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A spin-flip extension of the Fulton–Gouterman transformation

Matthias Rapp and Max Wagner

Institut für Theoretische Physik, Universität Stuttgart, Pfaffenwaldring 57, 70550 Stuttgart, Germany

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Abstract. In the present paper an extended version of the Fulton–Gouterman transformation (FGT) is applied to an archetypical electron–phonon system which also involves electronic spin (the two-sites–two-spins–one-particle (221 model)). Two alternatives of the Fulton–Gouterman concept are given: one corresponding to the original transformation (non-exponential) and one to an exponential form. The latter appears to be interesting with regard to recent discussions of squeezing and anti-squeezing effects in superconductivity since it offers itself to a multi-particle generalization. Some remarks on the physical background are also made.

1. Introduction

For many years the theoretical treatment of coupled electron–phonon systems has been of great interest and recently it has regained new actuality in the fields of superconductivity and quantum diffusion. Among the multitude of methods and techniques which have been invoked to handle this problem one seems to be of particular importance since it allows for an exact diagonalization of the coupled system with respect to the electronic subspace. It thus reduces the problem to that of an effective dynamics in the vibrational subspace. This method was first introduced in the seminal paper of Fulton and Gouterman (FG) [1]. In this paper an electronic two-level system coupled to oscillators and which displays an inversion symmetry is considered. The authors introduced a unitary transformation (FGT) which exploits this symmetry in an optimal way such that an electronic diagonalization is achieved. For the details of this transformation we refer to the original paper [1] and also to a book by one of us [2]. The first to apply this method for a numerically accurate solution of the vibrational subproblem were Shore and Sander [3].

Although at a first glance the FGT seems to be somewhat exotic, since the effective Schrödinger-like equations in the vibrational subspace (FG equations (FGE)) embody reflection operators, it has the invaluable advantage of displaying, in a lucid topological way, the dominant antagonistic tendencies in the exact solution [4, 5]. This was also first noted by Shore and Sander [3] who have shown that the vibrational part of the ground-state wavefunction has the tendency, depending on the strength of the electronic transfer term, to disintegrate its structure into two parts. Physically, this amounts to a competition between the tendency to reduce the electronic mobility, which is established by the electron–phonon coupling term (polaron effect, Debye–Waller screening, etc), and the ‘propagative tendency’ in the form of a reflection established by the transfer term [6–9]. Later, another effect of these antagonistic tendencies was found for more elevated states (‘exotic states’ [7]), which provided the possibility of a semiquantitative understanding of the retarded luminescence

phenomenon [7, 8]. Recently, the ground state of the above-mentioned two-level system has been approached by use of a squeezing transformation [10], allowing a modified trial function of the vibrational part of the total wavefunction to be constructed, which may also be advantageously discussed within the FG frame.

The FG method can be generalized to all spatially symmetric systems [11] which are governed by an Abelian group and for which the electronic base constitutes a regular representation. In particular, this generalization has been applied to the translational group [11], e.g. to crystalline exciton-phonon systems [12]. In translationally invariant and cyclic systems the FGT amounts to a kind of 'discretized' Lee-Low-Pines transformation (LLPT), i.e. to a specification of the LLPT for one-band electron systems in the Wannier representation. More details will be given in a forthcoming paper.

For future applications it seems desirable, on the one hand, to employ the FG concept to one-electron-phonon systems containing spin-flip terms in their Hamiltonians. An example of this kind, a spin-flip extension of the original FG Hamiltonian, is considered in this paper. On the other hand, it would seem attractive to apply the method to multi-electron systems which makes it necessary to introduce FG-type transformations of an exponential form. This will be studied in future papers.

In the spin-extended version of the original FG problem considered in this paper we describe an electron with spin and an orbital two-state base which is coupled to a bath of harmonic oscillators. This model, which we call the two-sites-two-spins-one-particle model (the 221 model), is presented in section 2. For reference, the original FG problem is described briefly in section 3. In section 4 the sequence of unitary manipulations which eventually leads to a complete diagonalization of the 221-Hamiltonian with respect to the electronic subspace is presented. Since the 221-Hamiltonian displays an Abelian symmetry, it may also be electronically diagonalized by means of group theory. To achieve this we make use of results from a previous paper by one of us [11] in section 5. In section 6 we elaborate somewhat on the physical background and possible practical applications. Finally, results and perspectives are given in section 7.

In our presentation, the unitary transformation of an operator A and a wavefunction Ψ will respectively be denoted as

$$T : A \equiv \tilde{A} = U^\dagger A U \quad T : \Psi \equiv \tilde{\Psi} = U^\dagger \Psi \quad (1)$$

where U is a unitary operator, $U^\dagger = U^{-1}$, which will be written most often in the exponential form $U = \exp S$, where S is an anti-Hermitian operator, $S^\dagger = -S$. We emphasize that all operators are given as functions of the original basic dynamical variables of the coupled system. Combined transformations $T_b : T_a : A$ are understood as

$$T_b : T_a : A = U_b^\dagger U_a^\dagger A U_a U_b. \quad (2)$$

2. The two-sites-two-spins-one-particle model (221 model)

The 221 model is represented by the Hamiltonian

$$H = H_{\text{ph}} + A(Q) \sum_{\sigma=\uparrow,\downarrow} (c_{1\sigma}^\dagger c_{2\sigma} + c_{2\sigma}^\dagger c_{1\sigma}) + B(Q) \sum_{\sigma=\uparrow,\downarrow} (c_{2\sigma}^\dagger c_{2\sigma} - c_{1\sigma}^\dagger c_{1\sigma}) \\ + C(Q) \sum_{\rho \neq \sigma=\uparrow,\downarrow} (c_{1\rho}^\dagger c_{2\sigma} + c_{2\rho}^\dagger c_{1\sigma}) + D(Q) \sum_{\rho \neq \sigma=\uparrow,\downarrow} (c_{2\rho}^\dagger c_{2\sigma} - c_{1\rho}^\dagger c_{1\sigma}). \quad (3)$$

We assume the Hamiltonian to be invariant against the symmetry operations E (unity operator) and I (reflection operator in Cartesian space) of the reflection group $\{E, I\}$ and we introduce dynamical variables for the electronic and oscillatory subsystems respectively, which may be characterized with respect to the symmetry operations. The electronic creation and annihilation operators $\{c_{i\sigma}^\dagger, c_{i\sigma}, i = 1, 2; \sigma = \uparrow, \downarrow\}$ are assigned to the electronic base vectors $|i\sigma\rangle = c_{i\sigma}^\dagger|\text{vac}\rangle$ and display the property

$$\begin{aligned} I c_{1\sigma} &= c_{2\sigma} I & I |1\sigma\rangle &= |2\sigma\rangle I_Q \\ I c_{2\sigma} &= c_{1\sigma} I & I |2\sigma\rangle &= |1\sigma\rangle I_Q \end{aligned} \quad \text{and correspondingly} \quad (4)$$

The vibrational coordinates $Q_{m,p}$ and $P_{m,p}$ are chosen as 'symmetry coordinates', i.e. they are related to a specific parity p ($p = 1$ for odd, $p = 2$ for even), such that

$$I Q_{m,p} = (-1)^p Q_{m,p} I \quad (5)$$

where $m = 1, \dots, M_p$ (M_p : multiplicity of representation $\Gamma^{(p)}$). The phonon Hamiltonian reads

$$H_{\text{ph}} = \frac{1}{2} \sum_{m,p} (P_{m,p}^\dagger P_{m,p} + \Omega_{m,p}^2 Q_{m,p}^\dagger Q_{m,p}). \quad (6)$$

