

THE PARTICLE-PHOTON INTERACTION IN SYSTEMS DESCRIBED BY MODEL HAMILTONIANS IN SECOND QUANTIZATION

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A simple replacement rule which allows the direct formulation in second quantization of the particle-photon interaction Hamiltonian in the velocity gauge is presented. We show that a transformation of the creation and annihilation operators of the particle Hamiltonian gives the interaction Hamiltonian. The rule is valid for both local and nonlocal Hamiltonians and gives the correct expression of the photon self-interaction term A^2 also when only a projection of the particle Hamiltonian is taken into account. As an example the replacement rule is applied to the exciton operators in order to derive the polariton Hamiltonian.

Keywords: A. quantum wells, D. optical properties, D. radiation effects, E. nonlinear optics.

1. INTRODUCTION

FOR SYSTEMS described by many-body Hamiltonians, a reduction to effective Hamiltonians H_0 is often necessary to perform explicit calculations. Effective Hamiltonians may contain nonlocal potentials and in many cases are appropriate projections of the exact Hamiltonian on an incomplete set of states. In this case the second quantization formalism is very useful allowing the expression of a Hamiltonian as a sum of matrix elements of a complete basis set. In fact, choosing the appropriate basis set and taking into account the relevant part of the sum one obtains the projection of the Hamiltonian which has to be diagonalized.

In the Hamiltonian formulation, the particle-field interaction depends on the choice of the gauge. Of course all physical results must be independent of this choice. Starace [1] has shown that the standard form of the interaction $-(e/mc)\mathbf{A} \cdot \mathbf{p}$ between electrons and the radiation field is no longer valid for Hamiltonians with nonlocal potentials, and in order to preserve the gauge invariance of multiphoton transition rates it has been shown in [2] that correction terms due to the nonlocal character of the potentials have to be added

to the standard electron-photon interaction. The interaction Hamiltonian in the length gauge, in the dipole approximation $-q\mathbf{x} \cdot \mathbf{E}(0)$, is not affected by the presence of nonlocal potentials, this gauge however cannot be conveniently used in the quantum theory of systems in which the field is strongly modified by the interaction with the material system as in the case of the strong correlation of the polarization of a crystal with photons giving rise to polaritons.

In this paper we present a simple and general replacement rule for the creation and annihilation operators of the particle system which allows the direct formulation of the interaction Hamiltonian in the velocity gauge. We show that the particle Hamiltonian in interaction with the field can be obtained by considering a unitary transformation of the unperturbed Hamiltonian. This allows the direct formulation in second quantization of the particle-photon interaction by means of a simple and general replacement rule for the creation and annihilation operators of the particle system. In this scheme, once the appropriate projection of the Hamiltonian has been chosen by selecting a set of states, it is sufficient to apply the rule to the corresponding operators in order to obtain the correct form of the Hamiltonian H_0 .

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In many cases terms of the exact Hamiltonian which have been neglected in order to obtain a model Hamiltonian are introduced after the diagonalization of H_0 and are evaluated in the framework of perturbation theory. The rule allows the evaluation of the effect of the field in such terms; in fact the rule is also valid for non-diagonal terms in the Hamiltonian and for terms which describe two particle scattering events.

2. THE INTERACTION WITH THE ELECTROMAGNETIC FIELD OF SYSTEMS WITH NONLOCAL POTENTIALS

In the velocity gauge, the interaction Hamiltonian of a systems of N charged particles in the electromagnetic field is obtained by replacing the kinetic energy of the particles

$$\sum_{i=1}^N \frac{p_i^2}{2m}$$

by the expression

$$\sum_{i=1}^N \frac{1}{2m} \left(\mathbf{p}_i - \frac{q}{c} \mathbf{A}(\mathbf{x}_i) \right)^2,$$

where the sum is over the particles, q is the value of the charge, c the velocity of light and $\mathbf{A}(\mathbf{x})$ is the vector potential of the electromagnetic field. The interaction Hamiltonian thus obtained is given by

$$H_I = -\frac{q}{mc} \sum_i \mathbf{A}(\mathbf{x}_i) \cdot \mathbf{p}_i + \frac{q^2}{2mc^2} \sum_i \mathbf{A}^2(\mathbf{x}_i). \quad (1)$$

