REVIEWS OF TOPICAL PROBLEMS

Two electron problem and the nonlocal equations of electrodynamics

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Contents

Abstract. A survey is offered for the current knowledge of nonlocal electrodynamic equations which in some cases (e.g., in solving boundary value problems in optics) can replace Maxwell's equations. The nonlocal equations are derived using the semi-classical or quantum-electrodynamic approaches. The former involves an expansion of retarded potentials in appropriate parameters and a subsequent transition, to terms of order v^2/c^2, to quantum mechanical operators in the Lagrangian of a system of moving charges. The latter approach is to consider second- and third-order quantum electrodynamic effects for two hydrogen-like atoms arbitrarily far apart. Various nonlocal equations are derived for the propagation of photons and electromagnetic waves in spin systems, dielectrics, and metals, taking into account a variety of quantum transitions and intermediate states in the spectrum of the interacting atoms. By combining nonlocal field equations with relevant constitutive equations, a number of typical boundary-value optical problems are solved for semi-infinite media, superthin films, and for objects whose linear dimensions are much smaller than the light wavelength.

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1. Introduction

The Coulomb e^2/r law determines the energy of interaction between two electrons whose separation is less than the characteristic spatial scale of the problem under study and whose velocities v are negligible compared to the speed of light c (i.e., $v/c \to 0$). An example is a system of neutral atoms (van der Waals interaction) separated by a distance smaller than the characteristic wavelengths λ_0 in the spectrum of the interacting atoms ($r \ll \lambda_0$). Depending on the multipolarity of the quantum transitions between the atomic states, the

operator of the interatomic interaction is in this case proportional to $1/r^3$ (dipole-dipole interaction), or to $1/r^4$ (dipole-quadrupole interaction), etc. The energy of interaction between two neutral atoms in their *S* states is known to be proportional to $1/r^6$ [1]. If, however, one of the atoms is in the ground state and the other in an excited state, the potential energy of interaction is proportional to $1/r^3$ [1], whereas the time for the energy transfer between the atoms is finite [2] and far exceeds the time t_p a photon takes to travel the distance r ($t_p = r/c$).

For separations $r > \lambda_0$, the Coulomb law is insufficient to describe the interaction of atoms and, together with the Coulomb interaction, one must consider the retarded interaction, which depends on the speed of light and vanishes for $c \rightarrow \infty$.

The effect of the retarded interaction on a system of moving electrical charges was analyzed within the framework of classical electrodynamics by Born and Ewald (see [3]). In Refs [4, 5], the Lagrangian of a system of moving interacting charges is obtained to an accuracy v^2/c^2. The incorporation of retardation effects into the system of interacting atoms made it necessary to somewhat revise the existing theoretical methods. The Lagrangian and Hamiltonian for a system of interacting atoms were derived in Refs [6 – 8] to an accuracy v^2/c^2.

In the studies of Refs [9] and [10], methods of quantum electrodynamics were used to analyze the interaction between free electrons taking into account the retardation effect. In the language of quantum electrodynamics, the retarded interaction is due to the exchange of virtual transverse photons between the charges, whereas the Coulomb interaction results from the exchange of longitudinal and scalar photons [11]. The case of two interacting electrons in the helium atom was examined by Breit [11, 12]. The Breit operator has the form

$$B = \frac{e^2}{r} - \frac{e^2}{2r}\left[\boldsymbol{\alpha}_1\boldsymbol{\alpha}_2 + \frac{(\boldsymbol{\alpha}_1\mathbf{r})(\boldsymbol{\alpha}_2\mathbf{r})}{r^2}\right], \tag{1.1}$$

where $\boldsymbol{\alpha}_1$ and $\boldsymbol{\alpha}_2$ are the Dirac matrices for the electrons [11]. The first term in Eqn (1.1) is the electron-electron Coulomb interaction, whereas the second introduces corrections for the motion of the electrons and for the electron spins. The application of the Breit operator is limited to not-too-large electron separations, and this is the reason why the problem of two electrons belonging to two atoms an arbitrary distance apart had to be revisited in the early 1970s, when multi-atomic systems subject to radiation fields came under close scrutiny. A corresponding quantum-mechanical treatment for hydrogen-like atoms was first given in Ref. [13] and later in Ref. [7]. In the former, the operator of the dipole-dipole interaction for the atomic electrons is derived in the electric dipole approximation taking into account only the electrons' orbital degrees of freedom. The more general analysis of Ref. [13] involves second-order quantum electrodynamics effects, including the virtual exchange of photons of all polarizations, and leads to the operator

$$U^{(2)} = e^2 \exp\left(\frac{\mathrm{i}\omega_0 a}{c}\right)\left\{\frac{1}{r} + \frac{a(\boldsymbol{\alpha}_2\mathbf{n})}{r^2} - \frac{(\boldsymbol{\alpha}_1\boldsymbol{\alpha}_2) + (\boldsymbol{\alpha}_1\mathbf{n})(\boldsymbol{\alpha}_2\mathbf{n})}{2r}\right.$$
$$\left. - \frac{a^2}{2}\frac{(\boldsymbol{\alpha}_1\boldsymbol{\alpha}_2) - 3(\boldsymbol{\alpha}_1\mathbf{n})(\boldsymbol{\alpha}_2\mathbf{n})}{r^3}\right\}, \tag{1.2}$$

where a is the interatomic separation, $\mathbf{n} = \mathbf{r}/r$, and ω_0 is an eigenfrequency in the spectrum of the interaction atoms. The first term in Eqn (1.2) is the Coulomb interaction between the two atomic electrons, and the remaining terms correct for the motion of the bound electrons separated by a distance large compared to $\lambda_0 = 2\pi c/\omega_0$. For $a \rightarrow 0$, the operators (1.1) and (1.2) are identical. Below we will give a detailed derivation of the operator (1.2) and discuss its physical meaning.

The next step in solving the two-electron problem was to take the field of real photons into account. The specific examples considered were free electrons [14, 15] and two electrons in a helium-like atom with an assumed nuclear field [16]. In Refs [6, 7], first a semiclassical and then a quantum mechanical approach were applied to the interaction between two electrons belonging to their respective hydrogen-like atoms arbitrarily far apart; the analysis involved third-order quantum electrodynamical effects, described in part by the operator (1.2). The importance of this work is that it provided formulas which described polarizing fields in a system of two hydrogen-like atoms and which made it possible to derive nonlocal equations for the propagation of photons in various media for various types of quantum transitions and intermediate states in the spectrum of the interacting atoms. In the present review, various nonlocal equations will be presented and their physical meanings will be discussed in detail. Because of the complexity of Maxwell's equations, and because of their locality property, these equations do not always lead to a proper solution. In this review we consider certain types of problems amenable to a treatment using nonlocal equations. It can be said that nonlocal equations have made a breakthrough in the solution of a number of major problems in classical, nonlinear, and quantum optics, such as (1) the derivation of formulas for the refractive index inside and at the surface of a medium, (2) the derivation of formulas for optical field amplitudes for various observation points inside and outside optical media, (3) the choice of appropriate boundary conditions, (4) the construction of the theory of the transition layer on the surface of an optical medium, and (5) the solution of a number of problems in the optics of small objects less than the light wavelength across.

The plan of this review is as follows. We start by considering the third-order quantum electrodynamic effects needed for describing interactions between two hydrogen-like atoms arbitrarily far apart. Based on the solution obtained, we present a method for deriving various nonlocal equations for various degrees of freedom of atomic electrons. We then re-derive the nonlocal equations semiclassically and also derive various nonlocal equations of the electrodynamics of alternating classical and quantum fields. We conclude by presenting solutions to certain optical problems to demonstrate the advantages of the nonlocal equations over Maxwell's equations.

2. Effective interaction energy matrix for two hydrogen-like atoms arbitrarily far apart

Feynman diagrams for the interaction of two electrons in an external field are shown in Fig. 1.

Integrating over the time and over the frequencies and wave vectors of virtual photons in the *S* matrix yields the following effective interaction energy matrix for two atomic electrons ($\hbar = c = 1$):
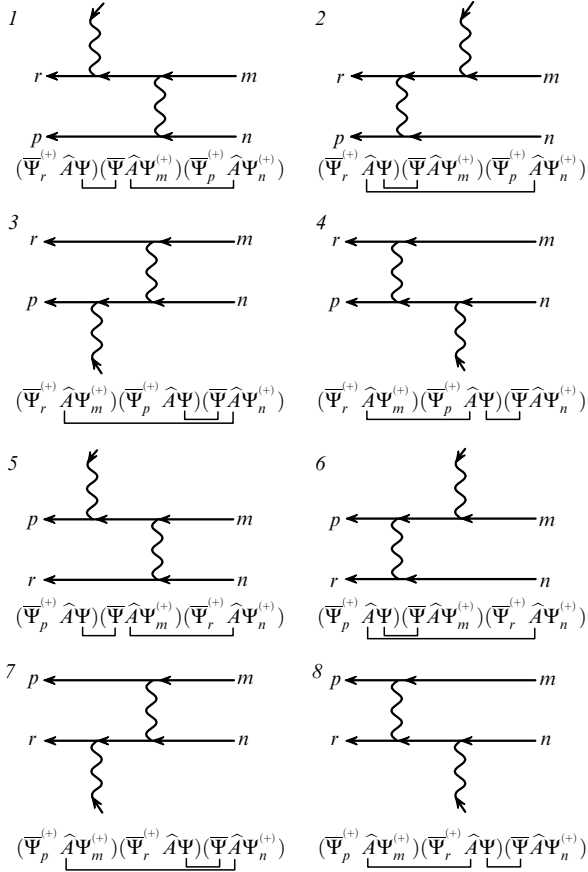
Figure 1. Feynman diagrams for the $e^- e^-$ interaction of two atoms with the emission or absorption of a photon.

$$U_{i \to f}^{(3)} = e^3 \int d\mathbf{r}' \, d\mathbf{r}'' \, d\mathbf{r}''' \left\{ -\frac{1}{|\mathbf{r}'' - \mathbf{r}'''|} \right.$$

$$\times \sum_{l_\pm} \frac{\exp\{i|\omega_n^{(+)} - \omega_p^{(+)}||\mathbf{r}'' - \mathbf{r}'''|\}}{\omega_l(1 - i0) + \omega - \omega_r^{(+)}} \overline{\Psi}_r^{(+)}(\mathbf{r}') \widehat{A}(\mathbf{r}') \Psi_l(\mathbf{r}')$$

$$\times \overline{\Psi}_l(\mathbf{r}'') \gamma_\mu'' \Psi_m^{(+)}(\mathbf{r}'') \overline{\Psi}_p^{(+)}(\mathbf{r}''') \gamma_\mu''' \Psi_n^{(+)}(\mathbf{r}''') - \frac{1}{|\mathbf{r}' - \mathbf{r}'''|}$$

$$\times \sum_{l_\pm} \frac{\exp\{i|\omega_n^{(+)} - \omega_p^{(+)}||\mathbf{r}' - \mathbf{r}'''|\}}{\omega_l(1 - i0) - \omega - \omega_m^{(+)}}$$

$$\times \overline{\Psi}_r^{(+)}(\mathbf{r}') \gamma_\mu' \Psi_l(\mathbf{r}') \overline{\Psi}_l(\mathbf{r}'') \, \widehat{A}(\mathbf{r}'') \Psi_m^{(+)}(\mathbf{r}'') \overline{\Psi}_p^{(+)}(\mathbf{r}''') \gamma_\mu''' \Psi_n^{(+)}(\mathbf{r}''')$$

$$- \frac{1}{|\mathbf{r}' - \mathbf{r}'''|} \sum_{l_\pm} \frac{\exp\{i|\omega_r^{(+)} - \omega_m^{(+)}||\mathbf{r}' - \mathbf{r}'''|\}}{\omega_l(1 - i0) + \omega - \omega_p^{(+)}}$$

$$\times \overline{\Psi}_r^{(+)}(\mathbf{r}') \gamma_\mu' \Psi_m^{(+)}(\mathbf{r}') \overline{\Psi}_p^{(+)}(\mathbf{r}'') \, \widehat{A}(\mathbf{r}'') \Psi_l(\mathbf{r}'') \overline{\Psi}_l(\mathbf{r}''') \gamma_\mu''' \Psi_n^{(+)}(\mathbf{r}''')$$

$$- \frac{1}{|\mathbf{r}' - \mathbf{r}'''|} \sum_{l_\pm} \frac{\exp\{i|\omega_r^{(+)} - \omega_m^{(+)}||\mathbf{r}' - \mathbf{r}''|\}}{\omega_l(1 - i0) - \omega - \omega_n^{(+)}} \, \overline{\Psi}_r^{(+)}(\mathbf{r}') \gamma_\mu'$$

$$\times \Psi_m^{(+)}(\mathbf{r}') \overline{\Psi}_p(\mathbf{r}'') \gamma_\mu'' \Psi_l(\mathbf{r}'') \overline{\Psi}_l(\mathbf{r}''') \, \widehat{A}(\mathbf{r}''') \Psi_n^{(+)}(\mathbf{r}''')$$

$$+ \frac{1}{|\mathbf{r}'' - \mathbf{r}'''|} \sum_{l_\pm} \frac{\exp\{i|\omega_r^{(+)} - \omega_n^{(+)}||\mathbf{r}'' - \mathbf{r}'''|\}}{\omega_l(1 + i0) + \omega - \omega_p^{(+)}} \, \overline{\Psi}_p^{(+)}(\mathbf{r}') \, \widehat{A}(\mathbf{r}')$$

$$\times \Psi_l(\mathbf{r}') \overline{\Psi}_l(\mathbf{r}'') \gamma_\mu'' \Psi_m^{(+)}(\mathbf{r}'') \overline{\Psi}_r^{(+)}(\mathbf{r}''') \gamma_\mu''' \Psi_n^{(+)}(\mathbf{r}''')$$

$$+ \frac{1}{|\mathbf{r}' - \mathbf{r}'''|} \sum_{l_\pm} \frac{\exp\{i|\omega_n^{(+)} - \omega_r^{(+)}||\mathbf{r}' - \mathbf{r}'''|\}}{\omega_l(1 - i0) - \omega - \omega_m^{(+)}} \, \overline{\Psi}_p^{(+)}(\mathbf{r}') \gamma_\mu' \Psi_l(\mathbf{r}')$$

$$\times \overline{\Psi}_l(\mathbf{r}'') \widehat{A}(\mathbf{r}'') \Psi_m^{(+)}(\mathbf{r}'') \overline{\Psi}_r^{(+)}(\mathbf{r}''') \gamma_\mu''' \Psi_n^{(+)}(\mathbf{r}''')$$

$$+ \frac{1}{|\mathbf{r}' - \mathbf{r}'''|} \sum_{l_\pm} \frac{\exp\{i|\omega_p^{(+)} - \omega_m^{(+)}||\mathbf{r}' - \mathbf{r}'''|\}}{\omega_l(1 - i0) + \omega - \omega_r^{(+)}} \, \overline{\Psi}_p^{(+)}(\mathbf{r}') \gamma_\mu'$$

where $\omega_n^{(+)}$, $\omega_m^{(+)}$ ($\omega_p^{(+)}$, $\omega_r^{(+)}$) are the frequencies of the initial (final) electron states, $\Psi_{m(n)}^{(+)}$ are the solutions of the Dirac equation for a positive-frequency electron, $\overline{\Psi}_{p(r)}^{(+)} = \Psi_{p(r)}^{(+)*} \gamma_4$, $\Psi_{p(r)}^{(+)*}$ is the conjugate wave function, $\gamma_4 = \beta$, $\gamma_j = -i\beta\alpha_j$ ($j = 1, 2, 3$), and

$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix}, \quad \beta = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix},$$

where $\boldsymbol{\sigma}$ is the Pauli matrix. Primes on the radius vector $\mathbf{r}$ and the γ matrices indicate various wave functions of the interacting particles (note that differently primed γ matrices commute with each other). The summation in Eqn (2.1) runs over the complete set of positive- and negative-frequency intermediate states. We will regard negative-frequency electron states as positron states without explicitly changing to a positron wave function involving a charge conjugate transformation, the reason being that a superposition of states with opposite charges cannot produce the general solution of the Dirac equation [11]. The solution we will employ in this paper is

$$\Psi = \sum_r a_r \psi_r^{(+)} + \sum_r b_r^+ \psi_r^{(-)},$$

$$\overline{\Psi} = \sum_r a_r^+ \overline{\psi}_r^{(+)} + \sum_r b_r \overline{\Psi}_r^{(-)}, \tag{2.1a}$$

where a_r, a_r^+, b_r, b_r^+ are the second quantized operators of the electron-positron field. A transition from $S_{i \to f}^{(3)}$ to the effective interaction energy (2.1) was performed using the equation

$$S_{i \to f}^{(3)} = -2\pi i U_{i \to f}^{(3)} \, \delta\big(\omega_r^{(+)} - \omega_m^{(+)} - \omega + \omega_p^{(+)} - \omega_n^{(+)}\big), \tag{2.2}$$

where the sign of the optical photon frequency ω indicates that in the vector potential operator $\widehat{\mathbf{A}} = \sum_\mu \gamma_\mu A_\mu$ we have separated out the negative-frequency part proportional to the destruction operator for photon of a given mode.

Consider the first term in Eqn (2.1) and separate out in it the factor accounting for the electron-electron exchange of virtual photons. First, we write the distance between the electrons as

$$|\mathbf{r}'' - \mathbf{r}'''| \approx a\left(1 + \frac{\mathbf{a}\Delta\xi}{a^2} + \frac{M_1}{a}\right), \tag{2.3}$$

where $a = |\mathbf{a}'' - \mathbf{a}'''|$ is the atomic separation, $\Delta\xi = \xi'' - \xi'''$, with ξ'', ξ''' denoting the distances of the electrons from their respective nuclei, and $M_1 = M_1(\mathbf{a}, \Delta\xi)$ includes higher-order correction terms in $\Delta\xi/a$. We change to a system of units in which $c \neq 1$ and assume that

$$\frac{1}{c}\big|\omega_n^{(+)} - \omega_p^{(+)}\big| \, \frac{\Delta\xi}{a} \ll 1. \tag{2.4}$$

Assuming that the internuclear separations lie in the wide range $|\Delta\xi| \leqslant a \leqslant \infty$ we then obtain

$$\frac{1}{|\mathbf{r}'' - \mathbf{r}'''|} \exp\left\{\frac{\mathrm{i}}{c}|\omega_n^{(+)} - \omega_p^{(+)}||\mathbf{r}'' - \mathbf{r}'''|\right\}$$

$$= \frac{1}{a}\exp\left\{\frac{\mathrm{i}}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}\frac{1}{1 + (\mathbf{a}\Delta\xi)/a^2 + M_1/a}$$

$$\times \left\{1 + \frac{\mathrm{i}}{c}|\omega_n^{(+)} - \omega_p^{(+)}|\left(\frac{\mathbf{a}\Delta\xi}{a} + M_1\right)\right.$$

$$\left. - \frac{1}{2c^2}(\omega_n^{(+)} - \omega_p^{(+)})^2\left(\frac{\mathbf{a}\Delta\xi}{a} + M_1\right)^2\right\}. \tag{2.3a}$$

If we now eliminate the frequencies from Eqn (2.3a) by using the equations $H'''\psi_n^{(+)}(\mathbf{r}''') = \omega_n^{(+)}\psi_n^{(+)}(\mathbf{r}''')$, we see that the following transformation takes place:

$$\frac{1}{|\mathbf{r}'' - \mathbf{r}'''|}\exp\left\{\frac{\mathrm{i}}{c}|\omega_n^{(+)} - \omega_p^{(+)}||\mathbf{r}'' - \mathbf{r}'''|\right\}$$

$$\Rightarrow \frac{1}{a}\exp\left\{\frac{\mathrm{i}}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}\left\{\left[1 + \frac{\mathbf{a}\Delta\xi}{a^2} + \frac{M_1}{a}\right]^{-1}\right.$$

$$\left. + \frac{\mathrm{i}}{c}[f_1, H'''] + \frac{1}{2c^2}[H'', [H''', f_2]]\frac{\omega_n^{(+)} - \omega_p^{(+)}}{\omega_l - \omega_m^{(+)}}\right\}, \quad (2.5)$$

where

$$f_1 = a - \frac{a^2}{|\mathbf{r}'' - \mathbf{r}'''|}, \quad f_2 = a|\mathbf{r}'' - \mathbf{r}'''| + \frac{a^3}{|\mathbf{r}'' - \mathbf{r}'''|} - 2a^2. \tag{2.6}$$

As can be seen from Eqn (2.3a), our analysis, along with the expansion in powers of $1/c$, also involves an expansion in the small parameter $\Delta\xi/a$. Whereas in the former expansion we retain terms up to quadratic, the latter is carried out to an arbitrarily high order because the function M_1 contains all the necessary higher correction terms — this is easily seen by substituting the expansion (2.3) into Eqn (2.6) and comparing Eqns (2.5) and (2.3a). Thus, the interaction of two atomic electrons of arbitrary multipolarities will be accounted for throughout the entire discussion.

The Hamiltonians for the individual atoms with their nuclei at rest are

$$H'' = c\boldsymbol{\alpha}''\mathbf{p}'' + \gamma_4''mc^2 - \frac{Z_1e^2}{\xi''},$$

$$H''' = c\boldsymbol{\alpha}'''\mathbf{p}''' + \gamma_4'''mc^2 - \frac{Z_2e^2}{\xi'''}, \tag{2.7}$$

where $\mathbf{p}''$, $\mathbf{p}'''$ are the momentum operators of the electron and Z_1, Z_2 are the charges of the point-like nuclei. Next we obtain the commutation relations for Eqn (2.5) assuming the atomic nuclei to be at rest. For $\hbar \neq 1$ we have

$$[f_1, H'''] = -\mathrm{i}\hbar ca^2\frac{(\boldsymbol{\alpha}''\mathbf{n})}{|\mathbf{r}'' - \mathbf{r}'''|^2},$$

$$[H'', [H''', f_2]] = ac^2[\boldsymbol{\alpha}''\mathbf{p}'', [\boldsymbol{\alpha}'''\mathbf{p}''', |\mathbf{r}'' - \mathbf{r}'''|]]$$

$$+ a^3c^2\left[\boldsymbol{\alpha}''\mathbf{p}'', \left[\boldsymbol{\alpha}'''\mathbf{p}''', \frac{1}{|\mathbf{r}'' - \mathbf{r}'''|}\right]\right]. \tag{2.8}$$

Thus, the operator accounting for the exchange of virtual photons in the matrix (2.1) takes the form

$$B_{1l}(\mathbf{r}'', \mathbf{r}''') = e^2 \exp\left\{\frac{\mathrm{i}}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}\left\{\frac{1 - (\boldsymbol{\alpha}''\boldsymbol{\alpha}''')}{|\mathbf{r}'' - \mathbf{r}'''|}\right.$$

$$+ a\frac{(\boldsymbol{\alpha}''\mathbf{n})}{|\mathbf{r}'' - \mathbf{r}'''|^2} + \frac{1}{2}R_{1l}\left(\frac{(\boldsymbol{\alpha}''\boldsymbol{\alpha}''') - (\boldsymbol{\alpha}''\mathbf{n})(\boldsymbol{\alpha}'''\mathbf{n})}{|\mathbf{r}'' - \mathbf{r}'''|}\right.$$

$$\left.\left. - a^2\frac{(\boldsymbol{\alpha}''\boldsymbol{\alpha}''') - 3(\boldsymbol{\alpha}''\mathbf{n})(\boldsymbol{\alpha}'''\mathbf{n})}{|\mathbf{r}'' - \mathbf{r}'''|^3}\right)\right\}, \tag{2.9}$$

where

$$R_{1l} = \frac{\omega_n^{(+)} - \omega_p^{(+)}}{\omega_l - \omega_m^{(+)}}, \qquad \mathbf{n} = \frac{\mathbf{r}'' - \mathbf{r}'''}{|\mathbf{r}'' - \mathbf{r}'''|}.$$

In the special case of resonant photon exchange $R_{1l} = 1$, and the operator (2.8) goes over into the corresponding operator of Ref. [4]. For $a \to 0$ and $R_{1l} = 1$, Eqn (2.9) is identical to the Breit operator [11]. To understand the physical meaning of the expansion (2.5), note that in obtaining a Lagrangian accurate to $(v/c)^2$ for a system of charges with a continuous energy spectrum [4] one employs a unified time scale R/c, with R being the charge separation. This is the interaction transfer time between the charges, and the retarded potentials can be expanded in powers of R/c only if the charge distribution does not change appreciably in the time R/c. This is of course a very stringent condition if extended systems are considered. In the derivation of the Breit operator for a system of two electrons with a discrete energy spectrum (see [11]), the expansion parameter was taken to be the quantity $\omega_0r/c \ll 1$, where ω_0 is the characteristic frequency in the spectrum of the interacting electrons, and r is the distance between the electrons. Thus, along with the interaction transfer time $T_e = r/c$, the characteristic time $T_a = 2\pi/\omega_0$ was introduced. In this case, $2\pi T_e \ll T_a$, i.e., the electron density in the system of two interacting, moving electrons changes substantially during the transfer of the interaction. It is clear that this condition is satisfied when electron-electron separations are not too large — in helium-like atoms, for example. Because the expansion parameter is taken to be the quantity (2.4), the expansion (2.5) is valid for two atomic electrons arbitrarily far apart. The interaction transfer time $T_e = a/c$ in this case is much longer than the characteristic intra-atomic time scale T_a. This leads to an additional retardation in the interaction of the electrons, which is precisely the effect described by the additional terms in the operator (2.9).

