orbitals A,B, and the Coulomb repulsion $V_{ia,jb}$ for electrons on different sites.

For the strong on-site repulsions and one electron per site, the ground state subspace C_0 includes only the states with single occupied sites: $|a^\sigma\rangle$, where $\sigma = \uparrow, \downarrow$ denotes the spin and $a = A$ or B. Due to the orbital and spin degeneracy the ground state space C_0 is 4^{N_S}-fold degenerate for N_S sites in the system. The excited eigenspace C_1 includes the empty sites, the double occupied sites ($|a^\uparrow a^\downarrow\rangle$, $|a^\sigma b^\zeta\rangle$, where $a \neq b$ and $\zeta = \uparrow, \downarrow$), the triple occupied sites ($|a^\uparrow a^\downarrow b^\sigma\rangle$, where: $a \neq b$), and the full sites ($|A^\uparrow A^\downarrow B^\uparrow B^\downarrow\rangle$). The ground state space C_0 is separated from the subspace C_1 of excited states with an energy gap: Δ_{min}, being the lowest of the intrasite electron repulsion parameters. We assume that the formal criterion for the convergence of a perturbation calculation is fulfilled (i.e., the parameters of T are much smaller than Δ_{min}). Our aim here is to derive the second order effective Hamiltonian for the model (37), in the lowest energy subspace C_0.

Following the general procedure described above we first rewrite the perturbation T in terms of the Hubbard operators, and next split this term into diagonal and nondiagonal contributions, as in Eq. (3). In the result we obtain the relevant parameters of the perturbation, viz.

$$\begin{aligned} v_{ij,+1}^{(A^\sigma B^\zeta,\, B^\zeta)(0,\, A^\sigma)} &= \quad t_{iA,jA}, \\ v_{ij,+1}^{(A^\uparrow A^\downarrow,\, A^{-\sigma})(0,\, A^\sigma)} &= \sigma\, t_{iA,jA}, \\ v_{ij,+1}^{(A^\zeta B^\sigma,\, A^\zeta)(0,\, B^\sigma)} &= -\, t_{iB,jB}, \\ v_{ij,+1}^{(B^\uparrow B^\downarrow,\, B^{-\sigma})(0,\, B^\sigma)} &= \sigma\, t_{iB,jB}, \end{aligned} \tag{38}$$

for $p = +1$ and in a similar way for $p = -1$. The upper indices of the parameters $v_{jm,+1}^{\alpha\beta}$ in Eq. (38) refer to actions of the corresponding products of Fermi-like operators transferring between the site states. We note that beyond the parameters $v_{jm,+1}^{\alpha\beta}$ listed in Eq. (38) there are other parameters of the perturbation T, which are, however, not important for the derivation of the effective second order Hamiltonian with the above defined ground state subspace C_0.

Since the subspace C_0 includes only the states with singly occupied sites, all the contributions of $h^{[2]}$ from Eq. (19) with the boson-like or fermion-like Hubbard operators can be neglected in the present case. In the result our second order contribution to the effective Hamiltonian reduces to the third term from the right-hand side of Eq. (19). As follows from the number of the relevant parameters from Eq. (38) and two possible values of $p = \pm 1$, the parameter $J_{ij}^{\kappa\lambda}$ of $h^{[2]}$, given by Eq. (20), includes $2 \times 4 \times 4 = 32$ possible contributions [some of them vanishing due to the values of the numerical factors defined in Eq. (17)]. The obtained effective Hamiltonian projected to the subspace C_0 takes the form

$$\begin{aligned} \tilde{H}^{[2]} = &- \sum_{ija,\sigma\zeta} \sigma\zeta \left(\frac{t_{ia,ja}\, t_{ja,ia}}{U_{aa}} \right) L_i^{a^\sigma,a^\zeta}\, L_j^{a^{-\sigma},a^{-\zeta}} \\ &- \sum_{\substack{ij\sigma\zeta \\ ab(a\neq b)}} \left(\frac{t_{ia,ja}\, t_{ja,ia}}{2U_{ab}} \right) L_i^{a^\sigma,a^\sigma}\, L_j^{b^\zeta,b^\zeta} \\ &+ \sum_{\substack{ij\sigma\zeta \\ ab(a\neq b)}} \left(\frac{t_{ia,ja}\, t_{jb,ib}}{2U_{ab}} \right) L_i^{a^\sigma,b^\zeta}\, L_j^{b^\zeta,a^\sigma} + h^{[1]}. \end{aligned} \tag{39}$$

Above, the term $h^{[1]}$ includes the last two contributions to the perturbation Hamiltonian from Eq. (37), due to the intersite Coulomb interactions and the energy difference between the orbitals A,B, and written in terms of the Hubbard operators.

