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1997 J. Phys. A: Math. Gen. 30 2811

(http://iopscience.iop.org/0305-4470/30/8/025)

J. Phys. A: Math. Gen. **30** (1997) 2811–2823. Printed in the UK PII: S0305-4470(97)79817-2

Multiband Fulton–Gouterman transformation and its application to electron–phonon systems

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Received 25 November 1996

Abstract. We first generalize the Fulton–Gouterman transformation to a multi-band and exponential form. While the exponential form admits the consideration of multiparticle systems, which will be done in future papers, here we treat the multi-band form in its application to an archetypical electron–phonon Hamiltonian, which contains the Fröhlich one as a particular limiting case. Several generalizations will be outlined.

1. Introduction

Unitary transformations are an efficient and well known means to bring a physical problem into a somewhat easier form, which often enlightens new insights into the problem considered. Perhaps the most famous unitary transformation is that of Fröhlich in the theory of superconductivity. But many others have been used in the past decades. A transformation of particular fascination in electron–phonon and exciton–phonon problems is that of Lee *et al* (LLPT) [1]. Its attraction lies in the fact that it diagonalizes Hamiltonians of Fröhlich type with regard to the electronic subspace. The original LLPT is a one-electron transformation and formulated in the first quantized form. It was devised for translationally invariant systems and owes its diagonalizing property to the invariance of a generalized momentum operator. If the LLPT is applied to more general forms of electron–phonon Hamiltonians it loses its diagonalization property, as will be seen in a forthcoming paper.

A transformation of similar qualities as the LLPT is that of Fulton and Gouterman (FGT) [2] and its generalization given by one of us [3]. Also in this transformation electronic diagonalization is achieved for electron–phonon or exciton–phonon systems provided they are governed by an Abelian symmetry group (or subgroup), and that the electronic base establishes a regular representation of the group (or subgroup). The main virtue of the FGT lies on the one hand in the fact that the Schrödinger-like equations (FG equations) in the phononic subspace, to which the dynamics is reduced after the FGT has been performed, display favourable topological features. Specifically, they convince in a lucid manner the basic antagonistic tendencies inherent in the dynamics of the coupled systems. The FGT has been applied in the field of quantum diffusion [4] as well as in excitonic problems [5–9]. For example, it has been a guide to find new types of exciton–phonon states of high energy ('exotic states') which are of squeezed nature in the phonon subspace and in which the exciton is liberated to free motion, again in contrast to the conventional selftrapping in the low-energy states [7]. This provides a semi-quantitative description of the retarded luminescence phenomenon [10, 11] and also has an impact on energy transport problems.

0305-4470/97/082811+13$19.50

On the other hand the generalized Fulton–Gouterman transformation (GFGT) has the virtue that in translationally invariant systems it displays its diagonalizing quality for electron–phonon Hamiltonians of more general forms than that of the Fröhlich type, provided the electronic Hilbert space is restricted to a single band ('regular base' of the Abelian group).

The main purpose of the present study is to analyse the implications of an FGT-type transformation, if a multi-band electronic Hilbert space is chosen. If this base is taken in such a manner that it diagonalizes the pure electronic part of the Hamiltonian, it is of particular interest to discuss the remaining band–band transition terms initiated after the multi-band FGT (GFGT) has been performed.

This paper is organized as follows. In the second section we introduce the multi-band generalization of the FGT and present the explicit transformation behaviour of the basic operators and wavefunctions. In the third section we apply the GFGT to the most general Hamiltonian that satisfies Abelian symmetry. In section 4 we give the GFGT operator and the respective transformed expressions for the particular case of translational invariance. We also present the exponentional form of the GFGT, which will be the starting point for multi-particle calculations in future papers. In section 5 an archetypical translationally invariant electron–phonon Hamiltonian is presented which contains the Fröhlich Hamiltonian as a particular limiting case. The GFGT is applied and the transformed expressions are given explicitly. In section 6 we present the phonon equations due to the transformed Hamiltonian of section 5, which are called the Fulton–Gouterman equations (FGE). Also a perturbation theory of the considered FGE is presented. In section 7 we give an illustrative presentation of the topological meaning of the FGE by means of a simple translationally invariant system. Finally, in section 8 we collect the results and the perspectives of the present work.

2. Generalized Fulton–Gouterman transformation (GFGT)

The original transformation of Fulton and Gouterman has been devised for two-states electron–phonon systems of inversion symmetry [2]. Later, one of us [3] generalized this transformation to systems of Abelian symmetry. The very essence of the GFGT is to exploit the symmetry of a system as far as possible. This transformation has proven very effective in exciton–phonon problems and others [4–11]. In particular it has given new insight to the coupled dynamics of the two subsystems ('squeezed' and 'anti-squeezed' states, etc). We will present here merely the essential ingredients and the generalization to an electronic multi-band form to establish the notation. For further details we refer to the original work of [3].

In a system of Abelian symmetry the group elements R_r $(r = 0, \ldots, N)$ commute, $R_r R_s = R_s R_r$, whence the irreducible representations Γ are one-dimensional and their number equals the number of group elements $N + 1$: $\Gamma = 0, 1, \ldots, N$. The characters are denoted as $\chi(\Gamma, r)$, where $|\chi(\Gamma, r)| = 1$, and the identity element E is taken to be $E = R_0$, i.e. $\chi(\Gamma, 0) = 1$. We further note $(R_r)^{-1} = (R_r)^{\dagger}$ and $\chi^*(\Gamma, R_r) \equiv \chi^*(\Gamma, r) = \chi(\Gamma, (R_r)^{-1})$.

The appropriate electronic complete orthonormal base (CONB) of a system of Abelian symmetry is most conveniently taken in the form of irreducible base sets. We introduce a CONB in the electronic subspace $\{|\Gamma, \lambda\rangle\}$ where λ is some band index, such that

$$\langle \Gamma, \lambda | \Gamma', \lambda' \rangle = \delta_{\Gamma,\Gamma'} \cdot \delta_{\lambda,\lambda'} \tag{1}$$

$$\sum_{\Gamma,\lambda} |\Gamma, \lambda\rangle\langle \Gamma, \lambda| = I^{(el)} \tag{2}$$

$I^{(el)}$ representing the electronic unity operator. We further note that the unitary operator

R_r may be written as a product, $R_r = R_r^{(el)} \cdot R_r^{(ph)}$, where $R_r^{(el)}$ and $R_r^{(ph)}$ refer to the two subsystems respectively.