For systems described by many body Hamiltonians, a reduction to effective Hamiltonians is necessary in order to perform explicit calculations. Effective Hamiltonians may contain nonlocal potentials which can always be expressed as a local momentum dependent operator [2]

$$H_0 = \sum_i H_0(\mathbf{x}_i, \mathbf{p}_i) = \sum_i \frac{p_i^2}{2m} + \sum_i V(\mathbf{x}_i, \mathbf{p}_i), \quad (2)$$

where, for simplicity, we have considered only one particle operators. In this case the particle Hamiltonian in the presence of an electromagnetic field takes the form

$$H = \sum_i \frac{1}{2m} \left(\mathbf{p}_i - \frac{q}{c} \mathbf{A}(\mathbf{x}_i) \right)^2 + \sum_i V(\mathbf{x}_i, \mathbf{p} - \frac{q}{c} \mathbf{A}(\mathbf{x}_i)) \quad (3)$$

as pointed out by Starace [1]. In order to treat the interaction by perturbation theory, following [2], the

Hamiltonian (3) is expanded into a power series in the vector potential. The interaction Hamiltonian obtained up to the second order in the vector potential, in the dipole approximation is

$$H_I = -\frac{q}{c} \sum_i \mathbf{A}(\mathbf{x}_i = 0) \cdot \mathbf{v}_i + \frac{1}{2i\hbar} \frac{e^2}{c^2} \sum_i [\mathbf{A}(\mathbf{x}_i = 0) \cdot \mathbf{x}_i, \mathbf{A}(\mathbf{x}_i = 0) \cdot \mathbf{v}_i], \quad (4)$$

where \mathbf{v}_i is the i th electron velocity given by

$$\mathbf{v}_i = \frac{1}{i\hbar} [\mathbf{x}_i, H_0]$$

and the commutation relation

$$\frac{\partial g(\mathbf{x}, \mathbf{p})}{\partial \mathbf{p}} = \frac{1}{i\hbar} [\mathbf{x}, g(\mathbf{x})]$$

has been used to express the coefficients of the power series in terms of commutators. Expression (4) is valid for both local and nonlocal potentials, in fact for local potentials $\mathbf{v}_i = \mathbf{p}_i/m$ and equation (4) reduces to the usual form of the interaction (1).

It is possible to derive equation (4) as well as higher order terms in a different way which will be useful when introducing the second quantization formalism. To this aim we consider the relation

$$e^{if(\mathbf{x})} g(\mathbf{x}, \mathbf{p}) e^{-if(\mathbf{x})} = g\left(\mathbf{x}, \mathbf{p} - \frac{\partial f(\mathbf{x})}{\partial \mathbf{x}}\right), \quad (5)$$

which can be proved by applying the commutation rules between the coordinate and its conjugate momentum [4].

If the interaction of the particles with the magnetic field is negligible, we can define a scalar function $\chi(\mathbf{x})$ such that

$$\frac{\partial \chi(\mathbf{x})}{\partial \mathbf{x}} = \mathbf{A}(\mathbf{x}). \quad (6)$$

In the standard dipole approximation, the scalar function $\chi(\mathbf{x}) = \mathbf{x} \cdot \mathbf{A}(0)$ does the job. We can perform the minimal coupling replacement by applying the following unitary transformation to the unperturbed Hamiltonian H_0

$$UH_0U^{-1}, \quad (7)$$

where

$$U(\mathbf{x}) = \exp\left(\frac{iq}{c\hbar} \sum_i \chi(\mathbf{x}_i)\right). \quad (8)$$

By applying the relations (5) and (6) we obtain

$$UH_0U^{-1} = \sum_i H_0\left(\mathbf{x}_i, \mathbf{p}_i - \frac{q}{c} \mathbf{A}(\mathbf{x})\right) = H. \quad (9)$$