The obtained effective Hamiltonian for the two-orbital model includes two types of terms. The first term corresponds to the usual kinetic exchange: It describes the effective Heisenberg-like spin-spin interaction for two sites being in the same orbital state a. The next two terms on the right-hand side of Eq. (39) can be called an orbital exchange. These term contribute to the energy of the system only if the two sites are in the different orbital state. The effective Hamiltonian can be also written in an alternative way using the spin operators as well as the corresponding orbital pseudospin operators and indeed such the form is more commonly used in considering orbital ordering in solids (see, e.g., Eq. (2.8) in Ref. 44).

For the sake of interest in mesoscopic physics it is instructive to discuss shortly exact solution of the Hamiltonian (39) for a system of two multiorbital quantum dots. We find that depending on the model parameters there are three possible ground states of the model. The first two states are spin singlets, driven by the kinetic exchange term. In such the states the both dots are in the same, either A or B, orbital state depending on the values of the hopping $t_{ia,ja}$ and the Coulomb energy parameters U_{aa}. The third possible lowest energy solution is the either odd or even superposition (depending on the sign of the hopping parameters) of different orbital states on the two dots. This state is four-fold degenerate with respect to the possible spin values and we call it a state orbital singlet for short. One can also note that the spin degeneracy of the orbital singlet can be removed if one includes a small interorbital hopping. The orbital singlet will be favored over the spin singlets if $\min_a(U_{aa}) > U_{AB}$. A similar influence on the relative stability of the spin and orbital singlets have also intersite repulsion, present in $h^{[1]}$, whereas the energy difference between the orbitals $\Delta\varepsilon$ favors the spin singlet over the orbital singlet.

V. SUMMARY AND CONCLUSION

In this paper we applied the perturbation method defined with the help of the canonical transformation to a system of strongly interacting fermions. The considered system can be viewed as a collection of multiorbital units: atoms, molecules, lattice sites, or quantum dots. The model Hamiltonian is

generally defined with the help of the Hubbard operators and consists of two parts. The unperturbed part being the sum of the single site terms is diagonal in many-body site states, and its spectrum is assumed to split into well separated bands. The intersite part of the Hamiltonian, describing hopping of fermions, was treated as a perturbation.

Using the properties of the Hubbard operators we presented a recursive procedure of elimination of the interband terms from the canonically transformed Hamiltonian for an arbitrary order of the perturbation calculation. Explicit derivations were performed for the effective second order Hamiltonian, which was represented in a new and compact form in terms of the Hubbard operators. The obtained effective model can be used both to study the low energy excitations in the system as well as the dynamics of energy excitations in the high energy bands. We also carried out the procedure to the third order of perturbation and obtained the effective Hamiltonian for the special case of uniform occupancy of the sites in the ground state in the triangular systems.

The efficiency of the presented perturbational procedure was exemplified in more detail for two specific cases. The first one concerned with the single orbital Hubbard model in the presence of imperfections and the flux of the magnetic field. In this case we obtained the second order effective Hamiltonian for an arbitrary lattice and arbitrary number of fermions per site. For the triangular systems with the uniform occupation of the sites in the ground state, we applied the general effective third order Hamiltonian to obtain some new results for the exchange couplings of the resulting spin model. The second case is concerned with an extension of the Hubbard model to a problem of two nondegenerate orbitals per site. For this model we obtained the effective second order Hamiltonian and we discussed its various possible solutions for the two site system.