By definition the Abelian base vectors have the transformation properties

$$R_r^{(el)}|\Gamma, \lambda\rangle = \chi(\Gamma, r)|\Gamma, \lambda\rangle \tag{3}$$

and correspondingly

$$\langle\Gamma, \lambda|(R_r^{(el)})^\dagger = \chi^*(\Gamma, r)\langle\Gamma, \lambda|. \tag{4}$$

From the irreducible base we may deduce a generalized 'Wannier' base $\{|r, \lambda\rangle_W\}$, which is also a CONB,

$$|r, \lambda\rangle_W = \frac{1}{\sqrt{N+1}} \sum_\Gamma \chi(\Gamma, r)|\Gamma, \lambda\rangle \tag{5}$$

and the inversion reads

$$|\Gamma, \lambda\rangle = \frac{1}{\sqrt{N+1}} \sum_r \chi^*(\Gamma, r)|r, \lambda\rangle_W. \tag{6}$$

Using definition (5) we may generate the set of Wannier functions from a single one:

$$|r, \lambda\rangle_W = R_r^{(el)}|0, \lambda\rangle_W \tag{7}$$

$$\langle r, \lambda|_W = \langle 0, \lambda|_W (R_r^{(el)})^\dagger. \tag{8}$$

In the phonon subspace we introduce irreducible canonical coordinates $\{Q_\Gamma, P_\Gamma\}$, such that

$$R_r^{(ph)} Q_\Gamma = \chi(\Gamma, r) Q_\Gamma R_r^{(ph)} \tag{9}$$

$$R_r^{(ph)} P_\Gamma = \chi^*(\Gamma, r) P_\Gamma R_r^{(ph)}. \tag{10}$$

Naturally, here we could also consider several irreducible branches (necessitating a branch index μ), but for simplicity of notation we refrain from doing so. Likewise we introduce corresponding canonical variables of 'Wannier' type

$$Q_r = \frac{1}{\sqrt{N+1}} \sum_\Gamma \chi(\Gamma, r) Q_\Gamma \tag{11}$$

$$P_r = \frac{1}{\sqrt{N+1}} \sum_\Gamma (\chi(\Gamma, r))^* P_\Gamma. \tag{12}$$

This establishes the background for introducing the GFGT. It is administered by the operators

$$U_{\mathrm{GFG}} = \frac{1}{\sqrt{N+1}} \sum_{\substack{r,\Gamma \\ \lambda}} \chi^*(\Gamma, r)|r, \lambda\rangle_W \langle\Gamma, \lambda|R_r^{(ph)} \tag{13}$$

$$U_{\mathrm{GFG}}^\dagger = \frac{1}{\sqrt{N+1}} \sum_{\substack{r,\Gamma \\ \lambda}} \chi(\Gamma, r)|\Gamma, \lambda\rangle\langle r, \lambda|_W (R_r^{(ph)})^\dagger. \tag{14}$$

We note that the GFG-operators are written in a 'mixed' projective form, referring both to the irreducible base $|\Gamma, \lambda\rangle$ and to the Wannier base $|r, \lambda\rangle_W$. This presentation most directly leads to diagonalization with respect to the electronic subsystem. For later discussion it is advantageous, however, to write U_{GFG} also in a pure Wannier form. This is achieved if equation (6) is employed. Then

$$U_{\mathrm{GFG}} = \sum_{r,\lambda} |r, \lambda\rangle_W \langle r, \lambda|_W R_r^{(ph)} \tag{15}$$

$$U_{\mathrm{GFG}}^\dagger = \sum_{r,\lambda} |r, \lambda\rangle_W \langle r, \lambda|_W (R_r^{(ph)})^\dagger. \tag{16}$$

The basic transformation properties are given by

$$U_{\mathrm{GFG}}^{\dagger}|r,\lambda\rangle_W = \frac{1}{\sqrt{N+1}}\sum_{\Gamma}\chi(\Gamma,r)|\Gamma,\lambda\rangle(R_r^{(ph)})^{\dagger} = |r,\lambda\rangle_W(R_r^{(ph)})^{\dagger} \tag{17}$$

$$\langle r,\lambda|_W U_{\mathrm{GFG}} = \frac{1}{\sqrt{N+1}}\sum_{\Gamma}\chi^*(\Gamma,r)\langle\Gamma,\lambda|R_r^{(ph)} = \langle r,\lambda|_W R_r^{(ph)} \tag{18}$$

$$T_{\mathrm{GFG}} : |r,\lambda\rangle_W\langle r',\lambda'|_W \equiv U_{\mathrm{GFG}}^{\dagger}|r,\lambda\rangle_W\langle r',\lambda'|_W U_{\mathrm{GFG}} = |r,\lambda\rangle_W\langle r',\lambda'|_W(R_r^{(ph)})^{\dagger}R_{r'}^{(ph)} \tag{19}$$

$$T_{\mathrm{GFG}} : Q_{\Gamma} = \sum_{r,\lambda}\chi^*(\Gamma,r)Q_{\Gamma}|r,\lambda\rangle_W\langle r,\lambda|_W \tag{20}$$

$$T_{\mathrm{GFG}} : F^{(ph)}(Q,P) = \sum_{r,\lambda}|r,\lambda\rangle_W\langle r,\lambda|_W(R_r^{(ph)})^{\dagger}F^{(ph)}(Q,P)R_r^{(ph)} \tag{21}$$

where the notation

$$T_{\mathrm{GFG}} : A = (U_{\mathrm{GFG}})^{\dagger}AU_{\mathrm{GFG}} \tag{22}$$

has been used.

