Optics in the Multipole Approximation: From Atomic Systems to Solids

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In honor of Professor M. O. Scully's 60th birthday.

Starting from the microscopic light-matter interaction in form of the minimal coupling Hamiltonian, the multipole approximation for the optical response of localized electrons in atomic systems is extended to delocalized electrons in solids. A spatial averaging procedure is used to derive the electromagnetic sources for macroscopic Maxwell's equations as well as the corresponding many particle Hamiltonian on a coarse grained length scale. The results are illustrated for semiconductor bulk material up to quadrupole moments for the interband transitions, where gauge invariant equations of motion for the optical response are obtained.

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I. INTRODUCTION

The response of matter to electromagnetic radiation in the optical range, i.e. frequencies of about $\omega \approx 10^{14} - 10^{15} \text{ Hz}$, is usually determined by the electronic properties of the material. The motion of electrons is governed by the laws of quantum mechanics which allows us to obtain the electronic states via the electronic wavefunctions $|n\rangle$ and their corresponding energies ϵ_n , where n is the quantum number. Knowing the electronic states, the electromagnetic field induced response is determined by the probability for the transition of electrons between different states $|n\rangle \leftrightarrow |n'\rangle$ whereby energy $\hbar\omega_{nn'} = |\epsilon_n - \epsilon_{n'}|$ is absorbed or emitted [1]. These transitions yield the material polarization which acts as the source term in Maxwell's equations. In turn, Maxwell's equations determine the observables, such as electric and magnetic fields as well as their field correlations [2].

If the electromagnetic fields are treated classically and the matter quantum-mechanically, the corresponding theory is called semiclassical. There are two important self-consistency problems that are relevant for the semiclassical optics of solids which especially manifest themselves on ultrashort time and space scales:

i) The self-consistent transition from a microscopic (atomic) to a macroscopic (mesoscopic) theory which can be used to analyze spatially inhomogeneous excitation and multipoles on a length scale above the atomic scale but still below the wavelength of light. This part of the theory is connected with spatial averages explicitly or implicitly done in experiments [3].

ii) Solid state quantum mechanics is usually formulated in terms of electromagnetic potentials in Coulomb gauge instead of the total electromagnetic fields which occur

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in Maxwell's equations. Thus, in a rigorous way this solid-state theory is not consistent with direct solutions of Maxwell's equations.

Both self-consistency problems are only of academic interest for far-field optics where the measured fields are fully transversal [1,4], however, they become relevant when dealing with near-field optics where the full spatial and temporal dynamics of Maxwell's equations close to nanostructured electromagnetic sources is required [5]. In view of the continuing device miniaturization [6] and their near-field characterization [7] this appears to be a shortcoming of the present theory, especially if small scale structures are treated at the level of microscopic material models [8].

This paper addresses both self-consistency problems for the semiclassical treatment of the solid state and proposes a possible treatment via a macroscopically averaged many-particle Hamiltonian for the solid. To have a close analogy with well known concepts, the established theory for atomic systems is simultaneously formulated in a similar notation. A bulk semiconductor is used as model system to illustrate the proposed treatment of solids.

II. MICROSCOPIC MINIMAL COUPLING HAMILTONIAN

To have a common microscopic approach to the optics of atomic systems and solids, the analysis starts from the minimal coupling Hamiltonian [9]. In the minimal coupling Hamiltonian, the microscopic electromagnetic fields $\mathbf{E}(r, t)$ and $\mathbf{B}(r, t)$ enter in form of the transversal (T) vector potential $\mathbf{A}(r, t)$ and the longitudinal (L) scalar potential $\Phi(r, t)$. In the following the Coulomb gauge $\nabla \cdot \mathbf{A} = 0$ and cgs units are used:

$$\mathbf{E} = \mathbf{E}_L + \mathbf{E}_T = -\nabla\Phi(r, t) - \frac{1}{c_0}\partial_t\mathbf{A}(r, t), \quad (1)$$

$$\mathbf{B} = \nabla \times \mathbf{A}(r, t), \quad (2)$$

where r denotes the space vector on the microscopic, i.e. the atomic space scale.

For a many-electron system such as atomic systems or solids, the proper treatment is best done using the method of second quantization. The contributions to the minimal coupling Hamiltonian $H = H_0 + H_1 + H_2 + H_3$ in second quantization read [9]:

$$H_0 = \sum_{1,2} \langle 1 | \frac{\mathbf{p}^2}{2m_0} + V(r) | 2 \rangle a_1^\dagger a_2, \quad (3)$$

$$H_1 = \sum_{1,2} \langle 1 | -\frac{e}{c_0 m_0} \mathbf{A}(r, t) \cdot \mathbf{p} + \frac{e^2}{2m_0 c_0^2} A^2(r, t) | 2 \rangle a_1^\dagger a_2, \quad (4)$$

$$H_2 = e \sum_{1,2} \langle 1 | \Phi(r, t) | 2 \rangle a_1^\dagger a_2, \quad (5)$$

$$H_3 = \int d^3r (E_T^2(r, t) + B^2(r, t) - E_L^2(r, t)). \quad (6)$$

Here, the material operators are the Fermionic electron operators $a_1, a_1^\dagger$ which annihilate and create electrons in the electronic quantum state $|1\rangle$. As usual, the Hamiltonian contains a single particle part (momentum p and bare electron mass m_0) with the potential $V(r)$ of the nuclei which themselves are assumed to be fixed at some equilibrium position. Thus, for reasons of clarity, no atomic motion or phonon excitations in the solid are taken into account. The electron-photon interaction is expressed in terms of the transverse vector potential A , with e being the elementary charge and c_0 the vacuum light velocity. The longitudinal electron-electron interaction occurs via the scalar potential Φ . In the following, the states $|n\rangle$ are conveniently chosen as the single electron states $\langle r|1\rangle$ in the potential $V(r)$:

$$H_0(r, p)\langle r|1\rangle = \varepsilon_1\langle r|1\rangle. \quad (7)$$

A. Atomic Systems

In localized atomic systems, e.g. in atoms embedded in a solid state matrix, the electronic wavefunction

$$\langle r - R_0|1\rangle = \Psi_1(r - R_0), \quad (8)$$

is typically confined to a range of several Angströms to nanometers. The atomic nucleus is assumed to rest at the position R_0 , so that $r - R_0$ is the relative coordinate of the electron-nucleus motion.