It is known that the characteristic frequencies of atomic electrons are in the range from a few megahertz to 10^9 megahertz if the optical transitions and the fine and superfine splits of the atomic levels are taken into account. A question arises in this connection as to how to choose a characteristic intra-atomic time scale T_a. For the retardation effects to be fully incorporated into the electron interaction, it is clear that the highest — i.e., optical — frequencies must be taken as characteristic frequencies in the spectrum of the interacting electrons. As is shown below, the operator (2.9) accounts for various types of interaction between two electrons, and the presence of a unified time scale T_a explains the fact that, for example, the spin-spin interaction of the electrons has additional retardation terms beyond those in Ref. [11] — even though spin transitions are radio-frequency ones.

We turn now to those matrix elements in Eqn (2.1) which correspond to the remaining Feynman diagrams of

Fig. 1. The repetition of the procedure already used to pass from frequencies to operators yields operators B_{sl} ($s = 2, 3, \ldots, 8$), analogous to the operator B_{1l}. These contain coefficients that determine the differences between the initial, intermediate, and final frequencies, and also involve various retardation factors according to the location of the wave functions in Eqn (2.1). Noting that the third-order effects in Fig. 1 obey the conservation law (2.2) and that for the emission of a real photon the sign of the frequency ω in Eqn (2.2) should be reversed, we obtain the following equation for the energies of the interacting electrons:

$$E_r^{(+)} - E_m^{(+)} + E_p^{(+)} - E_n^{(+)} \pm \hbar\omega = 0. \qquad (2.10)$$

Based on this conservation law, the following quantum transition schemes can be recognized (we separate two states, e. g., p and n, in the spectrum of the interacting atoms and assume that the initial atomic states n and m are the same):

A. As a result of the exchange of virtual photons of frequency $\omega'' = \omega_n^{(+)} - \omega_p^{(+)}$, one — say, the first — atom makes a transition to an intermediate state ω_l and then returns to the initial state $E_m^{(+)}$, i.e., $E_r^{(+)} = E_m^{(+)}$. The second atom changes its quantum state, $E_n^{(+)} \rightarrow E_p^{(+)}$, with the consequence that one photon is emitted or absorbed by the two-atom system. The retardation factor for this particular interaction type is $\exp\{(i/c)|\omega_n^{(+)} - \omega_p^{(+)}|a\}$. The emission (absorption) of a real photon may not spatially coincide with the location of the atom undergoing the quantum transition $E_n^{(+)} \rightarrow E_p^{(+)}$. This scheme corresponds to diagrams *1, 2* and *7, 8* in Fig. 1.

B. Diagrams *1–8* represent transitions involving the emission or absorption of a double energy photon. Both atoms change their states, i.e., $E_r^{(+)} \neq E_m^{(+)}$ and $E_p^{(+)} \neq E_n^{(+)}$ in this case.

C. For $E_r^{(+)} = E_m^{(+)}$, when only one of the atoms changes its state by emitting or absorbing a real photon with energy $\hbar\omega = |E_p^{(+)} - E_n^{(+)}|$, an interaction with a unity retardation factor is possible. This is allowed by diagrams *3–6*.

The nature of the quantum transitions in schemes *A*, *B*, and *C* is determined by the properties of the operators B_{sl} and $\widehat{A}$, by the properties of the wave functions, and also by the energy level schemes of the atomic electrons.

3. Transformation into two-component wave functions

We can pass to two-component wave functions by using the following transformations [11]:

$$\Psi_n^{(+)} = \begin{pmatrix} \varphi_n^{(+)} \\ \dfrac{\boldsymbol{\sigma}\mathbf{p}}{2mc}\varphi_n^{(+)} \end{pmatrix}, \quad \varphi_n^{(+)} = \left(1 - \frac{p^2}{8m^2c^2}\right)\Phi_n^{(+)}. \quad (3.1)$$

Let us apply these transformations to calculating the matrix element

$$\left\langle \Psi_l^*(\mathbf{r}'')\Psi_p^{(+)*}(\mathbf{r}''')|B_{1l}|\Psi_m^{(+)}(\mathbf{r}'')\Psi_n^{(+)}(\mathbf{r}''')\right\rangle \qquad (3.2)$$

in the first term of the matrix (2.1). We first consider the matrix element of the operator $1/|\mathbf{r}'' - \mathbf{r}'''|$ in Eqn (2.9). Taking the positive-energy states in Eqn (3.2) as intermediate

states and using Eqn (3.1) we obtain

$$
\begin{aligned}
e^2 \exp&\left\{\frac{i}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\} \\
&\times \int \left\{ \Phi_l^{(+)*}(\mathbf{r}'')\Phi_p^{(+)*}(\mathbf{r}''')\Phi_m^{(+)}(\mathbf{r}'')\Phi_n^{(+)}(\mathbf{r}''') \right. \\
&- \frac{1}{8m^2c^2}\Phi_l^{(+)*}(\mathbf{r}'')\Phi_p^{(+)*}(\mathbf{r}''')\Phi_m^{(+)}(\mathbf{r}'')\left[\mathbf{p}''^2\Phi_n^{(+)}(\mathbf{r}''')\right] \\
&- \frac{1}{8m^2c^2}\Phi_l^{(+)*}(\mathbf{r}'')\Phi_p^{(+)*}(\mathbf{r}''')\Phi_n^{(+)}(\mathbf{r}''')\left[\mathbf{p}''^2\Phi_m^{(+)}(\mathbf{r}'')\right] \\
&+ \frac{1}{8m^2c^2}\Phi_l^{(+)*}(\mathbf{r}'')\left[\mathbf{p}'''^2\Phi_p^{(+)}(\mathbf{r}''')\right]^*\Phi_m^{(+)}(\mathbf{r}'')\Phi_n^{(+)}(\mathbf{r}''') \\
&+ \left. \frac{1}{8m^2c^2}\left[\mathbf{p}''^2\Phi_n^{(+)}(\mathbf{r}'')\right]^*\Phi_p^{(+)*}(\mathbf{r}''')\Phi_m^{(+)}(\mathbf{r}'')\Phi_n^{(+)}(\mathbf{r}''') \right\} \\
&\times \frac{d\mathbf{r}''\,d\mathbf{r}'''}{|\mathbf{r}'' - \mathbf{r}'''|},
\end{aligned}
\qquad (3.3)
$$

ignoring the third- and the higher-order terms in $1/c$. We now have to transform Eqn (3.3) into

$$\int \Phi_l^{(+)*}(\mathbf{r}'')\Phi_p^{(+)*}(\mathbf{r}''')V_1^{(+)}\Phi_m^{(+)}(\mathbf{r}'')\Phi_n^{(+)}(\mathbf{r}''') \equiv (V_1^{(+)})_{lp,mn}, \qquad (3.4)$$

and, in doing so, to find the explicit form of the operator $V_1^{(+)}$. To transform Eqn (3.3) into Eqn (3.4), it is necessary to integrate by parts. For this, we choose the origin of the coordinate system to be the point a'' and refer the coordinates $\mathbf{r}'$ and $\mathbf{r}''$ to the first and $\mathbf{r}'''$ to the second atom. We take into account, further, that the atomic wave functions do not overlap and that they vanish at infinity. This means that, unlike the case of Ref. [11], the powers of $1/|\mathbf{r}'' - \mathbf{r}'''|$ do not go to infinity for $\mathbf{r}''$ and $\mathbf{r}'''$ within the spatial limits of the interacting atoms. With these considerations in mind we obtain the following operator:

$$V_1^{(+)} = e^2 \exp\left\{\frac{i}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}\frac{1}{r}, \quad r = |\mathbf{r}'' - \mathbf{r}'''|. \quad (3.5)$$

This operator is an analogue of — and in the limit $a \rightarrow 0$ is identical to — the usual Coulomb interaction of two electrons arbitrarily far apart. We consider next the matrix elements of the remaining terms of the operator B_{1l} using the two-component wave functions $\Phi_n^{(+)}$. Substituting wave functions (3.1) into the matrix (2.1) and taking the matrix product in the integrand we find that it suffices to replace φ by Φ in all the resulting terms because they already contain the factor $1/c^2$. Repeating now the transformation already used in deriving the operator (3.5) and separating out the operators containing various powers of $1/r$ we find that the operator proportional $1/r^3$ has the form [17]

$$
\begin{aligned}
V_{2l}^{(+)} = \exp&\left\{\frac{i}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}\frac{e^2\hbar^2}{4m^2c^2}\left\{\frac{1}{r^3}[(\boldsymbol{\sigma}''\boldsymbol{\sigma}''') \right. \\
&- 3(\boldsymbol{\sigma}''\mathbf{n})(\boldsymbol{\sigma}'''\mathbf{n})] + R_{1l}\frac{a^2}{r^5}\left[15(\boldsymbol{\sigma}''\mathbf{n})(\boldsymbol{\sigma}'''\mathbf{n}) - 9(\boldsymbol{\sigma}''\boldsymbol{\sigma}''')\right]\right\}.
\end{aligned}
$$
$$(3.6)$$

Operator (3.6) goes over to the spin-spin interaction operator for two electrons as $a \rightarrow 0$ [11]. For electrons arbitrarily far apart, in accord with the condition (2.4), there arises an

additional retardation determined by the retardation factor $\exp\left\{(i/c)|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}$ and also by the additional terms in Eqn (3.6). In the special case of two electrons resonantly interacting without either emitting or absorbing a photon, we have $R_{1l} = 1$. After some manipulation, the operator proportional to $1/r$ takes the form [17]

$$V_{3l}^{(+)} = \exp\left\{\frac{i}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}\frac{e^2}{m^2c^2}\left\{\left(\frac{1}{2}R_{1l} - 1\right)\right.$$

$$\times \frac{1}{r}(\mathbf{p''}\mathbf{p'''}) - \frac{1}{2}R_{1l}\frac{1}{r}\mathbf{n}(\mathbf{np''})\mathbf{p'''}$$

$$\left. - \frac{1}{2}R_{1l}\frac{a^2}{r^3}\left[(\mathbf{p''}\mathbf{p'''}) - 3\mathbf{n}(\mathbf{np''})\mathbf{p'''}\right]\right\}$$

$$+ \exp\left\{\frac{i}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}\frac{e^2a}{mc}\frac{1}{r^2}(\mathbf{np''}). \tag{3.7}$$

Since in the limit $a \to 0$ and $R_{1l} = 1$ this goes over to the corresponding retarded interaction operator for two electrons in a helium-like atom [11], we will refer to operator (3.7) as to the *retarded interaction operator* for two electrons arbitrarily far apart. For $R_{1l} = 1$ (resonant interaction of two electrons), operator (3.7) can be obtained from the classical Hamiltonian function for a system of atoms by replacing the electron momenta with the corresponding momentum operators [6]. In the matrix elements of the operator B_{1l}, the terms proportional to $1/r^2$ have the form [17]

$$V_{4l}^{(+)} = \exp\left\{\frac{i}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}\frac{e^2\hbar}{4m^2c^2}\left\{\left(\frac{1}{2}R_{1l} - 1\right)\right.$$

$$\times \frac{1}{r^2}\left(2\boldsymbol{\sigma''}[\mathbf{np'''}] - 2\boldsymbol{\sigma'''}[\mathbf{np''}]\right) + \frac{1}{2}R_{1l}\frac{1}{r^2}$$

$$\times \left(2\boldsymbol{\sigma''}[\mathbf{np'}] + 2\boldsymbol{\sigma'''}[\mathbf{np'''}] - 2\boldsymbol{\sigma''}[\mathbf{np'}] - 2\boldsymbol{\sigma'''}[\mathbf{np'''}]\right)$$

$$\left. + 3R_{1l}\frac{a^2}{r^4}\boldsymbol{\sigma''}[\mathbf{np''}]\right\}. \tag{3.8}$$

In the limit $a \to 0$ and $R_{1l} = 1$ this goes over to the corresponding spin-orbit interaction operator for two electrons in the Breit operator [11], and we therefore refer to operator (3.8) as to the *spin-orbit retarded interaction operator* for two atomic electrons arbitrarily far apart.

We next consider the interaction of two electrons by means of the field of virtual photons, taking into account only the orbital degrees of freedom. The operator for this portion of the interaction is $V_{1l}^{(+)} + V_{3l}^{(+)}$. As mentioned earlier, the derivation of the interaction operator of two atoms takes into account transitions of arbitrary multipolarity in atomic spectra. Retaining only terms linear in ξ'' and ξ''' in the expansion of the functions $1/r$ and $1/r^2$ we obtain the operator [17]

$$V_{1l}^{(+)} + V_{3l}^{(+)} = \exp\left\{\frac{i}{c}|\omega_n^{(+)} - \omega_p^{(+)}|a\right\}$$

$$\times \left\{\frac{(\mathbf{d''}\mathbf{d'''}) - 3(\mathbf{nd''})(\mathbf{nd'''})}{a^3} + \frac{e}{mc}\frac{(\mathbf{d'''}\mathbf{p''}) - 3(\mathbf{nd'''})(\mathbf{np''})}{a^2}\right.$$

$$+ \frac{e^2}{m^2c^2}\left[\left(\frac{1}{2}R_{1l} - 1\right)\frac{(\mathbf{p''}\mathbf{p'''})}{a} - \frac{1}{2}R_{1l}\frac{(\mathbf{np''})(\mathbf{np'''})}{a}\right.$$

$$\left.\left. - \frac{1}{2}R_{1l}\frac{(\mathbf{p''}\mathbf{p'''}) - 3(\mathbf{np''})(\mathbf{np'''})}{a}\right]\right\}, \tag{3.9}$$

where $\mathbf{n} = \mathbf{a}/a$ and $\mathbf{d''} = e\boldsymbol{\xi''}$, $\mathbf{d'''} = e\boldsymbol{\xi'''}$ are the electric dipole operators of individual atoms. The operator (3.9) describes the electric dipole-dipole interaction of two arbitrarily spaced neutral atoms, one of which makes a transition to a certain intermediate state — when the interaction (3.9) is part of the process of emission (or absorption) of a real photon. In the special case of two atoms interacting without emitting or absorbing a real photon, operator (3.9) corresponds to the second-order quantum electrodynamics effect, for which the energy conservation equation $E_r^{(+)} - E_m^{(+)} + E_p^{(+)} - E_n^{(+)} = 0$ is satisfied. In this case we must set $R_{1l} = 1$ in the operator (3.9), thus converting it into the corresponding operator of Ref. [11].

4. Role of an external field in the interaction of two atomic electrons

Let us use the transformation (3.1) to go over from the wave function Ψ to the two-component wave functions Φ in the matrix elements of the type

$$\langle \overline{\Psi}_r^{(+)}(\mathbf{r'})|e\widehat{A}'(\mathbf{r'})|\Psi_l(\mathbf{r'})\rangle, \tag{4.1}$$

in the matrix (2.1). The matrix elements (4.1) of the vector potential of the external field determine the interaction of two atomic electrons with the field of real photons. Consider first matrix elements (4.1) for the cases in which atomic electrons make their transitions via positive-energy intermediate states. Performing some necessary manipulations on the matrix element (4.1) for the atomic transition from the state $\Phi_l^{(+)}(\mathbf{r'})$ to $\Phi_r^{(+)}(\mathbf{r'})$, we separate out the following transition operator:

$$R_1^+ = eA_4'(\mathbf{r'}). \tag{4.2}$$

The terms proportional to $1/c$ have the form

$$R_2^+ = -\frac{ie}{2mc}(\mathbf{p'}\mathbf{A'}) - \frac{ie}{2mc}(\mathbf{A'}\mathbf{p'}) - \frac{i\hbar e}{2mc}(\boldsymbol{\sigma'}\mathbf{H'}), \tag{4.3}$$

where $\mathbf{A'}(\mathbf{r'})$ is the vector potential operator for the external field at the electron position described by the radius vector $\mathbf{r'}$, and $\mathbf{H'}(\mathbf{r'})$ is the corresponding magnetic field strength operator ($[\nabla'\mathbf{A'}] = \mathbf{H'}$). The operator R_3^+ containing the second power of $1/c$ is

$$R_3^+ = -\frac{e\hbar^2}{8m^2c^2}(\Delta'A_4') - \frac{i\hbar e}{4m^2c^2}(\nabla'A_4')\mathbf{p'}$$

$$+ \frac{e}{4m^2c^2}(\boldsymbol{\sigma'}\mathbf{p'})A_4'(\boldsymbol{\sigma'}\mathbf{p'}). \tag{4.4}$$

The terms of order $1/c^3$ in the transition operator are of the form

$$R_4^+ = \frac{e}{16m^3c^3}\left\{i(\boldsymbol{\sigma'}\mathbf{A'})(\boldsymbol{\sigma'}\mathbf{p'})\mathbf{p'^2} - i\mathbf{p'^2}(\boldsymbol{\sigma'}\mathbf{A'})(\boldsymbol{\sigma'}\mathbf{p'})\right.$$

$$+ \hbar\sum_\alpha\sigma'_\alpha\left(\boldsymbol{\sigma'}\frac{\partial\mathbf{A'}}{\partial x'_\alpha}\right)\mathbf{p'^2} + i\boldsymbol{\sigma'}(\boldsymbol{\sigma'}\mathbf{A'})\mathbf{p'^3}$$

$$\left. - \hbar\sum_\alpha\sigma'_\alpha\mathbf{p'^2}\left(\boldsymbol{\sigma'}\frac{\partial\mathbf{A'}}{\partial x'_\alpha}\right) - i\boldsymbol{\sigma'}\mathbf{p'^2}(\boldsymbol{\sigma'}\mathbf{A'})\mathbf{p'}\right\}, \tag{4.5}$$

where $\alpha = x, y, z$. Each of the transition operators obtained can cause an atomic electron to make a quantum transition from a certain intermediate state $\Phi_l^{(+)}$ to a final state $\Phi_r^{(+)}$ provided the electron has come to this intermediate state through its exchange of virtual photons with an electron in the

other atom. Note that the potential A'_4 of the quantized external radiation field is zero in this case. For a static external field, the term (4.2) differs from zero, and the interaction between electrons belonging to two different atoms then occurs via the field of virtual photons, in which process the conservation law

$$E_r^{(+)} - E_m^{(+)} + E_p^{(+)} - E_n^{(+)} = 0 \,.$$

is obeyed.

4.1 Inclusion of negative-energy intermediate states

The effective interaction energy matrix (2.1) involves a summation over the negative-energy intermediate states of the interacting electrons, implying that some of the interaction energy is due to the influence of positronic states in the spectrum of the electrons. This influence enters indirectly through the electrons' intermediate virtual states, both the initial and final states being positive-energy electronic ones. The positronic intermediate states can be included by going over to a two-component wave function in Eqn (2.1), with the use of wave functions of the form

$$\Psi_l^{(-)} = \begin{pmatrix} -\dfrac{(\boldsymbol{\sigma}\mathbf{p})}{2mc}\chi_l^{(-)} \\ \chi_l^{(-)} \end{pmatrix}, \quad \chi_l^{(-)} = \left(1 - \dfrac{\mathbf{p}^2}{8m^2c^2}\right)\Phi_l^{(-)} \,.$$
(4.6)

Note that there is no need, in this procedure, to go over to the positronic wave function (which contains the charge conjugate transformation [11]). Instead, we proceed as follows to account for the intermediate positronic states.

We note, first, that the photon energy $\hbar\omega$ is much lower than the energy of the electron field and that the electron energies differ little from their rest values. Accordingly in (2.1), we set

$$\hbar\left[\omega_l(1 - \mathrm{i}0) + \omega - \omega_r^{(+)}\right] \approx -2mc^2$$
(4.7)

etc.

Second, we introduce the projection operators

$$\Lambda'_- = \frac{mc^2 - H'}{2mc^2}, \quad \Lambda''_- = \frac{mc^2 - H''}{2mc^2} \,,$$
(4.8)

which have the properties that

$$\Lambda'_- \Psi_l(\mathbf{r}') = \Psi_l^{(-)}(\mathbf{r}'), \quad \Lambda''_- \Psi_l(\mathbf{r}'') = \Psi_l^{(-)}(\mathbf{r}'') \,.$$
(4.9)

Then, applying the transformation of Section 2, we can isolate the following operator from the first and second terms in the matrix (2.1):

$$P_{1l}^+ = \frac{e}{2mc^2}\left(\gamma'_4\gamma'_\delta A'_\delta \Lambda'_- B_{1l} + B_{2l}\Lambda''_-\gamma''_4\gamma''_\delta A''_\delta\right),$$
(4.10)

where $\delta = 1,2,3,4$ and A'_δ is the four-vector potential operator for the external field at the position of the electron described by the radius vector $\mathbf{r}'$.