3. Application to a general electron–phonon Hamiltonian

We now turn to the application of the GFGT to an electron–phonon Hamiltonian. The most general form satisfying the assumed Abelian symmetry

$$R_r H = H R_r \tag{23}$$

may be written as

$$H = \sum_r R_r H_0 (R_r)^{\dagger} \qquad R_r = R_r^{(el)}R_r^{(ph)} \tag{24}$$

where

$$H_0 = \sum_{\substack{s\\ \lambda,\lambda'}}|0,\lambda\rangle_W\langle s,\lambda'|_W h^{(ph)}(s,\lambda\lambda') \tag{25}$$

$$= \sum_{\substack{s\\ \lambda,\lambda'}}|0,\lambda\rangle_W\langle 0,\lambda'|_W(R_s^{(el)})^{\dagger}h^{(ph)}(s,\lambda\lambda') \tag{26}$$

and $h^{(ph)}(s,\lambda\lambda')$ is an arbitrary Hamiltonian form in the phonon subspace. Onto this Hamiltonian we apply the GFGT,

$$T_{\mathrm{GFG}} : H \equiv U_{\mathrm{GFG}}^{\dagger}HU_{\mathrm{GFG}} \tag{27}$$

$$= \sum_{\substack{r\\ \lambda,\lambda'}}|r,\lambda\rangle_W\langle r,\lambda'|_W(R_s^{(el)})^{\dagger}h^{(ph)}(s,\lambda\lambda')R_s^{(ph)} \tag{28}$$

$$= \sum_{\substack{\Gamma\\ \lambda,\lambda'}}\sum_{s}|\Gamma,\lambda\rangle\langle\Gamma,\lambda'|h^{(ph)}(s,\lambda\lambda')\chi^*(\Gamma,s)R_s^{(ph)}(Q,P) \tag{29}$$

where the orthonormality relations for the Abelian characters $\chi(\Gamma,r)$

$$\frac{1}{N+1}\sum_{\Gamma}(\chi(\Gamma,r))^*\chi(\Gamma,r') = \delta_{rr'} \tag{30}$$

$$\frac{1}{N+1}\sum_{r}(\chi(\Gamma,r))^*\chi(\Gamma',r) = \delta_{\Gamma\Gamma'} \tag{31}$$

have been employed. Expression (29) is seen to be diagonal in the electronic projectors with respect to the irreducible representation, whence the transformed eigenfunctions may be written in the form

$$T_{\mathrm{GFG}} : \tilde{\Psi}^{(\Gamma)} \equiv \tilde{\Psi}^{(G)} = \sum_{\lambda} |\Gamma\lambda\rangle \Phi^{(\Gamma\lambda)}(Q) \tag{32}$$

which, inserted in the Schrödinger equation pertaining to Hamiltonian (29), leads to the generalized Fulton–Gouterman equations

$$\sum_{\substack{s \\ \lambda'}} \chi^*(\Gamma, s) h^{(ph)}(s, \lambda\lambda') R_s^{(ph)}(Q, P) \Phi^{(\Gamma\lambda')}(Q) = E^{(\Gamma)} \Phi^{(\Gamma\lambda)}(Q) \tag{33}$$

which strictly refers to the phonon subspace $\{Q\}$ only. In particular, if band-coupling terms ($\lambda' \neq \lambda$) are absent or discarded, one ends up with a single Schrödinger equation in the phonon subspace characterized by $\Gamma\lambda$, i.e. by the irreducible representation Γ and the electronic band λ. Thus, if $h^{(ph)}(s, \lambda\lambda') = 0$ for $\lambda \neq \lambda'$, we have

$$\sum_{s} \chi^*(\Gamma, s) h^{(ph)}(s, \lambda\lambda) R_s^{(ph)}(Q, P) \Phi^{(\Gamma\lambda)}(Q) = E^{(\Gamma)} \Phi^{(\Gamma\lambda)}(Q). \tag{34}$$

We add a final supplementary note. If a pure electronic Hamiltonian is considered, i.e. a special form of (25), in which $h^{(ph)}$ is taken as a constant,

$$H_0 = \sum_{\substack{s \\ \lambda,\lambda'}} |0, \lambda\rangle_W \langle s, \lambda'|_W C_{s,\lambda\lambda'} \tag{35}$$

we find

$$\begin{aligned} H_{el} &= \sum_r R_r H_0 R_r^\dagger \\ &= \sum_{\substack{r,s \\ \lambda,\lambda'}} C_{s,\lambda\lambda'} |r, \lambda\rangle_W \langle r, \lambda'|_W (R_s^{(el)})^\dagger \qquad (36) \\ &= \sum_{\substack{s,\Gamma \\ \lambda,\lambda'}} |\Gamma, \lambda\rangle \langle \Gamma, \lambda'| C_{s,\lambda\lambda'} (\chi(\Gamma, s))^* \qquad (37) \end{aligned}$$

the GFGT of which reads

$$T_{\mathrm{GFG}} : H_{el} = \sum_{\substack{s,\Gamma \\ \lambda,\lambda'}} |\Gamma, \lambda\rangle \langle \Gamma, \lambda'| C_{s,\lambda\lambda'} (\chi(\Gamma, s))^* R_s^{(ph)}(Q, P). \tag{38}$$

This shows that, via the operators $R_s^{(ph)}(Q, P)$, the purely electronic nature is lost in the transformed picture. This has been illustrated already in equation (19), where the electronic projectors have acquired a vibrational factor after this transformation. If the electronic base $\{|\Gamma\lambda\rangle\}$ is chosen as eigen-base of the electronic part of the Hamiltonian, we have $C_{s,\lambda\lambda'} = C_{s,\lambda}\delta_{\lambda\lambda'}$, and then

$$H_{el} = \sum_{\Gamma\lambda} E^{(el)}(\Gamma\lambda) |\Gamma\lambda\rangle \langle \Gamma\lambda| \tag{39}$$

$$E^{(el)} = \sum_{s} C_{s,\lambda} (\chi(\Gamma, s))^*. \tag{40}$$

Inverting the latter expression by means of the orthogonality relations of the characters (30) leads to

$$C_{s,\lambda} = \frac{1}{N+1} \sum_{\Gamma} \chi(\Gamma, s) E^{(el)}(\Gamma\lambda) \tag{41}$$

and inserting this into (38) we find

$$T_{\mathrm{GFG}} : H_{el} = \sum_{\Gamma\lambda} |\Gamma, \lambda\rangle\langle\Gamma, \lambda| \frac{1}{N+1} \sum_{\Gamma', s} (\chi(\Gamma, s))^* \chi(\Gamma', s) E^{(el)}(\Gamma', \lambda) R_s^{(ph)}(Q, P). \tag{42}$$