Relation (9) makes it possible to expand H in terms of the function $\chi(\mathbf{x})$. By applying the following operator property [3] between the two non-commuting operators

$$e^{\lambda B} A e^{-\lambda B} = A + [B, A] + \frac{\lambda^2}{2!} [B, [B, A]] + \dots,$$

we obtain

$$H = H_0 + \frac{iq}{c\hbar} \left[\sum_i \chi(\mathbf{x}_i), H_0 \right] + \left(\frac{iq}{c\hbar} \right)^2 \left[\sum_i \chi(\mathbf{x}_i), \left[\sum_i \chi(\mathbf{x}_i), H_0 \right] \right] + \dots \quad (10)$$

By inserting $\chi(\mathbf{x}) = \mathbf{x} \cdot \mathbf{A}(0)$ into this expression one can easily show the equivalence of the interaction terms in equation (10) with expression (4). As a concluding remark to this section, we would like to stress that the gauge invariance of the expression (9) is straightforward. Under a gauge transformation the total Hamiltonian H becomes

$$H' = U' U H_0 U^{-1} U'^{\dagger} + i\hbar \dot{U}' U'^{\dagger}, \quad (11)$$

by choosing $U' = U^{\dagger}$ the transformed Hamiltonian, in the dipole approximation, is the particle-photon Hamiltonian in the length gauge $H' = H_0 - q \sum_i \mathbf{x}_i \cdot \mathbf{E}(\mathbf{x}_i = 0)$.

3. THE PARTICLE-PHOTON INTERACTION HAMILTONIAN IN SECOND QUANTIZATION

The particle Hamiltonian (2) in its second quantization form is

$$H_0 = \int \Psi^{\dagger}(\mathbf{x}) \left(\frac{p^2}{2m} + V(\mathbf{x}, \mathbf{p}) \right) \Psi(\mathbf{x}) d^3x, \quad (12)$$

with the field operators satisfying the boson or fermion commutation rules. Expanding the field operator $\Psi(\mathbf{x})$ and its conjugate $\Psi^{\dagger}(\mathbf{x})$ in terms of this basis set

$$\Psi(\mathbf{x}, t) = \sum_n c_n(t) \phi_n(\mathbf{x}),$$

$$\Psi^{\dagger}(\mathbf{x}, t) = \sum_n c_n^{\dagger}(t) \phi_n^*(\mathbf{x}),$$

where $c_n(t)$ and $c_n^{\dagger}(t)$ are operators with their own commutation relations, the Hamiltonian takes the form

$$H_0 = \sum_{m,n} c_m^{\dagger} c_n \langle \phi_m | H_0 | \phi_n \rangle. \quad (13)$$

If ϕ_n are eigenstates of H_0 with eigenvalues $\hbar\omega_n$ the Hamiltonian (13) becomes

$$H_0 = \sum_n \hbar\omega_n c_n^{\dagger} c_n. \quad (14)$$

The particle Hamiltonian in the presence of an electromagnetic field can be expressed in second quantization as

$$H = \int \Psi^{\dagger}(\mathbf{x}) U(\mathbf{x}) \left(\frac{p^2}{2m} + V(\mathbf{x}, \mathbf{p}) \right) U^{-1}(\mathbf{x}) \Psi(\mathbf{x}) d^3x \\ = \sum_{n,n'} c_n^{\dagger} c_{n'} \langle \phi_n | U H_0 U^{-1} | \phi_{n'} \rangle \quad (15)$$

by introducing the complete set of states $|\phi_m\rangle$ we obtain the expression

$$H_0 = \sum_{m,n} \langle \phi_m | H_0 | \phi_n \rangle \left\{ \sum_{m'} \langle \phi_{m'} | U | \phi_m \rangle c_{m'}^{\dagger} \right\} \\ \times \left\{ \sum_{n'} \langle \phi_n | U^{-1} | \phi_{n'} \rangle c_{n'} \right\}. \quad (16)$$