The remaining terms in the matrix (2.1) can be combined pairwise in a similar way. We now transform operator (4.10) using the wave functions (3.1) for the initial and final electronic states. The quantities R_{1l} and R_{2l} already contain the factor $1/(2mc^2)$, and the operators B_{1l} and B_{2l} are therefore of reduced form. We next perform the matrix multiplication of the operators in Eqn (4.10) and multiply out the wave functions of the interacting electrons. Keeping terms of order $1/c$ and integrating by parts where necessary,

the operator (4.10) becomes

$$\begin{aligned}
P_1^+ = &\frac{e^3}{2mc^2}\exp\left\{\frac{\mathrm{i}}{c}\left|\omega_n^{(+)} - \omega_p^{(+)}\right|a\right\}\left\{-\frac{\mathrm{i}\hbar}{2mc}\frac{a}{r^2}(\boldsymbol{\sigma}''\mathbf{n})\right. \\
&\times \sum_\alpha \sigma'_\alpha\frac{\partial A'_4}{\partial x'_\alpha} - \frac{1}{2mc}(\boldsymbol{\sigma}'\mathbf{p}')(\boldsymbol{\sigma}''\mathbf{n})\frac{a}{r^2}A'_4 + \frac{\mathrm{i}\hbar}{4m^2c^2} \\
&\times (\boldsymbol{\sigma}'\mathbf{A}')(\boldsymbol{\sigma}''\mathbf{p}'')\frac{1}{r}(\boldsymbol{\sigma}'\mathbf{A}')\boldsymbol{\sigma}''\sum_\alpha\sigma'''_\alpha\boldsymbol{\sigma}'''\left(\frac{1}{r}\frac{\partial A'''_4}{\partial x'''_\alpha} + \frac{n_\alpha}{r^2}\right) \\
&- \mathrm{i}(\boldsymbol{\sigma}'\mathbf{A}')(\boldsymbol{\sigma}''\mathbf{n})\frac{a}{r^2} + \frac{\mathrm{i}}{2mc}(\boldsymbol{\sigma}'\mathbf{A}')(\boldsymbol{\sigma}''\boldsymbol{\sigma}''')(\boldsymbol{\sigma}'''\mathbf{p}''')\frac{1}{r} \\
&+ \frac{\mathrm{i}}{2mc}(\boldsymbol{\sigma}'\mathbf{A}')(\boldsymbol{\sigma}'\mathbf{p}')\frac{1}{r} + \frac{1}{2mc}(\boldsymbol{\sigma}'\mathbf{n})(\boldsymbol{\sigma}''\mathbf{p}'')\frac{a}{r^2}A'_4 \\
&- \frac{1}{2mc}\frac{a}{r^2}(\boldsymbol{\sigma}'\mathbf{n})(\boldsymbol{\sigma}''\mathbf{p}'')A''_4 + \frac{\hbar}{2mc}\sum_\alpha(\boldsymbol{\sigma}''\mathbf{A}'')\sigma'_\alpha\frac{n_\alpha}{r^2} \\
&+ \frac{\mathrm{i}}{2mc}\frac{1}{r}(\boldsymbol{\sigma}''\mathbf{p}'')(\boldsymbol{\sigma}''\mathbf{A}'') + \frac{\mathrm{i}}{2mc}\frac{1}{r}\boldsymbol{\sigma}'(\boldsymbol{\sigma}''\mathbf{A}'')\boldsymbol{\sigma}'''(\boldsymbol{\sigma}'''\mathbf{p}''') \\
&\left.- \mathrm{i}\frac{a}{r^2}(\boldsymbol{\sigma}'\mathbf{n})(\boldsymbol{\sigma}''\mathbf{A}'')\right\},
\end{aligned}$$
(4.11)

where $\boldsymbol{\sigma}'$, $\boldsymbol{\sigma}''$ operate on the spin wave functions of the electron in the first atom and $\boldsymbol{\sigma}'''$ on those of the electron in the second one. We now use the identities

$$\boldsymbol{\sigma}'''(\boldsymbol{\sigma}'''\mathbf{p}''') = -(\boldsymbol{\sigma}'''\mathbf{p}''')\boldsymbol{\sigma}''' + \mathbf{p}''' \,,$$

$$(\boldsymbol{\sigma}'''\mathbf{p}''')\frac{1}{r} = \frac{1}{r}(\boldsymbol{\sigma}'''\mathbf{p}''') - \mathrm{i}\hbar\frac{(\mathbf{n}\boldsymbol{\sigma}''')}{r^2} \,,$$

$$(\boldsymbol{\sigma}''\mathbf{A}'')(\boldsymbol{\sigma}''\boldsymbol{\sigma}''')(\boldsymbol{\sigma}'''\mathbf{p}''') = (\mathbf{A}''\mathbf{p}''') + \mathrm{i}\boldsymbol{\sigma}'''[\mathbf{A}''\mathbf{p}''']$$
$$- \mathrm{i}\mathbf{p}'''[\mathbf{A}''\boldsymbol{\sigma}''] + \sum_{\alpha\neq\beta}\sigma''_\alpha\sigma'''_\alpha A''_\beta p'''_\beta - \sum_{\alpha\neq\beta}\sigma''_\alpha p'''_\alpha A''_\beta \sigma'''_\beta \,. \quad (4.12)$$

and assume that the two atomic electrons interact in an alternating external field and that the vector potential operator $\mathbf{A}$ satisfies the Lorentz condition $\partial A_\mu/\partial x_\mu = 0$, $\mu = 1, 2, 3, 4$. In this case we can set

$$A'_4 = 0, \qquad \frac{\partial A''_4}{\partial x} = 0 \,.$$
(4.13)

and the operator (4.11) then assumes the form

$$\begin{aligned}
P_1^+ = &\frac{e^3}{2mc^2}\exp\left\{\frac{\mathrm{i}}{c}\left|\omega_n^{(+)} - \omega_p^{(+)}\right|a\right\}\left\{-2\mathrm{i}(\mathbf{A}''\mathbf{n}'')\frac{a}{r^2}\right. \\
&+ 2\boldsymbol{\sigma}''[\mathbf{A}''\mathbf{n}]\frac{a}{r^2} + \frac{\hbar}{mc}(\mathbf{A}''\mathbf{n})\frac{1}{r^2} + \frac{\mathrm{i}\hbar}{mc}\boldsymbol{\sigma}''[\mathbf{A}''\mathbf{n}]\frac{1}{r^2} \\
&+ \frac{\mathrm{i}}{2mc}\frac{1}{r}(\mathbf{p}''\mathbf{A}'') - \frac{1}{2mc}\frac{1}{r}\boldsymbol{\sigma}''[\mathbf{p}''\mathbf{A}''] + \frac{\mathrm{i}}{mc}\frac{1}{r}(\mathbf{A}''\mathbf{p}''') \\
&\left.- \frac{\mathrm{i}}{2mc}\frac{1}{r}\left(\sum_{\alpha\neq\beta}\sigma''_\alpha p'''_\alpha A''_\beta p'''_\beta - \sum_{\alpha\neq\beta}\sigma''_\alpha p'''_\alpha A''_\beta \sigma'''_\beta\right)\right\} \,. \quad (4.14)
\end{aligned}$$

In an analogous fashion we transform the remaining terms in the matrix (2.1) and the corresponding operators for the negative-energy intermediate states. The meaning of these operators (which we denote by P_2^+, P_3^+ and P_4^+) will be discussed later.

4.2 Positive-energy intermediate states

Consider now the sum over the positive-energy intermediate states (i.e., electron states l_+) in the effective interaction energy matrix. If we transform the matrix elements by using the approximate wave functions (3.1) in the same way as in

Sections 3 and 4, then the first two terms in the matrix (2.1) — those which correspond to diagrams *1* and *2* in Fig. 1 and which have identical retardation factors $\exp\{(\mathrm{i}/c)|\omega_n^{(+)} - \omega_p^{(+)}|a\}$ — take the form

$$A_{i \to f}^{(3)} = \sum_{s,s'} \sum_{l_+} \left\{ \frac{\langle \Phi_r^{(+)}|R_s^+|\Phi_l^{(+)}\rangle\langle \Phi_l^{(+)}\Phi_p^{(+)}|V_{s'l1}^{(+)}|\Phi_m^{(+)}\Phi_n^{(+)}\rangle}{\hbar[\omega_l(1-\mathrm{i}0) + \omega - \omega_r^{(+)}]} \right.$$
$$\left. + \frac{\langle \Phi_r^{(+)}\Phi_p^{(+)}|V_{s'l2}^{(+)}|\Phi_l^{(+)}\Phi_n^{(+)}\rangle\langle \Phi_l^{(+)}|R_s^+|\Phi_m^{(+)}\rangle}{\hbar[\omega_l(1-\mathrm{i}0) - \omega - \omega_m^{(+)}]} \right\}, \quad (4.15)$$

where the operators $V_{s'l1}^{(+)}$ and $V_{s'l2}^{(+)}$ are obtained from the operators B_{1l} and B_{2l}, respectively. Terms of order higher than third in $1/c$ will be dropped in the products of various matrix elements in Eqn (4.15). The remaining terms in Eqn (2.1) will be of similar form. Let us denote them by $B_{i \to f}^{(3)}$, $C_{i \to f}^{(3)}$, and $D_{i \to f}^{(3)}$. All the terms we have separated out in the matrix (2.1) have different meanings, as will be shown below.

5. Polarizing fields in a system of hydrogen-like atoms emitting or absorbing photons

Contained as factors in the operators (4.3), (4.4), and (4.5) for the first-order effects are atomic and field operators which cause quantum transitions to occur between the atoms and photonic states at one and the same point of observation. For two arbitrarily separated interacting atoms, with operators of the form (4.14) and matrix elements (4.15), the situation is different. In various terms of Eqn (4.14) one can separate out (a) an operator which acts at a point of observation and (b) an operator which acts at the position of the other atom, the one forming the polarizing field. The polarizing field in this case is that of virtual rather than real photons. Similarly, matrix elements of the type (4.15) contain the dependence on the coordinates of the two atoms, one of which is at the point of observation.

Consider now the polarizing fields that form according to scheme *C* of Section 2.1 — a scheme in which, due to the exchange of virtual photons and the emission (or absorption) of one real photon, only one of the atoms changes its state. Assume that the position of the first atom is described by the radius vector $\mathbf{r}_1$ (coordinates $\mathbf{r}'$ and $\mathbf{r}''$), and the position of the second atom, by $\mathbf{r}_2$ (coordinates $\mathbf{r}'''$). The initial state of the first atom is labeled by the index m with energy $E_m^{(+)}$, and that of the second, by n with energy $E_n^{(+)}$. As a result of virtual photon exchange, the first atom finds itself in an intermediate state $E_l^{(+)}$ or $E_l^{(-)}$ and then returns to its initial state, i.e., $E_r^{(+)} = E_m^{(+)}$. Whereas at the location of the first atom one real photon is absorbed, the second atom changes its quantum state and makes a transition to a level $E_p^{(+)} > E_n^{(+)}$. Such a transition scheme corresponds to the first term in the matrix (2.1) and to diagram *1* (see Fig. 1). A similar situation exists with regard to the second term in Eqn (2.1) — that for the Feynman diagram *2* in Fig. 1 — when a photon is absorbed not at the position of the atom that makes the transition $E_n^{(+)} \to E_p^{(+)}$ but rather at the location of the other atom, which forms the polarizing field. The remaining elements of the matrix (2.1) (whose diagrams are *3–8*) do not participate in the formation of the polarizing field in this scheme. Indeed, for $r = m$ each of these terms contains zero matrix elements of the type $\langle \Psi_r^{(+)}|\gamma_\mu'|\Psi_m^{(+)}\rangle$.

We proceed to write the vector potentials of the polarizing fields explicitly, using Eqns (4.14) and (4.15). For this it is necessary to distinguish a particular type of transition between the states of the interacting atoms — orbital transitions, for example. Then, in accord with Eqn (4.3), the following Hamiltonian operator is obtained for first-order effects for either of the atoms:

$$H_1''' = -\frac{e}{mc}\,\mathbf{p}'''\mathbf{A}''', \quad (5.1)$$

where $\mathbf{A}'''$ is the operator for the vector potential of the external field at the position of the second atom. With the help of the operator (4.14) we can write the Hamiltonian operator in the form

$$H_2''' = -\frac{e}{mc}\,\mathbf{p}'''\mathbf{A}^{(\mathrm{p})},$$
$$\mathbf{A}^{(\mathrm{p})} = -\frac{e^2}{2mc^2}\,\frac{1}{r}\exp\left(\frac{\mathrm{i}}{c}\,\omega_0 a\right)\mathbf{A}'', \quad (5.2)$$

where $\omega_0 = \omega_p^{(+)} - \omega_n^{(+)}$ is the frequency of the $p \to n$ transition. We shall refer to the polarizing field $\mathbf{A}^{(\mathrm{p})}$ as to the *positronic polarizing field*. The polarizing field (5.2) is due to the process in which a photon disappears at the location of the second atom and is absorbed at the location of the first one. The remaining terms in Eqn (4.14) describe the orbital quantum transitions and differ from the operator (5.2) in their physics. The $\mathbf{p}''\mathbf{A}''$ term, in particular, corresponds to the positronic polarizing field, but with a photon disappearing and being absorbed at one and the same point. Thus, the inclusion of positronic intermediate states in the interaction of two atomic electrons gives rise to an additional Hamiltonian, responsible for the interaction of atomic electrons with an external field. As a result, one should add a positronic polarizing field $\mathbf{A}^{(\mathrm{p})}$ to the external field $\mathbf{A}'''$ when considering the interaction of a system of atomic electrons with an external field.

We next consider the role of the interaction (4.15) via positive-energy intermediates states, taking only orbital quantum transitions into account. Inserting the operators (4.3) and (3.9) into Eqn (4.15) yields the following interaction Hamiltonian [17]:

$$H_3''' = -\frac{e}{mc}\,\mathbf{p}'''\mathbf{A}^{(\mathrm{e})}, \quad (5.3)$$

where $\mathbf{A}^{(\mathrm{e})}$, the vector potential of the electronic polarizing field, is given by

$$\mathbf{A}^{(\mathrm{e})} = \exp\left(\frac{\mathrm{i}}{c}\,\omega_0 a\right)\sum_{l_+}\left(\frac{(\mathbf{p}'_{rl}\mathbf{A}')}{\hbar[\omega_l(1-\mathrm{i}0) + \omega - \omega_r^{(+)}]}\right.$$
$$\times \left\{\frac{e}{\mathrm{i}m\omega_0}\frac{\mathbf{d}''_{lm} - 3(\mathbf{d}''_{lm}\mathbf{n})\mathbf{n}}{a^3} + \frac{e^2}{\mathrm{i}m^2 c\omega_0}\frac{\mathbf{p}''_{lm} - 3(\mathbf{p}''_{lm}\mathbf{n})\mathbf{n}}{a^2}\right.$$
$$+ \frac{e^2}{m^2 c^2}\left[\left(\frac{1}{2}R_{1l} - 1\right)\frac{1}{a}\,\mathbf{p}''_{lm} - \frac{1}{2}R_{1l}\frac{(\mathbf{np}''_m)\mathbf{n}}{a}\right.$$
$$\left.\left. - \frac{1}{2}R_{1l}\frac{\mathbf{p}''_{lm} - 3(\mathbf{np}''_{lm})\mathbf{n}}{a}\right]\right\} + \frac{1}{\hbar[\omega_l(1-\mathrm{i}0) - \omega - \omega_m^{(+)}]}$$
$$\times \left\{\frac{e}{\mathrm{i}m\omega_0}\frac{\mathbf{d}'_{rl} - 3(\mathbf{d}'_{rl}\mathbf{n})\mathbf{n}}{a^3} + \frac{e^2}{\mathrm{i}m^2 c\omega_0}\frac{\mathbf{p}'_{rl} - 3(\mathbf{p}'_{rl}\mathbf{n})\mathbf{n}}{a^2}\right.$$
$$+ \frac{e^2}{m^2 c^2}\left[\left(\frac{1}{2}R_{2l} - 1\right)\frac{1}{a}\,\mathbf{p}'_{rl} - \frac{1}{2}R_{2l}\frac{(\mathbf{np}'_{rl})\mathbf{n}}{a} - \frac{1}{2}R_{2l}\right.$$
$$\left.\left.\times \frac{\mathbf{p}'_{rl} - 3(\mathbf{np}'_{rl})\mathbf{n}}{a}\right]\right\}(\mathbf{p}''_{rl}\mathbf{A}'')\right), \quad (5.4)$$

with

$$R_{1l} = \frac{\omega_n^{(+)} - \omega_p^{(+)}}{\omega_l - \omega_m^{(+)}} , \qquad R_{2l} = \frac{\omega_n^{(+)} - \omega_p^{(+)}}{\omega_l - \omega_r^{(+)}} .$$

The electronic polarizing field (5.4) is written in the electrical dipole approximation using operator (3.9) to account for the virtual photon exchange. Note that the operators $\mathbf{A}'$ and $\mathbf{A}''$ in Eqn (5.4) were taken out of the matrix element sign in this approximation. The electronic polarizing field (5.4) is formed by two atoms, one of which finds itself in its initial quantum state as a result of the sequence of quantum transitions under study. This means that in Eqn (5.4) we can separate out the average value of the electric dipole moment of this atom in a certain state $r = m$. First-order perturbation theory approximates the averaged dipole moment $\mathbf{d}'_m$ as [2]

$$\mathbf{d}'_m = \frac{e}{mc\hbar} \exp(-\mathrm{i}\omega t) \sum_{l_+} \left\{ \frac{\mathbf{d}'_{ml}(\mathbf{p}'_{lm}\mathbf{A}'_0)}{\omega_{lm} - \omega + (\mathrm{i}/2)(\Gamma_l + \Gamma_m)} \right.$$
$$\left. + \frac{(\mathbf{p}'_{ml}\mathbf{A}'_0)\,\mathbf{d}'_{lm}}{\omega_{lm} + \omega - (\mathrm{i}/2)(\Gamma_l + \Gamma_m)} \right\}, \tag{5.5}$$

where ω_{lm} is the transition frequency, $\Gamma_{l(m)}^{-1}$ is the lifetime of the state $l(m)$, and $\mathbf{A}' = \mathbf{A}'_0 \exp(-\mathrm{i}\omega t)$. Note that in Eqn (5.5) we only left the negative-frequency term, responsible for the photon absorption process leading to the polarizing field (5.4). Using Eqn (5.5) and suppressing the index m, Eqn (5.4) becomes

$$\mathbf{A}^{(e)} = \frac{c}{\mathrm{i}\omega_0} \frac{[\mathbf{d}'] - 3([\mathbf{d}']\mathbf{n})\mathbf{n}}{a^3} + \frac{e}{\mathrm{i}m\omega_0} \frac{[\mathbf{p}'] - 3([\mathbf{p}']\mathbf{n})\mathbf{n}}{a^2}$$
$$- \frac{\mathrm{i}\omega_0}{c} \frac{([\mathbf{d}']\mathbf{n})\mathbf{n}}{a} - \frac{e}{mc} \frac{[\mathbf{p}']}{a} , \tag{5.6}$$

where the notation $[\dots]$ indicates an average taken at the time $t' = t - (a/c)(\omega_0/\omega)$. Here we have made use of the isotropy property of the atoms, which implies that the quantities $(\mathbf{p}'_{rl}\mathbf{A}')\,\mathbf{d}'_{lm}$ and $\mathbf{p}'_{rl}(\mathbf{d}'_{lm}\mathbf{A}')$ are equal. We next transform from the vector potentials (5.2) and (5.3) of the polarizing fields to the corresponding electric and magnetic field strengths using the Lorentz condition [11]

$$\frac{\partial A_\mu}{\partial x_\mu} = 0$$

For a radiation field proportional to $\exp(-\mathrm{i}\omega t)$ we have

$$\mathbf{E}^{(e)} = \frac{\mathrm{i}\omega}{c} \mathbf{A}^{(e)} , \qquad \mathbf{E}^{(p)} = \frac{\mathrm{i}\omega}{c} \mathbf{A}^{(p)} , \tag{5.7}$$

where the vector potentials $\mathbf{A}^{(e)}$ and $\mathbf{A}^{(p)}$ are proportional to $c_{\mathbf{k}\lambda}^{\mathrm{v}}$, the creation operator for a photon of wave vector $\mathbf{k}$ and polarization index $\lambda = 1, 2$ at the position of the atom producing the polarizing field (the atom-polarizer) in the process when the quantum transition $p \rightarrow n$ takes place at the position of the other atom (the atom-observer), arbitrarily separated from the atom-polarizer.

Let us now consider the case in which the frequency ω of the external radiation field is close to one of the frequencies $\omega_{lm} > 0$. Then the mean momentum is

$$[\mathbf{p}'] = \frac{\mathrm{i}m}{e} \omega_0[\mathbf{d}'] ,$$

and the operator for the strength of the electronic polarizing field is, from Eqn (5.6),

$$\mathbf{E}^{(e)} = \operatorname{rot} \operatorname{rot} \frac{[\mathbf{d}]}{a} , \tag{5.8}$$

where the differentiation is carried out with respect to the coordinates of the observation point a'''. The magnetic field strengths $\mathbf{H}^{(e)}$ and $\mathbf{H}^{(p)}$ are determined in a similar way using the standard vector potential – strength relation for the magnetic field (see Ref. [11]).

5.1 Integral equations for photon propagation in an electric dipole optical medium

Let us introduce the dipole (α_d) and momentum (α_p) atomic polarizabilities by means of the relations

$$\mathbf{d}' = \alpha_d \mathbf{A}' , \qquad \mathbf{p}' = \alpha_p \mathbf{A}' , \tag{5.9}$$

where $\mathbf{d}'$ and $\mathbf{p}'$ are the average, second-order perturbation theory values of the dipole moment and momentum in the state m. From Eqn (5.5) one can determine the dipole polarizability of an isotropic atom. To find the momentum polarizability α_p, the matrix elements $\mathbf{d}'_{lm}$ and Eqn (5.5) should be replaced by the matrix elements $\mathbf{p}'_{lm}$ of the momentum operator. Then the vector potential of the electronic polarizing field assumes the form

$$\mathbf{A}^{(e)} = \left\{ \alpha_d \frac{c}{\mathrm{i}\omega_0} \frac{\mathbf{e} - 3(\mathbf{e}\mathbf{n})\mathbf{n}}{a^3} + \alpha_p \frac{e}{\mathrm{i}m\omega_0} \frac{\mathbf{e} - 3(\mathbf{e}\mathbf{n})\mathbf{n}}{a^2} \right.$$
$$\left. - \alpha_d \frac{\mathrm{i}\omega_0}{c} \frac{(\mathbf{e}\mathbf{n})\mathbf{n}}{a} - \frac{e}{mc} \alpha_p \frac{\mathbf{e}}{a} \right\} [A'] \equiv \mathbf{K}_{\mathrm{e}}(a', a''')[A'] , \tag{5.10}$$

where $\mathbf{e}$ is the unit vector along the field $\mathbf{A}'$.

A transformation to an N-atom system can be carried out by summing the vector potentials, Eqns (5.2) and (5.10), of the polarizing fields created by $N - 1$ atoms at the position of the atom with a radius vector $\mathbf{a}''$. To assess the role of the electronic and positronic polarizing fields in such a system, terms in Eqn (5.10) proportional to $1/a$ must be compared with the vector potential (5.2). In doing this, it should be taken into account that, whereas the electronic field contains the polarizabilities α_d and α_p, which depend on the random distribution of eigenfrequencies ω_0 due to non-uniform broadening, the positronic field is independent of this broadening — the reason for why, provided certain conditions in the N-atom system, the electronic and positronic polarizing fields may become comparable in magnitude.

We turn our discussion to the optical medium. We assume the medium to be continuous and therefore introduce the concentration N/V to describe the distribution of atoms in it. Furthermore, we assume that the polarizing fields (5.10) and (5.2) are proportional not to the external field but rather to the field inside the medium. This allows us to write the following integral equation for the electrical field strength operator [17]:

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_I(\mathbf{r}, t) + \int \frac{N}{V} \mathbf{K}_{\mathrm{e}}(\mathbf{r}, \mathbf{r}') E\left(\mathbf{r}', t - \frac{R}{c}\right) \mathrm{d}V'$$
$$+ \int \frac{N}{V} \mathbf{K}_{\mathrm{p}}(R) E\left(\mathbf{r}', t - \frac{R}{c}\right) \mathrm{d}V' , \tag{5.11}$$

where $\mathbf{r}$ is the radius vector of the observation point; $\mathbf{r}'$ is the radius vector of a point within the medium or on its surface; $\mathbf{E}_I(\mathbf{r}, t)$ is the external electric field strength operator, represented as a superposition of plane waves with coordinate-independent amplitudes $c_{\mathbf{k}\lambda}^{\mathrm{v}}$ [11]; $R = |\mathbf{r} - \mathbf{r}'|$; and

$$\mathbf{K}_{\mathrm{p}}(R) = -\frac{e^2}{2mc^2} \frac{1}{R} . \tag{5.12}$$

An integral equation for the magnetic field strength operator can be obtained in a similar way by applying the rot operator to the vector potentials (5.10) and (5.2).

If the point of observation is outside the medium, then the integral in Eqn (5.11) is performed over the whole of the medium. If the point of observation is within the medium, it is necessary first to exclude a small sphere of radius L_0 with the atom inside. In the special case in which there is no positronic polarization in the medium and only one eigenfunction in the atomic spectrum is singled out, Eqn (5.11) is identical to the integro-differential equation of classical optics [18] if one transforms from operators to the corresponding classical fields in Eqn (5.10).

5.2 Integral equation for photon propagation in a system of electron spins

Let us consider one more example of an integral equation obtainable with the proposed method. Unlike the preceding case, we will consider only the spin degrees of freedom of the electrons in two one-electron atoms positioned arbitrarily far apart. This is relevant, for example, to magnetooptics, inversionless lasers, laser cooling of atoms, etc. We will assume spin transitions to take place between atomic states separated by the optical frequency ω_0. Such transitions may occur independently of the electric dipole transitions of Section 5.1.

We will apply operators (3.6) and

$$\left(R_2^{(+)}\right)_\sigma = -\frac{\mathrm{i}\hbar e}{2mc}\left(\boldsymbol{\sigma}'\mathbf{H}'\right) \tag{5.13}$$

to describe polarizing fields in a system of electron spins and will consider the same quantum transition scheme as that employed in Section 5.1.