The quantities $A(Q), B(Q), C(Q)$ and $D(Q)$ are assumed to depend on the oscillatory coordinates $\{Q_{m,p}\}$ only; a generalization to momentum coordinates is straightforward. Invariance of H with respect to $\{E, I\}$ requires

$$\begin{aligned} I A(Q) &= A(Q) I & I B(Q) &= -B(Q) I \\ I C(Q) &= C(Q) I & I D(Q) &= -D(Q) I \end{aligned} \quad (7)$$

The reflection operator I has the property

$$I^2 = 1 \quad (8)$$

and may be written as

$$I = I_{\text{el}} \cdot I_Q \quad (9)$$

where I_{el} and I_Q pertain to the electronic and vibrational subsystems, respectively. The reflection operator I_Q in $\{Q, P\}$ -space may be written in the form

$$I_Q = \exp \left[-i\pi \sum_{m(p=1)} (P_{m,p}^\dagger P_{m,p} + \Omega_{m,p}^2 Q_{m,p}^\dagger Q_{m,p} - \frac{1}{2}) \right] \quad (10)$$

and has the property

$$I_Q f(Q_{m,p}) = f((-1)^p Q_{m,p}). \quad (11)$$

In addition we introduce the spin-flip operator I_σ

$$\begin{aligned} I_\sigma c_{i\uparrow} &= c_{i\downarrow} I_\sigma & I_\sigma |i\uparrow\rangle &= |i\downarrow\rangle I_\sigma \\ I_\sigma c_{i\downarrow} &= c_{i\uparrow} I_\sigma & I_\sigma |i\downarrow\rangle &= |i\uparrow\rangle I_\sigma \end{aligned} \quad \text{and correspondingly} \quad (12)$$

and a 'spin-flip parity' $p_\sigma = 1, 2$ for this operator, such that for a spin-parity-ordered function we have

$$I_\sigma |\Psi^{(p_\sigma)}\rangle = (-1)^{p_\sigma} |\Psi^{(p_\sigma)}\rangle. \quad (13)$$

The restriction to one particle leads to the particle conservation law

$$\sum_{i=1}^2 (c_{i\uparrow}^\dagger c_{i\uparrow} + c_{i\downarrow}^\dagger c_{i\downarrow}) = 1. \quad (14)$$

3. Background: Fulton–Gouterman transformation (FGT)

For reference and lucidity we briefly describe the original FG problem which is a spinless model. It is represented by the Hamiltonian

$$H = H_{\text{ph}} + a(Q)(c_1^\dagger c_2 + c_2^\dagger c_1) + b(Q)(c_2^\dagger c_2 - c_1^\dagger c_1) \quad (15)$$

where H_{ph} is again of the form (6) and symmetry requires the relations

$$\begin{aligned} I c_1 = c_2 I & & I |1\rangle = |2\rangle I_Q \\ I c_2 = c_1 I & \text{and correspondingly} & I |2\rangle = |1\rangle I_Q \end{aligned} \quad (16)$$

and for the Q -dependent functions $a(Q), b(Q)$

$$I a(Q) = a(Q) I \quad I b(Q) = -b(Q) I. \quad (17)$$

Particle conservation (for one particle) reads

$$\sum_{i=1}^2 c_i^\dagger c_i = 1. \quad (18)$$

Hamiltonian (15) can be diagonalized with respect to the electronic problem by a unitary transformation, the so-called FGT (T_{FG}), which is characterized by the unitary operator

$$U_{\text{FG}} = \frac{(1 + I_Q)}{2\sqrt{2}}(c_1^\dagger c_2 + c_2^\dagger c_1 + c_2^\dagger c_2 - c_1^\dagger c_1) + \frac{(1 - I_Q)}{2\sqrt{2}}(1 + c_2^\dagger c_1 - c_1^\dagger c_2). \quad (19)$$

We emphasize at this point that the form (19) displays the unitarity property only, if we postulate a one-particle conservation law. This necessitates that an application of U_{FG} in the specific form (19) to non-particle-number-conserving operators like c_i , etc, is not permitted. For further details we refer to the book by one of us [2]. This restriction is not needed if we write U_{FG} in an exponential form, $U_{\text{FG}} = \exp[S_{\text{FG}}]$, $S^\dagger = -S$, where

$$S_{\text{FG}} = i \frac{\pi}{4\sqrt{2}}(1 + I_Q)(c_1^\dagger c_2 + c_2^\dagger c_1 + c_2^\dagger c_2 - c_1^\dagger c_1 - 1/\sqrt{2}) + \frac{\pi}{8}(1 - I_Q)(c_2^\dagger c_1 - c_1^\dagger c_2). \quad (20)$$

Under the one-particle provision this coincides with expression (19), but it may also be used for particle numbers which do not equal one. For later applications (e.g. in superconductivity) this will be a crucial point. The transformation of an operator A and of a wavefunction Ψ is defined in equation (1). Using this relation the transformed fundamental operators are found to be

$$\begin{aligned} T_{\text{FG}} : c_1 &= i \left(\frac{1 + I_Q}{2\sqrt{2}} \right) (c_2 - c_1) + \left(\frac{1 - I_Q}{2\sqrt{2}} \right) (c_1 - c_2) \\ T_{\text{FG}} : c_2 &= i \left(\frac{1 + I_Q}{2\sqrt{2}} \right) (c_1 + c_2) + \left(\frac{1 - I_Q}{2\sqrt{2}} \right) (c_1 + c_2) \\ T_{\text{FG}} : Q_{m,1} &= Q_{m,1}(c_2^\dagger c_1 + c_1^\dagger c_2) \\ T_{\text{FG}} : P_{m,1} &= P_{m,1}(c_2^\dagger c_1 + c_1^\dagger c_2) \\ T_{\text{FG}} : Q_{m,2} &= Q_{m,2} \\ T_{\text{FG}} : P_{m,2} &= P_{m,2} \end{aligned} \quad (21)$$

and the basic constituents of the wavefunctions transform like

$$\begin{aligned}
 T_{FG} : |1\rangle f(Q) &= \frac{1}{\sqrt{2}}(|2\rangle - I_Q|1\rangle) f(Q) \\
 T_{FG} : |2\rangle f(Q) &= \frac{1}{\sqrt{2}}(|2\rangle + I_Q|1\rangle) f(Q)
 \end{aligned}
 \tag{22}$$

where the ket-vectors $|p\rangle$ with $p = 1, 2$ are given by $|p\rangle = c_p^\dagger|\text{vac}\rangle$ and $f(Q)$ is any phonon function. The transformed Hamiltonian displays a diagonalized form in the electronic subspace

$$T_{FG} : H = \tilde{H}_{FG} = \sum_{p=1}^2 h_{FG}^{(p)}(Q) c_p^\dagger c_p
 \tag{23}$$

where

$$h_{FG}^{(p)}(Q) = H_{ph} + (-1)^p I_Q a(Q) + b(Q).
 \tag{24}$$

Hence the new (transformed) eigenfunctions have the form

$$\tilde{\Psi}_{FG}^{(p)} = |p\rangle \Phi^{(p)}(Q) \quad p = 1, 2
 \tag{25}$$

where the $\Phi^{(p)}$'s satisfy Schrödinger-like eigenvalue equations which we have called the FGE

$$h_{FG}^{(p)}(Q) \Phi_n^{(p)}(Q) = E_n^{(p)} \Phi_n^{(p)}(Q).
 \tag{26}$$

We receive the eigenfunctions $\Psi_{FG}^{(p)}$ of the original Hamiltonian (15) by transforming $\tilde{\Psi}_{FG}^{(p)}$ back to the original picture

$$\Psi_{FG}^{(p)} = U_{FG} \tilde{\Psi}_{FG}^{(p)} = \frac{1}{\sqrt{2}}[|2\rangle + (-1)^p |1\rangle I_Q] \Phi_n^{(p)}(Q)
 \tag{27}$$

such that

$$I \Psi_{FG}^{(p)} = (-1)^p \Psi_{FG}^{(p)}
 \tag{28}$$

with $p = 1, 2$ denoting odd and even parity, respectively. We stress that the phonon functions $\Phi^{(p)}(Q)$ attached to the even- and odd-parity total eigenfunctions $\Psi_{FG}^{(p)}$ do not themselves display the parity property in Q -space; this is only true for the full eigenfunctions $\Psi_{FG}^{(p)}$.