Usually, the typical extension of the atomic wavefunction is much smaller than the light wavelength at an optically induced transition between two electronic states Ψ_1, Ψ_2 . This is the basis for the multipole approximation [1,2] which consists of the expansion of matrix elements of spatially weakly varying functions $f(r)$, like the optical field, at the origin of the atomic system:

$$\int d^3r \Psi_1^*(r - R_0) f(r) \Psi_2(r - R_0) \approx \delta_{1,2} f(R_0) + \int d^3r \Psi_1^*(r) \mathbf{r} \cdot \nabla_{R_0} f(R_0) \Psi_2(r) + \dots \quad (9)$$

Furthermore, the electronic states are usually well separated by their energy difference $\varepsilon_1 - \varepsilon_2$, so that isolated optical transitions between the electronic states can be excited.

B. Solids

Solids are build up of many atoms forming a periodic crystal lattice where the wavefunction overlap of electrons from different atoms must be taken into account. Therefore, in a perfect solid (i.e. not disorderd) the electronic wavefunction is extended over the entire system. The smallest periodic unit of the crystal lattice is the

so called elementary cell Ω_0 with a typical extension of several Angströms. Since the development of bulk crystal properties typically requires thousands of atoms and more [10], the spatial extension of the electronic wavefunction in such solids exceeds the wavelength of light and the simple multipole approximation, Eq.(9), becomes questionable.

According to Bloch's theorem the wavefunction of an electron in an extended crystal lattice can be written as a lattice periodic part (varying on the length scale of the atomic unit cell, i.e. several Angström) multiplied by an envelope function. For infinitely extended bulk material, the eigenfunctions of the single particle Hamiltonian H_0 , Eq.(3), are the Bloch-functions:

$$\langle r|1\rangle = \frac{1}{\sqrt{V}} u_{\lambda_1 k_1}(r) e^{ik_1 \cdot r} \quad (10)$$

where $u_{\lambda k}$ is the lattice periodic part with the band index λ and the wavenumber k . The corresponding electronic eigenenergies occur in the form of forbidden and allowed energy regions, the energy bands $\varepsilon_{k\lambda}$. At a later stage in this paper we illustrate our results for the case of interband transitions in a semiconductor where optical excitation lifts electrons from the initially filled valence into the initially empty conduction band.

III. SPATIAL AVERAGES

Microscopic Maxwell's equations:

$$\nabla \times \mathbf{E}(r, t) = -\frac{1}{c_0} \partial_t \mathbf{B}(r, t) \quad (11)$$

$$\nabla \times \mathbf{B}(r, t) = \frac{1}{c_0} \partial_t \mathbf{E}(r, t) + \frac{4\pi}{c_0} \mathbf{j}_{mic}(r, t) \quad (12)$$

$$\nabla \cdot \mathbf{E}(r, t) = 4\pi \rho_{mic}(r) \quad (13)$$

$$\nabla \cdot \mathbf{B}(r, t) = 0 \quad (14)$$

can be derived from the minimal coupling Hamiltonian and contain microscopic material sources, i.e. the current $\mathbf{j}_{mic}$ and the density ρ_{mic} , varying on atomic scales [9]:

$$\rho_{mic}(r) = e \sum_{1,2} a_1^\dagger a_2 \langle 1|r \rangle \langle r|2 \rangle \quad (15)$$

$$\mathbf{j}_{mic}(r) = \frac{e}{m_0} \sum_{1,2} a_1^\dagger a_2 \langle 1|r \rangle \left(\mathbf{p} - \frac{e}{c_0} \mathbf{A}(r) \right) \langle r|2 \rangle \quad (16)$$

However, for most practical applications, macroscopic Maxwell's equations [11]:

$$\nabla \times \mathbf{E}(R, t) = -\frac{1}{c_0} \partial_t \mathbf{B}(R, t), \quad (17)$$

$$\nabla \times \mathbf{B}(R, t) = \frac{1}{c_0} \partial_t \mathbf{E}(R, t) + \frac{4\pi}{c_0} \mathbf{j}(R, t), \quad (18)$$

$$\nabla \cdot \mathbf{E}(R, t) = 4\pi \rho(R), \quad (19)$$

$$\nabla \cdot \mathbf{B}(R, t) = 0 \quad (20)$$

are solved. In contrast to the microscopic Maxwell's equations, these macroscopic equations contain the macroscopic current $\mathbf{j}(R)$ and density $\rho(R)$, determined by the spatial average over their microscopic counterparts [3]. Schematically we can write

$$f(R) = \int d^3r g(r) f_{mic}(r + R), \quad (21)$$

where $f_{mic}(r)$ can be the microscopic charge density or the vector components of the microscopic current, respectively.

The averaging procedure, Eq.(21), transforms the quantity $f_{mic}(r)$ varying on the atomic scale into the quantity $f(R)$ which varies on the coarse grained macroscopic scale R . $g(r)$ is a normalized averaging function which varies on the atomic scale. Sometimes, $g(r)$ is chosen to be constant over a typical average volume Ω around the macroscopic position R :

$$f(R) = \frac{1}{\Omega} \int_{\Omega} d^3r f_{mic}(r + R). \quad (22)$$

Note that the average, Eq.(21) is automatically done in experiments, typically by the finite resolution of some component of the experimental apparatus.