Substituting operators (3.6) and (5.13) into the matrix (4.15) we obtain the following interaction operator:

$$H_3''' = -\frac{\hbar e}{2mc}\left(\boldsymbol{\sigma}'''\,\mathbf{H}^{(e)}\right), \tag{5.14}$$

where the magnetic field strength is

$$\mathbf{H}^{(e)} = \exp\left(\frac{\mathrm{i}}{c}\omega_0 a\right)\frac{e^2\hbar^2}{4m^2c^2}\sum_{l_+}\Bigg(\frac{(\boldsymbol{\sigma}_{rl}\,\mathbf{H}')}{\hbar\left[\omega_l(1-\mathrm{i}0)+\omega-\omega_r^{(+)}\right]}$$
$$\times\left\{\frac{\boldsymbol{\sigma}_{lm}''-3(\boldsymbol{\sigma}_{lm}''\,\mathbf{n})\mathbf{n}}{r^3}+15\frac{a^2}{r^5}\,R_{1l}(\boldsymbol{\sigma}_{lm}''\,\mathbf{n})\mathbf{n}-9\frac{a^2}{r^5}\,R_{1l}\,\boldsymbol{\sigma}_{lm}''\right\}$$
$$+\frac{1}{\hbar\left[\omega_l(1-\mathrm{i}0)-\omega-\omega_m^{(+)}\right]}\left\{\frac{\boldsymbol{\sigma}_{rl}'-3(\boldsymbol{\sigma}_{rl}'\,\mathbf{n})\mathbf{n}}{r^3}\right.$$
$$\left.+15\frac{a^2}{r^5}\,R_{2l}(\boldsymbol{\sigma}_{rl}'\,\mathbf{n})\mathbf{n}-9\frac{a^2}{r^5}\,R_{2l}\,\boldsymbol{\sigma}_{rl}'\right\}\Bigg). \tag{5.15}$$

This field is the electronic polarizing field, i.e., it is due to the electronic intermediate states only. As can be seen from Eqn (4.14), a positronic polarizing field does not exist for the transition scheme adopted.

We proceed by separating out in Eqn (5.15) the average values of the spin magnetic moments of the atom-polarizer, using a formula analogous to the first-order perturbation theory result (5.5). For the average values of the spin variables in the state m we introduce the notation

$$\boldsymbol{\sigma}_m' = (\alpha_\sigma)_m\mathbf{H}', \qquad \mathbf{q}_m' = (\alpha_q)_m\mathbf{H}', \tag{5.16}$$

where $(\alpha_\sigma)_m$ is the spin polarizability in the state m; $\mathbf{q}_m'$ is the average value, for the state m, of the operator $\mathbf{q}'$ with matrix elements $\mathbf{q}_{lm}' = \boldsymbol{\sigma}_{lm}'/\omega_{lm}$; and $(\alpha_q)_m$ is the corresponding polarizability. The polarizing field (5.15) then takes the form

$$\mathbf{H}^{(e)} = \mu_{\mathrm{B}}\bigg\{\alpha_\sigma\,\frac{\mathbf{h}-3(\mathbf{hn})\mathbf{n}}{r^3}-15\omega_0\alpha_q\,\frac{a^2}{r^5}\,(\mathbf{hn})$$
$$+9\omega_0\alpha_q\,\frac{a^2}{r^5}\,\mathbf{h}\bigg\}[H'] \equiv \mathbf{K}_\sigma(\mathbf{a}',\mathbf{a}''')[H'], \tag{5.17}$$

where μ_{B} is the Bohr magneton and $\mathbf{h}$, the unit vector along the magnetic field strength $\mathbf{H}'$. A transition to the integral equation for photon propagation in a continuous optical medium is performed in the same way as in Section 5.1., i.e., by replacing the external field in Eqn (5.17) by the field inside the medium. The magnetic field strength operator for spin transitions then takes the following form for an external or internal point of observation $\mathbf{r}$ at the time t [7]:

$$\mathbf{H}(\mathbf{r},t) = \mathbf{H}_I(\mathbf{r},t) + \int\frac{N}{V}\,\mathbf{K}_\sigma(\mathbf{r},\mathbf{r}')H\left(\mathbf{r}',t-\frac{R}{c}\right)\mathrm{d}V', \tag{5.18}$$

where $\mathbf{H}_I(\mathbf{r},t)$ is the external magnetic field strength operator.

The results of the preceding analysis can be divided into two main categories, namely, (a) those related to the solution of the major quantum electrodynamics problem of two interacting electrons and (b) those concerning the application of this solution to the derivation of integral field equations for problems in optics.

The interaction of two electrons belonging to two respective, arbitrarily separated atoms at rest is treated as a third-order quantum electrodynamics effect, part of it being virtual photon exchange involving various positive- and negative-energy intermediate states. With this approach, it proves possible to classify various transitions schemes that lead to the emission or absorption of a photon in a system of two interacting hydrogen-like atoms. The scheme we have discussed above in detail is that in which one of the atoms is an atom-polarizer and the other, an atom-emitter (atom-absorber).

Let us discuss here the advisability of transforming to integral field equations from the corresponding differential equations. Although the integral equations describing the electric and magnetic field strengths in classical optics [18] are in effect equivalent to Maxwell's equations, they enable a rigorous derivation of the Lorentz–Lorentz formula and make it possible to solve a number of important optical problems. However, this powerful method has had very few applications thus far. A possible reason is that the well-known integral equations of classical optics have a sense only for isotropic, non-magnetic dielectric media. It is therefore of great interest to derive the corresponding integral equations for a wider class of problems — and this is precisely what the present paper is concerned with. The problem we addressed first was that of two electrons in the framework of quantum electrodynamics. The study of the interaction of two electrons in a radiation field reveals various types of quantum transition. In particular, as pointed out above, we can separate out the positronic polarizing field, which in some cases cannot be neglected compared with its electronic counterpart. In view of the correspondence between Eqn (5.11) and the classical integral equation, it can be argued that the positronic polarizing field can be interpreted as an

additional current in Maxwell's equations [19]. The electronic polarizing field in the electric dipole approximation is well known in optics: this is a field of dipoles which is entirely due to electronic states in the spectrum of interacting atoms. The positronic polarizing field, on the other hand, arises when positronic states are taken into account — not the real but virtual ones, whose existence does not require that the energy conservation law be obeyed. We thus see that the study of the two-electron problem in the framework of quantum electrodynamics not only contributes to the development of the method of integral equations but also reveals new mechanisms for the emission and absorption of real photons in a system of interacting atoms.

Two types of interaction between an atom and a radiation field are discernable. In one of these, real photons pass from excited atoms to unexcited ones, the radiation transfer time being determined by the distance between the atoms. This type of interaction can be taken as the basis for deriving integral equations for radiation transport, which are used, for example, in the optics of turbulent media [20]. The second type of interaction involves the concept of the polarizing field, a field considered in this review as a third-order quantum electrodynamics effect. As indicated above, a photon disappears at the location of one of the interacting atoms and is absorbed at the location of the other. A similar situation exists of course if one considers the emission of a photon in a system of two interacting atoms. An important point about this type of interaction is that a photon disappears and becomes absorbed at the same instant of time, implying that the polarizing field forms instantaneously due to the continuous exchange of virtual photons between the atomic electrons [otherwise, the conservation law (2.10) would be violated]. Note that the retardation factor $\exp[(i/c)\omega_0 a]$ in the various elements of matrix (2.1) only indicates that the effective interaction energy is a periodic function of the interatomic separation.

6. Semiclassical derivation of nonlocal equations

The quantum electrodynamical derivation of nonlocal equations in Section 5 is based on isolating the mean of the induced multipole moment of an atom from a polarizing field. For electric dipole transitions, the electronic polarizing field produced at a certain point of observation is proportional to the mean of the induced dipole moment of the atom-polarizer (this moment is $\alpha\mathbf{E}$, where α is the electronic polarizability of the atom-polarizer, and $\mathbf{E}$ is the electric field strength at the location of this atom). Taking this into account, one can employ a quasiclassical method to derive nonlocal equations of electrodynamics. Then, if one uses the electric dipole approximation, it is necessary that in the expression for the electric dipole field [18],

$$\mathbf{E}_1 = \text{rot rot}\, \frac{[\mathbf{d}_2]}{R_{12}},$$

the dipole moment $\mathbf{d}_2$ be written as $\alpha\mathbf{E}_2$, where $\mathbf{E}_1$ and $\mathbf{E}_2$ are the electric field strengths at the locations of the atom-observer and the atom-polarizer, respectively, R_{12} is the distance between the point dipoles, and $[\ldots]$ indicates that the quantity in question is taken at the time $t - R_{12}/c$. The essential point to be noted here is that the semiclassical method should be considered to be complementary to the quantum electrodynamical method because only this latter

yields a correct scheme of the transitions that produce polarizing fields. From this point of view, the electric dipole field of classical optics [18] appears as a field of virtual, rather than real, photons.

In what follows, a number of nonlocal equations derived semiclassically for classical alternating fields will be considered.

6.1 Integral equations for the propagation of electromagnetic waves in dielectrics

Eqn (5.11) for a classical field $\mathbf{E}(\mathbf{r}, t)$ can be derived as follows. Write down the Hamiltonian function for a system of N atoms at rest using retardation potentials to account for the interatomic interaction [4]. Expand the retardation potentials in powers of the small parameter (2.4), retain only the terms of the order of v^2/c^2, and assume the induced dipole moments of the atoms to be proportional to the electric field strength $\mathbf{E}$, the coefficient being the atomic polarizability α. Then, if only electronic polarizing fields are considered, we obtain [18]

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_I(\mathbf{r}, t) + \int \text{rot rot}\, \frac{\mathbf{P}(\mathbf{r}', t - R/c)}{R}\, dV', \qquad (6.1)$$

where $\mathbf{P} = (N/V)\alpha\mathbf{E}$ is the polarization vector of the medium, $R = |\mathbf{r} - \mathbf{r}'|$, $\mathbf{r}$ is the point of observation, $\mathbf{r}'$ is a point inside the medium or on its surface, and $\mathbf{E}_I$ is the electric field of the external wave at the point of observation at the time t.

Eqn (6.1) has been formulated for a continuous dielectric consisting of identical atoms. Following the same line of argument, Eqn (6.1) can be modified to describe, for example, activated, discrete-continuous, or nonlinear dielectrics if a more general phenomenological law is used to describe the polarization vector as a function of field $\mathbf{E}$ inside the medium. For activated dielectrics with a continuous distribution of resonant and nonresonant atoms with respective polarizalibilities α_R and α_{NR}, we have the integral equation [21]

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_I(\mathbf{r}, t) + \int \text{rot rot}\, \frac{1}{R}\left(\frac{N_R}{V}\right)\alpha_R\, \mathbf{E}\left(\mathbf{r}', t - \frac{R}{c}\right) dV'$$

$$+ \int \text{rot rot}\, \frac{1}{R}\left(\frac{N_{NR}}{V}\right)\alpha_{NR}\mathbf{E}\left(\mathbf{r}', t - \frac{R}{c}\right) dV', \quad (6.2)$$

where (N_R/V) and (N_{NR}/V) are the corresponding concentrations of resonant and non-resonant atoms.

In a discrete-continuous dielectric, we take into account a small region around the observation point, embraced by the so-called Lorentz sphere with the property that the atoms are distributed discretely inside it and continuously outside it. As will be shown below, the structure factor gives rise to the nearfield effect, which is taken into account by writing the nonlocal equation in the form [22]

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_I(\mathbf{r}, t) + \int_{V_1} \text{rot rot}\, \frac{\alpha}{R}\left(\frac{N}{V}\right)\mathbf{E}\left(\mathbf{r}', t - \frac{R}{c}\right) dV'$$

$$+ \sum_a \text{rot rot}\, \frac{[\mathbf{d}_a]}{R_a}, \qquad (6.3)$$

where V_1 is the volume of the dielectric occupied by the continuously distributed atoms; the summation runs over all discretely distributed atoms within the Lorentz sphere, with the induced dipole moments $\mathbf{d}_a$ depending on the retardation time $t - R_a/c$; R_a is the distance of atom a from the point of observation at the center of the Lorentz sphere.

The discrete-continuous properties of a dielectric close to its surface are conveniently studied by dividing the near-surface layer into separate sublayers. We will treat a superthin dielectric film as a system of N monolayers separated by a distance a_0, where a_0 is the lattice constant. Let us enclose the observation point $\mathbf{r} = x, y, z$, which can be either inside or outside the film, by a cylinder with the base radius $\varepsilon \gg a_0$. The axis of the cylinder is taken to pass through the point of observation parallel to the z axis. The atoms within the cylinder are treated as discretely distributed dipoles, and those outside form a set of continuously distributed layers. A change in the position of the observation point relative to the film's framing surfaces in the x', y' plane leads to a displacement of the cylinder as a whole in the same plane. Then the nonlocal equation for the electric field strength $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}) \exp(-\mathrm{i}\omega t)$ of frequency ω is written as the following system of N equations [23]:

$$\mathbf{E}_l(x, y) = \mathbf{E}_{Il}(x, y)$$
$$+ \frac{\alpha}{a_0^2} \sum_{j=1}^{N} \int_{\sigma}^{\Gamma_\infty} \left[\mathrm{rot\,rot}\, \mathbf{E}_j(x', y') G(R_j) \right]\big|_{z=z_l} \mathrm{d}s'$$
$$+ \alpha \sum_{j=1}^{N} \sum_{a} \left[\mathrm{rot\,rot}\, \mathbf{E}_j(x_a, y_a) G(R_{aj}) \right]\big|_{z=z_l},$$
$$l = 1, 2, \ldots, N, \qquad (6.4)$$

where the external field within the lth monolayer is given by

$$\mathbf{E}_{Il}(x, y) = \mathbf{A}_I(x, y, z), \quad \mathbf{E}_l(x, y) \equiv \mathbf{E}(x, y, z_l),$$
$$\mathbf{E}_I(\mathbf{r}, t) = \mathbf{A}_I(\mathbf{r}) \exp(-\mathrm{i}\omega t), \qquad (6.5)$$

α is the linear polarizability of the atoms of the medium, $G(R) = \exp(\mathrm{i}k_0 R)/R$, $k_0 = \omega/c$, and

$$R_j = [(x - x_j')^2 + (y - y_j')^2 + (z - z_j')^2]^{1/2},$$
$$R_{aj} = [(x - x_a')^2 + (y - y_a')^2 + (z - z_j')^2]^{1/2}.$$

The integral in Eqn (6.4) is over the x', y' plane except for the sphere σ of radius ε centered at the point (x, y, z_l), and the differentiation is carried out with respect to the coordinates of the observation point (x, y, z_l).

Now consider a plane wave incident on the film under study at an angle θ_I. Then

$$\mathbf{A}_I(\mathbf{r}) = \mathbf{E}_{0I} \exp(\mathrm{i}k_0 \mathbf{s}_I \mathbf{r}), \quad \mathbf{s}_I = (- \sin \theta_I, 0, - \cos \theta_I). \quad (6.6)$$

Further, because of the uniformity of the film surface, the solution of Eqn (6.4) can be written as

$$\mathbf{E}_l(x, y) = \mathbf{E}_l \exp(-\mathrm{i}k_0 \sin \theta_I x). \qquad (6.7)$$

Upon making some manipulations and noting that $z_l = -a_0(l - 1)$, Eqn (6.4) becomes

$$\mathbf{E}_l = \mathbf{E}_{Il} - \mathrm{i}2\pi C \frac{k_0 a_0}{\cos \theta_I} \sum_{j=1}^{l-1} \left[\mathbf{s}_I \times (\mathbf{s}_I \times \mathbf{E}_j) \right]$$
$$\times \exp \left[\mathrm{i}k_0 a_0 \cos \theta_I (l - j) \right] - \mathrm{i}2\pi C \frac{k_0 a_0}{\cos \theta_I}$$
$$\times \sum_{j=l+1}^{N} \left[\mathbf{s}_R \times (\mathbf{s}_R \times \mathbf{E}_j) \right] \exp \left[\mathrm{i}k_0 a_0 \cos \theta_I (j - l) \right]$$
$$+ C \sum_{j=1}^{N} \widehat{\Pi} \left(a_0(j - l) \right) \mathbf{E}_j, \qquad (6.8)$$

where $C = \alpha/a_0^3$, the tensor $\widehat{\Pi}$ is defined in the appendix (part A), and

$$\mathbf{s}_R = (- \sin \theta_I, 0, \cos \theta_I). \qquad (6.9)$$

To transform from Eqn (6.8) for a discretely-continuous dielectric medium into the corresponding wave propagation equation for a continuous medium, it is necessary to take the limit $a_0 \to 0$, thus obtaining

$$\mathbf{E}_l(z) = \mathbf{E}_{Il} - \mathrm{i}2\pi C \frac{k_0 a_0}{\cos \theta_I} \sum_{j=1}^{l-1} \left[\mathbf{s}_I \times (\mathbf{s}_I \times \mathbf{E}_j) \right]$$
$$\times \exp \left[\mathrm{i}k_0 a_0 \cos \theta_I (l - j) \right] - \mathrm{i}2\pi C \frac{k_0 a_0}{\cos \theta_I}$$
$$\times \sum_{j=l+1}^{N} \left[\mathbf{s}_R \times (\mathbf{s}_R \times \mathbf{E}_j) \right] \exp \left[\mathrm{i}k_0 a_0 \cos \theta_I (j - l) \right]$$
$$+ \pi C \frac{a_0}{\varepsilon} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix} \mathbf{E}_l. \qquad (6.10)$$

Upon converting the sum to an integral by replacing $\mathbf{E}_j$ by $\mathbf{E}(z)$, this becomes

$$\left[1 - \pi C \frac{a_0}{\varepsilon} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix} \right] \mathbf{E}(z) = \mathbf{A}_I(z)$$
$$- \mathrm{i} \frac{2\pi k_0}{\cos \theta_I} \int_{-h}^{z} \left[\mathbf{s}_R \times (\mathbf{s}_R \times \mathbf{P}(z')) \right]$$
$$\times \exp \left[\mathrm{i}k_0 \cos \theta_I (z - z') \right] \mathrm{d}z' - \mathrm{i} \frac{2\pi k_0}{\cos \theta_I}$$
$$\times \int_{z}^{0} \left[\mathbf{s}_I \times (\mathbf{s}_I \times \mathbf{P}(z')) \right] \exp \left[\mathrm{i}k_0 \cos \theta_I (z' - z) \right] \mathrm{d}z', \qquad (6.11)$$

where $\mathbf{P}(z) = C\mathbf{E}(z)$ is the polarization vector of the medium at a depth z.

Now we turn to the case of an s-polarized wave in a medium. Eqn (6.11) takes the form

$$\left[1 - \pi C \frac{a_0}{\varepsilon} \right] E^y(z) = A_I^y(z) + \mathrm{i} \frac{2\pi k_0}{\cos \theta_I} \int_{-h}^{0} P^y(z')$$
$$\times \exp \left[\mathrm{i}k_0 \cos \theta_I |z - z'| \right] \mathrm{d}z'. \qquad (6.12)$$

Noting that $\varepsilon = (3/4)a_0$ and applying the Lorentz – Lorentz formula

$$n^2 = \frac{1 + (8\pi/3)C}{1 - (4\pi/3)C} \qquad (6.13)$$

for the refractive index n of the medium, we find that

$$1 - \pi C \frac{a_0}{\varepsilon} = \frac{3}{n^2 + 2}, \qquad (6.14)$$

showing that the right-hand side of Eqn (6.12) contains the y-component of the macroscopic field in the medium. For an external wave incident normal to the boundary, Eqn (6.12) yields

$$\left[1 - \frac{4\pi}{3} C \right] E^y(z) = A_I^y(z)$$
$$+ \mathrm{i}2\pi k_0 \int_{-h}^{0} P^y(z') \exp \left[\mathrm{i}k_0 |z - z'| \right] \mathrm{d}z'. \qquad (6.15)$$

This equation, combined with the Bloch equations [24] that include the local field effect [25, 26], is used to describe Dicke's

optical super-radiation in superthin films of resonant atoms [27] and is also useful in solving a number of boundary value problems of nonlinear resonance optics for semi-infinite dielectric media [28 – 30]

6.2 Equations for the propagation of electromagnetic fields in quadrupole and magnetodipole media

In order to include higher-order multipole moments of the atoms making up the medium, higher powers of electronic displacements relative to atomic nuclei must be retained when expanding the retarded potentials that determine the Lagrangian of a system of moving interacting charges [8]. Here we shall follow Refs [31 – 33] when writing down nonlocal equations for microscopic fields with allowance for the quadrupole and magnetodipole contributions to the optical properties of the medium. Let $\mathbf{E}'$ and $\mathbf{H}'$ denote the strengths of the microscopic electrical and magnetic fields, respectively, at a certain point of observation $\mathbf{r}_l$ and let $\mathbf{m}(\mathbf{r}_j)$ and $\hat{q}(\mathbf{r}_j)$ be the magnetic dipole moment and the quadrupole moment tensor, respectively, of the jth atom. Then the equations for the fields take the form

$$\mathbf{E}'(\mathbf{r}_l) = \mathbf{E}_I(\mathbf{r}_l) + \sum_{j \neq l} \left[\nabla \times \nabla \times \mathbf{d}(\mathbf{r}_j) G(R_{jl}) \right.$$
$$\left. - \nabla \times \nabla \times \nabla \hat{q}(\mathbf{r}_j) G(R_{jl}) + ik_0 \nabla \times \mathbf{m}(\mathbf{r}_j) G(R_{jl}) \right], \quad (6.16)$$

$$\mathbf{H}'(\mathbf{r}_l) = \mathbf{H}_I(\mathbf{r}_l) + \sum_{j \neq l} \left[\nabla \times \nabla \times \mathbf{m}(\mathbf{r}_j) G(R_{jl}) \right.$$
$$\left. + ik_0 \nabla \times \nabla \times \nabla \hat{q}(\mathbf{r}_j) G(R_{jl}) - ik_0 \nabla \times \mathbf{d}(\mathbf{r}_j) G(R_{jl}) \right], \quad (6.17)$$

where

$$G(R_{jl}) = \frac{\exp(ik_0 R_{jl})}{R_{jl}}, \quad (6.18)$$

$R_{jl} = |\mathbf{r}_j - \mathbf{r}_l|$, and $\mathbf{E}_I(\mathbf{r})$ and $\mathbf{H}_I(\mathbf{r}_l)$ are the electric and magnetic field strengths of the external wave at the observation point, respectively.

A transition to the corresponding integral equations for the propagation of electromagnetic waves is performed [33] using the quantities

$$\widehat{Q} = \frac{N}{V} \hat{q}, \quad \mathbf{M} = \frac{N}{V} \mathbf{m}, \quad (6.19)$$

where N/V is the concentration of atoms in the optical medium.

6.3 Equations for the propagation of electromagnetic waves in a conducting medium

We will treat a conducting medium as a system of moving interacting charges, carrying out calculations to order v^2/c^2, where v is the velocity of the charge and c is the speed of light in vacuum. (In this approximation one can write down the Lagrangian function for an individual charge at a certain point of observation $\mathbf{r}$ at the time t [4, 8].) The scalar (φ_c) and vector ($\mathbf{A}_c$) potentials of the field produced by the moving charges then take the respective forms

$$\varphi_c(\mathbf{r}, t) = \sum_{j=1}^{N} \frac{e}{|\mathbf{r} - \mathbf{r}_j|},$$
$$\mathbf{A}_c(\mathbf{r}, t) = \sum_{j=1}^{N} \frac{e}{2c} \frac{1}{|\mathbf{r} - \mathbf{r}_j|} \left[\mathbf{v}_j + (\mathbf{v}_j \mathbf{n}_j) \mathbf{n}_j \right], \quad (6.20)$$

where $\mathbf{n}_j = (\mathbf{r} - \mathbf{r}_j)/|\mathbf{r} - \mathbf{r}_j|$, $\mathbf{r}_j$ is the radius vector of the jth electron in the medium, e is the electron charge, $\mathbf{v}_j$ is the velocity of the jth electron, and N is the number of electrons in the medium. Eqns (6.20) are obtained by expanding the retardation potentials in terms of $|\mathbf{r} - \mathbf{r}_j|/c$ under the assumption that the electron density varies slowly on the time scale of the problem [4].