To avoid notational confusion it seems advisable at this stage to emphasize a basic peculiarity in the practical formulation of unitary transformations. In the original picture, the basic electronic operators of our system are related to some specific positions in Cartesian space, e.g. the operator c_1 or the electronic orbit $|1\rangle$ pertains to a specific geometric 'site 1' such that by reflection we have $Ic_1 = c_2I$ or $I|1\rangle = |2\rangle I_Q$ (see (16)). As observed in (21), the transformation T_{FG} transmutes c_1 into a superposition of c_1 and c_2 . From this we recognize, therefore, that in the transformed picture the respective indices 1 and 2 no longer pertain to well defined geometrical positions in the original Cartesian space. Rather, they

now adopt the quality of symmetry properties, index 1 referring to an odd- and index 2 referring to an even-parity state (see (27)).

One of the principal virtues of the FGE is the topological display of the conflicting tendencies inherent in most coupled electron-phonon systems. One of their terms, the $a(Q)$ term, is connected with the reflection operator and tends to separate the vibrational wavefunction into two parts in Q -space. This was first analysed numerically by Shore and Sander [3]. More generally this influence can be described as a tendency to enlarge the spatial extension of the vibrational wavefunction, which in recent times in the field of superconductivity and quantum diffusion has been approximated by a 'squeezed state' in P -space (i.e. an 'anti-squeezed' state in Q -space) [10]. But, as noted from the act of reflection, it is rather a tendency towards spatial separation within the vibrational functions $\Phi_m^{(\mu)}$ than a tendency towards broadening. The countereffect is represented by the $b(Q)$ terms and, in the most simple case, is a 'polaron' effect, i.e. a tendency to reduce the mobility of the electron, increasing its effective mass and dressing it with a phonon cloud.

In the aforementioned book [2] a list of alternatives to the FGT is given. The simplest one is discussed in [13]. It reads

$$U_{\text{MFG}} = \frac{1}{\sqrt{2}}[1 + I_Q(c_2^\dagger c_1 - c_1^\dagger c_2)] = \exp[S_{\text{MFG}}] \quad (29)$$

$$S_{\text{MFG}} = \frac{1}{4}\pi I_Q(c_2^\dagger c_1 - c_1^\dagger c_2). \quad (30)$$

Using equation (1) again we find the properties

$$\begin{aligned} T_{\text{MFG}} : c_1 &= \frac{1}{\sqrt{2}}(c_1 - I_Q c_2) \\ T_{\text{MFG}} : c_2 &= \frac{1}{\sqrt{2}}(c_2 + I_Q c_1) \\ T_{\text{MFG}} : Q_{m,1} &= Q_{m,1}(c_1^\dagger c_2 + c_2^\dagger c_1) \\ T_{\text{MFG}} : P_{m,1} &= P_{m,1}(c_1^\dagger c_2 + c_2^\dagger c_1) \\ T_{\text{MFG}} : Q_{m,2} &= Q_{m,2} \\ T_{\text{MFG}} : P_{m,2} &= P_{m,2} \\ T_{\text{MFG}} : |1\rangle f(Q) &= \frac{1}{\sqrt{2}}(|1\rangle + I_Q |2\rangle) f(Q) \\ T_{\text{MFG}} : |2\rangle f(Q) &= \frac{1}{\sqrt{2}}(|2\rangle - I_Q |1\rangle) f(Q). \end{aligned} \quad (31)$$

The transformed form of Hamiltonian (15) is

$$T_{\text{MFG}} : H = \sum_{j=1}^2 h_{\text{MFG}}^{(j)}(Q) c_j^\dagger c_j \quad (32)$$

where

$$h_{\text{MFG}}^{(j)}(Q) = H_{\text{ph}} + (-1)^j [-I_Q a(Q) + b(Q)]. \quad (33)$$

In the following discussion we will rely fully on this modified form of the FGT (MFGT) since the calculation proves much simpler than the one with the original form given by expression (44). Since $T_{\text{MFG}}: H$ is again diagonal with respect to the electronic problem, the new (transformed) eigenfunctions have a simple product form $\tilde{\Psi}_{\text{MFG}}^{(j)} = |j\rangle \hat{\Phi}^{(j)}(Q)$, $j = 1, 2$.

Again we obtain two Schrödinger-like eigenvalue equations for $\hat{\Phi}^{(j)}(Q)$ which we call the modified FG equations (MFGE)

$$h_{\text{MFG}}^{(j)}(Q) \hat{\Phi}_n^{(j)}(Q) = \hat{E}_n^{(j)} \hat{\Phi}_n^{(j)}(Q) \tag{34}$$

and the original eigenfunctions $\Psi_{\text{MFG}}^{(j)}$ are given by

$$\Psi_{\text{MFG}}^{(j)} = U_{\text{MFG}} \tilde{\Psi}_{\text{MFG}}^{(j)} = \frac{1}{\sqrt{2}} [|2\rangle - (-1)^j |1\rangle I_Q] (I_Q)^j \hat{\Phi}_n^{(j)}(Q). \tag{35}$$

Comparing these equations with (27) we note that $j = 1$ now pertains to even parity ($p = 2$) and $j = 2$ to odd parity ($p = 1$).

4. Intuitive step-by-step construction

Our aim is to diagonalize the 221-Hamiltonian of section 2 with respect to the electronic subsystem by means of a unitary transformation which preferably should be in the form either of a single exponential or of a product of exponentials since then it is appropriate for a multi-particle generalization. To achieve this we rely on a step-by-step procedure, where the first two steps are straightforward extensions of the MFGT (31). We first choose a transformation U_a which would diagonalize Hamiltonian (3) if there were no spin-flip terms, i.e. we just supplement transformation (29), (30) by spin indices

$$U_a = \prod_{\sigma=\uparrow,\downarrow} U_\sigma U_\sigma = \exp[S_\sigma] = \frac{1}{\sqrt{2}} [1 + I_Q (c_{2\sigma}^\dagger c_{1\sigma} - c_{1\sigma}^\dagger c_{2\sigma})] \tag{36}$$

$$S_\sigma = \frac{1}{4} \pi I_Q (c_{2\sigma}^\dagger c_{1\sigma} - c_{1\sigma}^\dagger c_{2\sigma}). \tag{37}$$

The basic transformation properties are again given by (31), if appropriate spin indication is supplemented, and the transformed Hamiltonian (3) reads

$$\begin{aligned} T_a : H = H_{\text{ph}} + [-A(Q) I_Q + B(Q)] \sum_{\sigma=\uparrow,\downarrow} (c_{2\sigma}^\dagger c_{2\sigma} - c_{1\sigma}^\dagger c_{1\sigma}) \\ + [-C(Q) I_Q + D(Q)] \sum_{\rho \neq \sigma=\uparrow,\downarrow} (c_{2\rho}^\dagger c_{2\rho} - c_{1\rho}^\dagger c_{1\rho}). \end{aligned} \tag{38}$$