From the spatial average of the carrier density Eq.(15), and the current, Eq.(16), the usual multipole contributions in macroscopic Maxwell's equations are expected [3]:

$$\rho(R) = \rho_0(R) - \nabla_R \cdot \mathbf{P}(R) + \dots, \quad (23)$$

$$\mathbf{j}(R) = \mathbf{j}_0(R) + \dot{\mathbf{P}}(R) + \dots \quad (24)$$

Here, the monopole density ρ_0 and current $\mathbf{j}_0$ occur, as well as the polarization density $\mathbf{P}$ and polarization current $\dot{\mathbf{P}}$ as the first contributions within the multipole approximation.

Using Eq.(21) and the microscopic sources, Eqs. (15) and (16), the macroscopic quantities can be written as:

$$\rho(R) = e \sum_{1,2} a_1^\dagger a_2 \int d^3r g(r) \langle 1|r + R \rangle \langle r + R|2 \rangle, \quad (25)$$

$$\mathbf{j}(R) = \frac{e}{m_0} \sum_{1,2} a_1^\dagger a_2 \int d^3r g(r) \langle 1|\dot{\mathbf{r}} + R \rangle \left(\mathbf{p} - \frac{e}{c_0} \mathbf{A}(r + R) \right) \langle r + R|2 \rangle. \quad (26)$$

Now, the attempt is made to use Eqs.(25) and (26) to calculate a form similar to Eqs.(23) and (24). Because of their different wavefunctions, compare Eqs.(8) and (10), the spatial average for atomic systems and solids must be treated differently.

A. Atomic Systems

Using the well localized single-particle wavefunctions for electrons in an atomic system, Eq.(8), one obtains for the macroscopic density, Eq.(25):

$$\begin{aligned}
\rho(R) &= e \sum_{12} a_1^\dagger a_2 \int d^3 r g(r) \Psi_1^*(r - R_0 + R) \Psi_2(r - R_0 + R) \\
&= \sum_{12} a_1^\dagger a_2 e \int d^3 r \Psi_1^*(r) \Psi_2(r) \sum_n \frac{1}{n!} (-\mathbf{r} \cdot \nabla_R)^n g(R - R_0) \\
&= \rho_0(R) - \nabla \cdot \Pi(R), \tag{27}
\end{aligned}$$

where an expansion of the averaging function at the position of a typical nucleus is carried out. Here, the macroscopic monopole charge density ρ_0 and the generalized polarization Π are given by:

$$\rho_0(R) = e \sum_1 a_1^\dagger a_1 g(R - R_0), \tag{28}$$

$$\Pi(R) = \sum_{12} a_1^\dagger a_2 e \int d^3 r \mathbf{r} \Psi_1^*(r) \Psi_2(r) \sum_n \frac{1}{(n+1)!} (-\mathbf{r} \cdot \nabla_R)^n g(R - R_0). \tag{29}$$

Eqs.(28) and (29) represent the multipole expansion for atomic systems, usually obtained in first quantization ($g(R - R_0) \rightarrow \delta(\mathbf{R} - \mathbf{R}_0)$) [2]. The generalized polarization Π contains all higher multipole moments, its first contribution of the sum is the usual dipole polarization $\mathbf{P}$:

$$\mathbf{P}(R) = \sum_{12} a_1^\dagger a_2 \mathbf{d}_{12} g(R - R_0), \tag{30}$$

$$\mathbf{d}_{12} = e \int d^3 r \mathbf{r} \Psi_1^*(r) \Psi_2(r) \tag{31}$$

where $\mathbf{d}_{12}$ is the dipole moment of the atomic system for the optical transition between the states Ψ_1 and Ψ_2 . The corresponding approach for the microscopic current, Eq.(26), yields the macroscopic current.

B. Solids

Using the Bloch functions, Eq.(10), as single particle wavefunctions in a solid one obtains for the macroscopic carrier density, Eq.(25):

$$\rho(R) = \frac{e}{V} \sum_{\lambda_1 \lambda_2} \sum_{k_1 k_2} a_{\lambda_1 k_1}^\dagger a_{\lambda_2 k_2} \int d^3 r g(r) e^{-i(k_1 - k_2) \cdot (r + R)} u_{\lambda_1 k_1}^*(r + R) u_{\lambda_2 k_2}(r + R). \tag{32}$$

Splitting the spatial integration into a sum over the elementary cells Ω_n at the lattice-vector position R_n the matrix element occuring in Eq.(32) becomes:

$$\begin{aligned}
&\int d^3 r g(r) e^{-i(k_1 - k_2) \cdot (r + R)} u_{\lambda_1 k_1}^*(r + R) u_{\lambda_2 k_2}(r + R) \\
&= e^{-i(k_1 - k_2) \cdot R} \sum_{R_n} \int_{\Omega_n} d^3 r_n g(R_n + r_n) e^{-i(k_1 - k_2) \cdot (R_n + r_n)} u_{\lambda_1 k_1}^*(R_n + r_n + R) u_{\lambda_2 k_2}(R_n + r_n + R). \tag{33}
\end{aligned}$$

Using the Fourier-transform g_K of the averaging function $g(r)$, the matrix element can be written as:

$$e^{-i(k_1 - k_2) \cdot R} \sum_{R_n} \int_{\Omega_n} d^3 r_n \sum_K g_K e^{-iK \cdot (R_n + r_n)} e^{-i(k_1 - k_2) \cdot (R_n + r_n)} u_{\lambda_1 k_1}^*(r_n + R) u_{\lambda_2 k_2}(r_n + R), \tag{34}$$

where the translational invariance of the lattice periodic wavefunctions $u_{\lambda k}(r + R_n) = u_{\lambda k}(r)$ is used [10]. Next, Umklapp-processes:

$$\frac{1}{N} \sum_{R_n} e^{-i\mathbf{Q} \cdot \mathbf{R}_n} = \sum_{\bar{\mathbf{Q}}} \delta_{\mathbf{Q}, \bar{\mathbf{Q}}} \approx \delta_{\mathbf{Q}, 0}, \quad (35)$$