According to this expression, the Hamiltonian of a system of particles in an electromagnetic field in its second quantization form can be obtained directly from the unperturbed Hamiltonian (14) replacing the annihilation c_n and creation c_n^{\dagger} operators by the expressions

$$c_m \rightarrow \sum_n \langle \phi_m | U^{-1} | \phi_n \rangle c_n, \\ c_m^{\dagger} \rightarrow \sum_n \langle \phi_n | U | \phi_m \rangle c_n^{\dagger}. \quad (17)$$

The rule is valid for both local and nonlocal potentials and in order to obtain the interaction Hamiltonian up to the desired order in the vector potential one has to expand the unitary operator U :

$$c_m \rightarrow c_m + \frac{iq}{c\hbar} \sum_n \langle \phi_m | \chi(\mathbf{x}) | \phi_n \rangle c_n \\ + \frac{1}{2!} \left(\frac{iq}{c\hbar} \right)^2 \langle \phi_m | \chi^2(\mathbf{x}) | \phi_n \rangle + \dots \\ c_m^{\dagger} \rightarrow c_m^{\dagger} + \frac{-iq}{c\hbar} \sum_n \langle \phi_n | \chi(\mathbf{x}) | \phi_m \rangle c_n^{\dagger} \\ + \frac{1}{2!} \left(\frac{-iq}{c\hbar} \right)^2 \sum_n \langle \phi_n | \chi^2(\mathbf{x}) | \phi_m \rangle c_n^{\dagger} + \dots \quad (18)$$

This replacement rule represents the second quantization analogy of the substitution $\mathbf{p} \rightarrow \mathbf{p} - (e/c)\mathbf{A}$ of the particle momentum.

In order to write the exciton-photon Hamiltonian up to the second order in the operators by applying the rule (17), we have to consider the matrix elements

$$\langle \Phi_0 | \chi(\mathbf{x}) | \Phi_{n,\mathbf{k}} \rangle, \quad (23)$$

which can be expressed in terms of transition dipoles calculated between Bloch functions. We approximate the periodic part of the Bloch functions with their counterparts at band extrema, and in order to maintain the dipole approximation in the calculation of matrix elements without neglecting macroscopic variations of the field we do not use the standard dipole approximation $\chi(\mathbf{x}) = \mathbf{x} \cdot \mathbf{A}(0)$ but, following [10], we express the integral over the whole crystal as a sum of integrals over the elementary cells at \mathbf{R} and expand $\chi(\mathbf{x})$ at the point \mathbf{R} in each elementary cell. We obtain

$$\langle \phi_0 | \chi(\mathbf{x}) | \phi_{n,\mathbf{k}} \rangle = F_n(0) \langle u_v(\mathbf{x}) | \mathbf{x} \cdot \hat{\mathbf{e}} | u_c(\mathbf{x}) \rangle \frac{1}{N} \times \sum_{\mathbf{R}} \exp(i\mathbf{k} \cdot \mathbf{R}) A(\mathbf{R}),$$

where $F_n(\mathbf{R}) = \sum_{\mathbf{q}} \exp(i\mathbf{q} \cdot \mathbf{R}) G_n(\mathbf{q})$, $u_{c,v}(\mathbf{x})$ are the Bloch functions at the Γ point of the Brillouin zone and for simplicity only one field polarization $\hat{\mathbf{e}}$ has been considered. After the usual quantization of the photon vector potential in the dielectric of volume V

$$\mathbf{A}(\mathbf{x}) = \sum_{\mathbf{k}} \sqrt{\frac{2\pi\hbar v}{V k}} \mathbf{e}_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{x}) (A_{\mathbf{k}}^{(1)} + A_{-\mathbf{k}}^{(1)\dagger}), \quad (24)$$

we finally obtain

$$\langle \Phi_0 | \chi(\mathbf{x}) | \Phi_{n,\mathbf{k}} \rangle = \sqrt{\frac{2\pi\hbar v}{V k}} F_n(0) \langle u_v(\mathbf{x}) | \mathbf{x} \cdot \hat{\mathbf{e}}_{\mathbf{k}} | u_c(\mathbf{x}) \rangle \times (A_{-\mathbf{k}}^{(1)} + A_{\mathbf{k}}^{(1)\dagger}). \quad (25)$$

