Microscopic theory of linear light scattering from mesoscopic media and in near-field optics

Ole Keller
Institute of Physics, Aalborg University, Pontoppidanstræde 103, DK-9220 Aalborg Øst, Denmark
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On the basis of quantum mechanical response theory a microscopic propagator theory of linear light scattering from mesoscopic systems is presented. The central integral equation problem is transferred to a matrix equation problem by discretization in transitions between pairs of (many-body) energy eigenstates. The local-field calculation which appears from this approach is valid down to the microscopic region. Previous theories based on the (macroscopic) dielectric constant concept make use of spatial (geometrical) discretization and cannot in general be trusted on the mesoscopic length scale. The present theory can be applied to light scattering studies in near-field optics. After a brief discussion of the macroscopic integral equation problem a microscopic potential description of the scattering process is established. In combination with the use of microscopic electromagnetic propagators the formalism allows one to make contact to the macroscopic theory of light scattering and to the spatial photon localization problem. The quantum structure of the microscopic conductivity response tensor enables one to establish a clear physical picture of the origin of local-field phenomena in mesoscopic and near-field optics. The Huygens scalar propagator formalism is revisited and its generality in microscopic physics pointed out.

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I. INTRODUCTION

Theoretical studies of the light scattering from (nonmagnetic) condensed matter media often are carried out on the basis of macroscopic electrodynamics, and considerable insight in the underlying physics obtained using Green's function techniques. In the space ($\mathbf{r}$)-frequency (ω) domain one usually starts from the following propagator equation for the macroscopic electric field, $\mathbf{E}(\mathbf{r};\omega)$ [1–3]:

$$\mathbf{E}(\mathbf{r};\omega) = \mathbf{E}^{ext}(\mathbf{r};\omega) + \mathbf{E}^{SF}(\mathbf{r};\omega) - \mu_0\omega^2 \times \int_{\epsilon\to 0} \overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)\cdot\mathbf{P}(\mathbf{r}';\omega)d^3r', \quad (1)$$

where $\mathbf{E}^{ext}(\mathbf{r};\omega)$ is the external (incident) electric field driving the electrodynamics of the medium, and $\mathbf{P}(\mathbf{r};\omega)$ is the macroscopic polarization induced in the scattering regime. The dyadic electromagnetic propagator $\overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)$ is singular at $\mathbf{r}'=\mathbf{r}$, and the singularity is so strong that the integral over the singularity is only conditionally convergent [1]. The various choices of the geometry of the principal volume ϵ which excludes the singularity of $\overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)$, and which becomes infinitesimally small ($\epsilon\to 0$) at the end of the calculation as indicated, lead to the addition of different so-called self-field contributions, $\mathbf{E}^{SF}(\mathbf{r};\omega)$, to the prevailing electric field [2]. The self-field only contributes to the electric field inside the scattering medium. In the Appendix, I briefly discuss the issue of the singularity of the propagator, paying particular attention to the spherical contraction scheme. The macroscopic electric field can only be determined from Eq. (1) if the prevailing polarization distribution is known (and vice versa). In the linear regime of macroscopic electrodynamics traditionally one relates $\mathbf{P}(\mathbf{r};\omega)$ and $\mathbf{E}(\mathbf{r};\omega)$ via a phenomenological constitutive relation of the form

$$\mathbf{P}(\mathbf{r};\omega) = \epsilon_0\overleftrightarrow{\chi}(\mathbf{r};\omega)\cdot\mathbf{E}(\mathbf{r};\omega), \quad (2)$$

where $\overleftrightarrow{\chi}(\mathbf{r};\omega)$ is the macroscopic susceptibility of the medium. The tensorial form of $\overleftrightarrow{\chi}(\mathbf{r};\omega)$ and its space dependence allow one to include optical anisotropy and inhomogeneity in the formalism. In Eq. (2) the quantities $\mathbf{P}, \overleftrightarrow{\chi}$, and $\mathbf{E}$ relate to the same angular frequency, and this is so because we have assumed that the medium does not change its physical properties over time (translational invariance in time). It has also been assumed that the constitutive equation is local in space, i.e., the polarization at a given space point ($\mathbf{r}$) does only depend on the electric field in the same point [4]. By insertion of Eq. (2) into Eq. (1) one obtains the central propagator equation for analyses of linear light scattering in macroscopic electrodynamics, namely,

$$\mathbf{E}(\mathbf{r};\omega) = \mathbf{E}^{ext}(\mathbf{r};\omega) + \mathbf{E}^{SF}(\mathbf{r};\omega) - q_0^2\int_{\epsilon\to 0} \overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)\cdot\overleftrightarrow{\chi}(\mathbf{r}';\omega)\cdot\mathbf{E}(\mathbf{r}';\omega) \times d^3r', \quad (3)$$

where $q_0=\omega/c_0$ is the vacuum wave number of light. Since the macroscopic electric self-field, $\mathbf{E}^{SF}(\mathbf{r};\omega)$, at least approximately as we shall realize, can be expressed in terms of the macroscopic polarization which in turn relates to the electric field via Eq. (2), Eq. (3) is an integral equation from which the macroscopic electric field may be determined assuming $\overleftrightarrow{\chi}(\mathbf{r};\omega)$ [and $\overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)$] to be known. The integral equation in Eq. (3) can be used to study not only standard scattering problems in macroscopic electrodynamics but also hole and grating diffraction problems in physical optics, for example. Over the years various techniques have been employed to solve Eq. (3), or simpler (approximate) forms of integral equations for $\mathbf{E}(\mathbf{r};\omega)$. The most commonly used technique is a method of perturbation, in which successive

terms in the expansion are obtained by iteration (Born Series technique) [4]. Max Born used this technique in the context of quantum mechanical collision theory [5], and the scalar theory of optical scattering is from a mathematical point of view closely related to the scalar wave function scattering in quantum physics [4]. If the scattering of the external field is sufficiently weak usually only a few terms in the Born series expansion are needed. The Born approach is very illuminating from a physical point of view because it allows one to describe single, double, etc., and multiple scattering events in a direct manner [4,6]. In the given framework all self-sustaining solutions (eigenmodes) in the nonmagnetic medium under considerations in principle can be determined from the homogeneous integral equation one obtains by setting $\mathbf{E}^{ext}(\mathbf{r};\omega)=\mathbf{0}$ in Eq. (3). If the electromagnetic interaction of the external field with the particle system is strong it may be necessary to seek the exact solution to Eq. (3), and in most cases this means that one has to resort to numerical methods. The exact solution incorporates multiple scattering events to infinite order. The understanding of many macroscopic problems in near-field optics and in light scattering from mesocopic media (quantum dots, wires, and wells, e.g.) require exact solutions of Eq. (3). In near-field optics this is often so in situations where the external source and the medium are in close contact electromagnetically, and in mesoscopic systems the presence of only few electronic resonances in the optical frequency range of excitation often allows one to drive the system in resonance. Strong interaction of course makes the need for incorporation of nonlinear phenomena more urgent in general, and these phenomena are not included in Eq. (3).

Numerical solutions of Eq. (3) are based on a discretization procedure in direct space. Although the replacement of the integral by a sum of terms over volume elements so small that $\overleftrightarrow{\mathbf{G}}\cdot\overleftrightarrow{\chi}\cdot\mathbf{E}$ can be taken as a constant in each cell works well in many situations it is in general not the case in mesoscopic electrodynamics and near-field optics. Despite this fact one often comes across calculations where people have divided the integral into volume elements of linear extension (almost) comparable to atomic (unit cell) size. Such calculations often aim at understanding local-field effects and interface dynamics. As the need for a shrink in the cell volume size becomes more urgent the macroscopic susceptibility concept, $\overleftrightarrow{\chi}(\mathbf{r};\omega)$, tends to be meaningless [7–10]. Basically, one should always ask oneself the following question when using geometrical (spatial) diecretization procedures: how small do I dare to make the cell volume and still believe in the numerical result? Many researchers attempting to determine the local field in so-called "hot spots" have over the years applied macroscopic analyses [based on Eq. (3)] far beyond their limit of validity and often come up with the local-field enhancements orders of magnitude larger than estimated by quantum physics.