A second transformation U_b may be found if we take notice of the fact that the spin-flip-operator I_σ introduced in section 2 plays a somewhat similar role to that of the reflection operator I_{el} for the electronic orbitals. This suggests a kind of FGT for the spins

$$U_b = \prod_{j=1,2} U_j \quad U_j = \exp[S_j] = \frac{1}{\sqrt{2}} [1 + I_Q (c_{j\uparrow}^\dagger c_{j\downarrow} - c_{j\downarrow}^\dagger c_{j\uparrow})] \tag{39}$$

$$S_j = \frac{1}{4} \pi I_Q (c_{j\uparrow}^\dagger c_{j\downarrow} - c_{j\downarrow}^\dagger c_{j\uparrow}). \tag{40}$$

In passing, we note that this transformation would diagonalize a Hamiltonian which would not involve orbital transfer terms $1 \leftrightarrow 2$, i.e. if the $A(Q)$ - and $C(Q)$ -terms were absent in (3). The basic transformation properties of (39), (40) are given in appendix B. If the transformations T_a and T_b are applied to Hamiltonian (3) in succession we find

$$\begin{aligned} T_b : T_a : H = & H_{\text{ph}} - A(Q)I_Q [c_{2\uparrow}^\dagger c_{2\uparrow} + c_{2\downarrow}^\dagger c_{2\downarrow} - c_{1\uparrow}^\dagger c_{1\uparrow} - c_{1\downarrow}^\dagger c_{1\downarrow}] \\ & + B(Q)I_Q [c_{2\uparrow}^\dagger c_{2\downarrow} - c_{2\downarrow}^\dagger c_{2\uparrow} - c_{1\uparrow}^\dagger c_{1\downarrow} + c_{1\downarrow}^\dagger c_{1\uparrow}] \\ & + C(Q) [c_{2\uparrow}^\dagger c_{2\uparrow} - c_{2\downarrow}^\dagger c_{2\downarrow} - c_{1\uparrow}^\dagger c_{1\uparrow} + c_{1\downarrow}^\dagger c_{1\downarrow}] \\ & + D(Q) [c_{2\uparrow}^\dagger c_{2\downarrow} + c_{2\downarrow}^\dagger c_{2\uparrow} - c_{1\uparrow}^\dagger c_{1\downarrow} - c_{1\downarrow}^\dagger c_{1\uparrow}] \end{aligned} \quad (41)$$

and electronic diagonalization is not yet achieved.

Correspondingly to section 3 we note here that the 'spin' indices in the transformed picture (after the application of T_b , T_c or T_t to the electronic operators) are no longer related to the spins themselves but to mixed spin states which can be attached to odd or even spin parity (see equation (51)). For the 'orbital' indices in the transformed picture (T_a or T_t) the remark made in section 3 directly applies again.

A final transformation T_c , which achieves this goal, is provided by

$$U_c = \exp[S_c] = \frac{(1+I_Q)}{2} \sum_{j=1}^2 (c_{j\uparrow}^\dagger c_{j\uparrow} - c_{j\downarrow}^\dagger c_{j\downarrow}) + \frac{(1-I_Q)}{2} \sum_{j=1}^2 (c_{j\uparrow}^\dagger c_{j\downarrow} - c_{j\downarrow}^\dagger c_{j\uparrow}) \quad (42)$$

$$S_c = i\frac{\pi}{4}(1+I_Q) \left[\sum_{j=1}^2 (c_{j\uparrow}^\dagger c_{j\uparrow} - c_{j\downarrow}^\dagger c_{j\downarrow}) - 1 \right] + \frac{\pi}{4}(1-I_Q) \sum_{j=1}^2 (c_{j\uparrow}^\dagger c_{j\downarrow} - c_{j\downarrow}^\dagger c_{j\uparrow}) \quad (43)$$

and yields the electronically diagonalized form

$$\tilde{H} = T_c : T_b : T_a : H = T_t : H = \sum_{j=1}^2 \sum_{\tau=\uparrow,\downarrow} h_{\text{MFG}}^{(j\tau)}(Q) c_{j\tau}^\dagger c_{j\tau} \quad (44)$$

where the total transformation operator reads $U_t = U_a U_b U_c$,

$$h_{\text{MFG}}^{(j\tau)}(Q) = H_{\text{ph}} - (-1)^j A(Q)I_Q - (-1)^{j+\tau} B(Q) - (-1)^{j+\tau} C(Q)I_Q + (-1)^j D(Q) \quad (45)$$

and where, in the exponents, $\tau = 1$ is taken for $\tau = \uparrow$ and $\tau = 2$ for $\tau = \downarrow$. The transformed eigenfunctions are thus of the form

$$\tilde{\Psi}_n^{(j\tau)} = |j\tau\rangle \hat{\Phi}_n^{(j\tau)}(Q) \quad (46)$$

where $\hat{\Phi}_n^{(j\tau)}(Q)$ satisfies the FG-type equations

$$h_{\text{MFG}}^{(j\tau)}(Q) \hat{\Phi}_n^{(j\tau)}(Q) = E_n^{(j\tau)} \hat{\Phi}_n^{(j\tau)}(Q). \quad (47)$$

The basic properties of T_c and of the total transformation T_t are written in appendix B.

By means of the total transformation $U_t = U_a U_b U_c$ the eigenfunctions $\Psi_{\text{MFG}}^{(j\tau)}$ of the original Hamiltonian H (see (3)) are found from expression (46) and may be written in the form $\Psi_{\text{MFG}}^{(j\tau)} = U_t \tilde{\Psi}_{\text{MFG}}^{(j\tau)}$

$$\Psi_{\text{MFG}}^{(j\tau)} = \frac{1}{2}(I_Q)^{j+\tau} [(-1)^{j+\tau} |1\uparrow\rangle - (-1)^\tau I_Q |2\uparrow\rangle + (-1)^j |1\downarrow\rangle - I_Q |2\downarrow\rangle] \hat{\Phi}_n^{(j\tau)}(Q) \quad (48)$$

where $j = 1, 2$; $\tau = 1, 2$ and $\hat{\Phi}^{(j\tau)}(Q)$ are the eigenfunctions of the MFGE (47).

For later reference we note the symmetry behaviour of this solution. Applying the properties of the parity operators I and I_σ introduced in section 2 to expression (48) we get

$$I\Psi_{\text{MFG}}^{(j\tau)} = (-1)^{j+1}\Psi_{\text{MFG}}^{(j\tau)} \quad I_\sigma\Psi_{\text{MFG}}^{(j\tau)} = (-1)^\tau\Psi_{\text{MFG}}^{(j\tau)} \quad (49)$$

from which we derive the parity correspondences

$$j = 1 \stackrel{\Delta}{=} p = 2 \quad j = 2 \stackrel{\Delta}{=} p = 1 \quad (50)$$

and for ‘spin parity’

$$\tau \stackrel{\Delta}{=} p_\sigma. \quad (51)$$

5. Group-theoretical construction

In the preceding section we have found the electronic diagonalization by a sequence of three unitary transformations which have been successively established by physical intuition. There is also, however, a systematic way to construct such a type of unitary transformation.