The field created by atomic cores at the point of observation $\mathbf{r}$ will be described using the vectors $\mathbf{r}_{\alpha\beta} = \mathbf{a}_\alpha + \xi_{\alpha\beta}$, where $\mathbf{a}_\alpha$ are the radius vectors of the nuclei of the cores and $\xi_{\alpha\beta}$ are the radius vectors of the electrons relative to their nuclei. Then the scalar (φ_v) and vector ($\mathbf{A}_v$) potentials of the atomic cores take the respective forms [8]

$$\varphi_v(\mathbf{r}, t) = \sum_{\beta=1}^{n_\alpha} \sum_{\alpha=1}^{N_A} \frac{e_\beta}{|\mathbf{r} - \mathbf{r}_{\alpha\beta}|}, \quad (6.21)$$

$$\mathbf{A}_v(\mathbf{r}, t) = \sum_{\beta=1}^{n_\alpha} \sum_{\alpha=1}^{N_A} \frac{e_\beta}{2c} \frac{1}{|\mathbf{r} - \mathbf{r}_{\alpha\beta}|} \left[\mathbf{v}_{\alpha\beta} + \mathbf{n}_{\alpha\beta}(\mathbf{n}_{\alpha\beta}\mathbf{v}_{\alpha\beta}) \right]$$
$$+ \sum_{\beta=1}^{n_\alpha} \sum_{\alpha=1}^{N_A} \frac{e_\beta}{2c} \frac{|\mathbf{r} - \mathbf{a}_\alpha|^2}{|\mathbf{r} - \mathbf{r}_{\alpha\beta}|^2} \left[\mathbf{v}_{\alpha\beta} - 3\mathbf{n}_{\alpha\beta}(\mathbf{n}_{\alpha\beta}\mathbf{v}_{\alpha\beta}) \right]$$
$$- \sum_{\beta=1}^{n_\alpha} \sum_{\alpha=1}^{N_A} \frac{e_\beta |\mathbf{r} - \mathbf{a}_\alpha|}{|\mathbf{r} - \mathbf{r}_{\alpha\beta}|^2} \mathbf{n}_{\alpha\beta},$$

where n_α is the number of electrons in the αth atomic core, N_A is the number of atomic cores, and $\mathbf{n}_{\alpha\beta}$ is the unit vector directed from the point of observation $\mathbf{r}$ to the electron, whose position is described by the radius vector $\mathbf{r}_{\alpha\beta}$ (the βth electron in the αth atomic core). Eqns (6.21) are also derived from the retarded potentials, but this time they were expanded in terms of $(\mathbf{a}_\alpha/a_\alpha)(\xi_{\alpha\beta}/c)$ taking into account that the motion of atomic electrons is faster than the motion of those electrons between the atomic cores.

Substituting Eqns (6.20) and (6.21) into the Lagrangian for an individual charge and calculating it to order v^2/c^2 yields the Lagrangian for the entire system of charges, valid for an arbitrary multipolarity of the field of the atomic cores. Let us adopt the electric dipole approximation for the field of atomic cores by expanding the functions $|\mathbf{r} - \mathbf{r}_{\alpha\beta}|$ and $|\mathbf{r} - \mathbf{r}_{\alpha\beta}|^{-1}$ into power series, retaining only the terms linear in $\xi_{\alpha\beta}$ and noting that $\xi_{\alpha\beta} \ll |\mathbf{r} - \mathbf{a}_\alpha|$, i.e., that points of observation are far away from the atomic cores. After some manipulation, the scalar potential of the αth atom is found to be

$$\varphi_{v\alpha}(\mathbf{r}, t) = \frac{1}{|\mathbf{r} - \mathbf{a}_\alpha|} N_\alpha|e| + \frac{\mathbf{r} - \mathbf{a}_\alpha}{|\mathbf{r} - \mathbf{a}_\alpha|^3} \mathbf{d}_\alpha, \quad (6.22)$$

where N_α is the number of electrons given away by the αth atom, and $\mathbf{d}_\alpha = \sum e_\beta \xi_{\alpha\beta}$ is the electric dipole of the αth atom.

We proceed by transforming the vector potential $\mathbf{A}_{v\alpha}$ in the electric dipole approximation. After some manipulation we arrive at

$$\mathbf{A}_{v\alpha} = \frac{1}{c} \frac{\mathbf{d}_\alpha}{|\mathbf{r} - \mathbf{a}_\alpha|} - \frac{1}{c} \frac{(\mathbf{r} - \mathbf{a}_\alpha)\big((\mathbf{r} - \mathbf{a}_\alpha)\dot{\mathbf{d}}_\alpha\big)}{|\mathbf{r} - \mathbf{a}_\alpha|^3}$$
$$- \frac{\mathbf{r} - \mathbf{a}_\alpha}{|\mathbf{r} - \mathbf{a}_\alpha|^2} N_\alpha|e| - \frac{2(\mathbf{r} - \mathbf{a}_\alpha)\big((\mathbf{r} - \mathbf{a}_\alpha)\mathbf{d}_\alpha\big)}{|\mathbf{r} - \mathbf{a}_\alpha|^4} + \frac{\mathbf{d}_\alpha}{|\mathbf{r} - \mathbf{a}_\alpha|^2}.$$
$$(6.23)$$

Let us now invoke the condition of charge neutrality in the form

$$\sum_{\alpha=1}^{N_A} \frac{N_\alpha |e|}{|\mathbf{r} - \mathbf{a}_\alpha|} - \sum_{j=1}^{N} \frac{|e|}{|\mathbf{r} - \mathbf{r}_j|} = 0 . \tag{6.23a}$$

Then, using Eqs (6.22) and (6.23), differentiating (6.22) with respect to the coordinates of the observation point and taking the vectors $\mathbf{a}_\alpha$ to be time-independent, we find the strength of the electric field of the αth atomic core to be

$$\mathbf{E}_{v\alpha} = \mathrm{rot\,rot}\, \frac{[\mathbf{d}_\alpha]}{R_\alpha} , \tag{6.24}$$

where $R_\alpha = |\mathbf{r} - \mathbf{a}_\alpha|$ and the notation $[\ldots]$ indicates that the variable is taken at the retardation time $t - R_\alpha/c$.

In a similar way, the strength of the magnetic field of the αth atomic core is given by

$$\mathbf{H}_{v\alpha} = \mathrm{rot}\, \mathbf{A}_{v\alpha} = \left\{ \frac{1}{cR_\alpha^3} [\dot{\mathbf{d}}_\alpha] + \frac{1}{c^2 R_\alpha^2} [\ddot{\mathbf{d}}_\alpha] \right\} [\mathbf{u}_\alpha \times \mathbf{R}_\alpha] , \tag{6.25}$$

where $\mathbf{u}_\alpha$ is the unit vector along the electric dipole moment $\mathbf{d}_\alpha$.

We next apply formulas (6.20) to calculating the electric field $\mathbf{E}_c(\mathbf{r}, t)$ and magnetic field $\mathbf{H}_c(\mathbf{r}, t)$ produced by the conduction electrons at the point of observation $\mathbf{r}$ at the time t. With the use of the system's electrical neutrality property (6.23a), we obtain

$$\mathbf{E}_{cj}(\mathbf{r}, t) = -\frac{1}{c}\dot{\mathbf{A}}_{cj} = -\frac{e}{2c^2}\left[\mathbf{v}_j + (\mathbf{v}_j\, \mathbf{n}_j)\right]\frac{\partial}{\partial t}\left(\frac{1}{|\mathbf{r} - \mathbf{r}_j|}\right)$$
$$-\frac{e}{2c^2}\frac{1}{|\mathbf{r} - \mathbf{r}_j|}\left[\dot{\mathbf{v}}_j + (\dot{\mathbf{v}}_j\, \mathbf{n}_j)\mathbf{n}_j + (\mathbf{v}_j\, \dot{\mathbf{n}}_j)\mathbf{n}_j + (\mathbf{v}_j\, \mathbf{n}_j)\dot{\mathbf{n}}_j\right] . \tag{6.26}$$

Let us calculate the time derivatives

$$\frac{\partial}{\partial t}\frac{1}{|\mathbf{r} - \mathbf{r}_j|} = -\frac{1}{R_j^2}\dot{R}_j , \quad \dot{\mathbf{n}}_j = \frac{\dot{\mathbf{R}}_j}{R_j} - \frac{\mathbf{R}_j \dot{R}_j}{R_j^2} ,$$

where $R_j = |\mathbf{r} - \mathbf{r}_j|$. The derivative $\dot{\mathbf{R}}_j$ at a given observation point is the velocity $\mathbf{v}_j$ of the jth charge. The derivative $\dot{R}_j$ is obtained by differentiating the identity $R_j^2 = \mathbf{R}_j^2$. Eqn (6.26) then becomes

$$\mathbf{E}_{cj} = -3\frac{e}{2c^2}\frac{1}{R_j^2}\mathbf{n}_j(\mathbf{n}_j\, \mathbf{v}_j)^2 - \frac{e}{2c^2}\frac{1}{R_j}\dot{\mathbf{v}}_j$$
$$-\frac{e}{2c^2}\frac{1}{R_j}(\dot{\mathbf{v}}_j\, \mathbf{n}_j)\mathbf{n}_j + \frac{e}{2c^2}\frac{\mathbf{v}_j^2}{R_j^2}\mathbf{n}_j . \tag{6.27}$$

The magnetic field strength at the observation point $\mathbf{r}$ is now found from Eqn (6.20) to be

$$\mathbf{H}_{cj} = \mathrm{rot}\, \mathbf{A}_{cj} = \frac{e}{c}\frac{1}{R_j^2}[\mathbf{v}_j \times \mathbf{n}_j] . \tag{6.28}$$

To transform Eqns (6.25), (6.24), (6.27), (6.28) into the integral equations for the propagation of electromagnetic waves in a conducting medium, we make use of the continuity and self-consistency of the internal fields. This yields the following equation for the electric field strength in an optical medium [34]:

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_I(\mathbf{r}, t) + \int \mathrm{rot\,rot}\, \frac{\mathbf{P}_A(\mathbf{r}', t - R/c)}{R}\,\mathrm{d}V'$$
$$+ \int \mathbf{L}_E\left(\mathbf{r}', t - \frac{R}{c}\right)\mathrm{d}V' , \tag{6.29}$$

in which $\mathbf{E}_I(\mathbf{r}, t)$ is the strength of the electrical field of the external wave, and

$$\mathbf{L}_E = \frac{N}{V}\left\{ -\frac{3e}{2c^2}\frac{1}{R^2}(\mathbf{v}\mathbf{n})^2\mathbf{n} - \frac{e}{2c^2}\frac{1}{R}\dot{\mathbf{v}} \right.$$
$$\left. -\frac{e}{2c^2}\frac{1}{R}(\dot{\mathbf{v}}\mathbf{n})\mathbf{n} + \frac{e}{2c^2}\frac{1}{R^2}\mathbf{v}^2\mathbf{n} \right\} , \tag{6.29a}$$

where $\mathbf{n} = \mathbf{R}/R$ is the unit vector directed from the point of observation $\mathbf{r}$ to a certain point $\mathbf{r}'$ in the medium.

The integral equation for the magnetic field strength is obtained in a similar fashion to be [34]

$$\mathbf{H}(\mathbf{r}, t) = \mathbf{H}_I(\mathbf{r}, t) + \frac{1}{c}\int \mathrm{rot}\, \frac{\mathbf{P}_A(\mathbf{r}', t - R/c)}{R}\,\mathrm{d}V'$$
$$+ \frac{e}{c}\frac{N}{V}\int \frac{1}{R^2}[\mathbf{v} \times \mathbf{n}]\,\mathrm{d}V' , \tag{6.30}$$

where $\mathbf{H}_I$ is the strength of the magnetic field of the external wave.

If the point of observation $\mathbf{r}$ is outside the optical medium, the integration in Eqns (6.29) and (6.30) is performed over the entire medium. If the point of observation is within the medium, it is first necessary to exclude a small region bounded by a sphere σ_0 of small radius a in order to eliminate the divergence that arises as $R \to 0$.

Equations (6.29) and (6.30) are integro-differential equations for microscopic fields. By solving them it is possible to determine the microscopic field at various observation points within and outside the medium, using the appropriate constitutive equations to specify the field dependences of the vectors $\mathbf{P}_A$ and $\mathbf{v}$.

In Ref. [34], Eqn (6.29) was employed to explain the significant discrepancies between the theoretical and experimental behavior of the optical constants of silver over a wide range of wavelengths [18]. The vectors $\mathbf{P}_A$ and $\mathbf{v}$ in Eqn (6.29) were assumed to be linear in $\mathbf{E}$, and the nonlinear terms in function (6.29a) were dropped. Also, the atomic cores were treated as two-level quantum mechanical systems with oscillator strength f and lifetime Γ^{-1}, and the conduction electrons were considered to be classical particles with polarizability α_c and damping coefficient β. The results of Ref. [34] included a formula for the complex refractive index of a metal, a proof of the generalized Ewald–Oseen's extinction theorem for metals, a formula for the amplitudes of the reflected and refracted waves, and also values of f, Γ^{-1} and β, the latter being calculated from the formula $\beta = 4\pi\omega_\mathrm{p}^2\rho$, with ω_p the plasma frequency and ρ the electrical resistivity. Figures 2 and 3 show both the theoretical and experimental results as presented in Ref. [18] and the results obtained using the theory of Ref. [34].

Thus, by using the quantum electrodynamical and semiclassical methods, the quantized field Eqns (5.11) and (5.18) (allowing for the spin and orbital degrees of freedom of interacting atoms) and the classical field Eqns (6.1)–(6.4), (6.15), (6.19), (6.18), (6.29), and (6.30) have been derived. These equations are nonlocal in both space and time due to
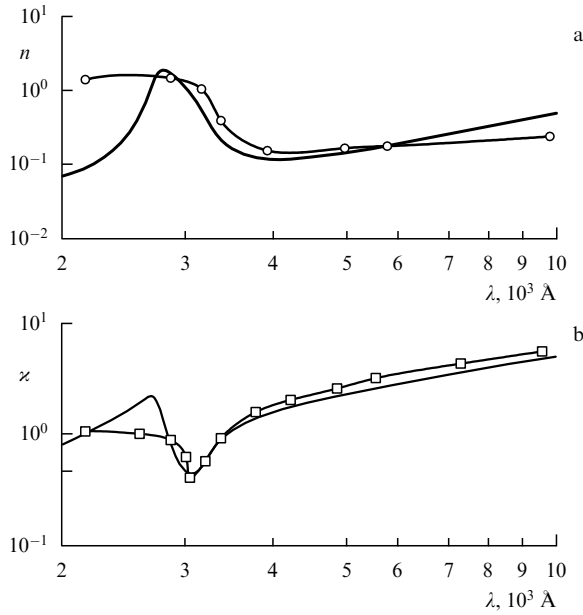
Figure 2. Measured (symbols) and predicted wavelength dependences of (a) the refractive index and (b) the absorption coefficient of metallic silver.
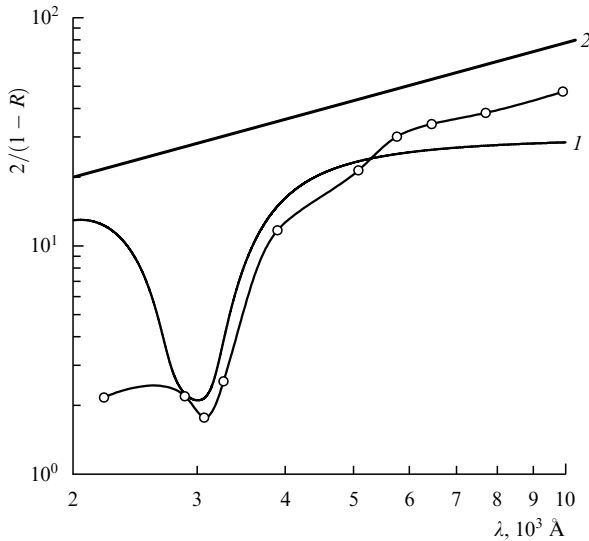

Figure 3. Measured (symbols) and predicted wavelength dependences of the reflection power of the vacuum-silver interface: *1*, present theory; *2*, theory of Ref. [18]; $f = 0.05$, $\Gamma = 10^{15}\,\text{s}^{-1}$, $\omega_0 = 7.95 \times 10^{15}\,\text{s}^{-1}$, $\beta = 4.17 \times 10^{15}\,\text{s}^{-1}$, $(N_A/V) = 5.86 \times 10^{22}\,\text{cm}^{-3}$, $N/V = 0.4(N_A/V)$

the fact that the field at a certain point of observation is coupled to the behavior of charges at other points in the medium. Below we will present solutions to a number of typical problems of classical and nonlinear optics, in which the nonlocality property of the equations of electrodynamics plays a crucial role.

7. Spontaneous radiation of an atom close to a vacuum-dielectric interface. The near-field effect

Let us consider the spontaneous radiation from a two-level atom as observed in the near-field at a distance $L \ll \lambda_0$ ($\lambda_0 = 2\pi c/\omega$, ω the frequency of the emitted photons) from

the surface Σ of a dielectric with polarizability α, concentration (N/V) and refractive index n_0. The solution of this boundary value problem of quantum optics was considered in Refs [35–42]. Let us have a look at the basic results of these papers. Ref. [35] presents the following expression for the amplitude of a quantum transition of an atom:

$$b(t) = \exp\left\{-K_0\left[1 + R\exp(2ik_0L)\right]t\right\}, \qquad (7.1)$$

where $K_0 = \pi d\omega/(\hbar Ac)$; A is the effective area of the surface; $k_0 = \omega/c$; and $R = -(n_0 - 1)/(n_0 + 1)$ is the reflection coefficient as given by the Fresnel formula for normally incident light.

From Eqn (7.1), one calculates the lifetime of the excited state of an atom

$$\left\{K_0[1 + R\cos(2k_0L)]\right\}^{-1},$$

as well as the frequency shift of an atom

$$K_0R\sin(2k_0L).$$

One further contribution of Ref. [35] is the derivation of the quantum analogue of the Ewald–Oseen extinction theorem well known in the classical optics of dielectrics [18]. Note that a dielectric was treated as a continuous optical medium in this derivation.

In solving the boundary value problem stated above we will take into account the discrete-continuous nature of the dielectric. We will assume that the point of observation $\mathbf{r}_0$ (Fig. 4) is embraced by a Lorentz sphere of radius L_0 and that the atoms of the dielectric are distributed in a discrete fashion within the sphere. Outside the sphere, it will be assumed that the atoms are distributed continuously, with the number of discretely distributed atoms varying with the location of the point of observation. As will be shown below, the presence of two spatial scales λ_0 and L_0 in the description of the spontaneous decay of an atom leads to the near-field effect. Within the closed Lorentz sphere, the field produced at the center of the sphere by the discrete distribution of atoms is zero whatever the symmetry of the distribution, as long as we consider only the Coulomb field — which varies as R_a^{-3}, where R_a is the distance from the ath atom inside the Lorentz sphere to its center. It can be shown by directly calculating the polarizing fields inside the Lorentz sphere that the retardation field proportional to $1/R_a$ is always nonzero at the center of the sphere, independently of the precise symmetry of the discretely distributed atoms. However, the near-field effect should be strongest when the Lorentz sphere is truncated, which occurs when the point of observation approaches the
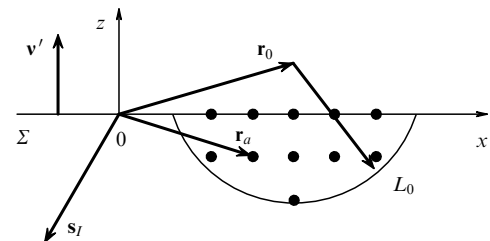

Figure 4. Vector scheme for the boundary value problem: Σ, the surface $z = 0$; $\mathbf{r}_0$, the position vector of the two-level atom; $\mathbf{s}_I$, unit vector along the direction in which a photon is emitted; L_0, radius of the Lorentz sphere; $\mathbf{v}'$, outer normal to the surface Σ.

surface Σ of the optical medium. It was this fact that was taken into account in the solution of Ref. [42].

We now apply the electronic polarizing field operator $\mathbf{A}^{(e)}$ (5.10) and write the interaction operator for a two-level atom near the surface in the form

$$H_1 = 2\frac{\omega_0}{c}\sum_{\mathbf{k}\lambda} \mathbf{d}_{\mathbf{k}\lambda}^{\text{eff}}\mathbf{A}_{\mathbf{k}\lambda}\, r_2 + \text{h.c.} \qquad (7.2)$$

Here ω_0 is the resonant frequency of the atom at position $\mathbf{r}_0$; the index $\mathbf{k}\lambda$ is for the photon mode with wave vector $\mathbf{k}$ and polarization $\lambda = 1, 2$; r_2 is the effective spin operator for a two-level atom, $r_i = (1/2)\sigma_i$ ($i = 1, 2, 3$), σ_i being the Pauli matrix; $\mathbf{A}_{\mathbf{k}\lambda}$ is the vector potential operator for the photon field at the location of the atom; and $\mathbf{d}_{\mathbf{k}\lambda}^{\text{eff}}$ is the effective dipole moment of the atom which is given by [42]

$$\mathbf{d}_{\mathbf{k}\lambda}^{\text{eff}} = d_0\mathbf{u}_d\left\{\frac{(N/V)\alpha}{(n_0^2-1)k_0^2}\,\text{rot rot}\,\mathbf{e}_{\mathbf{k}\lambda}(I_\Sigma - I_{\Sigma_0} + I_{\Sigma_a})\right.$$
$$\left. + \mathbf{S} + \mathbf{e}_{\mathbf{k}\lambda}\exp(\mathrm{i}\mathbf{k}\mathbf{r}_0)\right\}, \qquad (7.3)$$

where $\mathbf{u}_d$ is the unit vector along the atom's dipole moment, d_0 is the transition dipole moment of a resonant atom, $\mathbf{e}_{\mathbf{k}\lambda}$ is the unit polarization vector of a spontaneous photon of mode $\mathbf{k}\lambda$,

$$I_\Sigma = -2\pi\exp(\mathrm{i}k_0\mathbf{s}_R\mathbf{r})\,\frac{\sin(\theta_I - \theta_T)}{\sin\theta_T\cos\theta_T}\,, \qquad (7.4)$$

θ_I is the angle of incidence of a spontaneous photon at the surface Σ,

$$\mathbf{s}_R = (-\sin\theta_I,\, 0,\, \cos\theta_I) \qquad (7.5)$$

is the unit vector, and θ_T is the refraction angle of spontaneous photons ($n_0\sin\theta_T = \sin\theta_I$ because of the uniformity of the surface Σ). The quantity I_{Σ_0} is the surface integral over the circle formed by the intersection of the Lorentz sphere and the surface Σ; the quantity I_{Σ_d} is the surface integral over that portion of the Lorentz sphere which is inside the medium;

$$\mathbf{S} = \alpha\sum_a\left[\mathrm{i}(\mathbf{k}\mathbf{r}_a + k_0 R_a)\right]\left[\mathbf{e}_{\mathbf{k}\lambda}\left(\frac{\mathrm{i}k_0}{R_a^2} + \frac{k_0^2}{R_a}\right) - k_0^2\, n_a\frac{(\mathbf{e}_{\mathbf{k}\lambda}\mathbf{n}_a)}{R_a}\right.$$
$$\left. - 3\mathrm{i}k_0\mathbf{n}_a\,\frac{(\mathbf{e}_{\mathbf{k}\lambda}\,\mathbf{n}_a)}{R_a^2} + \frac{3\mathbf{n}_a(\mathbf{e}_{\mathbf{k}\lambda}\,\mathbf{n}_a) - \mathbf{e}_{\mathbf{k}\lambda}}{R_a^3}\right], \qquad (7.6)$$

$\mathbf{R}_a = \mathbf{r}_0 - \mathbf{r}_a$, $\mathbf{n}_a = \mathbf{R}_a/R_a$, and $\mathbf{r}_a$ is the radius vector of the ath atom inside the Lorentz sphere relative to the origin of coordinates (see Fig. 4).

Numerical estimates show that the dominant contribution to the effective dipole moment (7.3) comes from the discrete distribution portion of the quantity $\mathbf{S}$, i.e., from the last term in Eqn (7.6), which varies as $1/R_a^3$ and accounts for the Coulomb interaction of resonant atoms.

If the Hamiltonian (7.2) is used, the Heisenberg equations of motion for photon operators are [42]

$$c_{\mathbf{k}\lambda}(\mathbf{r}_0, t) = c_{\mathbf{k}\lambda}^v(\mathbf{r}_0, t) - \frac{\mathrm{i}}{\hbar}2\frac{\omega_0}{c}(d_{\mathbf{k}\lambda}^{\text{eff}})^*g_{\mathbf{k}\lambda}$$
$$\times \int r_2(t',\mathbf{r}_0)G_{\mathbf{k}\lambda}(t-t')\,\mathrm{d}t', \qquad (7.7)$$

where $g_{\mathbf{k}\lambda} = [2\pi\hbar c^2/(V_R\,\omega_{\mathbf{k}})]^{1/2}$; V_R is the quantization volume of the field; $\omega_{\mathbf{k}}$ is the frequency of a photon of mode $\mathbf{k}\lambda$;

$$G_{\mathbf{k}\lambda}(t-t') = \begin{cases} 0, & t < t', \\ \lim_{\varepsilon\to 0}\exp\{-\mathrm{i}(\omega_k - \varepsilon\mathrm{i})(t-t')\}, & t > t', \end{cases} \qquad (7.8)$$

is the retardation Green function [24]; and the photon operator

$$c_{\mathbf{k}\lambda}^v(\mathbf{r}_0, t) = c_{\mathbf{k}\lambda}(0)\exp\{\mathrm{i}(\mathbf{k}\mathbf{r}_0 - \omega_k t)\} \qquad (7.9)$$

corresponds to the free (vacuum) photon field.