To understand the linear electrodynamics of mesoscopic media as well as many aspects of near-field optics it is usually necessary to replace the macroscopic susceptibility $\overleftrightarrow{\chi}(\mathbf{r};\omega)$ by a microscopic two-point response tensor $\overleftrightarrow{\chi}(\mathbf{r},\mathbf{r}';\omega)$ realizing that the constitutive relation between the relevant polarization $\mathbf{P}$ (to be defined later) and the microscopic electric field $\mathbf{e}$ in general is nonlocal in space, that is [7–10]

$$\mathbf{P}(\mathbf{r};\omega)=\int \overleftrightarrow{\chi}(\mathbf{r},\mathbf{r}';\omega)\cdot\mathbf{e}(\mathbf{r}';\omega)d^3r'. \tag{4}$$

In the micro- and mesoscopic regimes a somewhat simplified version of the integral equation for the microscopic local field reads [10]

$$\begin{aligned}\mathbf{e}(\mathbf{r};\omega)=\mathbf{e}^{ext}(\mathbf{r};\omega)+\mathbf{e}^{SF}(\mathbf{r};\omega)\\ -q_0^2\int_{\epsilon\to 0}\int \overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)\cdot\overleftrightarrow{\chi}(\mathbf{r}',\mathbf{r}'';\omega)\cdot\mathbf{e}(\mathbf{r}'';\omega)\\ \times d^3r''d^3r',\end{aligned} \tag{5}$$

where $\mathbf{e}^{SF}(\mathbf{r};\omega)$ is the microscopic electric self-field in the given contraction scheme. Although the external electric field most often can be considered as a macroscopic quantity we allow it to be a microscopic quantity $(\mathbf{e}^{ext}(\mathbf{r};\omega))$ in Eq. (5). At a first glance it seems hopeless to solve a double integral equation like the one in Eq. (5), even by numerical methods, taking into account the complications already encountered with equations à *la* Eq. (3). One of the main objectives of this work is to show that this is not necessarily correct. The reason is associated with the fact that the microscopic susceptibility has a quite simple tensor product structures, i.e.,

$$\overleftrightarrow{\chi}(\mathbf{r},\mathbf{r}';\omega)=\sum_N a_N(\omega)\mathbf{B}_N(\mathbf{r};\omega)\mathbf{C}_N(\mathbf{r}';\omega). \tag{6}$$

The form in Eq. (6) relates to microscopic physics in an intimite and clear manner and allows one to make a discretization of Eq. (5) in pairs of energy eigenstates (index N) for the light-unperturbed mesoscopic medium as we shall see below. This kind of discretization is valid right down to the atomic domain and can thus be used with confidence. Another main objective of the present study is a quantitative analysis of the physics underlying Eq. (5), and similar integral equations of wider generality. In the course of our description we shall see how macroscopic integral equations such as Eq. (3) appear, and understand the basic premises for using such equations. It is also a goal of this paper to point out certain approximations made in most linear light scattering but not properly identified or even not generally known.

II. MICROSCOPIC POTENTIAL DESCRIPTION

The starting point for us is the microscopic Maxwell equations

$$\nabla\times\mathbf{e}(\mathbf{r},t)=-\frac{\partial}{\partial t}\mathbf{b}(\mathbf{r},t) \tag{7}$$

$$\nabla\times\mathbf{b}(\mathbf{r},t)=\mu_0\mathbf{J}(\mathbf{r},t)+c_0^{-2}\frac{\partial}{\partial t}\mathbf{e}(\mathbf{r},t), \tag{8}$$

$$\nabla\cdot\mathbf{e}(\mathbf{r},t)=\epsilon_0^{-1}\rho(\mathbf{r},t) \tag{9}$$

$$\nabla \cdot \mathbf{b}(\mathbf{r},t) = 0 \tag{10}$$

which relate the microscopic electric $(\mathbf{e}(\mathbf{r},t))$ and magnetic $(\mathbf{b}(\mathbf{r},t))$ fields to the microscopic charge $(\rho(\mathbf{r},t))$ and current $(\mathbf{J}(\mathbf{r},t))$ densities. The (contravariant) current density four-vector $\{J^{\mu}\}=(c_0\rho,\mathbf{J})$ is the only one appearing in the rigorous electrodynamic description of the microscopic particles of matter (in the following our mesoscopic scattering system). The microscopic Maxwell equations lead to the following well-known inhomogeneous wave equations for the microscopic fields in space $(\mathbf{r})$ time (t):

$$(\Box\overleftrightarrow{\mathbf{U}} - \nabla\nabla) \cdot \mathbf{e}(\mathbf{r},t) = \mu_0 \frac{\partial}{\partial t}\mathbf{J}(\mathbf{r},t), \tag{11}$$

$$\Box\mathbf{b}(\mathbf{r},t) = -\mu_0 \nabla \times \mathbf{J}(\mathbf{r},t) \tag{12}$$

where $\Box \equiv \nabla^2 - c_0^{-2}\partial^2/\partial t^2$ is the d'Alembertian operator and $\overleftrightarrow{\mathbf{U}}$ is the 3×3 unit tensor.

Important insight in the physics behind the light scattering process can be obtained in the so-called generalized potential description, which now will be discussed. The potential approach is based on the division of the microscopic current density into two parts, i.e.,

$$\mathbf{J}(\mathbf{r},t) \equiv \frac{\partial}{\partial t}\mathbf{P}(\mathbf{r},t) + \nabla \times \mathbf{M}(\mathbf{r},t) \tag{13}$$

where $\mathbf{P}(\mathbf{r},t)$ and $\mathbf{M}(\mathbf{r},t)$ are *generalized* polarization and magnetization vector fields, respectively [11]. I have stressed the word generalized because $\mathbf{P}(\mathbf{r},t)$ and $\mathbf{M}(\mathbf{r},t)$ are not the textbook quantities defined as the electric and magnetic dipole moments per unit volume [4,12]. Microscopic aspects of $\mathbf{P}(\mathbf{r},t)$ and $\mathbf{M}(\mathbf{r},t)$ are discussed in detail in Ref. [13]. The potential description is fruitful because it allows one to make contacts to (i) the spatial photon localization problem which appears in both photon wave mechanical (first-quantized) and field-theoretic (second-quantized) studies of the scattering process [15–18], (ii) the macroscopic scattering theory, (iii) response theories based on various choices for the microscopic permittivity and permeability tensors, and to see (iv) longitudinal and transverse electrodynamics in a broader perspective [19]. If one divides the vector fields $\mathbf{J},\mathbf{P}$, and $\mathbf{M}$ into their divergence-free and rotational-free parts, in the following called the transverse (subscript T) and longitudinal (subscript L) components, Eq. (13) splits into the relations

$$\mathbf{J}_L(\mathbf{r},t) = \frac{\partial}{\partial t}\mathbf{P}_L(\mathbf{r},t) \tag{14}$$

$$\mathbf{J}_T(\mathbf{r},t) = \frac{\partial}{\partial t}\mathbf{P}_T(\mathbf{r},t) + \nabla \times \mathbf{M}_T(\mathbf{r},t). \tag{15}$$