In this section we present this approach. For this purpose we use the results of a previous paper by one of us [11]. There, a generalization of the FG approach is given which exploits group-theoretical theorems [14–16] and allows for more general symmetries of the Hamiltonian than a simple mirror symmetry. We consider a set of N orthonormal one-particle functions $\{|r\rangle\}$ which are distributed in space in such a manner that they can be uniquely generated from one function within the set, say $|N\rangle$, by the N symmetry operations R_r of a group G

$$\begin{aligned} |r\rangle &= R_r|N\rangle & r &= 1, \dots, N & R_N &= E(\text{unity element}) \\ \langle r|r'\rangle &= \delta_{rr'} & \forall r, r' &= 1, \dots, N \\ R_r^\dagger &= R_r^{-1}. \end{aligned} \quad (52)$$

These functions then establish the regular representation of the group. We require that this group be Abelian

$$R_r R_{r'} = R_{r'} R_r \quad \forall r, r' = 1, \dots, N. \quad (53)$$

Then there are precisely N irreducible representations $\Gamma^{(\gamma)}$, $\gamma = 1, \dots, N$, all of which are one dimensional. We assume the Hamiltonian of the electron–phonon system is governed by the group G . Then the Hamiltonian of the vibrational subsystem can be written in the form

$$H_{\text{ph}} = \frac{1}{2} \sum_{m,\gamma} (P_{m,\gamma}^\dagger P_{m,\gamma} + \Omega_{m\gamma}^2 Q_{m,\gamma}^\dagger Q_{m,\gamma}) \quad (54)$$

where $Q_{m,\gamma}$ are symmetry adapted coordinates pertaining to the irreducible representation $\Gamma^{(\gamma)}$ and $m = 1, \dots, M_\gamma$ is the multiplicity index within one irreducible coordinate sequence. The interaction Hamiltonian W is written in the form

$$W = \sum_{r,s=1}^N |r\rangle \langle s| A_{rs}(Q) \quad (55)$$

where the functions A_{rs} are chosen in such a manner that $R_r W = W R_r$. The structural form of the eigenfunctions is then uniquely given by the Wigner formula

$$\Psi^{(\nu)} = N^{-1/2} \sum_{r=1}^N \chi^{(\nu)}(R_r) (R_r | N) R_r(Q) \Phi^{(\nu)}(Q). \quad (56)$$

$\Phi^{(\nu)}(Q) :=$ normalized phonon function

$R_r :=$ symmetry operation

$R_r(Q) :=$ symmetry operation in Q-space.

Instead of making recourse to the group-theoretically correct structural form of the eigenfunctions we can also apply a unitary transformation to the Hamiltonian $H = H_{\text{ph}} + W$ of the form [11]

$$U_{\text{FG}} = N^{-1/2} \sum_{r=1}^N \sum_{\gamma=1}^N \chi^{(\nu)}(R_r) |r\rangle \langle \gamma | R_r(Q) \quad (57)$$

$$U_{\text{FG}}^\dagger = N^{-1/2} \sum_{r=1}^N \sum_{\gamma=1}^N \chi^{(\nu)}(R_r)^* | \gamma \rangle \langle r | R_r^\dagger(Q).$$

If this transformation is applied to H the latter is diagonalized with respect to the electronic subsystem

$$U_{\text{FG}}^\dagger H U_{\text{FG}} = T_{\text{FG}} : H = \sum_{\gamma=1}^N | \gamma \rangle \langle \gamma | h_{\text{FG}}^{(\nu)}(P, Q) \quad (58)$$

where

$$h_{\text{FG}}^{(\nu)} = \left[H_{\text{ph}} + N^{-1} \sum_{r,s=1}^N \chi^{(\nu)}(R_r)^* \chi^{(\nu)}(R_s) [R_r^\dagger A_{rs}] R_r^\dagger(Q) R_s(Q) \right]. \quad (59)$$

The remaining vibrational Schrödinger problem is thus governed by the generalized FGE

$$h_{\text{FG}}^{(\nu)}(Q) \Phi^{(\nu)}(Q) = E^{(\nu)} \Phi^{(\nu)}(Q) \quad (60)$$

and the transformed eigenfunctions read

$$\tilde{\Psi}^{(\nu)} = | \gamma \rangle \Phi^{(\nu)}(Q). \quad (61)$$

Inverting the transformation, $\Psi^{(\nu)} = U_{\text{FG}} \tilde{\Psi}^{(\nu)}$, we receive just the Wigner expression (56), in which $\Phi^{(\nu)}(Q)$ is to be understood as a solution of (60).

We now apply the preceding formalism to our 221 model. We observe that the electronic base in this model can indeed be considered as a regular representation of an Abelian group. This becomes evident if we introduce the symmetry operations

$$\begin{aligned} R_1 &= I_\sigma I : \text{combined inversion-spin-flip operator} \\ R_2 &= I_\sigma : \text{spin-flip operator} \\ R_3 &= I : \text{inversion operator} \\ R_4 &= E : \text{unity operator.} \end{aligned} \quad (62)$$

By means of definition (52) we then have the electronic base-vector set $\{|r\rangle\}$

$$\begin{aligned} |1\rangle &= |1 \uparrow\rangle \\ |2\rangle &= |2 \uparrow\rangle \\ |3\rangle &= |1 \downarrow\rangle \\ |4\rangle &= |2 \downarrow\rangle. \end{aligned} \tag{63}$$

The group, which is established by the operations (62), is isomorphic to the group C_{2h} , which is Abelian. Its four irreducible representations can be assigned to the orbital and spin parities $p (= 1, 2)$ and $p_\sigma (= 1, 2)$ introduced in section 2

$$\gamma \hat{=} pp_\sigma = 11, 12, 21, 22 \tag{64}$$

and the characters are then given by

$$\begin{aligned} \chi^{(pp_\sigma)}(E) &= 1 \\ \chi^{(pp_\sigma)}(I) &= (-1)^p \\ \chi^{(pp_\sigma)}(I_\sigma) &= (-1)^{p_\sigma} \\ \chi^{(pp_\sigma)}(I_\sigma I) &= (-1)^{p+p_\sigma}. \end{aligned} \tag{65}$$

Inserting these in expression (56) we get the unitary transformation operator

$$U_{FG} = \frac{1}{2} \sum_{p, p_\sigma=1}^2 [(-1)^{p+p_\sigma} I_Q |1 \uparrow\rangle + (-1)^{p_\sigma} |2 \uparrow\rangle + (-1)^p I_Q |1 \downarrow\rangle + |2 \downarrow\rangle] \langle pp_\sigma| \tag{66}$$

and the transformed Hamiltonian (58) assumes the form

$$T_{FG} : H = \sum_{p, p_\sigma=1}^2 h_{FG}^{(pp_\sigma)}(Q) |pp_\sigma\rangle \langle pp_\sigma| \tag{67}$$

where

$$h_{FG}^{(pp_\sigma)}(Q) = H_{ph} + (-1)^p A(Q) I_Q + B(Q) + (-1)^{p+p_\sigma} C(Q) I_Q + (-1)^{p_\sigma} D(Q). \tag{68}$$

This leads to the FGE (see equation (60))

$$h_{FG}^{(pp_\sigma)}(Q) \Phi^{(pp_\sigma)}(Q) = E^{(pp_\sigma)} \Phi^{(pp_\sigma)}(Q) \tag{69}$$

and if this vibrational eigenvalue problem has been solved the total eigenfunction is found to be

$$\tilde{\Psi}^{(pp_\sigma)} = |pp_\sigma\rangle \Phi^{(pp_\sigma)}(Q). \tag{70}$$