The used form of the hopping term, Eq. (3), is universal enough to describe processes of electron transfer in very different materials. Aside from a typical one particle hopping in the narrow band systems[41,44] it can be applied to a correlated hopping in molecular crystals,[13,34] where the hopping rate depends on the electron occupation of the sites involved. The derived effective Hamiltonians can be useful for analysis of a complex magnetic and electric local polarization in multiferroics.[32,33] Moreover, it can be applied to nanostructures (e.g., coherently coupled quantum dots or molecules[25–28]) to study many-body physics out of equilibrium. Since the hopping Hamiltonian is not limited here to spin conserving processes it can also describe effects of a spin-orbit coupling due to an external electric field in molecular magnets.[30]

Finally we showed (in Appendix B) that the recursive formulation of the canonical perturbation theory can be extended to include the intersite density-density interactions into the model. The intersite term can be treated either as a perturbation or they can be included into the unperturbed part of the Hamiltonian, depending on the relative strength of these interactions in a system under consideration. This extension can be important for studying systems where the intersite Coulomb interactions are strong enough so that they can lead to appearance of nonuniform phases (i.e., various types of charge ordered states or phase separations).

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APPENDIX A: CANONICAL TRANSFORMATION

The idea of the recursive formulation of the canonical perturbation theory consists in eliminating the nondiagonal terms from the Hamiltonian order by order by means of a unitary transformation. The transformed Hamiltonian reads

$$\tilde{H} = \mathrm{e}^{iS} H \mathrm{e}^{-iS} = H + [iS, H] + \frac{1}{2!}\big[iS, [iS, H]\big] + \cdots . \tag{A1}$$

We assume that the generator S of the canonical transformation is expandable in powers of τ

$$iS = \sum_{N=1} iS^{[N]} \tau^N. \tag{A2}$$

Using Eq. (A2) in Eq. (A1) with $H = W + \tau T$, we obtain the expansion of the transformed Hamiltonian in powers of τ,

$$\tilde{H} = W + \sum_{N \geqslant 1} h^{[N]} \tau^N. \tag{A3}$$

where the first three terms are given by

$$\begin{aligned}
h^{[1]} &= [iS^{[1]}, W] + T, \\
h^{[2]} &= [iS^{[2]}, W] + [iS^{[1]}, T] + \frac{1}{2!}\big[iS^{[1]}, [iS^{[1]}, W]\big], \\
h^{[3]} &= [iS^{[3]}, W] + [iS^{[2]}, T] + \frac{1}{2!}\big[iS^{[1]}, [iS^{[2]}, W]\big] \\
&\quad + \frac{1}{2!}\big[iS^{[2]}, [iS^{[1]}, W]\big] + \frac{1}{2!}\big[iS^{[1]}, [iS^{[1]}, T]\big] \\
&\quad + \frac{1}{3!}\big[iS^{[1]}, \big[iS^{[1]}, [iS^{[1]}, W]\big]\big].
\end{aligned} \tag{A4}$$

The general form of the term of the series (A3) reads

$$\begin{aligned}
h^{[N]} &= [iS^{[N]}, W] + [iS^{[N-1]}, T] + \cdots \\
&\quad + \frac{1}{N!}[iS^{[1]}, \big[iS^{[1]}, \ldots, [iS^{[1]}, W]\big] \ldots], \text{ for: } N > 1.
\end{aligned} \tag{A5}$$

Now we require that the nondiagonal terms in the expansion (A3) vanish (i.e., that $h_p^{[N]} \equiv \sum_q P_{p+q} h^{[N]} P_q = \delta_{p,0} h_0^{[N]}$). This condition, together with Eq. (A5), makes the procedure of determining of the generator iS a recursive one. In particular, for the first order term we obtain

$$h_p^{[1]} = \big[iS_p^{[1]}, W\big] + T_p. \tag{A6}$$

From Eq. (A6) one gets the condition for the nondiagonal part of $iS^{[1]}$

$$\big[iS_p^{[1]}, W\big] = -T_p, \text{ for: } p \neq 0. \tag{A7}$$

The diagonal part of iS does not modify the nondiagonal contribution to $\tilde{H}$ and it is set to zero,[4] hence $h^{[1]} = T_0$. The second order contribution to $\tilde{H}$ reads

$$h_p^{[2]} = \left[iS_p^{[2]}, W\right] + \sum_r \left[iS_{p-r}^{[1]}, T_r\right] + \frac{1}{2!}\sum_r \left[iS_{p-r}^{[1]}, \left[iS_r^{[1]}, W\right]\right]. \tag{A8}$$