Eqn (7.7) must be supplemented by the following equations for the atomic operators [24]:

$$\dot{r}_1 = -\omega_0 r_2 + 2\frac{\omega_0}{\hbar c}\,d^{\text{eff}}Ar_3\,,$$
$$\dot{r}_2 = \omega_0 r_1\,,$$
$$\dot{r}_3 = -2\frac{\omega_0}{\hbar c}\,d^{\text{eff}}Ar_1\,, \qquad (7.10)$$

where

$$d^{\text{eff}}A = \sum_{\mathbf{k}\lambda}\mathbf{d}_{\mathbf{k}\lambda}^{\text{eff}}\mathbf{A}_{\mathbf{k}\lambda}\,, \qquad (7.10a)$$

and the quantity $\mathbf{d}_{\mathbf{k}\lambda}^{\text{eff}}$ depends on the position of the atom relative to the surface Σ, according to Eqn (7.3).

Using Eqns (7.10) and (7.7) within the framework of the adiabatic approximation [24] and applying the normal ordering of vacuum-averaged operators, we obtain the following expression for the lifetime τ of the excited state of an atom near the surface [42]:

$$\frac{1}{\tau} = 2\pi\left(\frac{\omega_0}{\hbar c}\right)^2\sum_{\mathbf{k}\lambda}|d_{\mathbf{k}\lambda}^{\text{eff}}|^2 g_{\mathbf{k}\lambda}^2[\delta(\omega_{\mathbf{k}} - \omega_0) - \delta(\omega_{\mathbf{k}} + \omega_0)]\,, \qquad (7.11)$$

where $\delta(x)$ is the Dirac delta function.

The energy shift of an atom near the surface is given by [42]

$$\delta = \left(\frac{\omega_0}{\hbar c}\right)\sum_{\mathbf{k}\lambda}\left(\frac{\mathcal{P}}{\omega_{\mathbf{k}} + \omega_0} - \frac{\mathcal{P}}{\omega_{\mathbf{k}} - \omega_0}\right)g_{\mathbf{k}\lambda}^2|d_{\mathbf{k}\lambda}^{\text{eff}}|^2\,, \quad (7.12)$$

with $\mathcal{P}/x$ denoting principal-value integration.

In the wave zone with respect to the surface Σ ($L \gg \lambda_0$), the effective dipole moment takes the form

$$d_{\mathbf{k}\lambda}^{\text{eff}} = d_0\mathbf{u}_d\left\{\frac{3}{2(n_0^2+2)}\frac{\sin(\theta_T - \theta_I)}{\sin\theta_T\cos\theta_I}\left[\mathbf{e}_{\mathbf{k}\lambda} - \mathbf{s}_R(\mathbf{s}_R\,\mathbf{e}_{\mathbf{k}\lambda})\right]\right.$$
$$\left. \times \exp(\mathrm{i}k_0\mathbf{s}_R\mathbf{r}_0) + \mathbf{e}_{\mathbf{k}\lambda}\exp(\mathrm{i}k_0 r_0)\right\}. \qquad (7.13)$$

instead of Eqn (7.3).

Using this relation, we can show that also in the case of normal incidence ($\theta_I = 0$) the expression for the lifetime of an excited atom agrees with the corresponding results of Ref. [35]. Numerical analysis shows that, due to the near-field effect, i.e., due to taking into account the structural factor of the dielectric near its surface, the near-field lifetime of an atom shows variations within 30% of its free space value.

Application of the nonlocal equation (5.11) together with equations (7.10) to the solution of the boundary-value problem of the spontaneous emission from an atom near a discrete-continuous dielectric makes it possible to describe the field of spontaneous photons not only at point $\mathbf{r}_0$ but also at any other observation point. This can be achieved [19, 42] by applying the quantum analogue of the Ewald–Oseen extinction theorem, which — unlike in Ref. [35] — enables one to solve the three-dimensional boundary value problem. The numerical analysis of the solution so obtained involves calculating certain surface integrals which also arise in boundary value problems for classical fields. Below we will consider some problems of classical and nonlinear optics, perform a detailed numerical analysis of their solutions, and apply the generalized Ewald–Oseen procedure.

8. Relation between the microscopic and macroscopic fields inside and at the surface of a discrete-continuous dielectric

The Fresnel formulas of the classical optics of dielectrics [18] can be derived with the help of the integral equation (6.1) using the Ewalds–Oseen procedure. The advantages of this approach compared to the traditional use of Maxwell's boundary conditions [18] are the rigorous derivation of the Lorentz–Lorentz formula and the proof of the extinction theorem, according to which the reflection and refraction of light occur at the infinitely thin — mathematical — boundary of the dielectric. Eqn (6.1) assumes that the dielectric is a continuous optical medium with a uniform concentration N/V of atoms of one species, all having the electron polarizability α. The concept of a continuous dielectric is based on the assumption that the field produced by discretely distributed dipoles within the Lorentz sphere [43] is zero for any type of symmetry including a chaotic distribution [18, 44]. An exception are dielectrics which consist of atoms of different species and have a mixed-type symmetry — as, for example, perovskite crystals [43].

Consider a dielectric made up of atoms of one species which have a certain type of symmetry in their distribution within the dielectric and on its surface. Assume that the point of observation is inside the dielectric and that the respective Lorentz sphere is closed (Fig. 5a). A direct calculation shows that the field created by the dipoles situated within the Lorentz sphere is zero at the center of the sphere for any type of symmetry — including a chaotic one — if, together with the Coulomb field of the dipoles, its retardation part, proportional to $1/R_a$, is also taken into account. The size of the Lorentz sphere is, however, much less than the light wavelength λ, i.e., $L_0 \ll \lambda$; therefore, the retardation part of the dipole field plays only a minor role. For example, as will be shown below, in the expression for the refractive index at observation points far away from the surface, the structural factor due to the retardation of the dipole field contributes about 1% of the refractive index value obtained within the conventional continuous dielectric concept. This is in stark contrast to the situation where the point of observation is in the near-field with respect to the surface of the dielectric (Fig. 5b, c). In such cases, the Lorentz sphere is truncated, the field produced by the dipoles at its center is zero whatever the symmetry of the dielectric, and the dipole field is dominated by its Coulomb part, proportional to $1/R_a^3$. Thus we see that in the optics of dielectrics the more general concept of a discrete-continuous dielectric should be employed.
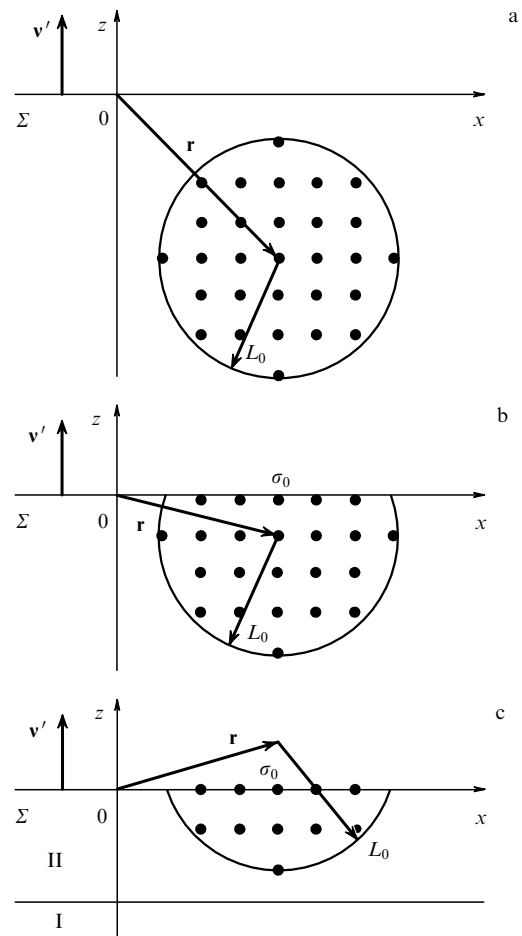

Figure 5. On the concept of a discrete-continuous dielectric: (a) the point of observation situated inside the dielectric in the wave zone with respect to the surface Σ of the dielectric; (b) near-field discrete-continuous dipole distribution for the refracted wave; (c) near-field discrete-continuous dipole distribution for the reflected wave. Regions II and I correspond to the near-field and wave zones of the dielectric, L_0 is the radius of the Lorentz sphere, σ_0 is the area of the circle formed by the intersection of the Lorentz sphere and the surface Σ, and $\mathbf{v}'$ is the normal to the surface Σ.

Let us consider the nonlocal equation [22]

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_I(\mathbf{r}, t) + \int_\sigma^\Sigma \operatorname{rot} \operatorname{rot} \frac{N\alpha\mathbf{E}(\mathbf{r}, t - R/c)}{R} \, \mathrm{d}V'$$
$$+ \sum_a \operatorname{rot} \operatorname{rot} \frac{\alpha\mathbf{E}(\mathbf{r}_a, t - R/c)}{R_a} \, . \quad (8.1)$$

It will be assumed that the dielectric is made up of atoms (molecules) of one species, with an electron polarizability α and concentration N. The symmetry of the dielectric is determined by the lattice sum over the dipoles within the Lorentz sphere. In Eqn (8.1), $\mathbf{E}_I$ is the external light wave; $\mathbf{E}(\mathbf{r}, t)$ is the microscopic field at the point of observation $\mathbf{r}$, which may lie inside or outside the dielectric, either in the wave zone or in the near zone with respect to its surface; and σ is the surface around the point of observation. The field $\mathbf{E}$, as will be shown below, differs from the macroscopic field $\mathbf{E}'$ at various observation points inside the dielectric and on its surface.

The macroscopic field obeys the Maxwell equations, in which the microscopic field is assumed to have been averaged in a certain way [18]. Let us establish the relation between

these two fields using Eqn (8.1) for the microscopic field $\mathbf{E}(\mathbf{r}, t)$. Within the framework of the continuous dielectric model this relation has the familiar form [18]

$$\mathbf{E} = \mathbf{E}' + \frac{4\pi}{3}\,\mathbf{P}\,. \tag{8.2}$$

Consider now the relation between the field $\mathbf{E}$ and $\mathbf{E}'$ within the discrete-continuous model at observation points inside or at the surface of a semi-infinite dielectric (see Fig. 5). Following the method of Ref. [43], we start by writing

$$\mathbf{E} = \mathbf{E}' + \mathbf{E}_{\mathrm{d}} + \mathbf{E}_L\,, \tag{8.3}$$

for optical fields. Here $\mathbf{E}_{\mathrm{d}}$ is the field produced by the dipoles inside the Lorentz sphere and $\mathbf{E}_L$ is the field at the surface of this sphere. Furthermore,

$$\mathbf{E}_{\mathrm{d}} = \mathbf{E}_0 + \mathbf{E}_1 + \mathbf{E}_2\,,$$

$$\mathbf{E}_0 = \frac{1}{N}\sum_a \frac{3\mathbf{n}_a(\mathbf{n}_a\mathbf{P}_a) - \mathbf{P}_a}{R_a^3}\,,$$

$$\mathbf{E}_1 = \frac{1}{N}\sum_a \frac{3\mathbf{n}_a(\mathbf{n}_a\dot{\mathbf{P}}_a) - \dot{\mathbf{P}}_a}{R_a^2 c}\,,$$

$$\mathbf{E}_2 = \frac{1}{N}\sum_a \frac{3\mathbf{n}_a(\mathbf{n}_a\ddot{\mathbf{P}}_a) - \ddot{\mathbf{P}}_a}{R_a c^2}\,, \tag{8.4}$$

where $\quad \mathbf{n}_a = \mathbf{R}_a/R_a, \quad \mathbf{R}_a = \mathbf{r} - \mathbf{r}_a, \quad \mathbf{P}_a = N\alpha\mathbf{E}_a$, $\mathbf{E}_a = \mathbf{E}(\mathbf{r}_a, t - R_a/c)$. Since the radius of the Lorentz sphere is small, i.e., $k_0 R_a \ll 1$, $k_0 = \omega/c$, where ω is the frequency of the optical field, we will assume that

$$\mathbf{E}_a \approx \mathbf{E}(\mathbf{r}_a, t) \approx \mathbf{E}(\mathbf{r}, t) \tag{8.5}$$

at the observation point $\mathbf{r}$ at the center of the Lorentz sphere. Then the index a of the polarization vector may be omitted in Eqn (8.4). Assuming the polarization vector to vary with time as

$$\mathbf{P}(\mathbf{r}, t) = \mathbf{P}(\mathbf{r})\exp(-\mathrm{i}\omega t)\,, \tag{8.6}$$

we have for the closed Lorentz sphere

$$\mathbf{E}_0 = \widehat{\beta}_0\mathbf{P}, \quad \mathbf{E}_1 = \widehat{\beta}_1\mathbf{P}, \quad \mathbf{E}_2 = \widehat{\beta}_2\mathbf{P}, \tag{8.7}$$

where the diagonal tensors $\widehat{\beta}_0$, $\widehat{\beta}_1$ and $\widehat{\beta}_2$ have the form

$$(\widehat{\beta}_0)_{ij} = \delta_{ij}\frac{1}{N}\sum_a \frac{3n_{ai}^2 - 1}{R_a^3}\,, \tag{8.8a}$$

$$(\widehat{\beta}_1)_{ij} = -\mathrm{i}\delta_{ij}\frac{k_0}{N}\sum_a \frac{3n_{ai}^2 - 1}{R_a^2}\,, \tag{8.8b}$$

$$(\widehat{\beta}_2)_{ij} = -\delta_{ij}\frac{k_0^2}{N}\sum_a \frac{n_{ai}^2 - 1}{R_a}\,, \tag{8.8c}$$

$$i, j = x, y, z.$$

By applying numerical analysis, the contributions from the fields $\mathbf{E}_0$, $\mathbf{E}_1$ and $\mathbf{E}_2$ to the dipole fields inside the Lorentz sphere can be examined for various types of symmetry, including chaotic. It turns out that the fields $\mathbf{E}_0$ and $\mathbf{E}_1$ are negligibly small. For a closed Lorentz sphere we have

$$\mathbf{E}_L = \frac{4\pi}{3}\,\mathbf{P} \tag{8.9}$$

using the approximation (8.5), so that in this case we obtain

$$\mathbf{E} = \mathbf{E}' + \left(\frac{4\pi}{3} + \beta_2\right)\mathbf{P}\,, \tag{8.10}$$

where the scalar quantity β_2 corresponds to the diagonal components of the tensor (8.8c) and is the structural factor of the dielectric. Clearly, the role of this structural factor increases for observation points inside the dielectric as the factor k_0^2/N increases — in rarefied optical media, for example.

Now consider the case of a truncated Lorentz sphere for observation points near the surface of the dielectric (Fig. 5b, c). In this case the Lorentz field takes the form

$$\mathbf{E}_L = \frac{2\pi}{3}\left[1 - \left(\frac{z}{L_0}\right)^3\right]\mathbf{P}\,, \tag{8.11}$$

and the components (8.7) of the dipole field inside the Lorentz sphere all differ from zero, the dominant role being played by the field $\mathbf{E}_0$, i.e., $\mathbf{E}_0 \gg \mathbf{E}_1, \mathbf{E}_2$. Then, using Eqn (8.11), we have [22]

$$\mathbf{E} = \mathbf{E}' + \left\{\frac{2\pi}{3}\left[1 - \left(\frac{z}{L_0}\right)^3\right] + \widehat{\beta}_0 + \widehat{\beta}_1 + \widehat{\beta}_2\right\}\mathbf{P}\,. \tag{8.12}$$

The role of the structural factor $\widehat{\beta}_0$ for observation points near the surface of a semi-infinite dielectric may be significant for condensed media.

Using relation (8.12) and two electric induction definitions known in macroscopic electrodynamics [18] it is possible to obtain a formula for the dielectric constant of the near-surface layer of a dielectric as a function of the depth at which the point of observation is located. In Ref. [22], the refractive index for the near-surface layer was obtained using Eqn (8.1) within the context of the microscopic theory. In the present review, all boundary-value problems are solved using the corresponding microscopic nonlocal equations for discrete-continuous dielectrics. Note that, as in the molecular theory of light reflection [44], this solution ignores the interatomic (intermolecular) interaction which affects the energy spectrum of the dielectric. The Lorentz sphere radius playing the role of the nonlocality parameter is determined either numerically or analytically [23]. In this review we will attribute any deviation from the Fresnel laws which is produced by a nonzero dipole field inside the Lorentz sphere to the near-field effect.

9. Microscopic theory of the transition layer on the ideal surface of a semi-infinite dielectric medium. The near-field effect

We consider here the boundary-value problem of the classical optics of dielectrics, with the polarization vector of the dielectric medium being a linear function of the field. Within the framework of the continuous dielectric model, the solution of this problem leads to the Fresnel laws [18]. Let us consider this problem within the discrete-continuous model (see Section 8).

It is a well known fact [45 – 51] that the Fresnel laws show anomalies for light reflection from a mirror surface of a real body. These anomalies manifest themselves most clearly in the vicinity of the complete polarization angle: here the p-component of the reflected wave is never completely extinguished and the light reflected from the surface is elliptically

polarized. These effects are observed experimentally for both clean and contaminated surfaces as well as for those subject to elastic deformations. Drude [52 – 54] explained these anomalies by assuming that at the interface between two media with refractive indices n_1 and n_2 there is a very thin transition layer, in which the refractive index changes smoothly from n_1 to n_2. According to Drude's phenomenological theory [53, 54], the effect of such a layer can be determined by revising Maxwell's boundary conditions, which were obtained on the assumption of a sharp interface. An important contribution of Drude's theory is that, from the intensity and phase of the p-component of the reflected wave for Brewster's angle, it proves to be possible to estimate the thickness of the transition layer. According to Drude, this thickness can be most conveniently estimated by measuring the ellipticity of the light reflected at Brewster's angle provided a wave linearly polarized at an angle of $45°$ to the incidence surface is incident on the reflecting surface. In this case we have the following formula for the reflective indices [54]:

$$\frac{r_{pp}}{r_{ss}} = \frac{\mathrm{i}k_0 h}{2} \frac{\sqrt{n_1^2 + n_2^2}}{n_1^2 - n_2^2} \, \eta \, , \qquad (9.1)$$

where $k_0 = \omega/c$ is the wave number in vacuum; h is the transition layer thickness;

$$\eta = \tilde{n}^2 + n_1^2 n_2^2 \left(\frac{1}{\tilde{n}^2} \right) - n_1^2 - n_2^2 \, ,$$

where a tilde above a symbol indicates an integral average for the quantity in question; and n, a function of the depth z, is the refractive index of the transition layer material.

The phenomenological theory of Drude involves averaged values of the dielectric constant of the medium in a very thin transition layer, but the concept of permittivity has little or no meaning if the layer thickness is comparable with the size of the particles it is made of. Consequently, a microscopic theory of the transition layer has been developed [55 – 57], which is based on the Ewald – Oseen's extinction theorem [18] and takes into account explicitly the non-uniform components of the wave field inside a cubic lattice as well as the fact that the distance for which lower-lying layers cease to contribute to the near-surface field is finite. In these papers it was shown, in particular, that if one takes into account the discrete nature of the medium then even in the absence of transition layers the reflection of light occurs as if the lattice were a continuous medium and as if there were a continuous transition layer on its surface.

A microscopic theory of the transition layer on the surface of a semi-infinite absorbing or non-absorbing dielectric is given in Ref. [58], which, unlike the papers mentioned above, makes no assumptions concerning the thickness of the transition layer but instead performs a self-consistent calculation of the field operating near the surface and in the bulk of the semi-infinite medium. Also, both the Coulomb and the retardation part of the dipole field were taken into account in the calculation of microscopic fields. All this made it possible to describe more accurately the transition layer properties due to the near-field effect [22].

9.1 Transition layer on the surface of a discrete-continuous dielectric

Consider a semi-infinite isotropic dielectric made up of atoms (molecules) of one species, with a coordinate-independent concentration N and a polarizability α. If the point of

observation is near the surface, which is the case we address first, then the Lorentz sphere is truncated and the area of its surface changes as the point of observation moves deep into the medium — with the result that a transition layer forms on the surface. To describe the layer, the same method as in Ref. [23] will be applied. We divide the near-surface region into a system of layers, all of which are parallel to the surface and in each of which the field can be considered independent of the coordinate z (Fig. 6). The microscopic field at an arbitrary observation point at time t can be determined by means of the following integral equation [which is a modification of Eqn (6.3)]:

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_I(\mathbf{r}, t) + \sum_{j=1}^{L} \int_{V_j} \mathrm{rot}\,\mathrm{rot}\, \frac{1}{R} N\alpha \mathbf{E}_j \left(\mathbf{r}', t - \frac{R}{c} \right) \mathrm{d}V'$$

$$+ \int_V \mathrm{rot}\,\mathrm{rot}\, \frac{1}{R} N\alpha \mathbf{E}_v \left(\mathbf{r}', t - \frac{R}{c} \right) \mathrm{d}V'$$

$$+ \sum_a \mathrm{rot}\,\mathrm{rot}\, \frac{1}{R_a} \alpha \mathbf{E} \left(\mathbf{r}_a, t - \frac{R_a}{c} \right) , \qquad (9.2)$$

where $\mathbf{E}_I(\mathbf{r}, t)$ is the electrical field strength of the external wave; $\mathbf{E}_j(\mathbf{r}, t)$ is the field strength in the jth layer; $\mathbf{E}_v(\mathbf{r}, t)$ is the field strength in the bulk; $R = |\mathbf{r} - \mathbf{r}'|$, $R_a = |\mathbf{r} - \mathbf{r}_a|$; $\mathbf{r}_a$ is the radius vector of the ath dipole inside the Lorentz sphere; and L is the number of layers near the surface. The differentiation in Eqn (9.2) is carried out with respect to the coordinates of the observation point, and the integration goes over the volume of the jth layer or over the inner part V of the semi-infinite dielectric (minus the sphere σ). The polarization vector $\mathbf{P}$ will be considered linear in $\mathbf{E}$, i.e., $\mathbf{P} = N\alpha \mathbf{E}$, with a certain prescribed polarizability α which is generally complex.
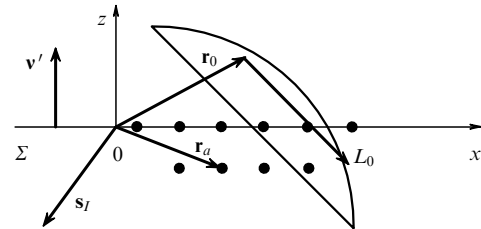

Figure 6. Transition layer on the boundary of a semi-infinite dielectric. The point of observation is in the Lth layer.

Assume that the incident wave is a monochromatic plane wave of frequency ω and wave vector $\mathbf{k}_I$, i.e.,

$$\mathbf{E}_I(\mathbf{r}, t) = \mathbf{E}_{0I} \exp(-\mathrm{i}\omega t) \exp(\mathrm{i}\mathbf{k}_I \mathbf{r}) \, . \qquad (9.3)$$

Assume further that the plane of incidence coincides with the xz plane, i.e., $\mathbf{k}_I$ has only x and z components:

$$\mathbf{k}_I = (k_{Ix}, 0, k_{Iz}) \, , \quad k_{Ix} = k_0 \sin\theta_I \, , \quad k_{Iz} = -k_0 \cos\theta_I \, ,$$

where θ_I is the angle of incidence. Then the field in each of the L layers can be written in the form

$$\mathbf{E}_j(\mathbf{r}) = \mathbf{E}_j \exp(-\mathrm{i}\omega t) \exp(\mathrm{i}k_{Ix} x) \, . \qquad (9.4)$$

The integrals over the monolayers in Eqn (9.2) are calculated by the method described in detail in Ref. [23], giving

$$\int_V \operatorname{rot} \operatorname{rot} \frac{1}{R} N\alpha \mathbf{E}_j \left(\mathbf{r}', t - \frac{R}{c} \right) \mathrm{d}V'$$

$$+ \sum_{z_a = z_j} \operatorname{rot} \operatorname{rot} \frac{1}{R_a} \alpha \mathbf{E}_j \left(\mathbf{r}_a, t - \frac{R_a}{c} \right)$$

$$= N\alpha \widehat{\Pi}(z - z_j) \mathbf{E}_j - \frac{\mathrm{i}2\pi N\alpha(z_{j-1} - z_j)}{k_0 \cos \theta_I}$$

$$\times \begin{cases} \left[\mathbf{k}_I [\mathbf{k}_I \mathbf{E}_j] \right] \exp \left[\mathrm{i}k_0 \cos \theta_I \, (z_j - z) \right], & z < z_j, \\ \left[\mathbf{k}_R [\mathbf{k}_R \mathbf{E}_j] \right] \exp \left[\mathrm{i}k_0 \cos \theta_I \, (z - z_j) \right], & z > z_j, \end{cases} \quad (9.5)$$

where $\mathbf{k}_R$ is the wave vector of the reflected wave: $\mathbf{k}_R = (k_0 \sin \theta_I, 0, k_0 \cos \theta_I)$. The form of the tensor $\widehat{\Pi}$ is given in part B of the appendix.