The bilinear exciton-photon Hamiltonian can be obtained by applying to the Hamiltonian of noninteracting excitons and photons

$$H_0 = \sum_{\mathbf{k}} \hbar v k A_{\mathbf{k}}^{(1)\dagger} A_{\mathbf{k}}^{(1)} + \sum_{n,\mathbf{k}} \hbar \omega_{n,\mathbf{k}} A_{n,\mathbf{k}}^{(2)\dagger} A_{n,\mathbf{k}}^{(2)} \quad (26)$$

the replacement rule

$$\begin{aligned} A_{n,\mathbf{k}}^{(2)} &\rightarrow A_{n,\mathbf{k}}^{(2)} - i \frac{C_{n,\mathbf{k}}^*}{\hbar \omega_{n,\mathbf{k}}} (A_{\mathbf{k}}^{(1)} + A_{-\mathbf{k}}^{(1)\dagger}), \\ A_{n,\mathbf{k}}^{(2)\dagger} &\rightarrow A_{n,\mathbf{k}}^{(2)\dagger} + i \frac{C_{n,\mathbf{k}}}{\hbar \omega_{n,\mathbf{k}}} (A_{-\mathbf{k}}^{(1)} + A_{\mathbf{k}}^{(1)\dagger}), \end{aligned} \quad (27)$$

with

$$C_{n,\mathbf{k}} = \hbar \omega_{n,\mathbf{k}} \frac{ie}{c\hbar} \sqrt{\frac{2\pi\hbar\omega}{V k}} F_n(0) \langle u_v(\mathbf{x}) | \mathbf{x} \cdot \hat{\mathbf{e}}_{\mathbf{k}} | u_c(\mathbf{x}) \rangle. \quad (28)$$

The polariton Hamiltonian is thus given by

$$\begin{aligned} H &= \sum_{\mathbf{k}} \hbar v k A_{\mathbf{k}}^{(1)\dagger} A_{\mathbf{k}}^{(1)} \\ &+ \sum_{n,\mathbf{k}} \hbar \omega_{n,\mathbf{k}} \left\{ A_{n,\mathbf{k}}^{(2)\dagger} + i \frac{C_{n,\mathbf{k}}}{\hbar \omega_{n,\mathbf{k}}} (A_{-\mathbf{k}}^{(1)} + A_{\mathbf{k}}^{(1)\dagger}) \right\} \\ &\times \left\{ A_{n,\mathbf{k}}^{(2)} - i \frac{C_{n,\mathbf{k}}^*}{\hbar \omega_{n,\mathbf{k}}} (A_{\mathbf{k}}^{(1)} + A_{-\mathbf{k}}^{(1)\dagger}) \right\}. \end{aligned} \quad (29)$$

The Hamiltonian (29), as it is well known [5], can be diagonalized introducing new Bose operators which are defined by

$$\begin{aligned} B_{\mathbf{k}}^{(m)} &= W_{\mathbf{k}}^{(m)} A_{\mathbf{k}}^{(1)} + \sum_n X_{n,\mathbf{k}}^{(m)} A_{(n),\mathbf{k}}^{(2)} + Y_{\mathbf{k}}^{(m)} A_{-\mathbf{k}}^{(1)\dagger} \\ &+ \sum_n Z_{n,\mathbf{k}}^{(m)} A_{n,-\mathbf{k}}^{(2)\dagger}. \end{aligned} \quad (30)$$