The L-T division of a vector field is unique in a given inertial system, and from a knowledge of $\mathbf{J}(\mathbf{r},t)$ the microscopic quantities $\mathbf{J}_T(\mathbf{r},t)$ and $\mathbf{J}_L(\mathbf{r},t)$ are determined. Apart from an unimportant constant the longitudinal part of the polarization is fixed by the microscopic charge density via

$$\rho(\mathbf{r},t) = -\nabla \cdot \mathbf{P}_L(\mathbf{r},t), \tag{16}$$

as one readily sees by combining Eq. (14) and the equation of continuity, $\nabla\cdot\mathbf{J}_L(\mathbf{r},t) + \partial\rho(\mathbf{r},t)/\partial t = 0$. The transverse quantities $\mathbf{P}_T(\mathbf{r},t)$ and $\mathbf{M}_T(\mathbf{r},t)$ are *not* uniquely determined since old $(\mathbf{P}_T^{OLD},\mathbf{M}_T^{OLD})$ and new $(\mathbf{P}_T^{NEW},\mathbf{M}_T^{NEW})$ sets related by

$$\mathbf{P}_T^{NEW}(\mathbf{r},t) = \mathbf{P}_T^{OLD}(\mathbf{r},t) + \nabla \times \mathbf{N}(\mathbf{r},t) \tag{17}$$

$$\mathbf{M}_T^{NEW}(\mathbf{r},t) = \mathbf{M}_T^{OLD}(\mathbf{r},t) - \frac{\partial}{\partial t}\mathbf{N}(\mathbf{r},t) \tag{18}$$

where $\mathbf{N}(\mathbf{r},t)$ is an arbitrary vector field, yet differentiable in space and time, lead to the same transverse current density $(\mathbf{J}_T^{NEW}=\mathbf{J}_T^{OLD}=\mathbf{J}_T)$ as one readily may realize. Due to the fact that only $\mathbf{M}_T$ enters the potential description $[\nabla\times\mathbf{M}=\nabla\times\mathbf{M}_T$ in Eq. (13)] one may choose $\mathbf{M}_L$ as appropriate (see Ref. [11]), and for instance take $\mathbf{M}_L=\mathbf{0}(\Rightarrow\mathbf{M}=\mathbf{M}_T)$.

On the micorscopic level only $\mathbf{J}$ and ρ are of physical significance. The longitudinal part ot the polarization is fixed by Eq. (16), and this part of the generalized polarization can be synthesized by adding up a (linear) distribution of point dipoles as described in Ref. [19], for instance. The nonuniqueness of the transverse part of the polarization geometrically can be associated to the fact that one can add to this distribution an arbitrary number of point dipoles arranged to form a closed curve in space, without affecting the prevailing microscopic charge and current densities. Various loops result in various choices for the transverse polarization and the individual loops have their own magnetic moment [19]. For a detailed discussion of this picture the reader is referred to Ref. [19], and references therein. Generalized potentials are also discussed in Ref. [9], and here the relation to linear response theory is treated.

Having decided on a given choice for $\mathbf{P}_T$ a *generalized* microscopic electric displacement $(\mathbf{d})$ field is defined by

$$\mathbf{d}(\mathbf{r},t) \equiv \epsilon_0\mathbf{e}_T(\mathbf{r},t) + \mathbf{P}_T(\mathbf{r},t), \tag{19}$$

where $\mathbf{e}_T(\mathbf{r},t)$ is the uniquely determined transverse part of the microscopic electric field. Since (apart from a trivial constant)

$$\epsilon_0\mathbf{e}_L(\mathbf{r},t) + \mathbf{P}_L(\mathbf{r},t) = \mathbf{0}, \tag{20}$$

as one may realize by combining Eqs. (9) and (16), the $\mathbf{D}$ field can also be written as

$$\mathbf{d}(\mathbf{r},t) = \epsilon_0\mathbf{e}(\mathbf{r},t) + \mathbf{P}(\mathbf{r},t). \tag{21}$$

The $\mathbf{d}$ field is a transverse vector field per definition, cf. Eq. (19).

In the potential formalism the dynamical microscopic field (Maxwell) equations take the flexible form

$$\nabla \times [\epsilon_0^{-1}\mathbf{d}(\mathbf{r},t)] + \frac{\partial}{\partial t}\mathbf{b}(\mathbf{r},t) = \epsilon_0^{-1}\nabla \times \mathbf{P}(\mathbf{r},t), \tag{22}$$

$$\nabla \times \mathbf{b}(\mathbf{r},t) - c_0^{-2}\frac{\partial}{\partial t}[\epsilon_0^{-1}\mathbf{d}(\mathbf{r},t)] = \mu_0 \nabla \times \mathbf{M}(\mathbf{r},t), \tag{23}$$

$$\nabla \cdot [\epsilon_0^{-1}\mathbf{d}(\mathbf{r},t)] = 0, \tag{24}$$

$$\nabla \cdot \mathbf{b}(\mathbf{r},t) = 0 \tag{25}$$

In photon wave mechanics based on the Riemann-Silberstein-Bialynicki energy wave function the combinations $\epsilon_0^{-1}\mathbf{d}(\mathbf{r},t) \pm ic_0\mathbf{b}(\mathbf{r},t)$ play an important role [14–18], and I have emphasized this by using $\epsilon_0^{-1}\mathbf{d}(\mathbf{r},t)$ and $\mathbf{b}(\mathbf{r},t)$ as primary microscopic field quantities in Eqs. (22)–(25). In the potential description the inhomogeneous wave equations for $\mathbf{d}(\mathbf{r},t)/\epsilon_0$ and $\mathbf{b}(\mathbf{r},t)$ become

$$\Box[\epsilon_0^{-1}\mathbf{d}(\mathbf{r},t)] = \nabla \times \left[\mu_0 \frac{\partial}{\partial t}\mathbf{M}(\mathbf{r},t) - \epsilon_0^{-1}\nabla \times \mathbf{P}(\mathbf{r},t)\right], \tag{26}$$

$$\Box\mathbf{b}(\mathbf{r},t) = -\mu_0 \nabla \times \left[\frac{\partial}{\partial t}\mathbf{P}(\mathbf{r},t) + \nabla \times \mathbf{M}(\mathbf{r},t)\right]. \tag{27}$$

III. ELECTROMAGNETIC PROPAGATOR FORMALISM

In the space-frequency domain where

$$\mathbf{J}(\mathbf{r};\omega) = -i\omega\mathbf{P}(\mathbf{r};\omega) + \nabla \times \mathbf{M}(\mathbf{r};\omega), \tag{28}$$

and

$$\mathbf{d}(\mathbf{r};\omega) = \epsilon_0\mathbf{e}_T(\mathbf{r};\omega) + \mathbf{P}_T(\mathbf{r};\omega)[=\epsilon_0\mathbf{e}(\mathbf{r};\omega) + \mathbf{P}(\mathbf{r};\omega)], \tag{29}$$

the inhomogeneous wave equation for the microscopic electric displacement field takes the form

$$(\nabla^2 + q_0^2)[\epsilon_0^{-1}\mathbf{d}(\mathbf{r};\omega)] = -i\mu_0\omega\mathbf{S}(\mathbf{r};\omega), \tag{30}$$

where

$$\mathbf{S}(\mathbf{r};\omega) = \nabla \times \left[\mathbf{M}(\mathbf{r};\omega) + i\frac{c_0^2}{\omega}\nabla \times \mathbf{P}(\mathbf{r};\omega)\right] \tag{31}$$

is the effective source current density. In integral form the relevant solution of Eq. (30) is

$$\epsilon_0^{-1}\mathbf{d}(\mathbf{r};\omega) = \epsilon_0^{-1}\mathbf{d}^{ext}(\mathbf{r};\omega) + i\mu_0\omega\int g(|\mathbf{r}-\mathbf{r}'|;\omega)\mathbf{S}(\mathbf{r}';\omega)d^3r', \tag{32}$$