where $\bar{\mathbf{Q}}$ is a vector of the reciprocal lattice, are neglected [10]. This approximation amounts to neglecting spatial frequencies on the scale of the inverse unit cell, i.e. no optical resolution better than the lattice constant is obtainable. Therefore, a coarse graining of the macroscopic coordinate at multiples of the lattice vector $R = m \cdot R_n$ can be chosen. Within these approximations the matrix element can be written as:

$$\begin{aligned} & \int d^3r g(r) e^{-i(\mathbf{k}_1 - \mathbf{k}_2) \cdot (r + \mathbf{R})} u_{\lambda_1 k_1}^*(r + \mathbf{R}) u_{\lambda_2 k_2}(r + \mathbf{R}) \\ & \approx N e^{-i(\mathbf{k}_1 - \mathbf{k}_2) \cdot \mathbf{R}} g_{\mathbf{k}_2 - \mathbf{k}_1} \int_{\Omega_0} d^3r u_{\lambda_1 k_1}^*(r) u_{\lambda_2 k_2}(r), \end{aligned} \quad (36)$$

where the integration runs over an elementary cell Ω_0 , and N is the number of elementary cells in the crystal. The averaged charge density, Eq.(32), is obtained as:

$$\begin{aligned} \rho(R) &= \frac{e}{\Omega_0} \sum_{\lambda_1 \lambda_2} \sum_{k_1 k_2} a_{\lambda_1 k_1}^\dagger a_{\lambda_2 k_2} e^{-i(\mathbf{k}_1 - \mathbf{k}_2) \cdot \mathbf{R}} g_{\mathbf{k}_2 - \mathbf{k}_1} \int_{\Omega_0} d^3r u_{\lambda_1 k_1}^*(r) u_{\lambda_2 k_2}(r) \\ &= e \sum_{\lambda_1 \lambda_2} \sum_{q, Q} a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q - \frac{Q}{2}} e^{-i\mathbf{Q} \cdot \mathbf{R}} g_{-\mathbf{Q}} \langle \lambda_1 q + \frac{Q}{2} | \lambda_2 q - \frac{Q}{2} \rangle, \end{aligned} \quad (37)$$

where a change of the summation variables to relative $k_1 - k_2 = Q$ and center of mass $q = \frac{k_1 + k_2}{2}$ coordinates has been performed. The macroscopic carrier density is given by the Fourier expansion of the quantum mechanically determined material amplitudes $a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q - \frac{Q}{2}}$, which can be calculated via the Heisenberg equations of motion using the minimal coupling Hamiltonian. Furthermore, their Fourier transform is weighted by the (experimental) resolution $g_{-\mathbf{Q}}$ of the averaging function and the Bloch integral:

$$\langle \lambda_1 q + \frac{Q}{2} | \lambda_2 q - \frac{Q}{2} \rangle = \frac{1}{\Omega_0} \int_{\Omega_0} d^3r u_{\lambda_1 q + \frac{Q}{2}}^*(r) u_{\lambda_2 q - \frac{Q}{2}}(r). \quad (38)$$

An appropriate expansion of the Bloch integral, as will be shown below for a bulk semiconductor, yields the multipole expansion for the solid.

A further simplification can be reached by assuming that the average is taken roughly over one elementary cell, so that on the macroscopic length scale:

$$g_{-\mathbf{Q}} = \frac{1}{N} \sum_{\mathbf{R}} g(\mathbf{R}) e^{-i\mathbf{Q} \cdot \mathbf{R}}, \quad (39)$$

$$g(\mathbf{R}) = \delta(\mathbf{R}), \quad g_{-\mathbf{Q}} = \frac{1}{V}. \quad (40)$$

In this approximation, the macroscopic carrier density, Eq.(25) and the macroscopic current density, Eq.(26), become:

$$\rho(R) = \frac{e}{V} \sum_{\lambda_1 \lambda_2} \sum_{q, Q} e^{-iQ \cdot R} \left(\lambda_1 q + \frac{Q}{2} \right) \left| \lambda_2 q - \frac{Q}{2} \right\rangle a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q - \frac{Q}{2}}, \quad (41)$$

$$\mathbf{j}(R) = \frac{e}{m_0 V} \sum_{\lambda_1 \lambda_2} \sum_{q, Q} e^{-iQ \cdot R} \left(\left(\lambda_1 q + \frac{Q}{2} \right) \mathbf{p} + \mathbf{q} \left| \lambda_2 q - \frac{Q}{2} \right\rangle - \frac{e}{c_0} A(R) \left(\lambda_1 q + \frac{Q}{2} \right) \left| \lambda_2 q - \frac{Q}{2} \right\rangle \right) a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q - \frac{Q}{2}}. \quad (42)$$

Macroscopic density and current are both represented by the Fourier-transform of the quantum mechanical transition amplitudes. The amplitudes are weighted by the Bloch integrals. Their expansion in a series around finite wavenumber values q or Q yields the multipole presentation for a solid.

IV. FIRST EXAMPLE: MULTIPOLE EXPANSION AT THE SEMICONDUCTOR BAND EDGE

As an illustrative example of our averaging procedure for solids, the multipole expansion for a bulk semiconductor for optical excitation close to its band edge is computed. For optical transitions around the band edge, only small wavenumber values q, Q are involved in Eqs. (41) and (42) [4]. To calculate the Bloch integrals in the limit of small wavenumbers, $k \cdot p$ theory [10] is adopted to expand the lattice periodic functions $u_{\lambda k}$. Unfortunately, for many semiconductors the valence band structure is degenerate at $k = 0$ and requires in general the solution of the Luttinger Hamiltonian. However, for the clarity of the presented approach and to obtain analytic results for a case study, only the nondegenerate situation is discussed here. For nondegenerate bands, $k \cdot p$ perturbation theory to second order for $u_{\lambda k} = \langle \tau | \lambda k \rangle$ yields:

$$\begin{aligned} |\lambda k\rangle = & \left(1 - \frac{1}{2} \sum_{\nu_1} \frac{|\langle \nu_1 | \mathbf{k} \cdot \mathbf{p} | \lambda \rangle|^2}{(\epsilon_{\nu_1} - \epsilon_\lambda)^2 m_0^2} |\nu_1\rangle \right) |\lambda\rangle \\ & + \sum_{\nu} \frac{\langle \nu | \mathbf{k} \cdot \mathbf{p} | \lambda \rangle}{(\epsilon_\lambda - \epsilon_\nu) m_0} |\nu\rangle \\ & + \sum_{\nu_1 \nu_2} \frac{\langle \nu_1 | \mathbf{k} \cdot \mathbf{p} | \nu_2 \rangle \langle \nu_2 | \mathbf{k} \cdot \mathbf{p} | \lambda \rangle}{(\epsilon_\lambda - \epsilon_{\nu_2})(\epsilon_\lambda - \epsilon_{\nu_1}) m_0^2} |\nu_1\rangle \\ & - \sum_{\nu_1} \frac{\langle \lambda | \mathbf{k} \cdot \mathbf{p} | \lambda \rangle \langle \nu_1 | \mathbf{k} \cdot \mathbf{p} | \lambda \rangle}{(\epsilon_{\nu_1} - \epsilon_\lambda)^2 m_0^2} |\nu_1\rangle. \end{aligned} \quad (43)$$

In the following we assume inversion symmetry of the crystal and neglect permanent momentum elements $\langle \lambda | \mathbf{p} | \lambda \rangle$.

Using Eq. (43) to calculate the Bloch integrals in Eq. (41) one obtains (after a lengthy but straightforward calculation) the macroscopic charge density up to second order in $k \cdot p$ theory:

$$\rho(R) = F_e(R) - F_h(R) - \nabla_R \cdot \left(\mathbf{P}(R) - \nabla_R \cdot \hat{\mathbf{Q}}(R) \right) \quad (44)$$

where the monopole density $F_{e/h}$ of electrons e and holes h , the dipole polarization vector $\mathbf{P}$ and quadrupole polarization tensor $\hat{\mathbf{Q}}$ occur. The detailed definition of these quantities is:

$$F_{e/h} = \frac{e}{V} \sum_{\mathbf{q}, \mathbf{Q}, \lambda} e^{-i\mathbf{Q} \cdot \mathbf{R}} \sigma_{\mathbf{qQ}}^{\lambda_e/h}, \quad (45)$$

$$\mathbf{P} = \frac{1}{V} \sum_{\mathbf{q}, \mathbf{Q}, \lambda} e^{-i\mathbf{Q} \cdot \mathbf{R}} \left(d_{\lambda_1 \lambda_2} - i\mathbf{q} \cdot \hat{\mathbf{B}}_{\lambda_1 \lambda_2}^{\lambda_1} - i\hat{\mathbf{B}}_{\lambda_1 \lambda_2}^{\lambda_2} \cdot \mathbf{q} \right) \sigma_{\mathbf{qQ}}^{\lambda_1 \lambda_2}, \quad (46)$$

$$\hat{\mathbf{Q}} = \frac{1}{V} \sum_{\mathbf{q}, \mathbf{Q}, \lambda} e^{-i\mathbf{Q} \cdot \mathbf{R}} \hat{\mathbf{B}}_{\lambda_1 \lambda_2}^{\lambda_2} \sigma_{\mathbf{qQ}}^{\lambda_1 \lambda_2}. \quad (47)$$

Here, the abbreviations

$$a_{c_1 \mathbf{q} + \frac{\mathbf{Q}}{2}}^\dagger a_{c_2 \mathbf{q} - \frac{\mathbf{Q}}{2}} = \sigma_{\mathbf{qQ}}^{\lambda_{c_1} \lambda_{c_2}} \quad (48)$$

have been introduced for transitions and occupations within the conduction bands ($\lambda_{c_1} = \lambda_{e_1}$) as well as

$$a_{\lambda_{v_1} \mathbf{q} + \frac{\mathbf{Q}}{2}}^\dagger a_{\lambda_{v_2} \mathbf{q} - \frac{\mathbf{Q}}{2}} = \delta_{v_1, v_2} \delta_{\mathbf{q} + \frac{\mathbf{Q}}{2}, \mathbf{q} - \frac{\mathbf{Q}}{2}} - \sigma_{\mathbf{qQ}}^{\lambda_{h_1} \lambda_{h_2}} \quad (49)$$

for transitions and occupations within the valence bands ($\lambda_{v_1} = \lambda_{h_1}$). Furthermore, we use

$$a_{\lambda_{v_1} \mathbf{q} + \frac{\mathbf{Q}}{2}}^\dagger a_{\lambda_{c_2} \mathbf{q} - \frac{\mathbf{Q}}{2}} = \sigma_{\mathbf{qQ}}^{\lambda_{h_1} \lambda_{c_2}} \quad (50)$$

for transitions between valence and conduction bands. Here, e denotes electronic, h hole- quantities [4].

The explicit microscopic expressions for the dipole components $d_{\lambda_1 \lambda_2}^\alpha$ and quadrupole-like moments $B_{\lambda_1 \lambda_2}^{\alpha\beta\lambda}$ read:

$$d_{\lambda_1 \lambda_2}^\alpha \equiv -ie \frac{\langle \lambda_1 | p^\alpha | \lambda_2 \rangle}{m_0(\epsilon_{\lambda_1} - \epsilon_{\lambda_2})}, \quad (51)$$

$$B_{\lambda_1 \lambda_2}^{\alpha\beta\lambda} \equiv e \sum_{\nu} \frac{\langle \lambda_1 | p^\alpha | \nu \rangle \langle \nu | p^\beta | \lambda_2 \rangle}{m_0^2(\epsilon_{\lambda_1} - \epsilon_{\lambda_2})(\epsilon_{\lambda} - \epsilon_{\nu})}. \quad (52)$$

ϵ_λ denotes the transition energies at the bandedge of the band λ and the integrals between the Bloch-functions $|\lambda\rangle$ at $\mathbf{k} = 0$ have to be taken over one elementary cell of the solid. Note, that the coefficients $d_{\lambda_1 \lambda_2}^\alpha$ and $B_{\lambda_1 \lambda_2}^{\alpha\beta\lambda}$ are directly related to the expressions for one- and two-photon absorption coefficients, respectively [13].