Using Eqs. (A7) and (A8) and the requirement of vanishing of the nondiagonal part in $h^{[2]}$ we obtain a condition to determine $S_p^{[2]}$,

$$\left[iS_p^{[2]}, W\right] = -\frac{1}{2}\sum_r \left[iS_{p-r}^{[1]}, T_r\right](1 + \delta_{r,0}), \tag{A9}$$

for $p \neq 0$. Equation (A9) leads to the second order contribution to $\tilde{H}$ in an explicitly diagonal form

$$h^{[2]} = \frac{1}{2}\sum_p \left[iS_{-p}^{[1]}, T_p\right]. \tag{A10}$$

For the third order term in $\tilde{H}$ we obtain in a similar way

$$h^{[3]} = \frac{1}{2}\sum_p \left[iS_{-p}^{[2]}, T_p\right] + \frac{1}{12}\sum_{pq(q\neq 0)} \left[iS_{-p}^{[1]}, \left[iS_{p-q}^{[1]}, T_q\right]\right]. \tag{A11}$$

The procedure can be effectively carried on to an arbitrary high order of the perturbation theory provided that one can find explicitly nondiagonal components of the generator iS from the recursive equations, like Eqs. (A7) and (A9). In general one has to compute the generator up to order $N-1$ to get the transformed Hamiltonian to order N. Note that in the present derivation we allow for arbitrary integer values of the interband index p in T_p, unlike the case of the single band Hubbard model, where $p = 0, \pm 1$ (Ref. 8). Such the extension may be necessary in application of the method for multiorbital models and for unperturbed Hamiltonians including intersite terms (see Appendix B).

APPENDIX B: CANONICAL PERTURBATION METHOD FOR MODELS WITH INTERSITE COULOMB INTERACTIONS

In what follows we show how the canonical perturbation method can be extended to treat models with intersite couplings, in particular density-density and spin-spin interactions. On the assumption that these terms do not mix different subspaces of the unperturbed Hamiltonian, they can be taken into account by a modification of the diagonal term of the perturbation T_0, viz.,

$$T_0 \to \sum_{\substack{(ij)\\ \alpha,\beta}} v_{ij,0}^{\alpha\beta} \overset{+}{F}{}_i^{\alpha} F_j^{\beta} + \sum_{\substack{(ij)\\ \alpha,\beta}} w_{ij}^{\alpha\beta} L_i^{\alpha} L_j^{\beta}. \tag{B1}$$

The second order effective Hamiltonian obtained in Sec. II B can be easily generalized to include the extra term, because it enters directly the first order contribution $h^{[1]}$ to $\tilde{H}$ (see Appendix A), whereas it does not modify $h^{[2]}$.

A principal criterion for convergence of a perturbation expansion is smallness of parameters of a perturbation part of a Hamiltonian with respect to energy of excitations of an unperturbed part. In the present case it can be written formally as

$$\max_{ij\alpha\beta,p} \left|v_{ij,p}^{\alpha\beta}/\Delta_{\min}\right| \ll 1, \quad \max_{ij\alpha\beta} \left|w_{ij}^{\alpha\beta}/\Delta_{\min}\right| \ll 1, \tag{B2}$$

where $\Delta_{\min}$ is the lowest energy separation between the different subspaces C_q of the unperturbed Hamiltonian. First of the above conditions can be well satisfied in the narrow band systems, however, the second one may be difficult to meet in the case of the intersite density-density Coulomb interactions. In the latter case one can reformulate the perturbation expansion by moving the intersite Coulomb term to the unperturbed Hamiltonian, for example,

$$W \to W = \sum_{i\alpha} E_i^{\alpha} L_i^{\alpha} \delta_{\alpha} + \sum_{\substack{(ij)\\ \alpha,\beta}} w_{ij}^{\alpha\beta} L_i^{\alpha} L_j^{\beta} \delta_{\alpha}\delta_{\beta}. \tag{B3}$$