Inverting the transformation by means of the unitary operator (66), $\Psi^{(pp_\sigma)} = U_{FG} \tilde{\Psi}^{(pp_\sigma)}$, we arrive at the original eigenfunction

$$\Psi_{FG}^{(p, p_\sigma)} = \frac{1}{2} [(-1)^{p+p_\sigma} I_Q |1 \uparrow\rangle + (-1)^{p_\sigma} |2 \uparrow\rangle + (-1)^p I_Q |1 \downarrow\rangle + |2 \downarrow\rangle] \Phi^{(p, p_\sigma)}(Q) \tag{71}$$

which has the symmetry properties

$$I \Psi_{\text{FG}}^{(pp\sigma)} = (-1)^p \Psi_{\text{FG}}^{(pp\sigma)} \quad I_\sigma \Psi_{\text{FG}}^{(pp\sigma)} = (-1)^{p_\sigma} \Psi_{\text{FG}}^{(pp\sigma)}. \quad (72)$$

The complete properties of the FGT are given by

$$\begin{aligned} U_{\text{FG}}^\dagger |i\rho\rangle \langle j\sigma| U_{\text{FG}} &= \frac{1}{4} \sum_{p,q=1}^2 \sum_{p_\sigma,q_\sigma=1}^2 (-1)^{ip+jq+\rho p_\sigma+\sigma q_\sigma} |pp_\sigma\rangle \langle qq_\sigma| \\ U_{\text{FG}} |pp_\sigma\rangle \langle qq_\sigma| U_{\text{FG}}^\dagger &= \frac{1}{4} \sum_{i,j=1}^2 \sum_{\rho,\sigma=1}^2 (-1)^{ip+jq+\rho p_\sigma+\sigma q_\sigma} |i\rho\rangle \langle j\sigma|. \end{aligned} \quad (73)$$

We stress again that i, j, ρ, σ have the meaning of orbital and spin indices in the original picture, while p, q, p_σ, q_σ indicate orbital and spin parities in the transformed picture.

6. Physical background

The 221 model should be considered as an archetype, just like its spinless analogue, which may sometimes govern the dynamics of molecular units in magnetic systems, if spin-flip interactions are involved. But it should be noted that the relevance of unitary transformations for prototype systems, even if they do not fully diagonalize the system, is of a broader scale for two major reasons. On the one hand they can often be performed up to a closed form and on the other hand sometimes the dynamics of more complicated and physically more realistic systems, possibly after some preceding manipulation, may be traced back to the dynamics of one such prototype system.

Specifically, our 221 model is possibly of use in high- T_c superconductors, where the transfer dynamics of an electron (or hole) between an oxygen and its copper neighbours may play a basic role in the ferromagnetic clustering process needed to establish metallic behaviour. In this case, the transfer mechanism involves a spin-flip process. Ferromagnetic clustering units of this type are presently considered, by many workers in the field, as the initial step in a percolative process which, after reaching a doping threshold, eventually allows for metallic behaviour in these cuprate materials although in the undoped form they are anti-ferromagnetic and isolating. For details we refer to the proceedings of a recent conference [17].

To show in a somewhat didactical manner how in the electron-phonon dynamics of a Cu-O unit our 221 model may be exploited, we characterize this unit by an electronic base of four states, $|1\sigma\rangle, |2\sigma\rangle$, i.e. a single orbital state with spins $\sigma = \uparrow, \downarrow$ per each ionic site, and by a single vibrational coordinate Q which describes the motion of the Cu-O unit against the surrounding. We introduce a fictitious formal inversion symmetry, such that $I|1\sigma\rangle = |2\sigma\rangle I$, $I_\sigma|i\sigma\rangle = |i, -\sigma\rangle I_\sigma$ and $IQ = -QI$. We note that these definitions do not necessitate that the Hamiltonian itself has inversion symmetry. The most simple version of the 221 model is then of the form

$$\begin{aligned} \hat{H} &= \frac{1}{2}(P^2 + \Omega^2 Q^2) + \sum_{i=1}^2 \sum_{\sigma=\uparrow,\downarrow} (\epsilon_i + \alpha_i Q) c_{i\sigma}^\dagger c_{i\sigma} \\ &+ T \sum_{\sigma=\uparrow,\downarrow} (c_{2\sigma}^\dagger c_{1\sigma} + c_{1\sigma}^\dagger c_{2\sigma}) + T_\sigma \sum_{\sigma=\uparrow,\downarrow} (c_{2\sigma}^\dagger c_{1,-\sigma} + c_{1\sigma}^\dagger c_{2,-\sigma}) \end{aligned} \quad (74)$$

where spin-flip dynamics are only considered in combination with orbital transfer. $\alpha_i Q$ and ϵ_i are the static and dynamical energies pertaining to the copper and oxygen ions respectively, while T and T_σ are the transfer constants with and without spin-flip. Both transfer terms can be viewed as being due to orbital overlap but other causes are also conceivable. If $\epsilon_1 \neq \epsilon_2$ and $\alpha_1 \neq -\alpha_2$ then the application of the FGT or the MFGT will also generate non-diagonal contributions in the transformed Hamiltonian, which then reads

$$T_t : \hat{H} = \sum_{j,\tau=1}^2 \hat{h}_{\text{MFG}}^{(j\tau)}(Q) c_{j\tau}^\dagger c_{j\tau} + H_{\text{nd}} \tag{75}$$

where $\hat{h}_{\text{MFG}}^{(j\tau)}(Q)$ is a special realization of $h_{\text{MFG}}^{(j\tau)}(Q)$ (see equation (45)) and has the form

$$\hat{h}_{\text{MFG}}^{(j\tau)}(Q) = \frac{1}{2}(P^2 + \Omega^2 Q^2) + \frac{1}{2}(\epsilon_1 + \epsilon_2) - \frac{1}{2}(-1)^{j+\tau}(\alpha_2 - \alpha_1)Q - (-1)^j T I_Q - (-1)^{j+\tau} T_\sigma I_Q. \tag{76}$$

H_{nd} pertains to the non-diagonal terms and is of the form

$$H_{\text{nd}} = \sum_{\tau=1}^2 \left[\frac{1}{2}(\epsilon_2 - \epsilon_1) I_Q (c_{1\tau}^\dagger c_{2\tau} + c_{2\tau}^\dagger c_{1\tau}) + \frac{1}{2}(\alpha_1 + \alpha_2) Q I_Q (-1)^\tau (c_{1\tau}^\dagger c_{2\tau} - c_{2\tau}^\dagger c_{1\tau}) \right]. \tag{77}$$

If $\epsilon_1 \approx \epsilon_2$ and $\alpha_1 \approx -\alpha_2$, the contribution of the non-diagonal Hamiltonian to the total energy eigenvalues vanishes ($H_{\text{nd}} \approx 0$). In this case, the non-diagonal parts of the transformed Hamiltonian may be treated as a small perturbation.

Both the original FGT as well as those of the present investigations are one-electron transformations and their utility is diminished if more than a single particle is involved. Therefore a multi-particle generalization is desirable. Such a generalization, however, is not easy to achieve if the one-particle transformation operator U is not given in an exponential form, since then the unitarity condition poses problems. Yet, these do not occur if U is written as $U = \exp[S]$, or as a product of exponentials, since then only anti-Hermiticity of $S (= -S^\dagger)$ must be required, which in the multi-particle case is also easily warranted. Looking back to the transformations of this study we note that the one found in section 4 is already in an exponential form

$$U_t = e^{S_a} e^{S_b} e^{S_c} \tag{78}$$

where the electronically bilinear operators S_a , S_b and S_c may be directly understood in a multi-particle sense as preserving their anti-Hermiticity. Since $[S_a, S_b] = 0$ and $[S_a, S_c] = 0$, but $[S_b, S_c] \neq 0$, we have two possibilities for writing U_t in an exponential way

$$U_t = e^{S_a + S_b} e^{S_c} \quad \text{or} \quad U_t = e^{S_b} e^{S_a + S_c}.$$

It should be noted, however, that the multi-particle application of this transformation is more complicated than in the one-particle case, since the conservation law (14) can no longer be used. We will discuss this further in a forthcoming paper.