Within the same approximation scheme we obtain for the current:

$$\mathbf{j}(\mathbf{R}) = -i \frac{1}{V} \sum_{\mathbf{q}, \mathbf{Q}, \lambda} e^{-i\mathbf{Q} \cdot \mathbf{R}} \sigma_{\mathbf{qQ}}^{\lambda_1 \lambda_2} (\epsilon_{\lambda_2} - \epsilon_{\lambda_1}) \left(d_{\lambda_1 \lambda_2} - i\hat{\mathbf{B}}_{\lambda_1 \lambda_2}^{\lambda_1} \cdot \left(\mathbf{q} + \frac{\mathbf{Q}}{2} \right) - i\left(\mathbf{q} - \frac{\mathbf{Q}}{2} \right) \cdot \hat{\mathbf{B}}_{\lambda_1 \lambda_2}^{\lambda_2} \right), \quad (53)$$

where for clarity off-resonant terms have been neglected in the rotating wave approximation.

To determine the quantum mechanical expectation values of the operators $\sigma_{\mathbf{qQ}}^{\lambda_1 \lambda_2}$, the Heisenberg equations of

motion for $a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q - \frac{Q}{2}}$ have to be calculated. However, before giving again an example for a bulk semiconductor, the minimal coupling Hamiltonian, has to be rewritten in the coarse grained spatial coordinate R to obtain a macroscopic Hamiltonian.

V. MACROSCOPIC MINIMAL COUPLING HAMILTONIAN FOR THE SOLID

The transition from the microscopic to the macroscopic space scale in the minimal coupling Hamiltonian is illustrated for the matrix element $\langle 1|\Phi(r)|2\rangle$ in Eq.(5). Using the solid state single particle wavefunctions, Eq.(10), the matrix element in Eq.(5) takes the form:

$$\langle 1|\Phi(r)|2\rangle = \frac{1}{V} \int dV e^{-i(k_1 - k_2) \cdot r} u_{\lambda_1 k_1}^*(r) u_{\lambda_2 k_2}(r) \Phi(r). \quad (54)$$

Again splitting the spatial integration into a sum and integration over the elementary cells, compare Sec.III B, we obtain:

$$\begin{aligned} \langle 1|\Phi(r)|2\rangle &= \frac{1}{V} \sum_R \int_{\Omega_0} d^3 r e^{-i(k_1 - k_2)(R+r)} \Phi(R+r) u_{\lambda_1 k_1}^*(r) u_{\lambda_2 k_2}(r) \\ &= \frac{1}{V} \sum_R e^{-i(k_1 - k_2)R} \int_{\Omega_0} d^3 r e^{-i(k_1 - k_2)r} u_{\lambda_1 k_1}^*(r) u_{\lambda_2 k_2}(r) \cdot \sum_K \Phi_K e^{-iK(R+r)} \end{aligned} \quad (55)$$

where we have used the periodicity of $u_{\lambda k}(r)$ and introduced the Fourier transform of the scalar potential Φ_K . Evaluating the lattice sum and neglecting Umklapp processes similar to the calculation in Sec.III B we get:

$$\langle 1|\Phi(r)|2\rangle = \frac{1}{V} \int d^3 R e^{-iQ \cdot R} \Phi(R) \langle \lambda_1 q + \frac{Q}{2} | \lambda_2 q - \frac{Q}{2} \rangle. \quad (56)$$

After these approximations, the Hamiltonian H_2 reads:

$$H_2 = \frac{e}{V} \sum_{\lambda_1 \lambda_2} \sum_{qQ} \int d^3 R e^{-iQ \cdot R} \Phi(R) \langle \lambda_1 q + \frac{Q}{2} | \lambda_2 q - \frac{Q}{2} \rangle a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q + \frac{Q}{2}}. \quad (57)$$

In Eq. (57) the microscopic r and the macroscopic R space dependence are separated. The dependence of r occurs in terms of the Bloch integrals and contains the optical selection rules in the Hamiltonian.

To show the self consistency of our approximations the macroscopic Hamiltonian is used now to determine the macroscopic sources ρ and j for Maxwell's equations. Explicitly, ρ and j are computed in the presence of an additional external potential $\Phi_{ext}(R)$, $A_{ext}(R)$, which implies $\Phi \rightarrow \Phi + \Phi_{ext}$ in H_2 and $A \rightarrow A + A_{ext}$ in H_1 [?]. In this case, the macroscopic carrier density $\rho(R)$ is calculated as:

$$\rho(R) = \frac{\delta H}{\delta \Phi_{ext}(R)} = \frac{e}{V} \sum_{\lambda_1 \lambda_2} \sum_{qQ} e^{-iQ \cdot R} \langle \lambda_1 q + \frac{Q}{2} | \lambda_2 q - \frac{Q}{2} \rangle a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q - \frac{Q}{2}}, \quad (58)$$

which is exactly the macroscopic density obtained from the spatial average of the microscopic sources, Eq. (41).