The requirement that H be diagonal in the polariton operators $B_{\mathbf{k}}^{(m)}$ leads to an eigenvalue problem, whose eigenenergies $E_{\mathbf{k}}^{(m)}$ are solutions of the equation

$$E^2 - (\hbar v k)^2 = -4(\hbar v k) E^2 \sum_n \frac{\hbar \omega_{n,\mathbf{k}} |C_{n,\mathbf{k}}|^2}{(\hbar \omega_{n,\mathbf{k}})^2 - E^2}. \quad (31)$$

It is important to notice that the velocity of light in the medium is given by $v = c/\sqrt{\epsilon_{\infty}}$ where the background dielectric constant ϵ_{∞} describes the effect of all the excitations of the system not included in the model; its value is therefore also dependent on the number of exciton levels explicitly included in the sum in equation (31).

Our replacement rule can also be applied to obtain the quantum Hamiltonian of polaritons in confined systems. Let us consider as an example a quantum well where the exciton is strongly confined in the z direction. Following [12], we consider the electron wavefunction in the envelope function approximation and calculate the transition matrix elements (23). The replacement rule thus obtained is

$$A_{\mathbf{q}}^{(2)\dagger} \rightarrow A_{\mathbf{q}}^{(2)\dagger} + \sum_{q_z} \frac{C_{\mathbf{q},q_z}}{\hbar \omega_{\mathbf{q}}} (A_{-\mathbf{q},q_z}^{(1)} + A_{\mathbf{q},q_z}^{(1)\dagger}) \quad (32)$$

with

$$C_{\mathbf{q},q_z} = \hbar \omega_{\mathbf{q}} \frac{ie}{c\hbar} \sqrt{\frac{2\pi\hbar v}{S k}} F_n(0) \langle u_v(\mathbf{x}) | \mathbf{x} \cdot \hat{\mathbf{e}}_{\mathbf{k}} | u_c(\mathbf{x}) \rangle I(q_z). \quad (33)$$

If the states of the basis set $|\phi_m\rangle$, are exact eigenstates of H_0 , expression (15) reduces to

$$H = \sum_m \hbar\omega_n \left(\sum_n \langle \phi_n | U | \phi_m \rangle c_n^\dagger \right) \times \left(\sum_{n'} \langle \phi_m | U^{-1} | \phi_{n'} \rangle c_{n'} \right). \quad (19)$$

So far we have only taken into account Hamiltonians which are summations of one particle operators, but the effect of particle-particle interactions are described by two particle operators of the form

$$\frac{1}{2} \sum_{i,j} V(\mathbf{r}_i, \mathbf{r}_j)$$

which in terms of creation and annihilation operators can be written as

$$\frac{1}{2} \sum_{l,m,i,j} V_{lmij} c_l^\dagger c_m^\dagger c_i c_j$$

with

$$V_{lmij} = \langle \phi_l(\mathbf{x}) \phi_m(\mathbf{x}') | V(\mathbf{x}, \mathbf{x}') | \phi_i(\mathbf{x}) \phi_j(\mathbf{x}') \rangle$$

and can be interpreted as describing two particle scattering events. In the presence of the field this term takes the form

$$\frac{1}{2} \sum_{l,m,i,j} [U(\mathbf{x})U(\mathbf{x}')V(\mathbf{x},\mathbf{x}')U^{-1}(\mathbf{x})U^{-1}(\mathbf{x}')]_{lmij} c_l^\dagger c_m^\dagger c_i c_j.$$

It is a straightforward matter to see that in this case, the coupling with the field is also described by replacing the creation and annihilation operators according to rule (17) by introducing the complete set of states $|\phi_m(\mathbf{x}), \phi_n(\mathbf{x}')\rangle$, and rearranging the factors.