4. GFGT in translational symmetry

One of the simplest Abelian groups is the translational group, the most famous cyclic group. There the three-dimensional group elements R_m may be written as a product of three one-dimensional translation elements:

$$R_{\boldsymbol{m}} = R_{m_1} R_{m_2} R_{m_3} \qquad m_i = a\left(0, \pm 1, \ldots, \pm\frac{N}{2}\right) \tag{43}$$

where a is the lattice vector and the index i represents the three spatial coordinates x, y, z. In this case we may turn to the Bloch notation of irreducible representations,

$$\boldsymbol{q} = (q_1, q_2, q_3) \qquad q_i = \frac{2\pi}{a(N+1)}\gamma_i \qquad \gamma_i = 0, \pm 1, \ldots, \pm\frac{N}{2} \tag{44}$$

and the characters are given by

$$\chi(\boldsymbol{q}, \boldsymbol{m}) = \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\boldsymbol{m}}. \tag{45}$$

The irreducible base functions are Bloch functions

$$|\Gamma, \lambda\rangle \equiv |\boldsymbol{q}\lambda\rangle = \mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{r}} u(\boldsymbol{q}\lambda; \boldsymbol{r}) \tag{46}$$

where $u(\boldsymbol{q}\lambda; \boldsymbol{r})$ is periodic:

$$R_{\boldsymbol{m}}^{(el)} : u(\boldsymbol{q}\lambda; \boldsymbol{r}) \equiv u(\boldsymbol{q}\lambda; \boldsymbol{r} - \boldsymbol{m}) \equiv u(\boldsymbol{q}\lambda; \boldsymbol{r}). \tag{47}$$

Given equation (45), we have

$$R_{\boldsymbol{m}}^{(el)} |\boldsymbol{q}\lambda\rangle = \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\boldsymbol{m}} |\boldsymbol{q}\lambda\rangle. \tag{48}$$

Applied to an arbitrary spatial function the translational elements have the property

$$R_{\boldsymbol{m}}^{(el)} f(\boldsymbol{r}) = f(\boldsymbol{r} - \boldsymbol{m}) R_{\boldsymbol{m}}^{(el)}. \tag{49}$$

The electronic operators may therefore be written in the form

$$R_{\boldsymbol{m}}^{(el)} = \exp\left[\frac{\mathrm{i}}{\hbar}\boldsymbol{m}\boldsymbol{p}\right] \tag{50}$$

where $\boldsymbol{p} = \mathrm{i}\hbar\boldsymbol{\nabla}$ is the electron momentum operator.

Now we introduce the Wannier companion $|\boldsymbol{m}\lambda\rangle_W$ to the Bloch base $|\boldsymbol{q}\lambda\rangle$ by means of the inter-relations

$$|\boldsymbol{m}\lambda\rangle_W = \frac{1}{(N+1)^{3/2}} \sum_{\boldsymbol{q}} \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\boldsymbol{m}} |\boldsymbol{q}\lambda\rangle \tag{51}$$

$$|\boldsymbol{q}\lambda\rangle = \frac{1}{(N+1)^{3/2}} \sum_{\boldsymbol{m}} \mathrm{e}^{+\mathrm{i}\boldsymbol{q}\boldsymbol{m}} |\boldsymbol{m}\lambda\rangle_W. \tag{52}$$

Both the Wannier and the Bloch bases are orthonormal bases:

$$\sum_{\boldsymbol{q}\lambda} |\boldsymbol{q}\lambda\rangle\langle\boldsymbol{q}\lambda| = I^{(el)} \tag{53}$$

$$\sum_{\boldsymbol{m}\lambda} |\boldsymbol{m}\lambda\rangle\langle\boldsymbol{m}\lambda| = I^{(el)} \tag{54}$$

where $I^{(el)}$ is the unity operator in the electronic subspace.

In the phonon subspace we have, in analogy to (51), Wannier partners Q_m and P_m to the phononic Bloch coordinates Q_q and P_q:

$$Q_m = \frac{1}{(N+1)^{3/2}} \sum_q \mathrm{e}^{-\mathrm{i}qm} Q_q \tag{55}$$

$$P_m = \frac{1}{(N+1)^{3/2}} \sum_q \mathrm{e}^{+\mathrm{i}qm} P_q. \tag{56}$$

The inversion is given by

$$Q_q = \frac{1}{(N+1)^{3/2}} \sum_m \mathrm{e}^{+\mathrm{i}qm} Q_m \tag{57}$$

$$P_q = \frac{1}{(N+1)^{3/2}} \sum_m \mathrm{e}^{-\mathrm{i}qm} P_m. \tag{58}$$

The phononic part of the translational operator applied to the phonon operators yields

$$R_m^{(ph)} Q_n = Q_{n+m} R_m^{(ph)} \tag{59}$$

$$R_m^{(ph)} P_n = P_{n+m} R_m^{(ph)} \tag{60}$$

$$R_m^{(ph)} Q_q = \mathrm{e}^{-\mathrm{i}qm} Q_q R_m^{(ph)} \tag{61}$$

$$R_m^{(ph)} P_q = \mathrm{e}^{\mathrm{i}qm} P_q R_m^{(ph)}. \tag{62}$$

Often it is advantageous to use the creation and annihilation operators $b_q^\dagger$ and b_q given by

$$b_q = \sqrt{\frac{\Omega(q)}{2\hbar}}\, Q_q^\dagger + \mathrm{i}\sqrt{\frac{1}{2\hbar\Omega(q)}}\, P_q \tag{63}$$

and we may therefore express the phononic operators Q_q and P_q as follows:

$$Q_q = \sqrt{\frac{\hbar}{2\Omega(q)}}(b_{-q} + b_q^\dagger) = Q_{-q}^\dagger \tag{64}$$

$$P_q = \frac{1}{\mathrm{i}}\sqrt{\frac{\hbar\Omega(q)}{2}}(b_q - b_{-q}^\dagger) = P_{-q}^\dagger. \tag{65}$$

