Geometrical and dynamical aspects of the correlated electron pair emission from ordered materials

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Abstract.

The scattering of N-body correlated systems from a non-overlapping multi-center periodic potential is formulated in terms of scattering path operators. An expansion of the total T-matrix in terms of scattering path operators is derived. Special attention is devoted to the separation of the geometrical arrangement of the scatterers from the internal correlation of the scattered compound. The role of internal correlation is illustrated in the case of the scattering of correlated electron pairs from a perfect crystal potential and the electron-pair excitation from core levels into the vacuum upon a photon absorption.

1. INTRODUCTION

The single particle scattering from an ordered multi-center potential is of a fundamental importance in solid state physics. Many structural and electronic properties of materials are deduced from the scattered electron flux, e.g. as done in low energy electron diffraction [1] and single photoemission [2]. Correspondingly, a number of reliable theoretical concepts have been put forward to deal with the single particle scattering. Of special importance for the present work is the scattering-path formalism [3, 4], as outlined below. In contrast, the scattering of correlated systems from crystals are much less understood. This is due to the fact that a number of material properties are adequately described within a single-particle picture and the non-separability introduced by the correlated scattered systems makes a theoretical treatment much more involved. It is the aim of this study to develop a systematic framework for the propagation of correlated compounds under the action of a non-overlapping muffin-tin crystal potential. This renders possible the treatment of correlated electron-pair scattering from a crystal potential. Such scattering states are needed for the description of two-orbital excitation upon photon absorption or charged particle impact.

2. THEORETICAL TREATMENT

At first, we formulate the single-particle scattering from a multi-center potential in terms of scattering path operators. The method is then generalized to correlated multi-particle scattering from a crystal potential.

Let us consider an effective one-particle system, characterized by a Hamiltonian H_0, being prepared in a defined state $|\phi\rangle$. The question of concern here is how the system evolves if subjected to the external perturbation W, i. e. what is the eigenstate $|\Psi\rangle$ of the Hamiltonian $H_0 + W$?. The calculational scheme for the answer is provided by the resovlent G_0 (the Green operator) of H_0 that leads to the integral equation

$$|\Psi^{\pm}(\epsilon)\rangle = |\phi(\epsilon)\rangle + G_0^{\pm}(\epsilon) W |\Psi^{\pm}(\epsilon)\rangle \tag{1}$$

where G_0^+ (G_0^-) is the advanced (retarded) Green function of the system H_0. Alternatively one can express the state $|\Psi^\pm(\epsilon)\rangle$ in terms of the unperturbed state $|\phi\rangle$ using the transition operator T

$$|\Psi^\pm(\epsilon)\rangle = \left[\mathbf{1} + G_0^\pm(\epsilon)T^\pm(\epsilon)\right]|\phi(\epsilon)\rangle. \tag{2}$$

From this equation it is apparent that the response of the system to the perturbation W is entirely described by the operator T. In fact T can be written in the integral form

$$T^+(\epsilon) = W + WG_0^\pm(\epsilon)T^\pm(\epsilon). \tag{3}$$

Thus, the problem is reduced to finding appropriate solution to the integral equation (3). Let us consider the case where W is a crystal potential consisting of a superposition of M individual non-overlapping core potentials w_i centered at sites $\mathbf{R}_i$ with domains Ω_i (in configuration space), i. e.

$$W = \sum_i^M w_i,\ \Omega_i \cap \Omega_j = 0,\ \forall j \neq i. \tag{4}$$

The T operator is then,

$$T = \sum_i^M (w_i + w_i G_0 T) = \sum_i^M Q_i \tag{5}$$

where (for brevity we omit the superscripts)

$$Q_i = w_i + w_i G_0 T = w_i + w_i G_0 Q_i + \sum_{j\neq i}^M w_i G_0 Q_j. \tag{6}$$

Noting that $T = (\mathbf{1} - WG_0)^{-1}W$, Eq.(6) can be rewritten as

$$Q_i = t_i + \sum_{j\neq i}^M t_i G_0 Q_j \tag{7}$$

where t_i is the single-site transition operator $t_i = w_i + w_i G_0 t_i$. Thus, combining Eq.(7) and Eq.(5) yields

$$T = \sum_i^M t_i + \sum_{j\neq i}^M t_i G_0 (t_j + w_j G_0 T) \tag{8}$$

Introducing the *scattering path operators*, τ^{ij} as

$$\tau^{ij} = t_i\delta_{ij} + \sum_{k\neq i}^M t_i G_0 \tau^{ik} = t_i\delta_{ij} + \sum_{k\neq i}^M \tau^{ik} G_0 t_i \tag{9}$$

and summing over j and comparing with Eq.(7) we can write

$$Q_i = \sum_j \tau^{ij} \tag{10}$$

$$T = \sum_i Q_i = \sum_{ij} \tau^{ij}. \tag{11}$$

The operator Q_i describes the transition of the state $|\phi\rangle$ under the action of w_i that is localized at the site $\mathbf{R}_i$ in the presence of all w_j, $j \neq i$. The operators τ^{kl}, first introduced by Gryorffy [3, 4], describes the transition of a state $|\phi'\rangle$ under the action of w_k, where $|\phi'\rangle$ evolves from $|\phi\rangle$ after being subjected to w_l. Hence, in Eq.(11) the transition of $|\phi\rangle$ due to the perturbation W is broken down into successive on-site transitions that are computationally more accessible.