Such a regrouping of the Coulomb interactions modifies the partition of the eigenspace of the unperturbed Hamiltonian into the subspaces C_q, corresponding to its various degenerate eigenvalues. Consequently, the resolution of the hopping Hamiltonian T into the interband terms T_p is also modified by the change of the unperturbed Hamiltonian. This can be exemplified in more detail by considering the spinless fermion model in one dimension

$$H = W + T = V\sum_i n_i\, n_{i+1} + t\sum_i (c_i^{\dagger} c_{i+1} + \text{H.c.}), \tag{B4}$$

where V denotes the intersite interaction and t is the hopping parameter. In this case the separation of T into the contributions T_p can be easily achieved with the help of projection operators $n_i^{\sigma} = \frac{1}{2}(1-\sigma) + \sigma\, n_i$ ($\sigma = \pm$), as follows:

$$T_p = t\sum_{i\sigma\sigma'} \left(n_{i-1}^{\sigma} c_i^{\dagger} c_{i+1}\, n_{i+2}^{\sigma'} + n_{i-1}^{\sigma'} c_{i+1}^{\dagger} c_i\, n_{i+2}^{\sigma}\right)\delta_{2p,\sigma-\sigma'}.$$

For the one-dimensional spinless fermion model the hopping term splits into three contributions, T_0 and $T_{\pm 1}$, and the unperturbed Hamiltonian has an equidistant spectrum. These features correspond to the ones of the Hubbard model on an arbitrary periodic lattice. In effect one can apply directly the general results of the perturbation expansion obtained by MacDonald *et al.*[8] to the spinless fermion model.

In the present case the hopping Hamiltonian can be cast into a form closely resembling the one used above, Eq. (3), if we introduce fermion-like operators by

$$\mathcal{F}_j^{\sigma\sigma'} = n_{j-1}^{\sigma} c_j\, n_{j+1}^{\sigma'}, \quad \overset{+}{\mathcal{F}}{}_j^{\sigma\sigma'} = n_{j-1}^{\sigma} c_j^{\dagger}\, n_{j+1}^{\sigma'}. \tag{B5}$$

In contrast to the previous case with the on-site unperturbed Hamiltonian, now we have the fermion-like operators (B5) as a product of three operators corresponding to the central site i

and its nearest neighbors. The above definition can be readily extended for the same model on an arbitrary d-dimensional lattice

$$\mathcal{F}_i^{(\sigma)} = c_i \prod_e n_{i+e}^{\sigma_e}, \tag{B6}$$

where the product goes over all vectors e connecting site i to its nearest neighbors and the vector index (σ) defined by $(\sigma) \equiv (\sigma_1,\sigma_2,\ldots)$ includes all σ indices assigned to the nearest neighbors of the central site i. Note, however, that for two- and three-dimensional lattices with each site having Z nearest neighbors, the hopping Hamiltonian is split into $2Z-1$ different contributions (i.e., the index p in T_p takes on the values: $-Z+1, -Z+2, \ldots, Z-2, Z-1$). Therefore, despite the fact that the unperturbed Hamiltonian in the d-dimensional lattice still has the equidistant spectrum, the formalism of the paper of MacDonald *et al.*[8] has to be extended to include more interband terms T_p.

Let us now consider a generalization of the definition of the fermion-like operator for a model with many orbital degrees of freedom per site and with the nearest neighbor density-density coupling included in the unperturbed Hamiltonian W as in Eq. (B3). A direct extension of the result (B6) for the present case reads

$$\mathcal{F}_i^{(\alpha)} = F_i^{\alpha_0} \prod_e L_{i+e}^{\alpha_e \delta_{\alpha_e}}, \tag{B7}$$

where $(\alpha)=(\alpha_0,\alpha_1,\ldots,)$ contains now vector indices α_j of the site i and all its nearest neighbors. The formulation of the canonical perturbation method presented in Appendix A is still valid with the hopping Hamiltonian redefined in terms of the new fermion-like operators. However, the specific form of the effective Hamiltonian is now more complicated than that in Sec. II B. In the present case the fermion-like operators (B7) include products of operators pertaining to a cluster of sites rather than a single site, and this modifies their commuting properties that determine the form of Eq. (19).

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