From Eqn (9.2) it follows that, in order to determine the field at an arbitrary point near the surface, it is necessary to know the behavior of the field in the bulk of the medium. Assume that the polarization wave in the bulk can be written in the form

$$\mathbf{P} = N\alpha \mathbf{E}_v(\mathbf{r}) \exp(-\mathrm{i}\omega t), \qquad (9.6)$$

in which the field strength in the bulk $\mathbf{E}_v(\mathbf{r})$ satisfies the equations

$$\nabla^2 \mathbf{E}_v(\mathbf{r}) + k_0^2 n^2 \mathbf{E}_v(\mathbf{r}) = 0, \quad \operatorname{div} \mathbf{E}_v = 0, \qquad (9.7)$$

where n is the refractive index of the medium. The refractive index determines the dispersion relation, i.e., the frequency dependence of the wave vector of the polarization wave, and is generally a complex quantity in our analysis. Applying the Gauss theorem and passing from the volume integral to a surface integral we obtain

$$\int_V \operatorname{rot} \operatorname{rot} \frac{1}{R} N\alpha \mathbf{E}_v \left(\mathbf{r}', t - \frac{R}{c} \right) \mathrm{d}V'$$

$$+ \sum_{z_a < z_L} \operatorname{rot} \operatorname{rot} \frac{1}{R_a} \alpha \mathbf{E}_v \left(\mathbf{r}_a, t - \frac{R_a}{c} \right)$$

$$= N\alpha \operatorname{rot} \operatorname{rot} \int_{\Sigma_L} \left(\mathbf{E}_v \frac{\partial G}{\partial v'} - G \frac{\partial \mathbf{E}_v}{\partial v'} \right) \mathrm{d}S' + N\alpha \hat{\beta} \, \mathbf{E}_v(\mathbf{r}, t). \qquad (9.8)$$

Here $G(R) = \exp(\mathrm{i}k_0 R)/R$, $\partial/\partial v'$ denotes differentiation along the outer normal to the surface Σ_L (see Fig. 6); the operator rot rot is taken out of the integral because the point of observation does not belong to the integration region; and $\hat{\beta}$ is the structural factor:

$$\hat{\beta} = \hat{\beta}_0 + \hat{\beta}_1 + \hat{\beta}_2,$$

$$(\hat{\beta}_0)_{ij} = \left(\int \mathrm{d}V' - \frac{1}{N} \sum \right) \frac{3n_i n_j - \delta_{ij}}{R^3} \exp(\mathrm{i}k_0 R) \exp(\mathrm{i}\mathbf{k}\mathbf{R}),$$

$$(\hat{\beta}_1)_{ij} = -\mathrm{i}k_0 \left(\int \mathrm{d}V' - \frac{1}{N} \sum \right) \frac{3n_i n_j - \delta_{ij}}{R^2} \exp(\mathrm{i}k_0 R) \exp(\mathrm{i}\mathbf{k}\mathbf{R}),$$

$$(\hat{\beta}_2)_{ij} = -k_0^2 \left(\int \mathrm{d}V' - \frac{1}{N} \sum \right) \frac{n_i n_j - \delta_{ij}}{R} \exp(\mathrm{i}k_0 R) \exp(\mathrm{i}\mathbf{k}\mathbf{R}),$$

$$(9.9)$$

where $i, j = x, y, z$, $\mathbf{n} = \mathbf{R}/R$, and $\mathbf{k}$ is the wave vector in the bulk of the medium,

$$\mathbf{k} = (k_{Ix}, 0, k_z), \quad k_z = -k_0 \sqrt{n^2 - \sin^2 \theta_I}.$$

With using Eqns (9.5) and (9.8), Eqn (9.2) for the field strength in an arbitrary layer near the surface can be written as [58]

$$\mathbf{E}_l = \mathbf{E}_{Il} + \mathrm{i}2\pi \frac{N\alpha}{k_{Iz}} \sum_{j=1}^{l-1} \left[\mathbf{k}_I [\mathbf{k}_I \mathbf{E}_j] \right] (z_{j-1} - z_j)$$

$$\times \exp \left[\mathrm{i}k_{Iz}(z_l - z_j) \right] + \mathrm{i}2\pi \frac{N\alpha}{k_{Iz}} \sum_{j=l+1}^{L} \left[\mathbf{k}_R [\mathbf{k}_R \mathbf{E}_j] \right]$$

$$\times (z_{j-1} - z_j) \exp \left[\mathrm{i}k_{Iz}(z_j - z_l) \right] + N\alpha \sum_{j=1}^{L} \widehat{\Pi}(z_l - z_j) \mathbf{E}_j$$

$$+ \frac{2\pi N\alpha}{k_{Iz}(k_{Iz} + k_z)} \exp(\mathrm{i}\mathbf{k}_R \mathbf{r}_l) \exp \left[\mathrm{i}(k_{Iz} + k_z)z_L \right]$$

$$\times \left[\mathbf{k}_R [\mathbf{k}_R \mathbf{E}_{0v}] \right] + N\alpha \hat{\beta}(z_l) \mathbf{E}_{0v}, \qquad (9.10)$$

where $l = \overline{1, L}$,

$$\mathbf{E}_{Il} = E_{0I} \exp \left[\frac{\mathrm{i}k_{Iz}(z_{l-1} + z_l)}{2} \right],$$

and $\mathbf{E}_{0v}$ is the amplitude of the field in the bulk.

Thus we see that, in order to determine the strength of the electromagnetic field at an arbitrary point of observation in the region near the surface, the field amplitude and the bulk refractive index of the medium are needed.

9.2 Refractive index of a discrete-continuous dielectric far from the boundary

Let us proceed to dividing the medium into layers deeper and deeper into the bulk until the Lorentz sphere around an arbitrary point in the bulk ceases to embrace the perturbed region near the surface. The field in the bulk of the medium satisfies Eqns (9.6) and (9.7). Upon applying the mathematical lemma proved in Ref. [23] and calculating integrals over the layers, Eqn (9.2) becomes

$$\mathbf{E}_v(\mathbf{r}) = \mathbf{E}_I(\mathbf{r}) - 2\pi \frac{\alpha N}{k_{Iz}^2} \exp(\mathrm{i}\mathbf{k}_I \mathbf{r}) \sum_{j=1}^{L} \left(\exp(-\mathrm{i}k_{Iz} z_{j-1}) \right)$$

$$- \exp(-\mathrm{i}k_{Iz} z_j)) \left[\mathbf{k}_I [\mathbf{k}_I \mathbf{E}_j] \right] + \frac{4\pi}{3} \frac{n^2 + 2}{n^2 - 1} \mathbf{E}_v(\mathbf{r})$$

$$+ N\alpha \operatorname{rot} \operatorname{rot} \int_{\Sigma_L} \left\{ \mathbf{E}_v \frac{\partial G}{\partial v'} - G \frac{\partial \mathbf{E}_v}{\partial v'} \right\} \mathrm{d}S'$$

$$+ N\alpha \hat{\beta} \mathbf{E}_v(\mathbf{r}). \qquad (9.11)$$

The tensor $\hat{\beta}$ here is given by Eqns (9.9), where the integrals are calculated over a closed sphere. Numerical calculations for various symmetry types of dipole distributions show that the components of the tensor $\hat{\beta}$ are generally nonvanishing, implying that our concept of a discrete-continuous medium opens new possibilities in using optical radiation to study atomic systems.

The terms in Eqn (9.11) can be divided into two groups. One of them forms a local equation which makes it possible to determine the refractive index for the points in the interior of the medium. If the tensor $\hat{\beta}$ can be presented as a scalar, one can separate out the following formula from this group [22]:

$$n^2 = \frac{1 + (8\pi/3)N\alpha - N\alpha\beta}{1 - (4\pi/3)N\alpha - N\alpha\beta}, \qquad (9.12)$$

which differs from the Lorentz–Lorentz formula only by the presence of the structure factor β [18].

The remaining terms in Eqn (9.11) form the nonlocal equation

$$\mathbf{E}_{0I} - 2\pi \frac{N\alpha}{k_{Iz}^2} \sum_{j=1}^{L} \left[\exp(-\mathrm{i}k_{Iz}z_{j-1}) - \exp(-\mathrm{i}k_{Iz}z_j) \right] \left[\mathbf{k}_I[\mathbf{k}_I\mathbf{E}_j] \right]$$

$$- \frac{2\pi N\alpha}{k_{Iz}(k_{Iz} - k_z)} \exp\left[\mathrm{i}(k_{Iz} - k_z)z_L \right] \left[\mathbf{k}_I[\mathbf{k}_I\mathbf{E}_{0v}] \right] = 0. \quad (9.13)$$

While this equation is analogous to Ewald–Oseen's extinction theorem [18], it is different in that, first, the meaning of the refractive index — given by Eqn (9.12) in our treatment — is different and, second, that the transition layer is taken into account. Eqns (9.7), (9.10), and (9.13) form a closed system which describes the behavior of the field near the surface and the amplitude of the field within the medium.

Figure 7 represents the solution of the system (9.7), (9.10), (9.13) for the case of an external wave incident normal to a semi-infinite medium. For not-too-large values of the product of the polarizability α by the concentration N ($\alpha N < 0{,}15$), calculations show that the transition layer thickness is determined mainly by the size of the Lorentz sphere and equals $1 - 2$ lattice constants (Fig. 7a) — thus justifying, for this particular case, the assumption, made in Ref. [55], that the field in the medium is well established within one atomic layer. For $\alpha N > 0.17$, however, the interaction between neighboring atomic layers becomes stronger, with the consequence that the layer thickness strongly depends on the parameter αN and rapidly increases as this parameter tends to $3/(4\pi)$ (Fig. 7b). Refractive indices $n > 3$ correspond to these values of αN. Such large values can be reached near

resonances where, however, absorption becomes an important factor.

9.3 Reflected wave field in the wave zone

Ref. [22] calculated the field of the reflected wave in the near zone relative to the surface of a discrete-continuous dielectric. It was shown that the reflected wave differs significantly from its the Fresnel counterpart, i.e., from the wave described by the continuous-medium Fresnel formula. This deviation from the Fresnel law was considered by the authors of Ref. [22] (one of whom is the present author) as a manifestation of the near-field effect. We now consider the field of a reflected wave far from the surface, taking into account the transition layer properties due to the near-field effect (see the discussion above). According to Eqn (9.2), at observation points outside the medium the reflected wave field $\mathbf{E}_R$ is given by

$$\mathbf{E}_R(\mathbf{r}, t) = \sum_{j=1}^{L} \mathrm{rot}\,\mathrm{rot} \int_{V_j} \frac{1}{R} \mathbf{P}_j\left(\mathbf{r}', t - \frac{R}{c} \right) \mathrm{d}V'$$

$$+ \mathrm{rot}\,\mathrm{rot} \int_{V} \frac{1}{R} \mathbf{P}_v\left(\mathbf{r}', t - \frac{R}{c} \right) \mathrm{d}V'. \quad (9.14)$$

The operator rot rot is taken out of the integral sign because the point of observation does not belong to the integration region. By calculating the volume integrals for each surface layer and for the bulk of the medium, we obtain

$$\mathbf{E}_R = 2\pi \frac{N\alpha}{k_{Iz}^2} \exp(\mathrm{i}\mathbf{k}_R\mathbf{r}) \sum_{j=1}^{L} \left(\exp(\mathrm{i}k_{Iz}z_{j-1}) \right.$$

$$\left. - \exp(\mathrm{i}k_{Iz}z_j) \right) \left[\mathbf{k}_R[\mathbf{k}_R\mathbf{E}_j] \right] + \frac{2\pi N\alpha}{k_{Iz}(k_{Iz} + k_z)} \exp(\mathrm{i}\mathbf{k}_R\mathbf{r})$$

$$\times \exp\left[\mathrm{i}(k_{Iz} + k_z)z_L \right] \left[\mathbf{k}_R[\mathbf{k}_R\mathbf{E}_{0v}] \right]. \quad (9.15)$$

Because of the non-exponential behavior of the field near the surface, Eqn (9.15) yields results somewhat different from those inferred from the usual Fresnel formulas. The behavior of the amplitude of the p-polarized wave field in the neighborhood of the Brewster angle is shown in Fig. 8, where the calculations based on the Fresnel formulas and on
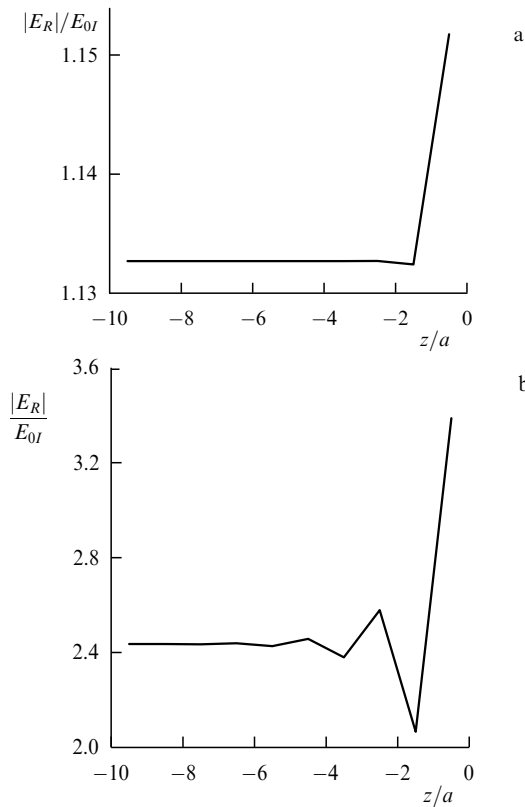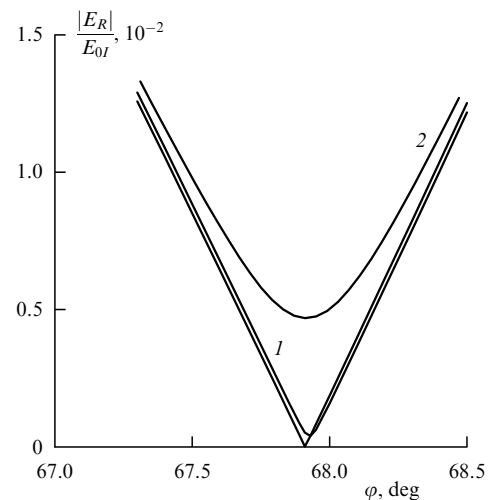
Figure 7. Amplitude $|E|$ of the microscopic field in a semi-infinite dielectric, for normal incidence. Parameters used in the calculations: $k_0 a = 0.005$, where a is the lattice constant; $N\alpha = 0.07$ (a), $N\alpha = 0.2$ (b).

Figure 8. Behavior of the amplitude of the reflected p-polarized wave near the Brewster angle. The calculations were done using Eqn (9.15) (curve *1*) and the formulas of Ref. [55] (curve *2*) for $N\alpha = 0.15$ and $k_0 a = 0.005$, with a the lattice constant.\

the formulas of Ref. [55] are also given for comparison. Although formula (9.15) gives a nonzero value for the amplitude of the reflected wave field, this value is an order of magnitude smaller than that obtained from the formulas of Ref. [55] and two orders of magnitude smaller than observed in experiment [44]. The discrepancy between the results of Ref. [55] and the theory of Ref. [58] is explained by the fact that the theory of Ref. [55] is actually somewhat inconsistent because it ignores the mutual influence of atoms in neighboring layers.

Based on the results obtained, it can be said that for materials with a refractive index $n < 2.5$, the near-field effect still contributes much less to the formation of the transition layer than other mechanisms do [44].

10. Optical probing of the electromagnetic field near the surface of a dielectric medium

In recent years there has been considerable interest in optic phenomena near the surface of various media at distances much less than the radiation wavelength (in the near-field). From a purely fundamental physics point of view, the near-field is of interest because it is in this region where the gradual formation of the reflected and transmitted waves takes place and where these waves show a behavior quite different from that in the far field.

On the other hand, a new area of applied research — the so-called scanning near-field optical microscopy — has received considerable attention in recent years [59 – 64]. Although various designs of optical near-field microscopes have been and are being developed, all of them are similar in that the optical response of the medium to an external influence is measured in the near-field. Therefore, the behavior of an electromagnetic field near a surface is of both fundamental and applied interest.

At present, the best resolution that can be achieved with a near-field optical microscope is of the order of a few nanometers, and the distance at which the surface under study is scanned is also of this order. We will demonstrate here that, with the knowledge of the behavior of an electromagnetic field near the surface of a dielectric, surface studies even with atomic-scale resolutions may become a reality.

The mathematical formalism we apply in this paper involves the use of integro-differential equations for the strength of the microscopic field [see Eqn (6.3)] and takes into account the discrete structure of the dielectric medium. It has been shown above that the optical properties of the surface, even in the absence of structural changes, may differ considerably from the bulk optical properties of the dielectric. However, many interesting aspects of the near-field behavior of an electromagnetic field have not been adequately studied. Below we solve the boundary value problem of linear classical optics relating to the interaction of an electromagnetic wave with a superthin dielectric film. It will be shown that all conclusions about the behavior of the field near the surface of a superthin film also remain valid for dielectric media of arbitrary thickness.

Let us consider a monochromatic wave with frequency ω and electric field strength vector $\mathbf{E}_I(\mathbf{r}, t) = \mathbf{A}_I(\mathbf{r}) \exp(-i\omega t)$ incident onto a dielectric film of thickness h. Above the film is a probe, which measures the electromagnetic field at some point in space (Fig. 9) and which is considered a single dipole with linear polarizability α_p. Such an experimental scheme,
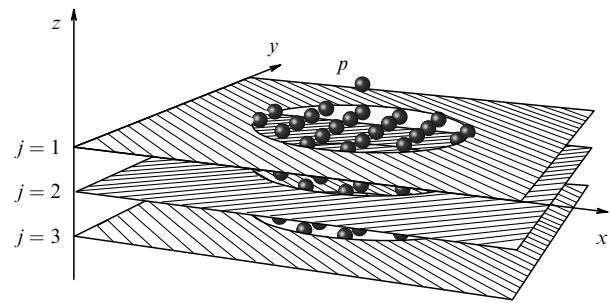

Figure 9. Model taking into account the discrete structure of a dielectric film. The point of observation $\mathbf{r}$, at the position of the atom acting as a measuring probe (p), is surrounded by a cylinder of radius ε, with its axis normal to the film surface. Atoms inside the cylinder are treated as discretely distributed dipoles; the film outside, as a set of continuous monolayers. The case $N = 3$ is given as an illustration.

with a single atom as a probe, can be realized in practice by, for example, using an atom in a magneto-optical trap [65]. Our concern here is to calculate the intensity of the electromagnetic field at the probe as this probe scans the film surface moving along the surface at a certain prescribed distance z above it.

In order to account for the discrete nature of the film, let us consider the following model. Let us treat the dielectric film as a system of N monolayers the distance a_0 apart, a_0 being the lattice constant (see Fig. 9). Consider a situation in which atoms in the film form a perfect crystal structure with a cubic symmetry (our analysis can easily be extended to include other types of symmetry). Enclose the observation point — which may be at the position of the probe as well as at the position of any of the atoms in the film — by a cylinder of radius $\varepsilon > a_0$, with the axis passing through the observation point parallel to the z axis of the coordinate system. The atoms inside the cylinder will be considered discretely distributed dipoles, and those outside the cylinder, a system of parallel, continuous atomic monolayers. As the point of observation moves in the (x, y) plane, the cylinder is displaced as a whole. With this model, in the stationary case the integro-differential equation for the strength of the local electric field $\mathbf{E}'(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}) \exp(-i\omega t)$ at the probe can be written in the form (6.4) (see Ref. [66])

$$\mathbf{E}(\mathbf{r}) = \mathbf{A}_I(\mathbf{r}) + \frac{\alpha}{a_0^2} \sum_{j=1}^{N} \int_{\sigma}^{\Gamma_\infty} \nabla_\mathbf{r} \times \nabla_\mathbf{r} \times \mathbf{E}_j(x', y') G(R_j) \, dS'$$

$$+ \alpha \sum_{j=1}^{N} \sum_{a_j} \nabla_\mathbf{r} \times \nabla_\mathbf{r} \times \mathbf{E}_j(x_a, y_a) G(R_{a_j}), \qquad (10.1)$$

where $\mathbf{r} = (x, y, z)$ is the position vector of the probe; α is the microscopic polarizability of the film atoms, assumed to be field-independent; $G(R) = \exp(ik_0 R)/R$ is the Green function of the Helmholtz equation; $k_0 = \omega/c$; $R_j = |\mathbf{r} - \mathbf{r}'_j|$; $\mathbf{r}'_j = (x', y', z_j)$; $R_{a_j} = = |\mathbf{r} - \mathbf{r}_{a_j}|$, and $\mathbf{r}_{a_j} = (x_{a_j}, y_{a_j}, z_j)$.

The integrals in Eqn (10.1) are taken over the (x', y') plane — except for the circle σ with the point of observation at the center. The last term in Eqn (10.1) accounts for the atoms inside the cylinder. We have also introduced the notation

$$\mathbf{E}_j(x, y) = \mathbf{E}(x, y, z_j), \qquad j = \overline{1, N}. \qquad (10.2)$$

Let $\mathbf{r}_{a_l} = (x_{a_l}, y_{a_l}, z_l)$ be the radius vector of a certain atom in the film's lth monolayer. Then for the local field at

this atom we have

$$\mathbf{E}_l(x_{a_l}, y_{a_l}) = \mathbf{E}_{Il}(x_{a_l}, y_{a_l}) + \alpha_p \nabla_{\mathbf{r}_{a_l}} \times \nabla_{\mathbf{r}_{a_l}} \times \mathbf{E}(\mathbf{r}) G(R_{a_l})$$

$$+ \frac{\alpha}{a_0^2} \sum_{j=1}^{N} \int_{\sigma}^{\Gamma_\infty} \nabla_{\mathbf{r}_{a_l}} \times \nabla_{\mathbf{r}_{a_l}} \times \mathbf{E}_j(x', y') G(R_{a_l j}) \, dS'$$

$$+ \alpha \sum_{j=1}^{N} \sum_{a_j} \nabla_{\mathbf{r}_{a_l}} \times \nabla_{\mathbf{r}_{a_l}} \times \mathbf{E}_j(x_a, y_a) G(R_{a_l a_j}), \quad l = \overline{1, N},$$

$$(10.3)$$

where $\mathbf{E}_{Il}(x_{a_l}, y_{a_l}) = \mathbf{A}_I(\mathbf{r}_{a_l})$, $R_{a_l} = |\mathbf{r}_{a_l} - \mathbf{r}|$, $R_{a_l j} = |\mathbf{r}_{a_l} - \mathbf{r}'_j|$, and $R_{a_l a_j} = |\mathbf{r}_{a_l} - \mathbf{r}_{a_j}|$. The problem is now that of finding a self-consistent solution to the system of equations (10.1), (10.3).

10.1 Field at the probe scanning the film surface
In order to achieve a reasonable accuracy in the calculation of the lattice sums in Eqns (10.1), (10.3), the region of discreteness must be large. We assume that, if the probe is situated at a distance of the lattice constant or higher above the surface, it does not significantly affect the electromagnetic field distribution in the film, even for fairly large polarizabilities α_p. The second term on the left of Eqn (10.3) can then be dropped, which considerably simplifies the solution of the system (10.1), (10.3) [23].