By comparison with the macroscopic Poisson equation $\nabla \nabla \cdot \Phi = -4\pi \rho(R)$ the macroscopic potential $\Phi(R)$ can be determined as:

$$\Phi(R) = \frac{e}{V} \sum_{1,2} e^{-iQ \cdot R} \frac{4\pi}{Q^2} \cdot \langle \lambda_1 q + \frac{Q}{2} | \lambda_2 q - \frac{Q}{2} \rangle a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q + \frac{Q}{2}} + \Phi_{ext}(R). \quad (59)$$

A similar evaluation of H_1 , Eq. (4), yields the macroscopic electron - transversal field interaction ($H_1 = H_1' + H_1''$):

$$\begin{aligned} H_1' &= \sum_{1,2} -\frac{e}{c_0 m_0} < 1 | A \cdot p | 2 > a_1^\dagger a_2, \\ &= -\frac{e}{c_0 m_0} \frac{1}{V} \int d^3 R e^{-iQ \cdot R} A(R) < \lambda_1 q + \frac{Q}{2} | p + q | \lambda_2 q - \frac{Q}{2} > a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q + \frac{Q}{2}} \\ H_1'' &= \frac{e^2}{2c_0^2 m_0} < 1 | A^2 | 2 > a_1^\dagger a_2 \\ &= \frac{e^2}{2c_0^2 m_0} \int d^3 R e^{-iQ \cdot R} b f A^2(R) < \lambda_1 q + \frac{Q}{2} | \lambda_2 q - \frac{Q}{2} > a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q + \frac{Q}{2}}, \quad (60) \end{aligned}$$

as well as the macroscopic current $j(R)$ via a functional derivative with respect to the external vector potential:

$$\begin{aligned} j(R) &= \frac{\delta H}{\delta A_{ext}(R)} \\ &= \frac{e}{m_0 V} \sum_{qQ} \sum_{\lambda} e^{-iQ \cdot R} \left(< \lambda_1 q + \frac{Q}{2} | p + q | \lambda_2 q - \frac{Q}{2} > - \frac{e}{c_0} A(R) < \lambda_1 q + \frac{Q}{2} | \lambda_2 q - \frac{Q}{2} > \right) a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q - \frac{Q}{2}}. \quad (61) \end{aligned}$$

The expression for the macroscopic current determined from the Hamiltonian agrees with the current determined from the spatial average procedure, Eq. (42).

Using Eqs. (59) and (60) the macroscopic Hamiltonian can be written as:

$$\begin{aligned} H &= \sum_{\lambda,k} \varepsilon_{\lambda k} a_{\lambda k}^\dagger a_{\lambda k} \\ &- \frac{1}{c_0 m_0} \sum_{qQ\lambda} < \lambda_1 q + \frac{Q}{2} | p + q | \lambda_2 q - \frac{Q}{2} > A_{-Q} a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q - \frac{Q}{2}} \\ &- \frac{e}{2c_0} \sum_{qQ\lambda} < \lambda_1 q + \frac{Q}{2} | \lambda_2 q - \frac{Q}{2} > A_{-Q}^2 a_{\lambda_1 q + \frac{Q}{2}}^\dagger a_{\lambda_2 q - \frac{Q}{2}} \\ &+ \frac{1}{2} \sum_{1234} V_{1234} a_1^\dagger a_2^\dagger a_3 a_4 + \int \frac{dV}{8\pi} (E_T^2(R) + B^2(R)) \quad , \quad (62) \end{aligned}$$

where the generalized Coulomb-interaction matrix V has been introduced:

$$\begin{aligned} V_{1234} &= V_{k_1 - k_3} \delta_{k_1 + k_2, k_3 + k_4} < \lambda_1 q + Q | \lambda_3 q > < \lambda_2 q' - Q | \lambda_4 q' > \\ V_{k_1 - k_3} &= \frac{4\pi e^2}{V |k_1 - k_3|^2}. \quad (63) \end{aligned}$$

The spatially averaged Hamiltonian contains the diagonal single-particle part, the electron-photon interaction where A_{-Q} is the Fourier-transform of the vector

potential, and the electron-electron interaction via the Coulomb potential.

Evaluating the Heisenberg equation of motion using the macroscopic Hamiltonian (62), it can be shown that the macroscopic current (61) fulfills the equation of continuity with the macroscopic carrier density, Eq.(58). This shows that a consistent level for the spatial average in the observables as well as in the description of the Hamiltonian is obtained.

The next step, to set up a self-consistent formulation of Hamiltonian and Maxwell's equations, is to rewrite the vector potential and the scalar potential, which both represent the transversal and the longitudinal fields, using the full fields. The problems connected with this procedure are discussed in the next section.

VI. TRANSVERSAL AND LONGITUDINAL VERSUS FULL FIELDS

Besides the spatial averaging, the second problem arising in the semiclassical treatment is that the equations of motion for the material quantities $\langle a_1^\dagger a_2 \rangle$, involve the minimal coupling Hamiltonian which depends on the electromagnetic potentials, not on the fields. Therefore, the system of material equations derived from the minimal coupling Hamiltonian and Maxwell's equations does not close on the level of the total electric and magnetic fields. Hence it cannot be treated by a direct numerical integration [11]. A formulation of Maxwell's theory in form of the wave equations for the transversal field or the vector potential, however, is problematic because for the numerical solution the sources have to be known at every space point to obtain the required projection on their transversal contribution.

For atomic systems, this self-consistency problem can be solved by the Poincare' transformation [?] which removes the potentials from the Hamiltonian in favor of the total electromagnetic fields. However, to the best of our knowledge, this can only be done for localized charge distributions. Another possibility to remove the potentials from the Hamiltonian which can also be applied for delocalized carriers in solids is to express the potentials approximately in terms of the fields without changing the gauge.

A typical example for this approach is to apply the slowly varying envelope approximation (SVEA) [2] for the vector potential:

$$A(r, t) \approx \frac{c_0}{i\omega_l} \bar{E}_T(r, t) e^{-i\omega_l t} \quad (64)$$

which should yield reliable results as long as the inter- and intraband transitions do not couple. Here, ω_l is the carrier frequency of the light and $\bar{E}_T$ is the slowly varying envelope of the electric field. In this case, by reconstructing the longitudinal field from $E_L = -\nabla_R \Phi$ and

combining it with the transversal part E_T , the total electric field $E(R)$ appears and the set of field and material equations is closed on the level of the total fields.