The translation of the annihilation operators amounts to

$$R_m^{(ph)} b_q = \mathrm{e}^{\mathrm{i}qm} b_q R_m^{(ph)}. \tag{66}$$

Since for cyclic symmetry $R_m^{(ph)}$ may now also be written in an exponential form,

$$R_m^{(ph)} \stackrel{\text{def}}{=} \exp\left[\frac{1}{2\hbar} m \sum_q q(Q_q P_q + P_q Q_q)\right] \stackrel{\text{def}}{=} \exp\left[-\mathrm{i}m \sum_q q b_q^\dagger b_q\right] \tag{67}$$

the GFG-operator (13) assumes a particularly simple form. For simplicity of notation we show this for a single 'regular' branch $\{P_q, Q_q\}$ of phonon coordinates, i.e. one which constitutes a 'regular' representation. A generalization to more than a single coordinate branch is straightforward. Then we define

$$U_{\mathrm{GFG}} = \exp\left[\frac{1}{2\hbar} \sum_{m\lambda,q} mq |m\lambda\rangle_W \langle m\lambda|_W (Q_q P_q + P_q Q_q)\right] \tag{68}$$

$$= \exp\left[-\mathrm{i} \sum_{m\lambda,q} mq |m\lambda\rangle_W \langle m\lambda|_W b_q^\dagger b_q\right]. \tag{69}$$

This expression will be the key to establishing the multiparticle form of the GFGT.

For application purposes we supplement the cyclic realization of the GFG-operator in the mixed projective form (13), reading

$$U_{\mathrm{GFG}} = \frac{1}{(N+1)^{3/2}} \sum_{\boldsymbol{m}\lambda,\boldsymbol{q}} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{m}} |\boldsymbol{m}\lambda\rangle_W \langle \boldsymbol{q}\lambda| R_{\boldsymbol{m}}^{(ph)} \tag{70}$$

and in the pure Bloch projection, reading

$$U_{\mathrm{GFG}} = \frac{1}{(N+1)^{3}} \sum_{\boldsymbol{m}\lambda,\boldsymbol{q}\boldsymbol{q}'} \mathrm{e}^{\mathrm{i}(\boldsymbol{q}-\boldsymbol{q}')\boldsymbol{m}} |\boldsymbol{q}'\lambda\rangle \langle \boldsymbol{q}\lambda| R_{\boldsymbol{m}}^{(ph)} \tag{71}$$

as well as in the pure Wannier projection, reading

$$U_{\mathrm{GFG}} = \sum_{\boldsymbol{m}\lambda} |\boldsymbol{m}\lambda\rangle_W \langle \boldsymbol{m}\lambda|_W R_{\boldsymbol{m}}^{(ph)} \tag{72}$$

where $R_{\boldsymbol{m}}^{(ph)}$ is defined in (67).

The basic transformation formulae are given by expressions (17)–(21), rewritten in the appropriate cyclic form:

$$U_{\mathrm{GFG}}^{\dagger} |\boldsymbol{m}\lambda\rangle_W = \frac{1}{(N+1)^{3/2}} \sum_{\boldsymbol{q}} \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\boldsymbol{m}} |\boldsymbol{q}\lambda\rangle (R_{\boldsymbol{m}}^{(ph)})^{\dagger} = |\boldsymbol{m}\lambda\rangle_W (R_{\boldsymbol{m}}^{(ph)})^{\dagger} \tag{73}$$

$$U_{\mathrm{GFG}}^{\dagger} |\boldsymbol{q}\lambda\rangle = \frac{1}{(N+1)^{3/2}} \sum_{\boldsymbol{m}} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{m}} |\boldsymbol{m}\lambda\rangle_W (R_{\boldsymbol{m}}^{(ph)})^{\dagger} \tag{74}$$

$$T_{\mathrm{GFG}} : Q_{\boldsymbol{q}} = \sum_{\boldsymbol{m}\lambda} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{m}} Q_{\boldsymbol{q}} |\boldsymbol{m}\lambda\rangle_W \langle \boldsymbol{m}\lambda|_W \tag{75}$$

$$T_{\mathrm{GFG}} : P_{\boldsymbol{q}} = \sum_{\boldsymbol{m}\lambda} \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\boldsymbol{m}} P_{\boldsymbol{q}} |\boldsymbol{m}\lambda\rangle_W \langle \boldsymbol{m}\lambda|_W \tag{76}$$

$$T_{\mathrm{GFG}} : Q_{\boldsymbol{m}} = \frac{1}{(N+1)^{3}} \sum_{\boldsymbol{n}\lambda,\boldsymbol{q}\boldsymbol{q}'} \mathrm{e}^{\mathrm{i}(\boldsymbol{q}-\boldsymbol{q}')\boldsymbol{m}} Q_{\boldsymbol{m}-\boldsymbol{n}} |\boldsymbol{q}'\lambda\rangle \langle \boldsymbol{q}\lambda| \tag{77}$$

$$T_{\mathrm{GFG}} : P_{\boldsymbol{m}} = \frac{1}{(N+1)^{3}} \sum_{\boldsymbol{n}\lambda,\boldsymbol{q}\boldsymbol{q}'} \mathrm{e}^{\mathrm{i}(\boldsymbol{q}-\boldsymbol{q}')\boldsymbol{m}} P_{\boldsymbol{m}-\boldsymbol{n}} |\boldsymbol{q}'\lambda\rangle \langle \boldsymbol{q}\lambda|. \tag{78}$$

5. Application to an archetypical translationally invariant electron–phonon Hamiltonian

In order to display all possible generalizations of the GFG we treat an archetypical electron–phonon Hamiltonian of translationally invariant nature. The Fröhlich Hamiltonian [13, 14] is contained as a particular limiting case of the given Hamiltonian which consists of four parts:

$$H = H_{el} + H_{ep} + H_{ph} + H_{anh}. \tag{79}$$

The electronic and the phonon–electron parts are generated by the transfer term T_{el} and a potential induced by the interatomic movement of the cores in direction $\boldsymbol{e}$:

$$H_{el} + H_{ep} = T_{el} + \sum_{\boldsymbol{m}} V(\boldsymbol{r} - (\boldsymbol{m} + \boldsymbol{e}Q_{\boldsymbol{m}})) \tag{80}$$

$$= T_{el} + \sum_{\boldsymbol{m}} V(\boldsymbol{r} - \boldsymbol{m}) + \sum_{\boldsymbol{m}} Q_{\boldsymbol{m}} \boldsymbol{e} \boldsymbol{\nabla} V(\boldsymbol{r} - \boldsymbol{m}) + \mathrm{O}(Q^2). \tag{81}$$