where $\mathbf{d}^{ext}(\mathbf{r};\omega) = \epsilon_0\mathbf{e}^{ext}(\mathbf{r};\omega)$ is the external displacement field, and with $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, and $R = |\mathbf{R}|$,

$$g(R;\omega) = \frac{\exp(iq_0R)}{4\pi R} \tag{33}$$

is the Huygens scalar propagator. To make the bridge to the dyadic propagator formalism mentioned in the Introduction Eq. (32) is rewritten in the form [10]

$$\epsilon_0^{-1}\mathbf{d}(\mathbf{r};\omega) = \epsilon_0^{-1}\mathbf{d}^{ext}(\mathbf{r};\omega) + \frac{1}{3i\epsilon_0\omega}\mathbf{S}(\mathbf{r};\omega) - i\mu_0\omega\int \overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)\cdot\mathbf{S}(\mathbf{r}';\omega)d^3r', \tag{34}$$

using spherical contraction around the singularity located at $\mathbf{r}' = \mathbf{r}$. Here, and in the following I omit for brevity to put an "$\epsilon \to 0$" at the foot of the integral sign. The self-field contribution is $\mathbf{S}(\mathbf{r};\omega)/(3i\epsilon_0\omega)$ in spherical contraction, and the dyadic (standard) propagator relates to the Huygens propagator via

$$\overleftrightarrow{\mathbf{G}}(\mathbf{R};\omega) = -(\overleftrightarrow{\mathbf{U}} + q_0^{-2}\nabla\nabla)g(R;\omega). \tag{35}$$

The minus sign in front of the parenthesis in Eq. (35) originates in a convention. The explicit from of $\overleftrightarrow{\mathbf{G}}(\mathbf{R};\omega)$ in spherical coordinates may be found in Ref. [20], e.g.

An important point should be emphasized here. Thus, due to the fact that the $\mathbf{S}(\mathbf{r}';\omega)$ is a transverse vector field, see Eq. (31), one may replace the standard propagator $\overleftrightarrow{\mathbf{G}}(\mathbf{R};\omega)$ by the transverse dyadic Green function [10,20]

$$\overleftrightarrow{\mathbf{G}}_T(\mathbf{R};\omega) = q_0^{-2}\nabla\nabla g(R;0) + \overleftrightarrow{\mathbf{G}}(\mathbf{R};\omega), \tag{36}$$

which explicit spherical coordinate form is given elsewhere [20]. The use of $\overleftrightarrow{\mathbf{G}}_T(\mathbf{R};\omega)$ instead of $\overleftrightarrow{\mathbf{G}}(\mathbf{R};\omega)$ in Eq. (34) underlines the fact that $\mathbf{d}(\mathbf{r};\omega)$ is a transverse vector field.

A particular choice namely $\mathbf{M}_T = \mathbf{0}$ is adequate in nonlocal light scattering studies where all magnetic properties are included in the microscopic "dielectric" response function, $\overleftrightarrow{\boldsymbol{\epsilon}}(\mathbf{r},\mathbf{r}';\omega)$, and for making the bridge to the macroscopic scattering theory for nonmagnetic media. It readily appears from Eq. (18) that such a choice is possible. Since $\nabla \times (\nabla \times \mathbf{P}) = -\nabla^2\mathbf{P}_T$, Eqs. (30) and (31) show that

$$(\nabla^2 + q_0^2)\mathbf{d}(\mathbf{r};\omega) = \nabla^2\mathbf{P}_T(\mathbf{r};\omega), \tag{37}$$

or equivalently, in the view of Eq. (19),

$$(\nabla^2 + q_0^2)\mathbf{e}_T(\mathbf{r};\omega) = -\epsilon_0^{-1}q_0^2\mathbf{P}_T(\mathbf{r};\omega). \tag{38}$$

Since Eq. (30) leads to Eq. (34), the relevant dyadic propagator solution of Eq. (37) is (in spherical contraction)

$$\mathbf{e}_T(\mathbf{r};\omega) = \mathbf{e}_T^{ext}(\mathbf{r};\omega) - \frac{1}{3\epsilon_0}\mathbf{P}_T(\mathbf{r};\omega) - \mu_0\omega^2\int \overleftrightarrow{\mathbf{G}}(\mathbf{R};\omega)\cdot\mathbf{P}_T(\mathbf{r}';\omega)d^3r'. \tag{39}$$

If one includes only electric-dipole (ED) contributions in $\mathbf{P}_T$, nonlocal phenomena are omitted and Eq. (39) is reduced to the macroscopic result

$$\mathbf{E}_T(\mathbf{r};\omega) = \mathbf{E}_T^{ext}(\mathbf{r};\omega) - \frac{1}{3\epsilon_0}\mathbf{P}_T^{ED}(\mathbf{r};\omega) - \mu_0\omega^2\int \overleftrightarrow{\mathbf{G}}(\mathbf{R};\omega)\cdot\mathbf{P}_T^{ED}(\mathbf{r}';\omega)d^3r'. \tag{40}$$

The inclusion of only electric-dipole terms in the transverse polarization field is stressed above by the superscript ED

added to $\mathbf{P}_T$. All magnetic scattering phenomena are still included in Eq. (39). Magnetic phenomena all are of spatially nonlocal nature [7,10]. Light scattering related to electric-quadrupole (EQ) and magnetic-dipole (MD) effects can be studied making a multipole expansion of $\mathbf{P}_T$, i.e., $\mathbf{P}_T=\mathbf{P}_T^{ED}+\mathbf{P}_T^{EQ/MD}+\cdots$.

In macroscopic light scattering investigations it is often (implicitly!) implied that the polarization field $\mathbf{P}^{ED}$ and the electric field $\mathbf{E}$ are transverse vector fields, and one often sees the macroscopic integral equation in the form displayed in Eq. (1) without the attached conditions $\boldsymbol{\nabla}\cdot\mathbf{E}(\mathbf{r};\omega)=\boldsymbol{\nabla}\cdot\mathbf{P}(\mathbf{r};\omega)=0$, $\mathbf{P}(\mathbf{r};\omega)=\mathbf{P}_T(\mathbf{r};\omega)$. In the most naive macroscopic approach $\mathbf{P}_T^{ED}$ is simply replaced (approximated) by $\mathbf{P}$, with the consequence that the self-field in Eq. (1) is taken as $\mathbf{E}^{SF}(\mathbf{r};\omega)=-\mathbf{P}(\mathbf{r};\omega)/(3\epsilon_0)$.

With the possible [see Eq. (17)] choice $\mathbf{P}_T=\mathbf{0}$, one gets $\boldsymbol{\nabla}\times\mathbf{M}=\mathbf{J}_T$ [see Eq. (15)], $\mathbf{S}=\mathbf{J}_T$ [Eq. (31)], and $\mathbf{d}=\epsilon_0\mathbf{e}_T$ [Eq. (19)], so that Eq. (34) attains the form

$$\mathbf{e}_T(\mathbf{r};\omega)=\mathbf{e}_T^{ext}(\mathbf{r};\omega)+\frac{1}{3i\epsilon_0\omega}\mathbf{J}_T(\mathbf{r};\omega)-i\mu_0\omega\int\overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)\cdot\mathbf{J}_T(\mathbf{r}';\omega)d^3r'. \quad (41)$$

possible with $\overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)$ replaced by $\overleftrightarrow{\mathbf{G}}_T(\mathbf{r}-\mathbf{r}';\omega)$, cf. the remark made immediately above Eq. (36). The integral equation in Eq. (41) can readily be obtained also from Eq. (39) noting that $\mathbf{J}_T(\mathbf{r};\omega)=-i\omega\mathbf{P}_T(\mathbf{r};\omega)$ when $\mathbf{M}_T(\mathbf{r};\omega)=\mathbf{0}$, cf. Eq. (15). The integral form in Eq. (41) has in recent years been used to study near-local phenomea attached to single-photon scattering from atoms, and the relation between spatial photon localization and the photon source problem [17,18]. Equation (41) is a good starting point for photon wave mechanical investigations of space-time photon localization, and on the basis of Eq. (41) the upgraduation of the first-quantized version of spatial photon localization to the second-quantized level is quite easily carried out.