VII. SECOND EXAMPLE: GAUGE INVARIANT OPTICAL RESPONSE AT THE SEMICONDUCTOR BAND EDGE

In this section, it is shown that within the slowly varying envelope approximation, Eq.(64), the rotating wave approximation, and at the level of the multipole approximation, Eq.(44), the material dynamics is driven by the full electric field, not only by its transversal part. Thus, the description using material and Maxwell's equations is self-consistent.

For this purpose, the Heisenberg equations of motion for the quantum mechanical averages $\langle \sigma_{k_1 k_2}^{vc} \rangle$ have to be calculated using the macroscopic Hamiltonian, Eq.(62):

$$-i\hbar \frac{\partial \langle \sigma_{k_1 k_2}^{vc} \rangle}{\partial t} = \langle [\sigma_{k_1 k_2}^{vc}, H] \rangle. \quad (65)$$

In general, Eq.(65) contains the full hierarchy of many body effects, containing mean-field (Hartree-Fock) contributions as well as Coulomb correlations [4]. For a semiconductor being initially in the ground state, the source for the material dynamics is the classical field. Here, to show that the full classical field (not only its transversal part) is the source term in the equation of motion, Eq.(65), the following calculations are restricted to the Hartree contribution of the longitudinal Coulomb interaction and the contribution due to the transversal vector potential. Evaluating Eq. (65) and keeping only the Hartree contribution, the equations for the interband transitions read:

$$\begin{aligned} -i\partial_t \langle \sigma_{k_1 k_2}^{vc} \rangle|_{source} &= \sum_{\lambda, q} \langle \sigma_{k_1 + q k_2}^{\lambda c} \rangle \langle \lambda k_1 + q | \left(e\Phi_{-q} - \frac{e}{c_0 m_0} (\mathbf{p} + \mathbf{k}_1 + \frac{q}{2}) \mathbf{A}_{-q} - \frac{e^2}{2c_0 m_0} \mathbf{A}_{-q}^2 \right) | v k_1 \rangle \\ &\quad - \sum_{\lambda, q} \langle \sigma_{k_1 k_2 - q}^{\nu \lambda} \rangle \langle \lambda k_2 | \left(e\Phi_{-q} - \frac{e}{c_0 m_0} (\mathbf{p} + \mathbf{k}_2 - \frac{q}{2}) \mathbf{A}_{-q} - \frac{e^2}{2c_0 m_0} \mathbf{A}_{-q}^2 \right) | \lambda k_2 - q \rangle. \end{aligned} \quad (66)$$

Using second order $k \cdot p$ -theory, Eq.(43), the matrix elements in the material equations can be expanded around the band edge. Applying additionally the slowly varying envelope equation, Eq.(64), and the rotating wave approximation (neglect of the second order term in the vector potential) the contribution resulting from the transversal interaction reads:

$$-i\partial_t \sigma_{k_1 k_2}^{vc}|_{trans} = \left(d_{cv} - i(k_2 \cdot \hat{\mathbf{B}}_{cv}^c + \hat{\mathbf{B}}_{cv}^v \cdot \mathbf{k}_1) \right) \cdot \mathbf{E}_{T k_1 - k_2}. \quad (67)$$

The longitudinal interaction contribution takes the form:

$$-i\partial_t \sigma_{k_1 k_2}^{vc}|_{long} = \left(d_{cv} - i(k_2 \cdot \hat{\mathbf{B}}_{cv}^c + \hat{\mathbf{B}}_{cv}^v \cdot \mathbf{k}_1) \right) \cdot \frac{(\mathbf{k}_1 - \mathbf{k}_2)(\mathbf{k}_1 - \mathbf{k}_2)}{(\mathbf{k}_1 - \mathbf{k}_2) \cdot (\mathbf{k}_1 - \mathbf{k}_2)} \cdot \mathbf{E}_{k_1 - k_2}. \quad (68)$$

From Eqs.(67) and (68), it can be recognized that the direct part of the Coulomb interaction and the interaction with the transversal field can be rewritten into the interaction with the Fourier-transform of the total field $E_{k_1-k_2}$:

$$-i\partial_t\sigma_{k_1k_2}^{vc}|_{source} = \left(d_{cv} - i(k_2 \cdot \hat{B}_{cv}^c + \hat{B}_{cv}^v \cdot k_1)\right) \cdot E_{k_1-k_2}. \quad (69)$$

The transformation into relative and center of mass coordinates yields:

$$-i\partial_t\sigma_{qQ}^{vc}|_{source} = \left(d_{cv} - i\left(q - \frac{Q}{2}\right) \cdot \hat{B}_{cv}^c - i\hat{B}_{cv}^v \cdot \left(q + \frac{Q}{2}\right)\right) \cdot E_{-Q}. \quad (70)$$

Eq. (70) shows that the material dynamics is determined by the dipole and the quadrupole contributions with respect to the total field. Note that in our approach there is no need to distinguish between electric and magnetic contributions [14]. Higher order moments occur in connection with a spatial derivative of the electric field and contain electric quadrupole and magnetic dipole radiation in the same notation.

VIII. CONCLUSIONS

The multipole approximation for atomic systems has been extended for the case of spatially resolved spectroscopy on macroscopic length scales, where delocalized electrons dominate the optical response. For this purpose, the multipole response has been traced back to Bloch integrals over the elementary cell of the crystal lattice, keeping simultaneously the spatial dependence of the material dynamics on a macroscopic length scale. The multipole expansion of the Bloch integrals shows that the dynamics of the material correlations is driven by the full optical field. Thus it is not necessary to project on its transversal part. This result is especially important for the numerical evaluations in nano-optics for resonantly excited materials, where the vectorial Maxwell's equations are solved simultaneously with a microscopic material theory, both formulated for the full optical field.

Future work in this field should avoid the rotating wave and the slowly varying envelope approximations to obtain results valid for pulse as short as the cycle of light. In addition, even if the approach developed for solids is illustrated for a bulk semiconductor, it can be extended for quantum confined structures using the envelope approximation under a quantum confinement situation.

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