4. THE POLARITON HAMILTONIAN AS AN EXAMPLE

When an electron is promoted from the valence to the conduction band the electron-electron interaction produces an electron-hole Coulomb attraction, which determines electron-hole hydrogen-like pair states called excitons. Exciton-polaritons are mixed photon exciton modes which result from the strong coupling between the excitons and the radiation field in semiconductors and insulators. The quantum photon-exciton interaction Hamiltonian was first introduced in 1958 by Hopfield [5]. In order to derive exciton states, one starts with the Hamiltonian

for the electronic system

$$H_{el} = \int d^3x \psi^\dagger(\mathbf{x}) H_L(\mathbf{x}) \psi(\mathbf{x}) + \frac{1}{2} \int d^3x d^3x' \psi^\dagger(\mathbf{x}) \psi^\dagger(\mathbf{x}') \frac{e^2}{|\mathbf{x} - \mathbf{x}'|} \psi(\mathbf{x}) \psi(\mathbf{x}'), \quad (20)$$

where $H_L(\mathbf{x})$ is the Hamiltonian of an electron in a periodic potential and the second term describes the Coulomb interaction between electrons. The electron field operators are expanded into the eigenfunctions of the one particle Hamiltonian, which, as is well known are Bloch functions. Then, neglecting the terms of the Coulomb interaction which do not conserve the number of pair excitations, it can be shown that the linear combinations of Slater determinants in which a Bloch electron has been promoted to the conduction band are the eigenfunctions of the approximated electron Hamiltonian

$$\Phi_{n,k} = \sum_q G_n(\mathbf{q}) \Phi_{v,q}^{c,k+q}. \quad (21)$$

The corresponding eigenvalues $\hbar\omega_{n,k}$ and the coefficients $G_n(\mathbf{q})$ are solutions of a secular problem [7, 8]. In equation (21) the spin has been neglected and a two band model has been considered. The Slater determinants $\Phi_{v,q}^{c,k+q}$ in equation (21) are obtained from the Slater determinant of the ground state, replacing the Bloch function $\psi_{v,k}$ of one electron in the valence band with the Bloch function $\psi_{c,k+q}$ of one electron in the conduction band [9]. It has been shown [2] that, owing to the neglected terms in the Coulomb interaction, the projection of the Coulomb potential included in the exciton Hamiltonian is nonlocal. In the microscopic theory of exciton-polaritons all the above approximations are retained and the Hamiltonian of N' electrons in a periodic potential is replaced by the exciton model Hamiltonian

$$H_{exc} = \sum_{n,k} \hbar\omega_{n,k} A_{n,k}^{(2)\dagger} A_{n,k}^{(2)}, \quad (22)$$

where n is a collective index labeling the hydrogen-like exciton levels, and $A_{n,k}^{(2)}$ and $A_{n,k}^{(2)\dagger}$ in a low excitation regime are Bose operators. Furthermore in many microscopic approaches to the polariton theory only one or a limited number of exciton levels are taken into account, the only effect of out of resonance excitons being a contribution to the background dielectric constant.

In this expression $\mathbf{k} = (\mathbf{q}, q_z)$ and $I(q_z)$ is the Fourier transform of the exciton confinement function. Owing to the breaking symmetry in one direction, the exciton of a given in-plane wave vector \mathbf{q} is coupled with the field modes with all available wavevector components q_z . The polariton Hamiltonian which is obtained by using equation (32) has already been considered [11, 12] and it has been shown that can be diagonalized by a general Hopfield transformation.

The replacement rule (18) allows us to directly obtain higher order terms in the exciton-photon interaction which describe non-linear optical effects and polariton-polariton interactions [13]. In order to derive explicit expressions for the non-linear terms we have to consider exciton-exciton matrix elements. Following the same procedure as for the ground state-exciton matrix elements (25), we obtain

$$\langle \Phi_{m, \mathbf{k}+\mathbf{q}} | \chi(\mathbf{x}) | \Phi_{n, \mathbf{k}} \rangle = \sqrt{\frac{2\pi\hbar v}{Vq}} \langle F_m(\mathbf{R}) | \mathbf{R} \cdot \hat{\mathbf{e}}_{\mathbf{q}} | F_n(\mathbf{R}) \rangle \times (A_{\mathbf{q}}^{(1)} + A_{-\mathbf{q}}^{(1)\dagger}). \quad (34)$$