IV. ASPECTS OF MICROSCOPIC RESPONSE THEORY

The microscopic Maxwell equations have led to an integral (propagator) relation between the transverse electric field $\mathbf{e}_T(\mathbf{r};\omega)$ and the transverse current density of the particle system $\mathbf{J}_T(\mathbf{r};\omega)$, viz. Eq. (41). The two unknown vector fields $\mathbf{E}_T(\mathbf{r};\omega)$ and $\mathbf{J}_T(\mathbf{r};\omega)$ can only be determined if another relation between the two can be established. In the macroscopic standard theory of light scattering the same kind of problem arises. The integral relation between $\mathbf{E}(\mathbf{r};\omega)$ and $\mathbf{P}(\mathbf{r};\omega)$ [Eq. (1)] here is supplemented by the constitutive equation in Eq. (2), and this supplement leads us to the standard integral equation in Eq. (3) for the macroscopic electric field. To solve this equation the $\mathbf{r}$ dependence of the macroscopic susceptibility $\overleftrightarrow{\chi}(\mathbf{r};\omega)$ must be known (postulated or found by a model calculation). In the microscopic theory the constitutive relation is no longer local in space, cf. Eq. (4). Instead one gets in the linear regime [10]

$$\mathbf{J}(\mathbf{r};\omega)=\int\overleftrightarrow{\boldsymbol{\sigma}}(\mathbf{r},\mathbf{r}';\omega)\cdot\mathbf{e}_T(\mathbf{r}';\omega)d^3r', \quad (42)$$

where $\overleftrightarrow{\sigma}(\mathbf{r},\mathbf{r}';\omega)$ is the so-called microscopic conductivity response function. The reader may wonder why only the transverse part of the microscopic electric field, $\mathbf{e}_T(\mathbf{r};\omega)$, and not the entire microscopic field, $\mathbf{e}(\mathbf{r};\omega)$, appears under the integral sign in Eq. (42). The reason is due to the fact that the longitudinal electric field can be eliminated as a dynamical variable in favor of the particle position coordinates [19]. In nonrelativistic microscopic electrodynamics $\overleftrightarrow{\sigma}(\mathbf{r},\mathbf{r}';\omega)$ most often is calculated on the basis of the (many-body) Schrödinger equation, or the Pauli equation if spin dynamics is of importance. The transverse part $\mathbf{J}_T(\mathbf{r};\omega)$ of the microscopic current density $\mathbf{J}(\mathbf{r};\omega)$ entering Eq. (42) can be obtained via

$$\mathbf{J}_T(\mathbf{r};\omega)=\int\overleftrightarrow{\delta}_T(\mathbf{r}-\mathbf{r}')\cdot\mathbf{J}(\mathbf{r}';\omega)d^3r', \quad (43)$$

where $\overleftrightarrow{\delta}_T(\mathbf{r}-\mathbf{r}')$ is the transverse delta-function tensor. By insertion of Eq. (43), with $\mathbf{J}(\mathbf{r}';\omega)$ given by Eq. (42), into Eq. (41) the basic integral equation for the transverse part of the microscopic electric field emerges.

Let us now assume that we know the energies and wave functions of the various stationary states of the mesoscopic medium. For a mesoscopic object, which linear dimensions are in the nanometer region (or smaller) and which geometrical form is simple, it is not out of scope to calculate these eigenenergies and wave functions with a sufficient accuracy for optical purposes, at least numerically. If one denotes the various stationary (many-body) states by $I,J,\ldots$, the associated eigenenergies by $E_I,E_J,\ldots$, and the probability that these states are occupied by $P_I,P_J,\ldots$ it can be shown that the conductivity tensor is given by $(I\neq J)$ [21]

$$\overleftrightarrow{\sigma}(\mathbf{r},\mathbf{r}';\omega)=\frac{2i}{\omega}\sum_{I,J}\left(\frac{\hbar\omega}{E_I-E_J}\right)\frac{P_J-P_I}{\hbar\omega+E_J-E_I}\mathbf{J}_{I\to J}(\mathbf{r})\mathbf{J}_{J\to I}(\mathbf{r}'), \quad (44)$$

if one starts from the (many-body) Schrödinger equation, and neglects irreversible damping mechanisms. To a certain extent these mechanisms can be included phenomenologically by adding suitable damping terms, $i\hbar/\tau_{JI}$, to the $\hbar\omega+E_J-E_I$ denominators. The spin contribution to $\overleftrightarrow{\sigma}(\mathbf{r},\mathbf{r}';\omega)$ can be calculated starting from the Pauli equation but in linear optics this contribution usually is negligible [22]. To include spin-orbit contributions to $\overleftrightarrow{\sigma}(\mathbf{r};\mathbf{r}';\omega)$ a relativistic calculation is required. In the present context where main principles are at focus there is no need to include the above-mentioned phenomena. The expression in Eq. (44) does include both so-called para- and diamagnetic terms, so that the scattering from dielectric, semiconducting, and metallic mesoscopic objects can be studied. The vectors $\mathbf{J}_{I\to J}(\mathbf{r})$ and $\mathbf{J}_{J\to I}(\mathbf{r}')$ in Eq. (44) denote the (many-body) transition current densities from state I to state $J(I\to J)$ and from J to $I(J\to I)$, respectively. In a single-particle description the transition current density from state A to state B is given by

$$\mathbf{J}_{A\to B}(\mathbf{r}) = \frac{e\hbar}{2mi}[\psi_A(\mathbf{r})\,\nabla\,\psi_B^*(\mathbf{r}) - \psi_B^*(\mathbf{r})\,\nabla\,\psi_A(\mathbf{r})], \quad (45)$$

where $\psi_A(\mathbf{r})$ and $\psi_B(\mathbf{r})$ are the time-independent parts of the wave functions belonging to the energy eigenstates A and B, and m, and $-e$ denote the electron mass and charge, respectively. In the following we shall need also the transverse part, $\mathbf{J}^T_{A\to B}(\mathbf{r})$, of $\mathbf{J}_{A\to B}(\mathbf{r})$, and this can be calculated from

$$\mathbf{J}^T_{A\to B}(\mathbf{r}) = \int \overleftrightarrow{\delta}_T(\mathbf{r}-\mathbf{r}')\cdot\mathbf{J}_{A\to B}(\mathbf{r}')d^3r', \quad (46)$$

cf. Eq. (43). The microscopic two-point susceptibility response tensor appearing in Eq. (5) is related to $\overleftrightarrow{\sigma}(\mathbf{r},\mathbf{r}';\omega)$ via

$$\overleftrightarrow{\chi}(\mathbf{r},\mathbf{r}';\omega) = \frac{i}{\epsilon_0\omega}\overleftrightarrow{\sigma}(\mathbf{r},\mathbf{r}';\omega), \quad (47)$$

remembering the choice $\mathbf{J}=\partial\mathbf{P}/\partial t$.

V. SOLUTION OF THE MICROSCOPIC SCATTERING PROBLEM

At this stage we are prepared to take up the central issue of this paper, namely the establishment of a self-consistent solution of the linear light scattering problem which does not invoke geometrical discretization, a principle most often used for macroscopic scattering but certainly of doubtful value for mesoscopic media.