Finally, it should be emphasized that the application of unitary transformations does not require the definition of a specific Hamiltonian since it refers to any dynamical variable which is established as a functional of the basic variables of a given Hilbert space. The latter may well be a subspace of the physically relevant total Hilbert space. Specifically, the electronic base (two orbitals, two spins), to which the transformations of this study are referring to, may be subunits of a larger physical system.

7. Results and conclusions

In this paper a spin-flip extension of the original two-site model of FG is considered. It consists of two orbital states of the electronic subsystem and includes spin-flip terms in the Hamiltonian. We denote this model as the 221 model. The electronic subsystem is coupled to a vibrational subsystem such that in Euclidean space the system is governed by the inversion symmetry group. The total symmetry group of the system, however, also includes spin-flip operations and is of dimension four; it is isomorphous to the group C_{2h} . Since this group is also Abelian, the generalization of the FG procedure can be employed. This is presented in detail. On the other hand, we present a sequence of transformations $U_t = U_a U_b U_c$ which in their combination also diagonalize the electronic subsystem. U_t constitutes a modification of the FGT U_{FG} . Its value lies in the fact that its factors U_a , U_b and U_c disentangle the diagonalization procedure and allow for an additional physical insight into the dynamics of the system.

Since the FG concept has proven useful for a large class of systems, where one particle (or exciton) is coupled to a vibrational subsystem, it is highly desirable to find a generalization of this concept for more than a single elementary particle or exciton. To achieve this, however, it is necessary to find an appropriate exponential form for the multi-particle unitary operator. Such a form, in general, is not provided by the group theoretical procedure presented in an earlier paper [11]. Therefore one of our principal motivations for the present work was aimed at finding such exponential form for an archetypical electron system which is important, e.g. in the present discussion about high- T_c superconductivity. This has been achieved in this paper. In a forthcoming paper we will apply the established unitary transformation to the multi-particle situation. An additional motivation for doing this is related to one of the most pronounced features of the FGT, which is the fact that it lays open the influence of the electronic coupling on the vibrational subsystem in a very lucid way, such that the vibrational parts of the total wavefunctions can be discussed easily. In particular the FGE are suggestive for finding topological properties of the vibrational functions. This is of considerable actuality, since recently effects relating to spatial alterations of the vibrational functions (squeezing, anti-squeezing) have been discussed both in the field of superconductivity and quantum diffusion.

Finally, we return to the relation between the FGT and the LLPT in polaron theory. For the latter we refer to the Pines article in the well known St Andrew's Lectures [18]. As mentioned in section 1, the FGT in translationally invariant systems may be viewed as a kind of 'discretized' LLPT. Since the LLPT in its original first quantized version is already written in exponential form, its transcription to a second quantized version, which is easily performed, already constitutes a legitimate multi-particle transformation.

For completeness we note that Lee *et al* [19] have supplemented their 'first' transformation by a 'second' one of oscillatory displacement character. The multi-particle transcription of this latter transformation does not pose difficulties and has been used by Lemmens *et al* [20] in the interacting polaron problem.

However, the full multi-particle application of the 'first' LLPT is rather complicated, since the respective commutator expansions, specifically that of the phonon coordinates and momenta, involve electron-electron interaction terms. On the other hand, the possibility of establishing an effective electronic correlation resulting from the original electron-phonon interaction provides a fascinating contrast to the conventional effective correlation derived from the famous Fröhlich transformation which is used in the BCS theory.

Appendix A. Search for the third transformation T_c

For our purpose we have to introduce a set of pseudo spin-operators including site numbers i

$$\sigma_{ix} = \frac{1}{2}(c_{i\uparrow}^\dagger c_{i\downarrow} + c_{i\downarrow}^\dagger c_{i\uparrow}) \quad \sigma_{iy} = \frac{1}{2i}(c_{i\uparrow}^\dagger c_{i\downarrow} - c_{i\downarrow}^\dagger c_{i\uparrow}) \quad \sigma_{iz} = \frac{1}{2}(c_{i\uparrow}^\dagger c_{i\uparrow} - c_{i\downarrow}^\dagger c_{i\downarrow}) \quad (80)$$

with commutation relations

$$[\sigma_{kx}, \sigma_{ly}] = i\delta_{kl}\sigma_{kz} \quad \text{cyclic} \quad (81)$$

$$[\sigma_{ix}, \sigma_{kx}] = [\sigma_{iy}, \sigma_{ky}] = [\sigma_{iz}, \sigma_{kz}] = 0 \quad \forall i \neq k \quad (82)$$

and

$$\sigma_{ix}^2 + \sigma_{2x}^2 = \sigma_{1y}^2 + \sigma_{2y}^2 = \sigma_{1z}^2 + \sigma_{2z}^2 = \frac{1}{4} \quad (83)$$

as particle conservation. Inserting (80) in the twice-transformed Hamiltonian (41) we obtain

$$T_b : T_a : H = H_{ph} - 4A(Q)I_Q(\sigma_{2x}^2 - \sigma_{1x}^2) + 2iB(Q)I_Q(\sigma_{2y} - \sigma_{1y}) + C(Q)(\sigma_{2z} - \sigma_{1z}) + 2D(Q)(\sigma_{2x} - \sigma_{1x}). \quad (84)$$

From the tables at the end of [2], we know the properties of the transformation

$$U = (1 + I_Q)\sigma_z + i(1 - I_Q)\sigma_y. \quad (85)$$

For example

$$\begin{aligned} T : \sigma_x^2 &= \sigma_x^2 = \frac{1}{4} \\ T : Q_{m,1}\sigma_z &= -2Q_{m,1}I_Q\sigma_x\sigma_y = -iQ_{m,1}I_Q\sigma_z \\ T : Q_{m,1}\sigma_x &= -2Q_{m,1}\sigma_x^2 = -\frac{1}{2}Q_{m,1}. \end{aligned} \quad (86)$$

In (84) similar terms to those in (86) appear. Therefore, the extended form of (85)

$$U_c = (1 + I_Q)(\sigma_{1z} + \sigma_{2z}) + i(1 - I_Q)(\sigma_{1y} + \sigma_{2y}) \quad (87)$$

should be able to diagonalize $T_b : T_a : H$ with respect to the electron problem.