Often it is advantageous to use the Fourier transformed potential $V(\boldsymbol{q}, \boldsymbol{G})$. Since the spatial coordinate $\boldsymbol{r}$ is continuous, we need an infinite number of $\boldsymbol{k}$-vectors in the Fourier space to

describe the system adequately. $\boldsymbol{q}$ defines the allowed $\boldsymbol{k}$-vectors of the first Brillouin zone while $\boldsymbol{G}$ is a reciprocal lattice vector

$$V(\boldsymbol{q},\boldsymbol{G}) = \frac{1}{(N+1)^{3/2}} \int_r \frac{\mathrm{d}^3\boldsymbol{r}}{a^3} \mathrm{e}^{-\mathrm{i}(q+G)r} V(\boldsymbol{r}) \tag{82}$$

$$V(\boldsymbol{r}) = \frac{1}{(N+1)^{3/2}} \sum_{\boldsymbol{q},\boldsymbol{G}} \mathrm{e}^{\mathrm{i}(q+G)r} V(\boldsymbol{q},\boldsymbol{G}) \tag{83}$$

where $\boldsymbol{q}$ is given in (44) and $\boldsymbol{G}$ is defined by

$$\boldsymbol{G} = (G_1, G_2, G_3) \qquad G_i = 2\pi \frac{g_i}{a} \qquad g_i = 0, \pm 1, \ldots, \pm\infty. \tag{84}$$

The pure electronic Hamiltonian reads

$$H_{el} = T_{el} + \sum_{\boldsymbol{m}} V(\boldsymbol{r} - \boldsymbol{m}) \tag{85}$$

and the electron–phonon Hamiltonian is given by means of (82):

$$H_{ep} = \mathrm{i} \sum_{\boldsymbol{q},\boldsymbol{G}} \boldsymbol{e}(\boldsymbol{q}+\boldsymbol{G}) \mathrm{e}^{\mathrm{i}(q+G)r} V(\boldsymbol{q},\boldsymbol{G}) Q_{-\boldsymbol{q}} + \mathrm{O}(Q^2). \tag{86}$$

Henceforth, we will neglect second- and higher-order terms in the phonon coordinates. Equation (86) may be divided into two parts:

$$H_{ep} = H_{ep}^{(N)} + H_{ep}^{(U)} \tag{87}$$

where $H_{ep}^{(N)}$ represents the *Normal* processes,

$$H_{ep}^{(N)} = \mathrm{i} \sum_{\boldsymbol{q}} \boldsymbol{e}\boldsymbol{q} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{r}} V(\boldsymbol{q}, \boldsymbol{G} = \boldsymbol{0}) Q_{-\boldsymbol{q}} \tag{88}$$

while $H_{ep}^{(U)}$ displays the *Umklapp* processes,

$$H_{ep}^{(U)} = \mathrm{i} \sum_{\boldsymbol{q},\boldsymbol{G}\neq\boldsymbol{0}} \boldsymbol{e}(\boldsymbol{q}+\boldsymbol{G}) \mathrm{e}^{\mathrm{i}(q+G)r} V(\boldsymbol{q},\boldsymbol{G}) Q_{-\boldsymbol{q}}. \tag{89}$$

The phonon Hamiltonian is formed by the harmonic part

$$H_{ph} = \tfrac{1}{2} \sum_{\boldsymbol{q}} [P_{\boldsymbol{q}}^\dagger P_{\boldsymbol{q}} + \Omega(\boldsymbol{q})^2 Q_{\boldsymbol{q}}^\dagger Q_{\boldsymbol{q}}] = \hbar \sum_{\boldsymbol{q}} \Omega(\boldsymbol{q}) (b_{\boldsymbol{q}}^\dagger b_{\boldsymbol{q}} + \tfrac{1}{2}) \tag{90}$$

and the lowest possible anharmonic one

$$H_{anh} = \sum_{\boldsymbol{q},\boldsymbol{q}',\boldsymbol{q}'',\boldsymbol{G}} K_{\boldsymbol{q}\boldsymbol{q}'\boldsymbol{q}''}^{(\boldsymbol{G})} Q_{\boldsymbol{q}} Q_{\boldsymbol{q}'} Q_{\boldsymbol{q}''} \delta(\boldsymbol{q} + \boldsymbol{q}' + \boldsymbol{q}'' - \boldsymbol{G}). \tag{91}$$

We now choose the eigen-base of H_{el} as an electronic base for our further calculations,

$$H_{el} |\boldsymbol{q}\lambda\rangle = E^{(el)}(\boldsymbol{q},\lambda) |\boldsymbol{q}\lambda\rangle. \tag{92}$$

The transformed electronic Hamiltonian is given by (42),

$$T_{\mathrm{GFG}} : H_{el} = \sum_{\boldsymbol{q}\lambda} |\boldsymbol{q},\lambda\rangle\langle\boldsymbol{q},\lambda| h_{el}(\boldsymbol{q},\lambda) \tag{93}$$

where

$$h_{el}(\boldsymbol{q},\lambda) = \frac{1}{(N+1)^3} \sum_{\boldsymbol{q}',\boldsymbol{m}} \mathrm{e}^{\mathrm{i}(q-q')m} E^{(el)}(\boldsymbol{q}',\lambda)\, R_{\boldsymbol{m}}^{(ph)}(Q,P) \tag{94}$$

$$= E^{(el)}\left(\boldsymbol{q} - \sum_{\boldsymbol{q}'} \boldsymbol{q}' b_{\boldsymbol{q}'}^\dagger b_{\boldsymbol{q}}', \lambda\right) \tag{95}$$

and where $E^{(el)}(\boldsymbol{q},\lambda)$ is introduced in equation (92). By means of the GFG given in (72) we calculate the transformed forms of the remaining parts of the considered Hamiltonian as follows:

$$T_{\mathrm{GFG}}: H_{ep}^{(N)} = \sum_{\boldsymbol{q},\lambda\lambda'} |\boldsymbol{q},\lambda\rangle\langle\boldsymbol{q},\lambda'|h_{ep}^{(N)}(\boldsymbol{q},\lambda\lambda') \tag{96}$$

where

$$h_{ep}^{(N)}(\boldsymbol{q},\lambda\lambda') = \sum_{\boldsymbol{n},\boldsymbol{q}'} \mathrm{i}e\boldsymbol{q}' V(\boldsymbol{q}',\boldsymbol{G}=\boldsymbol{0})\mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{n}}\langle\boldsymbol{0}\lambda|_W \mathrm{e}^{\mathrm{i}\boldsymbol{q}'\boldsymbol{r}}|\boldsymbol{n}\lambda'\rangle_W Q_{-\boldsymbol{q}'} R_{\boldsymbol{n}}^{(ph)}(Q,P) \tag{97}$$

and

$$T_{\mathrm{GFG}}: H_{ep}^{(U)} = \sum_{\boldsymbol{q},\lambda\lambda'} |\boldsymbol{q},\lambda\rangle\langle\boldsymbol{q},\lambda'|h_{ep}^{(U)}(\boldsymbol{q},\lambda\lambda') \tag{98}$$

where

$$h_{ep}^{(U)}(\boldsymbol{q},\lambda\lambda') = \sum_{\boldsymbol{n},\boldsymbol{q}'}\sum_{\boldsymbol{G}\neq\boldsymbol{0}} \mathrm{i}e(\boldsymbol{q}'+\boldsymbol{G})V(\boldsymbol{q}',\boldsymbol{G})\mathrm{e}^{\mathrm{i}\boldsymbol{q}\boldsymbol{n}}\langle\boldsymbol{0}\lambda|_W \mathrm{e}^{\mathrm{i}(\boldsymbol{q}'+\boldsymbol{G})\boldsymbol{r}}|\boldsymbol{n}\lambda'\rangle_W Q_{-\boldsymbol{q}'} R_{\boldsymbol{n}}^{(ph)}(Q,P) \tag{99}$$

and trivially

$$T_{\mathrm{GFG}}: H_{ph} = H_{ph} \tag{100}$$

$$T_{\mathrm{GFG}}: H_{anh} = H_{anh}. \tag{101}$$

Thus we are able to write the transformed Hamiltonian in the lucid form

$$T_{\mathrm{GFG}}: H = \sum_{\boldsymbol{q},\lambda\lambda'} |\boldsymbol{q},\lambda\rangle\langle\boldsymbol{q},\lambda'|h(\boldsymbol{q},\lambda\lambda') \tag{102}$$

where

$$h(\boldsymbol{q},\lambda\lambda') = h_{el}(\boldsymbol{q},\lambda)\delta_{\lambda\lambda'} + h_{ep}^{(N)}(\boldsymbol{q},\lambda\lambda') + h_{ep}^{(U)}(\boldsymbol{q},\lambda\lambda') + (H_{ph}+H_{anh})\delta_{\lambda\lambda'}. \tag{103}$$

6. The Fulton–Gouterman equations (FGEs)

Since we have now translated the Hamiltonian (79) into a form which is diagonal with respect to the $\boldsymbol{q}$-vector of the electronic sub-base $\{|\boldsymbol{q}\lambda\rangle\}$, the original Schrödinger equation is separated into a sequence of phononic equations, each of which, respectively, is related to one of the electronic $\boldsymbol{q}$-vectors:

$$\sum_{\lambda'} h(\boldsymbol{q},\lambda\lambda')\Phi^{(\boldsymbol{q}\lambda')}(Q) = E^{(\boldsymbol{q})}\Phi^{(\boldsymbol{q}\lambda)}(Q) \qquad \lambda = 0,\pm1,\pm2,\ldots. \tag{104}$$

These are called the Fulton–Gouterman equations (FGEs). The FGEs are totally equivalent to the Schrödinger equation in the transformed picture

$$(T_{\mathrm{GFG}}: H)\tilde{\Psi}^{(\boldsymbol{q})} = E^{(\boldsymbol{q})}\tilde{\Psi}^{(\boldsymbol{q})} \tag{105}$$

where due to the diagonal form of the transformed Hamiltonian the wavefunction $\tilde{\Psi}^{(\boldsymbol{q})}$ is given as a superposition of direct products of the electronic part $|\boldsymbol{q}\lambda\rangle$ and the phononic part $\Phi^{(\boldsymbol{q}\lambda)}(Q)$,

$$\tilde{\Psi}^{(\boldsymbol{q})} = \sum_{\lambda} |\boldsymbol{q}\lambda\rangle\Phi^{(\boldsymbol{q}\lambda)}. \tag{106}$$

We still are left with a summation over the electronic band index λ.

At this stage it appears useful to illustrate the remaining implications within the given set of FGEs. To this end we consider a perturbative approach, where the unperturbated problem is given by

$$h(\boldsymbol{q}, \lambda\lambda)\varphi_j^{(\boldsymbol{q}\lambda)} = \epsilon_j^{(\boldsymbol{q}\lambda)}\varphi_j^{(\boldsymbol{q}\lambda)} \tag{107}$$

j denoting the quantum number. We define the matrix elements

$$W_{jj'}^{(\lambda\lambda')} \equiv \left\langle \varphi_j^{(\boldsymbol{q}\lambda)} \middle| \sum_{\lambda''\neq\lambda} h(\boldsymbol{q}, \lambda\lambda'') \middle| \varphi_{j'}^{(\boldsymbol{q}\lambda')} \right\rangle \tag{108}$$

involving the perturbation operator

$$\sum_{\lambda''\neq\lambda} h(\boldsymbol{q}, \lambda\lambda''). \tag{109}$$

We thus receive the series for the energy

$$E_j^{(\boldsymbol{q}\lambda)} = \epsilon_j^{(\boldsymbol{q}\lambda)} + W_{jj}^{\lambda\lambda} + \sum_{\substack{\lambda'\neq\lambda \\ j}} \frac{|W_{jj'}^{\lambda\lambda'}|^2}{\epsilon_j^{(\boldsymbol{q}\lambda)} - \epsilon_{j'}^{(\boldsymbol{q}\lambda')}} + \mathrm{O}(|W|^3) \tag{110}$$

and for the phononic wavefunction

$$\Phi_j^{(\boldsymbol{q}\lambda)}(Q) = \varphi_j^{(\boldsymbol{q}\lambda)} + \sum_{\substack{\lambda'\neq\lambda \\ j}} \frac{W_{jj'}^{\lambda\lambda'}}{\epsilon_{j'}^{(\boldsymbol{q}\lambda')} - \epsilon_j^{(\boldsymbol{q}\lambda)}} \varphi_{j'}^{(\boldsymbol{q}\lambda')} + \mathrm{O}(|W|^2). \tag{111}$$