A. Physical structure of the prevailing transverse current density

With the abbreviation

$$\mathcal{A}_{IJ}(\omega) = \frac{2i\hbar}{E_I-E_J}\frac{P_J-P_I}{\hbar\omega+E_J-E_I} \quad (48)$$

the microscopic conductivity tensor is given by

$$\overleftrightarrow{\sigma}(\mathbf{r},\mathbf{r}';\omega) = \sum_{I,J}\mathcal{A}_{IJ}(\omega)\mathbf{J}_{I\to J}(\mathbf{r})\mathbf{J}_{J\to I}(\mathbf{r}'), \quad (49)$$

and it appears that it consists of weighted (weight factor $\mathcal{A}_{IJ}$) tensor products $(\mathbf{J}_{I\to J}(\mathbf{r})\mathbf{J}_{J\to I}(\mathbf{r}')\equiv\mathbf{J}_{I\to J}(\mathbf{r})\otimes\mathbf{J}_{J\to I}(\mathbf{r}'))$ [$\otimes$ is the dyadic (outer) product operator]. The two-coordinate $(\mathbf{r},\mathbf{r}')$ structure of a given tensor product is very simple, indeed. Thus the first factor, $\mathbf{J}_{I\to J}(\mathbf{r})$, is a function of $\mathbf{r}$ alone, and the second factor, $\mathbf{J}_{J\to I}(\mathbf{r}')$, depends only on $\mathbf{r}'$. As we shall realize below it is this property which allows one to go beyond the geometrical discretization scheme. Let us next introduce what may be called the double-transverse (TT) microscopic conductivity tensor

$$\overleftrightarrow{\sigma}_{TT}(\mathbf{r},\mathbf{r}';\omega) = \sum_{I,J}\mathcal{A}_{IJ}(\omega)\mathbf{J}^T_{I\to J}(\mathbf{r})\mathbf{J}^T_{J\to I}(\mathbf{r}'), \quad (50)$$

which involves only the transverse parts of the various transition current densities. With the help of $\overleftrightarrow{\sigma}_{TT}(\mathbf{r},\mathbf{r}';\omega)$ the spatially nonlocal relation between $\mathbf{J}_T(\mathbf{r};\omega)$ and $\mathbf{e}_T(\mathbf{r};\omega)$ can be written in the form

$$\mathbf{J}_T(\mathbf{r};\omega) = \int \overleftrightarrow{\sigma}_{TT}(\mathbf{r},\mathbf{r}';\omega)\cdot\mathbf{e}_T(\mathbf{r}';\omega)d^3r', \quad (51)$$

since

$$\int \mathbf{J}^L_{J\to I}(\mathbf{r})\cdot\mathbf{e}_T(\mathbf{r};\omega)d^3r = 0, \quad (52)$$

$\mathbf{J}^L_{J\to I}(\mathbf{r})=\mathbf{J}_{J\to I}(\mathbf{r})-\mathbf{J}^T_{J\to I}(\mathbf{r})$ being the longitudinal part of the transition current density $\mathbf{J}_{J\to I}(\mathbf{r})$.

By inserting Eq. (50) into Eq. (51) it appears that the transverse current density takes the form

$$\mathbf{J}_T(\mathbf{r};\omega) = \sum_{I,J}\mathcal{A}_{IJ}(\omega)\beta_{JI}(\omega)\mathbf{J}^T_{I\to J}(\mathbf{r}), \quad (53)$$

with the abbreviation

$$\beta_{JI}(\omega) = \int \mathbf{J}^T_{J\to I}(\mathbf{r})\cdot\mathbf{e}_T(\mathbf{r};\omega)d^3r. \quad (54)$$

The physical interpretation of Eq. (53) is very transparent. Hence it is seen that the transverse current density entering the basic integral equation in Eq. (41) consists of a weighted superposition of the transverse current densities which are related to the various electronic $I\to J$ transitions. The *spatial form* of the individual transverse current densities only depends on the wave functions entering the given $I\to J$ transition. The various $\mathbf{J}^T_{I\to J}(\mathbf{r})$'s are independent of the solution of the integral equation problem, and in a sense therefore serve as spatial constraints on the final solution. The strength with which a given $\mathbf{J}^T_{I\to J}(\mathbf{r})$ enters the prevailing $\mathbf{J}_T(\mathbf{r};\omega)$ is given by the frequency-dependent product $\mathcal{A}_{IJ}(\omega)\beta_{JI}(\omega)$. The presence of the factor $\hbar\omega+E_J-E_I$ in the denominator of $\mathcal{A}_{IJ}(\omega)$, shows, as expected that transitions which are near the electronic resonance condition

$$\hbar\omega + E_J - E_I = 0 \quad (55)$$

contribute more to $\mathbf{J}_T(\mathbf{r};\omega)$ than transitions far from resonance. At resonance one at least must include the irreversible damping originating in spontaneous emission. The factor $\mathcal{A}_{IJ}(\omega)$ also accounts for the fact that the individual weight factors must be proportional to the population probability difference P_J-P_I prevailing in the absence of the light excitation. The strength of a given $\mathbf{J}^T_{I\to J}(\mathbf{r})$ contribution to $\mathbf{J}_T(\mathbf{r};\omega)$ also depends on the strength with which the *opposite* electronic transition $J\to I$ is pumped. All transitions are pumped by the prevailing microscopic electric field $\mathbf{e}_T(\mathbf{r};\omega)$, and the pump strength of the $J\to I$ transition essentially is given by the work carried out per unit time by the field distribution $\mathbf{e}_T(\mathbf{r};\omega)$ on the given (transverse) transition current density distribution, $\mathbf{J}^T_{J\to I}(\mathbf{r})$. The afore-mentioned work per unit time is precisely given by the $\beta_{JI}(\omega)$ in Eq. (54). As long as the selfconsistent field $\mathbf{e}_T(\mathbf{r};\omega)$ has not been determined the $\beta_{JI}(\omega)$'s are *unknown numbers*.

All mesoscopic quantum systems interact with their surroundings and irreversible damping mechanisms therefore inevitably are present. If these mechanisms can be described via the inclusion of suitable phenomenological damping terms [as indicated in the text below Eq. (44)] the (many-

body) stationary states become quasistationary with finite lifetimes, τ_{JI}. Technically the real angular frequency (ω) now must be replaced by the complex quantity $\omega+i/\tau_{IJ}$ in the IJ transition. The separation techniques used in Secs. V B and V C to determine the local field is not affected by the inclusion of the irreversible damping times τ_{IJ}, because the tensor product structure of the conductivity tensor is retained. In most cases it will be necessary to include irreversible dampings. The damping parameters need not be so big in a mesoscopic system. Thus it might be sufficient in a two-level metallic quantum well system to include just one damping term for the (sub)interband transition and one for the free-electron-like intraband dynamics.

B. The Huygens propagator formalism revisited: Physical interpretation

Returning to the equivalence between Eqs. (32) and (34) it is obvious that the integral equation in Eq. (41) can be rewritten in the Huygens propagator form

$$\mathbf{e}_T(\mathbf{r};\omega) = \mathbf{e}_T^{ext}(\mathbf{r};\omega) + i\mu_0\omega \int g(|\mathbf{r}-\mathbf{r}'|;\omega)\mathbf{J}_T(\mathbf{r}';\omega)d^3r'. \tag{56}$$

For numerical purposes it is perhaps easier to use Eq. (56) than Eq. (41). In Eq. (41) appears a $|\mathbf{r}-\mathbf{r}'|^{-3}$ singularity in the Green's function $\overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)$ and with the use of this propagator and the self-field term $\mathbf{J}_T(\mathbf{r};\omega)/(3i\epsilon_0\omega)$ principal integration with spherical contraction must be used. With $\mathbf{J}_T(\mathbf{r};\omega)$ as the source current density one may use the transverse propagator $\overleftrightarrow{\mathbf{G}}_T(\mathbf{r}-\mathbf{r}';\omega)$ instead of $\overleftrightarrow{\mathbf{G}}(\mathbf{r}-\mathbf{r}';\omega)$. The transverse propagator exhibits a weaker ($|\mathbf{r}-\mathbf{r}'|^{-1}$) singularity [20], and use of $\overleftrightarrow{\mathbf{G}}_T(\mathbf{r}-\mathbf{r}';\omega)$ makes the integral absolutely convergent.