Here we derive a procedure analogous to the one outlined above for the case where the state $|\phi\rangle$ describes N interacting particles. The Hamiltonian of the system is $H = K + \sum_{j>i}^{N} u_{ij} + \sum_{k}^{M}\sum_{l}^{N} w_{kl} = U_{int} + W_{ext}$ where u_{ij} describes the interaction of particle i with particle j in the scattering compound, w_{kl} is the interaction potential of particle l with the potential centered at the site $\mathbf{R}_k$ and K is the kinetic energy operator. The idea now is to decouple dynamical properties due to W_{ext} from those due to W_{int}. To do that we define $w^k := \sum_l w_{kl}$ (the interaction of all N particles with the site $\mathbf{R}_k$) and assume $|\chi\rangle$ to be the eigenfuntion of $K + U_{int}$. As done in Eq.(6) we can write

$$q^k_{ext} = w^k + w^k G_0 q^k_{ext} + \sum_{l\neq k}^{M} w^k G_0 q^l_{ext} \tag{12}$$

with $T_{ext} = \sum_k^M q^k_{ext}$. Following the steps (7,8,11) one can write explicit expressions for the scattering path operator of the *whole* systems

$$\tau^{ij} = \bar{t}_i \delta_{ij} + \sum_{k\neq i}^{M} \bar{t}_i G \tau^{ik}_{ext} \tag{13}$$

where $\bar{t}_k = w^k + w^k G_0 \bar{t}_k$ is the scattering of the system from the single site k. Eq. (13) describes the scattering of the system, as a whole, from the multi-center potential W_{ext}. The internal response of the system due to the interaction U_{int} is described by $|\chi\rangle$ (the eigenfuntion of $K + U_{int}$. Noting that $|\chi\rangle$ can be written in the approximate form $|\chi\rangle = \prod_{j>i}^{N}(1 + u_{ij}g_{ij})|\phi\rangle$ where g_{ij} is the Green operator within the two-body potential u_{ij} and $|\phi\rangle$ is an asymptotic state of the operator K we substitute $|\chi\rangle$ and $T = \sum_{ij}\tau^{ij}$ into (2) and obtain the final expression for $|\Psi\rangle$. The physical picture underlying the above method is that the system as whole is considered as a quasi single particle that is scattered from the crystal potential W_{ext} after each scattering events the systems "relaxes" according to its internal degrees of freedom, as dictated by U_{int}.

For the treatment of the one-photon excitation of a correlated electron pair from a core level into the vacuum we assume a frozen core approximation for all degrees of freedom of the target except for those of the electron pair and apply the above method. The dipole transition amplitude $M_{fi}(\mathbf{k}_a, \mathbf{k}_b)$ for the emission of the two electrons with asymptotic wave vectors $\mathbf{k}_a$ and $\mathbf{k}_b$ reads [5]

$$|M_{fi}|^2 = \hat{\mathbf{e}} \cdot (\mathbf{k}_a + \mathbf{k}_b)\, \delta^{(3)}_{(\mathbf{q}_i - \mathbf{q}_f, \mathbf{g})} \mathcal{L} \tag{14}$$

where $\mathbf{q}_i = \mathbf{k}'_a + \mathbf{k}'_b$, $\mathbf{q}_f = \mathbf{k}_a + \mathbf{k}_b$ are the wave vectors of the pair's center-of-mass in the initial and final state, respectively, $\mathbf{g}$ is a bulk reciprocal lattice vector and $\hat{\mathbf{e}}$ is the polarization vector of the photon. The function $\mathcal{L}$ depends on the state from which the pair has been excited. From Eq. (14) two important conclusion are drawn which are the consequence of the above calculational scheme

a) According to the von Laue-like condition in Eq. (14), diffraction of the pair occurs when the center-of-mass momentum of the pair changes by a bulk reciprocal vector due to the emission process. This (and the selection rules stated below) is equivalent to assuming the pair as a quasi particle with momentum $\mathbf{k}_a + \mathbf{k}_b$ (the pair's center-of-mass momentum) and performing diffraction from the crystal potential

b) The selection rules can be summarized in the equation $\hat{\mathbf{e}} \cdot (\mathbf{k}_a + \mathbf{k}_b) = 0$, i. e. double-photo emission is forbidden if the momentum of the two-electron center-of-mass is perpendicular to the polarization vector $\hat{\mathbf{e}}$ or if $\mathbf{k}_a = -\mathbf{k}_b$. The influence of the internal degrees of freedom of the pair (the electronic repulsion) are described by the function $\mathcal{L}$.

For the description of the photo-double excitation from the valence band similar conclusions are drawn and are substantiated by experimental findings [6, 7].

3. CONCLUSIONS

Summarizing we presented a scattering path formalism for the scattering of correlated systems from a multi-center potential. The response of the system due to the internal correlation is disentangled from the scattering of the system as a whole from the external multi-center potential. The method is illustrated for the one photon excitations of an electron pair from core levels.

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