Rewritten in creation and annihilation operators we obtain

$$U_c = \frac{(1 + I_Q)}{2} \sum_{i=1}^2 (c_{i\uparrow}^\dagger c_{i\uparrow} - c_{i\downarrow}^\dagger c_{i\downarrow}) + \frac{(1 - I_Q)}{2} \sum_{i=1}^2 (c_{i\uparrow}^\dagger c_{i\downarrow} - c_{i\downarrow}^\dagger c_{i\uparrow}) \quad (88)$$

and its exponential form $U_c = \exp[S_c]$ with

$$S_c = i\frac{\pi}{4}(1 + I_Q) \left[\sum_{i=1}^2 (c_{i\uparrow}^\dagger c_{i\uparrow} - c_{i\downarrow}^\dagger c_{i\downarrow}) - 1 \right] + \frac{\pi}{4}(1 - I_Q) \sum_{i=1}^2 (c_{i\uparrow}^\dagger c_{i\downarrow} - c_{i\downarrow}^\dagger c_{i\uparrow}). \quad (89)$$

Appendix B. List of transformations

The basic properties of T_a are

$$\begin{aligned}
 T_a : c_{1\sigma} &= \frac{1}{\sqrt{2}}(c_{1\sigma} - I_Q c_{2\sigma}) \\
 T_a : c_{2\sigma} &= \frac{1}{\sqrt{2}}(c_{2\sigma} + I_Q c_{1\sigma}) \\
 T_a : Q_{m,1} &= Q_{m,1} I_Q \sum_{\sigma=\uparrow,\downarrow} (c_{2\sigma}^\dagger c_{1\sigma} - c_{1\sigma}^\dagger c_{2\sigma}) \\
 T_a : P_{m,1} &= P_{m,1} I_Q \sum_{\sigma=\uparrow,\downarrow} (c_{2\sigma}^\dagger c_{1\sigma} - c_{1\sigma}^\dagger c_{2\sigma}) \\
 T_a : Q_{m,2} &= Q_{m,2} \\
 T_a : P_{m,2} &= P_{m,2} \\
 T_a : |1\sigma\rangle f(Q) &= \frac{1}{\sqrt{2}}[|1\sigma\rangle + I_Q |2\sigma\rangle] f(Q) \\
 T_a : |2\sigma\rangle f(Q) &= \frac{1}{\sqrt{2}}[|2\sigma\rangle - I_Q |1\sigma\rangle] f(Q).
 \end{aligned} \tag{90}$$

The basic properties of T_b are

$$\begin{aligned}
 T_b : c_{i\uparrow} &= \frac{1}{\sqrt{2}}(c_{i\uparrow} + I_Q c_{i\downarrow}) \\
 T_b : c_{i\downarrow} &= \frac{1}{\sqrt{2}}(c_{i\downarrow} - I_Q c_{i\uparrow}) \\
 T_b : Q_{m,1} &= Q_{m,1} I_Q \sum_{i=1}^2 (c_{i\uparrow}^\dagger c_{i\downarrow} - c_{i\downarrow}^\dagger c_{i\uparrow}) \\
 T_b : P_{m,1} &= P_{m,1} I_Q \sum_{i=1}^2 (c_{i\uparrow}^\dagger c_{i\downarrow} - c_{i\downarrow}^\dagger c_{i\uparrow}) \\
 T_b : Q_{m,2} &= Q_{m,2} \\
 T_b : P_{m,2} &= P_{m,2} \\
 T_b : |i\uparrow\rangle f(Q) &= \frac{1}{\sqrt{2}}[|i\uparrow\rangle - I_Q |i\downarrow\rangle] f(Q) \\
 T_b : |i\downarrow\rangle f(Q) &= \frac{1}{\sqrt{2}}[|i\downarrow\rangle + I_Q |i\uparrow\rangle] f(Q).
 \end{aligned} \tag{91}$$

The basic properties of T_c are

$$\begin{aligned}
T_c : c_{i\uparrow} &= \frac{i}{2}(1 + I_Q)c_{i\uparrow} + \frac{1}{2}(1 - I_Q)c_{i\downarrow} \\
T_c : c_{i\downarrow} &= -\frac{i}{2}(1 + I_Q)c_{i\downarrow} - \frac{1}{2}(1 - I_Q)c_{i\uparrow} \\
T_c : Q_{m,1} &= Q_{m,1} \sum_{i=1}^2 (c_{i\uparrow}^\dagger c_{i\downarrow} + c_{i\downarrow}^\dagger c_{i\uparrow}) \\
T_c : P_{m,1} &= P_{m,1} \sum_{i=1}^2 (c_{i\uparrow}^\dagger c_{i\downarrow} + c_{i\downarrow}^\dagger c_{i\uparrow}) \\
T_c : Q_{m,2} &= Q_{m,2} \\
T_c : P_{m,2} &= P_{m,2} \\
T_c : |i \uparrow\rangle f(Q) &= \left[\frac{1 + I_Q}{2} |i \uparrow\rangle - \frac{1 - I_Q}{2} |i \downarrow\rangle \right] f(Q) \\
T_c : |i \downarrow\rangle f(Q) &= \left[-\frac{1 + I_Q}{2} |i \downarrow\rangle + \frac{1 - I_Q}{2} |i \uparrow\rangle \right] f(Q).
\end{aligned} \tag{92}$$

The basic properties of T_t are

$$\begin{aligned}
T_t : c_{1\uparrow} &= \frac{1}{4} \{ [(1 - I_Q) + i(1 + I_Q)](c_{1\uparrow} + c_{2\downarrow}) + [(1 - I_Q) - i(1 + I_Q)](c_{1\downarrow} + c_{2\uparrow}) \} \\
T_t : c_{1\downarrow} &= \frac{1}{4} \{ [(1 - I_Q) + i(1 + I_Q)](c_{2\downarrow} - c_{1\uparrow}) + [(1 - I_Q) - i(1 + I_Q)](c_{1\downarrow} - c_{2\uparrow}) \} \\
T_t : c_{2\uparrow} &= \frac{1}{4} \{ [(1 - I_Q) + i(1 + I_Q)](c_{2\uparrow} - c_{1\downarrow}) + [(1 - I_Q) - i(1 + I_Q)](c_{2\downarrow} - c_{1\uparrow}) \} \\
T_t : c_{2\downarrow} &= \frac{1}{4} \{ [(1 - I_Q) + i(1 + I_Q)](-c_{2\uparrow} - c_{1\downarrow}) + [(1 - I_Q) - i(1 + I_Q)](c_{1\uparrow} + c_{2\downarrow}) \} \\
T_t : Q_{m,1} &= Q_{m,1} I_Q [c_{2\uparrow}^\dagger c_{1\uparrow} - c_{2\downarrow}^\dagger c_{1\downarrow} - c_{1\uparrow}^\dagger c_{2\uparrow} + c_{1\downarrow}^\dagger c_{2\downarrow}] \\
T_t : P_{m,1} &= P_{m,1} I_Q [c_{2\uparrow}^\dagger c_{1\uparrow} - c_{2\downarrow}^\dagger c_{1\downarrow} - c_{1\uparrow}^\dagger c_{2\uparrow} + c_{1\downarrow}^\dagger c_{2\downarrow}] \\
T_t : Q_{m,2} &= Q_{m,2} \\
T_t : P_{m,2} &= P_{m,2} \\
T_t : |1 \uparrow\rangle f(Q) &= \frac{1}{2} [|1 \uparrow\rangle - |2 \uparrow\rangle I_Q - |1 \downarrow\rangle I_Q + |2 \downarrow\rangle] f(Q) \\
T_t : |2 \uparrow\rangle f(Q) &= \frac{1}{2} [|1 \uparrow\rangle I_Q + |2 \uparrow\rangle - |1 \downarrow\rangle - |2 \downarrow\rangle I_Q] f(Q) \\
T_t : |1 \downarrow\rangle f(Q) &= \frac{1}{2} [-|1 \uparrow\rangle + |2 \uparrow\rangle I_Q - |1 \downarrow\rangle I_Q + |2 \downarrow\rangle] f(Q) \\
T_t : |2 \downarrow\rangle f(Q) &= \frac{1}{2} [-|1 \uparrow\rangle I_Q - |2 \uparrow\rangle - |1 \downarrow\rangle - |2 \downarrow\rangle I_Q] f(Q).
\end{aligned} \tag{93}$$

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