A combination of Eqs. (53) and (56) results in the following integral equation for $\mathbf{e}_T(\mathbf{r};\omega)$:

$$\mathbf{e}_T(\mathbf{r};\omega) = \mathbf{e}_T^{ext}(\mathbf{r};\omega) + \sum_{I,J} \mathbf{F}_{IJ}(\mathbf{r};\omega)\beta_{JI}(\omega), \tag{57}$$

where

$$\mathbf{F}_{IJ}(\mathbf{r};\omega) = i\mu_0\omega\mathcal{A}_{IJ}(\omega) \int g(|\mathbf{r}-\mathbf{r}'|;\omega)\mathbf{J}_{I\to J}^T(\mathbf{r}')d^3r'. \tag{58}$$

The physical interpretation of Eqs. (57) and (58) is clear. Thus, the local field $\mathbf{e}_T(\mathbf{r};\omega)$ at $\mathbf{r}$ in Eq. (57) consists of the sum of the external field, $\mathbf{e}_T^{ext}(\mathbf{r};\omega)$, and the field, $\mathbf{F}_{IJ}(\mathbf{r};\omega)\beta_{JI}(\omega)$, generated by the various $I\to J$ transitions. The field created by a given $I\to J$ transition at the space point $\mathbf{r}$ originates in the microscopic transverse transition current density $\mathbf{J}_{I\to J}^T(\mathbf{r}')$ multiplied by the factor $i\mu_0\omega\mathcal{A}_{IJ}(\omega)\beta_{JI}(\omega)$. From each point $\mathbf{r}'$ in the $\mathbf{J}_{I\to J}^T(\mathbf{r}')$ distribution the field propagators outwards as described by the Huygens propagator, and the field propagation from $\mathbf{r}'$ to $\mathbf{r}$ is described by $g(|\mathbf{r}-\mathbf{r}'|;\omega)$ in ω space.

C. Last step: Conversion from integral to matrix equation

If the yet unknown field strengths $\beta_{JI}(\omega)$ appearing in Eq. (58) can be determined the transverse local field $\mathbf{e}_T(\mathbf{r};\omega)$ is readily found. To determine the $\beta_{JI}(\omega)$'s, Eq. (58) is inserted into Eq. (54). This leads to the following matrix equation for the unknown field strengths:

$$\beta_{JI}(\omega) - \sum_{O,P} N_{OP}^{JI}(\omega)\beta_{OP}(\omega) = H_{JI}(\omega), \tag{59}$$

where

$$N_{OP}^{JI}(\omega) \equiv i\mu_0\omega\mathcal{A}_{PO}(\omega) \int\int g(|\mathbf{r}-\mathbf{r}'|;\omega) \times \mathbf{J}_{J\to I}^T(\mathbf{r})\cdot\mathbf{J}_{P\to O}^T(\mathbf{r}')d^3r'd^3r, \tag{60}$$

and

$$H_{JI}(\omega) \equiv \int \mathbf{J}_{J\to I}^T(\mathbf{r})\cdot\mathbf{e}_T^{ext}(\mathbf{r};\omega)d^3r. \tag{61}$$

Let us take a closer look at the physics contained in the result of Eqs. (59)–(61). To each pair of electronic energy eigenstates there is associated two unknown matrix elements in Eq. (59). Thus for the arbitrary states O and P, the elements β_{OP} and β_{PO} describe the pumping of the $O\to P$ and $P\to O$ transitions by the local field $\mathbf{e}_T(\mathbf{r};\omega)$, respectively. Near the electronic resonance, given by $\hbar\omega+E_O-E_P=0$, β_{OP} elements usually will be much larger than β_{PO}, and in certain situations it may be justified to neglect β_{PO} (and other counter-resonant terms) completely. This has for instance turned out to be possible in nonlocal studies of the linear optical properties of few-level quantum wells [10]. Essentially, such an approximation is analogous to making the rotating wave approximation (RWA) in semiclassical and quantum optics [23]. The various sets of transitions, say $J\to I$ and $P\to O$, are coupled radiatively together, and this coupling manifests itself through the N_{OP}^{JI} factors given in Eq. (60). In this equation the radiation emitted the transverse transition current density $\mathbf{J}_{P\to O}^T(\mathbf{r}')$ at $\mathbf{r}'$ propagates to the space point $\mathbf{r}$ as described by the Huygens propagator $g(|\mathbf{r}-\mathbf{r}'|;\omega)$, and at $\mathbf{r}$ the radiation carries out a work on the transverse particle current density distribution $\mathbf{J}_{J\to I}^T(\mathbf{r})$ resulting in an electronic $J\to I$ transition. The local-field problem among the various transitions is solved in a selfconsistent manner by solving the matrix-equation problem in Eq. (59) for the unknown $\beta_{JI}(\omega)$'s. The matrix-equation set is inhomogeneous and the inhomogeneous terms are the various $H_{JI}(\omega)$'s. It appears that $H_{JI}(\omega)$ describes the work carried out per unit time on the $J\to I$ transition by the given external field $\mathbf{e}_T^{ext}(\mathbf{r};\omega)$, see Eq. (61). Once the various $\beta_{JI}(\omega)$'s have been determined from Eq. (59) insertion into (57) gives us the local field $\mathbf{e}_T(\mathbf{r};\omega)$. The procedure described above thus in principle has allowed us to obtain a selfconsistent solution to the linear light scattering problem without making use of the doubtful geometrical discretization. The principle used can be employed right down to the atomic domain. In practice (numerically) one can deal with matrix equations up to a certain dimension, which means that one can include only a limited number of (bound) transitions in a given calculation. It is not

least this limitation which makes the present theory of particular value for light scattering from mesoscopic (or microscopic) objects. In a mesoscopic system the number of bound states can be quite small, and the optical local-field calculation therefore carried out with matrices of relatively small dimensions. Near a given resonance perhaps only a few matrix elements are needed in the numerical calculation, but as always one has to worry about the importance of nonlinear effects near resonance.

D. Local-field resonances

In recent years the importance of local-field effects in mesoscopic optics and in near-field optics has been in focus, and many macroscopic studies have been carried out. From a survey of the literature one may get the impression that macroscopic approaches often overestimate the local-field enhancement, sometimes by orders of magnitude. There is hope that the present microscopic light scattering theory will enable us to obtain a more realistic estimate of local-field phenomena in mesoscopic and near-field optics. Neglecting local-field effects the resonances are those of the electronic transitions [given by Eq. (55)]. In the presence of local-field effects the resonance condition for the mesoscopic object under study is given by

$$\det\{\delta_{JI,OP} - N_{OP}^{JI}(\omega)\} = 0, \tag{62}$$

where $\det\{\cdots\}$ means the determinant of $\{\cdots\}$, and $\delta_{JI,OP}$ is the Kronecker delta. Once the various resonance (RES) frequencies (generic name: ω^{res}) have been obtained from Eq. (62) the relative values of the $\beta_{JI}^{res}(\omega^{res})$'s belonging to the given ω^{res} are obtained from the set of equations one gets by letting JI run through all (relevant) level combinations, i.e.,

$$\beta_{JI}^{res}(\omega^{res}) - \sum_{O,P} N_{OP}^{JI}(\omega^{res})\beta_{OP}^{res}(\omega^{res}) = 0. \tag{63}$$

With a knowledge of the $\beta_{JI}^{res}(\omega^{res})$ values (for the given ω^{res}) *the spatial form* of the resonant electric field $\mathbf{e}_T^{res}(\mathbf{r};\omega^{res})$ is given by

$$\mathbf{e}_T^{res}(\mathbf{r};\omega^{res}) = \sum_{I,J} \mathbf{F}_{IJ}(\mathbf{r};\omega^{res})\beta_{JI}^{res}(\omega^{res}). \tag{64}$$

The different resonant fields $\mathbf{e}_T^{res}(\mathbf{r};\omega^{res})$ can only be determined to within an arbitrary (space-independent) constant, cf. the fact that they represent the solutions to Eq. (57) in the absence of the external source field, $\mathbf{e}_T^{ext}(\mathbf{r};\omega)$.