In general these series tend to converge quickly, since the energies $\epsilon_j^{(\boldsymbol{q}\lambda)}$ in the denominators of the last terms in (110) and (111) are owing to different bands with the same $\boldsymbol{q}$-vector, which in most cases differ sufficiently. In view of that even the zeroth order wavefunctions will in general prove to be a rather good approximation.

7. Antagonistic physical tendencies in the FGEs

In this section we want to illustrate the topological aspects inherent in the FGEs. The antagonistic tendencies of the coupling and the electronic term in each FGE is easily seen in a simple translationally invariant model. For the sake of lucidity we limit our consideration to a one-dimensional one-band Hamiltonian of the Fröhlich type,

$$H = H_{ph} + H_{el} + H_{ep} \tag{112}$$

$$H_{ph} = \tfrac{1}{2}\sum_q [P_q^+ P_q + \Omega^2(Q) Q_q^+ Q_q] \tag{113}$$

$$H_{el} = -\sum_m T[|m\rangle\langle m+1| + |m+1\rangle\langle m|] \tag{114}$$

$$H_{ep} = \sum_{m,q} V_q Q_{-q} \mathrm{e}^{-\mathrm{i}qm} |m\rangle\langle m|. \tag{115}$$

The respective FG-operator has the form

$$U_{\mathrm{FG}} = \frac{1}{N^{1/2}} \sum_{m,q} \mathrm{e}^{-\mathrm{i}qm} |m\rangle\langle q| R_m^{(ph)}(Q, P) \tag{116}$$

with

$$R_m^{(ph)}(Q, P) = \exp\left[-\mathrm{i}m \sum_q q b_q^+ b_q \right] \tag{117}$$

as in (67). The transformed Hamiltonian then reads

$$T_{\mathrm{FG}} : H = \sum_{q} |q\rangle\langle q| h(q) \tag{118}$$

with

$$h(q) = H_{ph} - T(\mathrm{e}^{\mathrm{i}q} R_1^{(ph)}(Q, P) + \mathrm{e}^{-\mathrm{i}q} R_{-1}^{(ph)}(Q, P)) + \sum_{q'} V_{q'} Q_{-q'}. \tag{119}$$

The FGE

$$h(q)\Phi^{(q)}(Q) = E^{(q)}\Phi^{(q)}(Q) \tag{120}$$

constitutes an eigenvalue problem in the oscillatory subspace and is totally equivalent to the original full Schrödinger equation. In (119) the last term, i.e. the transformed coupling term, tends to provoke a displacement of the oscillatory equilibrium positions and exerts a polaronic (selftrapping) effect onto the electrons. On the other hand, the transformed transfer term in (119) tends to delocate the electron due to the phononic translational operator $R_1^{(ph)}(Q, P)$.

The combination of these antagonistic tendencies may become effective in a different manner for different energy regimes. In any case the most straightforward way to establish trial functions for the solution of equation (120) is by means of phononic unitary transformations,

$$\Phi^{(q)}(Q) = U(Q, P)\Phi^{(q,0)}(Q) \tag{121}$$

where $U^{+} = U^{-1}$ and where $\Phi^{(q,0)}(Q)$ may be taken as an eigenfunction of the undisturbed phonon Hamiltonian H_{ph}. The parameters inherent in the chosen unitary operator $U(Q, P)$ may be fixed by minimizing the energy of the ground state, or by applying the Peierls–Bogoliubov inequality.

In previous years several approaches to establishing solutions of single-band FGEs have been performed [5–11]. In simpler systems, like the dimer or trimer systems [5–7] and in the $E \times e$-Jahn-Teller problem [12] it turns out that there are two fundamental types of phononic wavefunctions. For low energies the mirror-image replica of the displaced oscillator function provokes an effective broadening of the wavefunction and thus is tantamount to a 'softening' of the vibrations. By contrast, at higher energies, other types of states also make their appearance, which amount to squeezed oscillatory undisplaced functions, i.e. the vibrations are getting 'harder'. For more details we refer to the originals. For extended electron–phonon systems the features described are present in a modified way [8, 9].

8. Results and perspectives

The main aims of the present work are, on the one hand, to formulate of the multi-band form of the generalized Fulton–Gouterman transformation (GFGT), and on the other hand, to establish an exponential form of the transformation operator.

In the past the single-band FGT has proven useful for the solution of many archetypical theoretical models. However, for extended electron–phonon or exciton–phonon systems a multi-band form is highly desirable. Equally desirable is an exponential form of the transformation operator, since this establishes the possibility to extend the formalism to more than a single electron (exciton). Both are achieved in the present paper. The extension to multi-electron solutions will be considered in a future work.

Another perspective of the multi-band form of the GFGT is the option to compare the virtues of this transformation with those of the famous Lee–Low–Pines transformation

(LLPT) [1]. In a forthcoming paper it will be shown that only in the case of linear electron–phonon coupling, and only if Umklapp-terms [15, 16] are neglected, the LLPT displays advantages over the GFGT, whereas for Umklapp processes as well as for a more general electron–phonon coupling the GFGT turns out to be preferable.

As regards the multi-particle formulation, one is confronted with the difficulty that the second-quantized version of the LLP operator is no longer unitary, whereas there is no such problem in the GFG case. Although in the multi-particle case electronic diagonalization is no longer achieved, the application of the GFGT seems to have a physical perspective for the recent revival of the bi-polaron problem in the context of high-T_C superconductivity [17], as well as for the polaron charge density waves [18–20].

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