By using the expression (34) we extend the replacement rule (27) including exciton-exciton transitions

$$A_{n, \mathbf{k}}^{(2)\dagger} \rightarrow A_{n, \mathbf{k}}^{(2)\dagger} + i \frac{C_{n, \mathbf{k}}}{\hbar\omega_{n, \mathbf{k}}} (A_{-\mathbf{k}}^{(1)} + A_{\mathbf{k}}^{(1)\dagger}) + i \sum_{\mathbf{q}, m} \frac{C_{m, \mathbf{k}, \mathbf{k}+\mathbf{q}}}{\hbar\omega_{m, \mathbf{k}, \mathbf{k}+\mathbf{q}}} A_{m, \mathbf{k}+\mathbf{q}}^{(2)\dagger} (A_{\mathbf{q}}^{(1)} + A_{\mathbf{q}}^{(1)\dagger}), \quad (35)$$

where $\omega_{m, \mathbf{k}, \mathbf{k}+\mathbf{q}} = \omega_{n, \mathbf{k}} - \omega_{m, \mathbf{k}+\mathbf{q}}$ and

$$C_{m, \mathbf{k}, \mathbf{k}+\mathbf{q}} = \hbar\omega_{m, \mathbf{k}, \mathbf{k}+\mathbf{q}} \frac{ie}{c\hbar} \sqrt{\frac{2\pi\hbar v}{Vq}} \times \langle F_m(\mathbf{R}) | \mathbf{R} \cdot \hat{\mathbf{e}}_{\mathbf{q}} | F_n(\mathbf{R}) \rangle.$$

Introducing the replacement rule (35) and the analogous replacement for $A_{n, \mathbf{k}}^{(2)}$ in the Hamiltonian of noninteracting excitons and photons (26), after some straightforward calculations, we obtain the terms of the third order in the exciton and photon operators

$$H^{NL} = i \sum_{\mathbf{k}, \mathbf{q}} \sum_{m, n} C_{m, \mathbf{k}, \mathbf{k}+\mathbf{q}} A_{m, \mathbf{k}+\mathbf{q}}^{(2)\dagger} A_{n, \mathbf{k}}^{(2)} (A_{\mathbf{q}}^{(1)} + A_{-\mathbf{q}}^{(1)\dagger}) + \sum_{\mathbf{k}, \mathbf{q}} \sum_{m, n} C_{n, \mathbf{k}}^* \frac{C_{m, \mathbf{k}, \mathbf{k}+\mathbf{q}}}{\hbar\omega_{m, \mathbf{k}, \mathbf{k}+\mathbf{q}}} A_{m, \mathbf{k}+\mathbf{q}}^{(2)\dagger} \times (A_{\mathbf{q}}^{(1)} + A_{-\mathbf{q}}^{(1)\dagger}) (A_{-\mathbf{k}}^{(1)} + A_{\mathbf{k}}^{(1)\dagger}) + \text{h.c.} \quad (36)$$

The first term of the nonlinear Hamiltonian (36) comes from the linear term in the interaction

Hamiltonian (4) proportional to \mathbf{A} , while the second term is a correction due to nonlocality which comes from the nonlinear term proportional to A^2 .

The nonlinear Hamiltonian (36) describes two photon absorption and sum frequency generation as discussed in [13, 14], where only the first term of equation (36) has been taken into account.

5. CONCLUSIONS

We have shown that a simple and general replacement rule, which is the second quantization analogy of the momentum minimal coupling replacement, allows us to directly obtain in the second quantization formalism the interaction Hamiltonian in the velocity gauge. The rule is also valid for nonlocal Hamiltonians and is particularly useful in obtaining the quantum Hamiltonian of systems such as polaritons where the length gauge cannot be conveniently used. Furthermore we have shown that the rule also applies to nondiagonal and anharmonic terms in the Hamiltonian of the particle system. When these terms are included in the system Hamiltonian corrective terms should be included in the interaction Hamiltonian.

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