VI. OUTLOOK

The central goal of the present theory was to establish a microscopic formalism for light scattering from mesoscopic media which allows one to go beyond the geometrical discretization scheme usually employed for instance in near-field optics, but generally of doubtful value on a length scale much shorter than the optical wavelength.

Perhaps the theory might enable one to obtain different insight also in classical diffraction theory. Classical diffraction theory is based on the Helmholtz-Kirchhoff integral theorem [24–26,4]. When this theorem is used in connection with studies of the light diffraction from a (small) hole in an opaque screen the main theoretical challenge is the establishment of a self-consistent solution to the boundary value problem for the field at the screen. An important step towards obtaining a rigorous solution to the hole diffraction problem was taken originally by Bethe [27] who investigated the diffraction from a hole of linear dimension much less than the wavelength of the electromagnetic field. The theory of Bethe is insufficient when it comes to an understanding of for instance the role of plasmon (polariton) effects in near-field hole diffraction. Usually modern problems as this are attacked on the basis of macroscopic diffraction (scattering) theory, and therefore the various shortcomings mentioned in this paper are inherent also in hole diffraction analyses. Since in particular the electronic excitations of matter in the vicinity of the hole play a role for the diffraction problem self-consistent quantum mechanical calculations carried out along the lines indicated in this paper may be feasible as I shall argue in a forthcoming paper devoted to the quantum theory of near-field aperture diffraction.

From a fundamental point of view a quantum optical understanding of single-photon transmission through a subwavelength hole can hardly be achieved without invoking quantum mechanical considerations of the field-matter interaction in the vicinity (near-field zone) of the hole. The present theory might be of importance also in this context.

APPENDIX: GREEN'S FUNCTION SINGULARITY AND SELF-FIELD

Due to the fact that the issue of the electromagnetic propagator singularity and the associated self-field plays an important role for the present work I here discuss the issue in some detail. The Appendix also is meant to serve the purpose of making the paper more accesible to the general reader. The final result of the analysis which is well known has been derived in various manners technically, cf. the works by Van Kranendonk and Sipe [1] and Yaghjian [2], and the present author [10]. A good preparation for studies of the aforementioned derivations can be obtained consulting Appendix V in the book by Born and Wolf [4]. Here a useful mathematical dilemma, used by Born and Wolf in a rigorous derivation of the Lorenz-Lorentz formula, is established. Since the essential parts of the analysis is the same for the macroscopic [2], semi-microscopic [1], and microscopic [10] problem, it is sufficient below to refer to microscopic electrodynamics. From a physical point of view the so-called spherical contraction scheme perhaps is the most important one, among other things because it relates directly to the isotropy of the speed of light in vacuum and is a natural scheme for studying light scattering from well-localized (mesoscopic, molecular, atomic) objects. The spherical contraction scheme is in focus below and the analysis given (in brief) deviates slightly from these found in Refs. [1,2,10].

The starting point for our analysis thus is the integral equation for the microscopic displacement field [Eq. (32)]. Although the Huygens scalar propagator, $g(R;\omega)$, exhibits a R^{-1} singularity at $\mathbf{r}' = \mathbf{r}$, the integral in Eq. (32) is absolutely

convergent and no (infinitesimally small) exclusion volume ϵ need to be introduced in Eq. (32). In order to replace the Huygens propagator by the dyadic standard propagator $\overleftrightarrow{\mathbf{G}}(\mathbf{R};\omega)$, we perform the operation $\boldsymbol{\nabla}\times(\boldsymbol{\nabla}\times)$ on both sides of Eq. (32). On the right hand side of the resulting equation we would like to change the order of integration and double differentiation. This cannot be done just like that. This perhaps is obvious because double differentiation under the integral sign [on $g(R;\omega)$] leads to the occurence of a R^{-3} singularity, and in the presence of such a singularity the integral is conditionally convergent only. By cutting out an infinitesimally small spherical volume ($\epsilon\rightarrow 0$) around $\mathbf{r}'=\mathbf{r}$, it is shown, e.g., in Ref. [4] that

$$\boldsymbol{\nabla}\times\left(\boldsymbol{\nabla}\times\int g(R;\omega)\mathbf{S}(\mathbf{r}';\omega)d^3r'\right) = \int_{\epsilon\rightarrow 0}\boldsymbol{\nabla}\times\{\boldsymbol{\nabla}\times[g(R;\omega)\mathbf{S}(\mathbf{r}';\omega)]\}d^3r' + \frac{2}{3}\mathbf{S}(\mathbf{r};\omega). \tag{A1}$$

To interchange the integral and differential operations [appearing on the left-hand side of Eq. (A1)] a high price in a sense has been paid: (i) the new integral is only conditionally convergent, and (ii) an extra term, $2\mathbf{S}/3$, must be added. The extra term is the one which evolves into the so-called (transverse) self-field contribution to the microscopic electric field. If instead of spherical contraction another contraction form had been chosen the self-field term would also have been different. Thus only the sum of the two terms on the right-hand side of Eq. (A1) can be ascribed a physical meaning basically. We have here paid the "high" price in order to put the standard propagator on the scene.

The usefulness of the lemma in Eq. (A1) is also related to the fact that it in certain cases allows one to reduce the geometrical size of what is considered to be the source of the field [10].

Since, first,

$$q_0^{-2}\boldsymbol{\nabla}\times[\boldsymbol{\nabla}\times(g\mathbf{S})] = -\overleftrightarrow{\mathbf{G}}\cdot\mathbf{S}, \tag{A2}$$

as one readily proves using the differential equation for the Huygens scalar propagator, i.e.,

$$(\nabla^2+q_0^2)g=0, \quad \mathbf{r}'\neq\mathbf{r} \tag{A3}$$

and the identity $\boldsymbol{\nabla}\times[\boldsymbol{\nabla}\times(\cdots)]=(\boldsymbol{\nabla}\boldsymbol{\nabla}-\overleftrightarrow{U}\nabla^2)(\cdots)$, and second,

$$\boldsymbol{\nabla}\times(\boldsymbol{\nabla}\times\mathbf{d}) = -\nabla^2\mathbf{d} = q_0^2\mathbf{d}+\frac{i\omega}{c_0^2}\mathbf{S}, \tag{A4}$$

where the first equality follows from the transverse nature of $\mathbf{d}(\mathbf{r};\omega)$ and the second from the inhomogeneous wave equation for $\mathbf{d}(\mathbf{r};\omega)$ [Eq. (30)], the final step in the analysis can be taken. Hence a combination of Eqs. (A2)–(A4), easily allows one to obtain Eq. (34) from Eq. (32), noting that the effective external source current density distribution $\mathbf{S}^{ext}(\mathbf{r};\omega)$ is located outside the domain of space in consideration [$(\nabla^2+q_0^2)\mathbf{d}^{ext}=\mathbf{0}$].

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