Let the external wave be plane,

$$\mathbf{A}_I(\mathbf{r}) = \mathbf{E}_{0I} \exp(ik_0 \mathbf{s}_I \mathbf{r}),$$

$$\mathbf{s}_I = (\sin\theta_I \cos\Phi_I, \sin\theta_I \sin\Phi_I, -\cos\theta_I), \quad (10.4)$$

where θ_I is the angle of incidence, and the azimuthal angle Φ_I determines the plane-of-incidence orientation relative to the basis vector a_{0y}. Ref. [23] considered the special case $\Phi_I = \pi$. Below we obtain the solution for arbitrary Φ_I. With the use of the uniformity property of the film surfaces, the solution for the local field in the medium can be written in the form

$$\mathbf{E}_l(x, y) = \mathbf{E}_l \exp\left[ik_0 \sin\theta_I(\cos\Phi_I x + \sin\Phi_I y)\right]. \quad (10.5)$$

Substituting (10.4) and (10.5) into (10.3) and using a technique similar to that described in Ref. [23] yields the following system of equations for observation points located at the atomic positions in the film:

$$\mathbf{E}_l = \mathbf{E}_{Il} - i2\pi C \frac{k_0 a_0}{\cos\theta_I} \sum_{j=1}^{l-1} \left[\mathbf{s}_I \times (\mathbf{s}_I \times \mathbf{E}_j)\right]$$

$$\times \exp\left[ik_0 a_0 \cos\theta_I (z_j - z_l)\right] - i2\pi C \frac{k_0 a_0}{\cos\theta_I}$$

$$\times \sum_{j=l+1}^{N} \left[\mathbf{s}_R \times (\mathbf{s}_R \times \mathbf{E}_j)\right] \exp\left[ik_0 a_0 \cos\theta_I (z_l - z_j)\right]$$

$$+ C \sum_{j=1}^{N} \widehat{\Pi}(z_l - z_j) \mathbf{E}_j, \quad l = \overline{1, N}, \quad (10.6)$$

where

$$\mathbf{E}_{Il} = \mathbf{E}_{0I} \exp(-ik_0 \cos\theta_I z_l), \quad l = \overline{1, N}, \quad (10.7)$$

$C = \alpha/a_0^3$, $\mathbf{s}_I$ is given by Eqn (10.4), and the vector $\mathbf{s}_R$ has the form

$$\mathbf{s}_R = (\sin\theta_I \cos\Phi_I, \sin\theta_I \sin\Phi_I, \cos\theta_I). \quad (10.8)$$

The explicit form of the tensor $\widehat{\Pi}$ is not reproduced here because of its cumbersome nature and is given in part A of the Appendix instead.

Once the system (10.6) has been solved, the field at the probe can be calculated using Eqn (10.1), which we rewrite in the form

$$\mathbf{E}(\mathbf{r}) = \mathbf{A}_I(\mathbf{r}) + \mathbf{E}_{0R}(\mathbf{r}) \exp(ik_0 \mathbf{s}_R \mathbf{r}), \quad z > 0, \quad (10.9)$$

where the reflected wave amplitude calculated with due account for the discrete nature of the medium is given by the expression [23]

$$\mathbf{E}_{0R}(\mathbf{r}) = -i2\pi C \frac{k_0 a_0}{\cos\theta_I} \sum_{j=1}^{N} \left[\mathbf{s}_R \times (\mathbf{s}_R \times \mathbf{E}_j)\right]$$

$$\times \exp[-ik_0 a_0 \cos\theta_I z_j] + C \exp[-ik_0 a_0 \cos\theta_I z]$$

$$\times \sum_{j=1}^{N} \widehat{\Pi}(x, y, z - z_j) \mathbf{E}_j, \quad (10.10)$$

in which the quantities $\mathbf{E}_j$, $j = \overline{1, N}$ are solutions of the system (10.6).

In Ref. [23], the field of the reflected (transmitted) wave was calculated for the point of observation located precisely above (below) some of the atoms in the film. Here we consider the more general case in which the point of observation takes an arbitrary position in the (x, y) plane.

As can be seen from Eqn (10.10), the reflected amplitude is generally a function of the coordinate $\mathbf{r}$ of the point of observation. The form of this function is determined by those terms in Eqn (10.10) that contain the tensor $\widehat{\Pi}$.

The dependence of the quantity $I = |\mathbf{E}|^2$, which is proportional to the intensity of the electromagnetic field at the probe, on the coordinates x, y is shown in Fig. 10 for the case of an s-polarized incident wave, in which

$$\mathbf{E}_{0I} = (\sin\Phi_I, -\cos\Phi_I, 0) E_{0I}. \quad (10.11)$$

The distance from the film surface z is taken to be one lattice constant. The number of monolayers is $N = 11$. The plots are presented for the case of normal incidence of the external wave ($\theta_I = 0$). In the case of oblique incidence, the field exhibits some particularities of its behavior inside and outside the film, which are due to the discrete distribution of atoms near the observation point and for which a separate analysis is needed.

As can be seen from Fig. 10, the intensity of the electromagnetic field is a periodic function of the coordinates x, y with a period of a_0, the lattice constant. Thus, the reflected wave at small distances from the surface is not a plane one.

The intensity minima correspond to atomic positions in the surface monolayer. The difference between the maximum and minimum intensity values is strongly dependent on the distance from the surface of the film, increasing as the distance decreases, and vice versa. Beyond $2a_0$, the intensity distribution in the (x, y) plane becomes all but uniform, i.e., the reflected wave becomes plane — a consequence of the fact that the terms in Eqn (10.10) containing the tensor $\widehat{\Pi}$ cease to contribute at such distances. Fig. 10a also shows that the image contrast in the direction normal to the polarization vector is much weaker. This agrees with the results of Ref. [62]. However, by rotating the polarization plane Φ_I, the image contrast can be significantly improved (Fig. 10c). It should be noted that, independently of the rotation angle Φ_I,
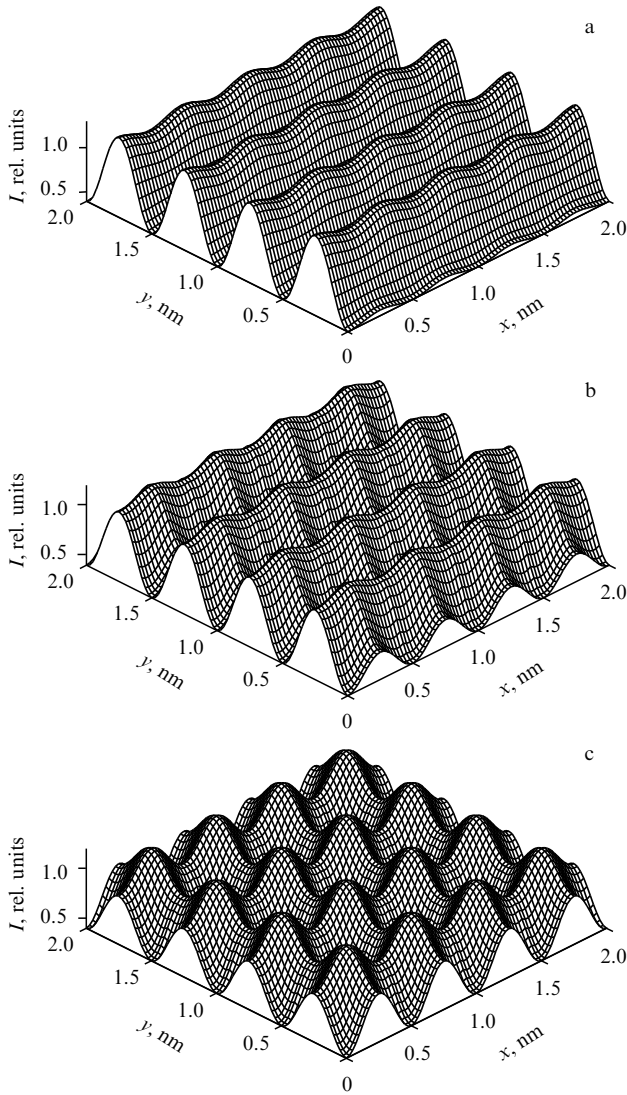
Figure 10. Electromagnetic field intensity at the probe as a function of the coordinates x, y. Calculations were done for $N = 11$, $k_0 a_0 = 0.005$, $C = 0, 2$, $a_0 = 0, 5$ nm, $z = a_0$, $\theta_I = 0°$; for (a) $\Phi_I = 0°$, (b) $\Phi_I = 30°$, (c) $\Phi_I = 45°$. The intensity distribution is a periodic function of x, y, with a period of a_0, the lattice constant. The intensity minima correspond to the positions of the atoms of the film.

Figure 11. Intensities (a) of the x-component and (b) of the z-component of the electromagnetic field ($I_x = |E_x|^2$, $I_y = |E_y|^2$) at the probe as functions of the coordinates x and y. Calculations were performed for the parameters: $N = 11$, $k_0 a_0 = 0.005$, $C = 0.2$, $a_0 = 0.5$ nm, $z = a_0$, $\theta_I = 0°$, $\Phi_I = 0°$. Although the external wave is polarized along the y axis, the near-field optical response of the medium contains all three components.

the field intensity above atoms and above the center of the unit cell remains unchanged.

One further aspect of the near-surface behavior of the electromagnetic field is that the reflected wave field always contains three spatial components, E_{Rx}, E_{Ry}, and E_{Rz}, even if the external wave is one-component. This is illustrated in Fig. 11, where the intensities of the x and z components of the electromagnetic field ($I_x = |E_x|^2$, $I_y = |E_y|^2$) at the probe are shown as functions of the coordinates x for a y-polarized external wave. Note that, whereas the total field intensity near the film surface oscillates with a period equal to the lattice constant, each of the components I_x, I_z is modulated with half that period; the y- component of the field near the surface is not shown because its behavior is virtually the same as that for the total intensity (see Fig. 10).

For the probe on the other side of the film ($z < -h$), the field is determined by the field of the transmitted wave, and all the near-field and the wave zone results for the reflected wave also remain valid in the case of the transmitted wave.
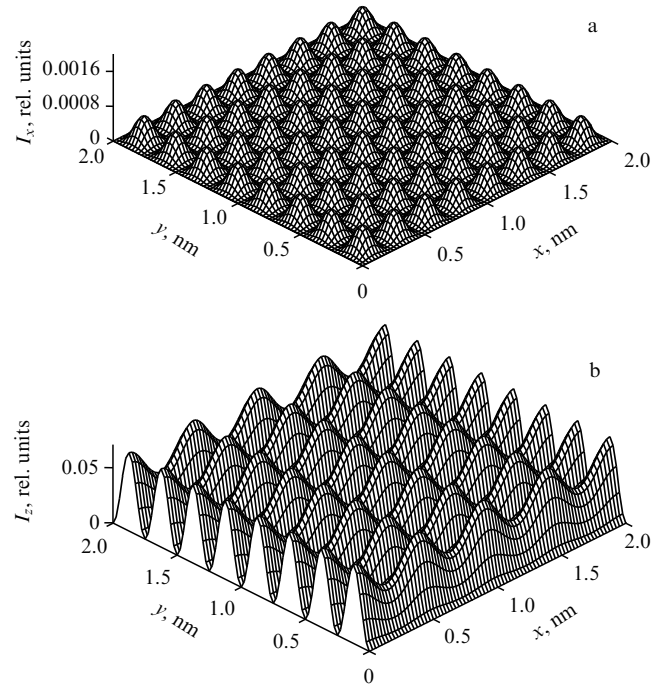
Thus we see that near the surface of the film the fields of the reflected and transmitted waves are not plane waves. One can also argue that the formation of the reflected and transmitted waves takes place at distances of the order of the lattice constant from the surface of the medium.

Thus, we have solved the boundary-value problem of linear classical optics, concerning the behavior of an electromagnetic field near the surface of a discretely structured dielectric medium [66]. It is shown that, at distances less than $2a_0$ from the surface (a_0 being the lattice constant), the behavior of the reflected and transmitted waves is fundamentally different from that in the far field.

In the literature, nobody usually goes beyond making the very general statement, not supported by any numerical estimates, that the formation of the reflected and transmitted waves takes place in a region less than the radiation wavelength in size (see, e.g., [44]). Our calculations show that this process actually occurs at a distance of about $2a_0$ from the surface.

Although the above discussion of the near-surface field behavior concerned a thin film of a dielectric, the same results hold for the case of extended media. In fact, since the surface effects we have discussed are observed at distances of no more than $2a_0$, they are clearly due to the near-surface layers only. The bulk layers can only give a constant contribution, of no consequence in the present context.

11. Optics of small objects and the near-field effect

The term *small object* will refer to a small group of atoms or molecules occupying a volume of linear dimensions less the light wavelength. Examples are atoms deposited on a

substrate, individual complexes in solid solutions, or recently observed aggregates [67]. The theory of linear molecular aggregates interacting with the field of a quasi-resonant light wave was discussed in Ref. [67] under the assumption that the fields at individual molecules are identical. Refs [68, 69] considered the interaction of a two-atom small object with the field of quasi-resonant radiation. Based on a combined system of equations for the field and atomic variables, it was shown that at near-resonant frequencies, the fields at individual atoms may differ considerably and that the wave-zone field due to the small object is strongly dependent on the interatomic separation and on how the axis of the small object is oriented relative to the propagation direction of the external light wave. In the approach of Refs [68, 69], it is the near-field effect which causes the field to behave in this manner inside and outside the small object.

11.1 Two atoms in the field of a weak quasi-resonant plane wave

The microscopic field of the light wave $\mathbf{E}(\mathbf{r}, t)$ at the point of observation $\mathbf{r}$ at the time t is determined by the following equation:

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_I(\mathbf{r}, t) + \sum_{j=1}^{2} \operatorname{rot} \operatorname{rot} \frac{\mathbf{p}_j(t - R_j/c)}{R_j} , \qquad (11.1)$$

where $\mathbf{E}_I(\mathbf{r}, t)$ is the electric field strength in the external light wave which travels at the velocity of light c and $\mathbf{p}_j$ is the induced dipole moment of the jth atom, which we take to be a linear function of the field $\mathbf{E}(\mathbf{r}_j, t - R_j/c)$ at the position of the jth atom. The distance R_j is $|\mathbf{r} - \mathbf{r}_j|$, where $\mathbf{r}_j$ is the radius vector of the jth atom relative to the origin positioned at the center of one of the atoms — say, the first atom. The differentiation in Eqn (11.1) is carried out with respect to the coordinates of the observation point. In the special case in which the observation point coincides with the position of one of the atoms, Eqn (11.1) reduces to a system of two equations for the unknowns $\mathbf{E}(\mathbf{r}_1, t)$ and $\mathbf{E}(\mathbf{r}_2, t)$. Once these are found, the field at other points of observation can also be found from Eqns (11.1).

To proceed further, it is necessary to supplement Eqn (11.1) with equations for the atomic variables. Let us treat atoms as Lorentz oscillators [24]. In this case the vector of the induced dipole moment $\mathbf{p}_j$ takes the form

$$\mathbf{p}_j = e(\mathbf{u}_j - i\mathbf{v}_j) \exp(-i\omega t) + \text{c.c.}, \qquad j = 1, 2, \qquad (11.2)$$

where e is the electron charge, and ω is the frequency of the oscillator. The quantities $\mathbf{u}_j$ and $\mathbf{v}_j$ depend on the position of the atom and also vary with time — because the atomic eigenfrequencies ω_1 and ω_2 differ from the frequency ω of the field of the external light wave. This variation, however, is slow if the differences $\omega - \omega_1$ and $\omega - \omega_2$ are small. In this case, the following inequalities hold:

$$|\dot{\mathbf{u}}_j| \ll \omega|\mathbf{u}_j|, \quad |\ddot{\mathbf{u}}_j| \ll \omega^2|\mathbf{u}_j|, \quad |\dot{\mathbf{v}}_j| \ll \omega|\mathbf{v}_j|, \quad |\ddot{\mathbf{v}}_j| \ll \omega^2|\mathbf{v}_j| . \qquad (11.3)$$

In view of the above conditions, the equation of motion of the jth dipole

$$\ddot{\mathbf{p}}_j + \frac{2}{\tau_0} \dot{\mathbf{p}}_j + \omega_j^2 \mathbf{p}_j = \frac{e^2}{m} \mathbf{E}(\mathbf{r}_j, t) \qquad (11.4)$$

(m is the electron mass, $1/\tau_0$ the fractional energy damping rate of an isolated dipole) becomes

$$\frac{\partial}{\partial t}(\mathbf{u}_j - i\mathbf{v}_j) = \left(-i\Delta_j - \frac{1}{T} \right)(\mathbf{u}_j - i\mathbf{v}_j) + i\varkappa_0 \mathbf{E}_{0j}, \qquad (11.5)$$

where $\varkappa_0 = e/(m\omega)$; the quantity $\mathbf{E}_{0j}$ follows from the expression for the field

$$\mathbf{E}(\mathbf{r}_j, t) = \mathbf{E}_0(\mathbf{r}_j) \exp(-i\omega t) + \text{c.c.} \qquad (11.6)$$

as $\mathbf{E}_{0j} = \mathbf{E}_0(\mathbf{r}_j)$; $1/T$ is the total oscillator damping rate, which may differ from that of an isolated oscillator; and $\Delta_j = \omega_j - \omega$ is the detuning from the resonance.

Eqns (11.5) and (11.1) form a closed system of equations in which the mutual influence of the field and the atoms is included in a self-consistent manner. Note that [1]

$$\operatorname{rot} \operatorname{rot} \frac{[\mathbf{p}_j]}{R_j} \equiv \frac{3([\mathbf{p}_j]\mathbf{n}_j)\mathbf{n}_j - [\mathbf{p}_j]}{R_j^3} + \frac{3([\dot{\mathbf{p}}_j]\mathbf{n}_j)\mathbf{n}_j - [\dot{\mathbf{p}}_j]}{cR_j^2}$$
$$+ \frac{([\ddot{\mathbf{p}}_j]\mathbf{n}_j)\mathbf{n}_j - [\ddot{\mathbf{p}}_j]}{c^2 R_j} , \qquad (11.7)$$

where the symbol $[\ldots]$ indicates that the quantity is taken at time $t - R/c$, $\mathbf{n}_j = \mathbf{R}_j/R_j$. In the special case in which the point of observation coincides with the position of an atom, R_j is equal to the interatomic separation R. The first term in Eqn (11.7) corresponds to the dipole's Coulomb field, and the remaining terms describe the retarded dipole field at the point of observation j. The field (11.7) is the polarizing field of the jth dipole and has a meaning different from that of the scattered field [17]. Below we examine the spatial distribution of the Coulomb and retarded polarizing fields at various observation points, assuming that the two dipoles interact self-consistently and using a stationary solution of Eqn (11.5). Consider first the electric field of the light wave inside a small object.

If we take the origin to be at point $\mathbf{r}_1$, then we have $\mathbf{r}_1(0, 0, 0)$ and $\mathbf{r}_2(0, R, 0)$. With this choice of the coordinate system, we obtain the following system of equations for the unknown fields $\mathbf{E}(\mathbf{r}_1, t)$ and $\mathbf{E}(\mathbf{r}_2, t)$ at the position of each of the atoms for $i \neq j$:

$$\mathbf{E}(\mathbf{r}_i, t) = \mathbf{E}_I(\mathbf{r}_i, t) + \frac{3[p_j^y]\mathbf{y}_0 - [\mathbf{p}_j]}{R^3} + \frac{3[\dot{p}_j^y]\mathbf{y}_0 - [\dot{\mathbf{p}}_j]}{cR^2}$$
$$+ \frac{[\ddot{p}_j^y]\mathbf{y}_0 - [\ddot{\mathbf{p}}_j]}{c^2 R} , \qquad (11.8)$$

where $\mathbf{y}_0$ is the unity vector along the y axis.

Suppose the external field to have the form

$$\mathbf{E}_I(\mathbf{r}_i, t) = \mathbf{E}_{0I} \exp\left[i(\mathbf{k_0}\mathbf{r}_i - \omega t)\right] + \text{c.c.}, \qquad (11.9)$$

where $\mathbf{E}_{0I}$ is a constant amplitude, and $\mathbf{k}_0$ is the wave vector whose magnitude is $k_0 = \omega/c$. The induced dipole moments and the field at points $\mathbf{r}_1$ and $\mathbf{r}_2$ are given by Eqns (11.2) and (11.6), where $\mathbf{p}_{0j} = e(\mathbf{u}_j - i\mathbf{v}_j)$ and $\mathbf{E}_{0j}$ are complex quantities. Substituting Eqns (11.9), (11.2), and (11.6) into Eqn (11.1) and separating out equally oscillating factors, we obtain for a stationary solution of Eqn (11.5) the equality

$$\mathbf{p}_{0j} = \alpha_j \mathbf{E}_{0j}, \qquad (11.10)$$

where

$$\alpha_j = \frac{e^2}{m} \frac{1}{\omega_j^2 - \omega^2 - 2\mathrm{i}\omega/T} \tag{11.11}$$

is the polarizability of the jth atom [24].

Substituting Eqn (11.10) into the system (11.8), after some algebra, we obtain the following system of coupled equations:

$$p_{01}^y = \alpha_1 \left\{ E_{0I}^y + 2G p_{02}^y \exp(\mathrm{i}k_0 R) \right\},$$

$$p_{02}^y = \alpha_2 \left\{ E_{0I}^y \exp(\mathrm{i}\mathbf{k}_0\mathbf{R}) + 2G p_{01}^y \exp(\mathrm{i}k_0 R) \right\}, \tag{11.12}$$

$$p_{01}^\beta = \alpha_1 \left\{ E_{0I}^\beta - F p_{02}^\beta \exp(\mathrm{i}k_0 R) \right\},$$

$$p_{02}^\beta = \alpha_2 \left\{ E_{0I}^\beta \exp(\mathrm{i}\mathbf{k}_0\mathbf{R}) - F p_{01}^\beta \exp(\mathrm{i}k_0 R) \right\}, \tag{11.13}$$

$$\beta = x, z,$$

where

$$G = \frac{1}{R^3} - \mathrm{i}\frac{k_0}{R^2}, \qquad F = G - \frac{k_0^2}{R}. \tag{11.14}$$

The systems of algebraic equations, (11.12), (11.13), are linear and can therefore be solved by any standard method, giving the following expressions for the unknowns [69]:

$$p_{01}^y = \alpha_1 \frac{1 + 2\alpha_2 G \exp[\mathrm{i}(k_0 R + \mathbf{k}_0\mathbf{R})]}{1 - 4\alpha_1\alpha_2 G^2 \exp(\mathrm{i}2k_0 R)} E_{0I}^y,$$

$$p_{01}^\beta = \alpha_1 \frac{1 - \alpha_2 F \exp[\mathrm{i}(k_0 R + \mathbf{k}_0\mathbf{R})]}{1 - \alpha_1\alpha_2 F^2 \exp(\mathrm{i}2k_0 R)} E_{0I}^\beta,$$

$$\tag{11.15}$$

$$p_{02}^y = \alpha_2 \frac{\exp(\mathrm{i}\mathbf{k}_0\mathbf{R}) + 2\alpha_1 G \exp(\mathrm{i}k_0 R)}{1 - 4\alpha_1\alpha_2 G^2 \exp(\mathrm{i}2k_0 R)} E_{0I}^y,$$

$$p_{02}^\beta = \alpha_2 \frac{\exp(\mathrm{i}\mathbf{k}_0\mathbf{R}) - \alpha_1 F \exp(\mathrm{i}k_0 R)}{1 - \alpha_1\alpha_2 F^2 \exp(\mathrm{i}2k_0 R)} E_{0I}^\beta.$$

The corresponding expressions for the field strengths can readily be obtained using Eqn (11.10).

Thus, we have obtained the solution of the self-consistent problem for the case in which the external field is produced by a plane wave of frequency ω.

Before we write down formulas for the field at each atom, let us make one further simplification.

Let the eigenfrequencies of the atoms be all identical, $\omega_1 = \omega_2 = \omega_0$. Then $\alpha_1 = \alpha_2 = \alpha$, and from Eqn (11.15) we obtain the following expressions for the complex amplitude of the field at each of the atoms:

$$E_{01}^y = \frac{1 + 2\alpha G \exp[\mathrm{i}(k_0 R + \mathbf{k}_0\mathbf{R})]}{1 - 4\alpha^2 G^2 \exp(\mathrm{i}2k_0 R)} E_{0I}^y,$$

$$E_{01}^\beta = \frac{1 - \alpha F \exp[\mathrm{i}(k_0 R + \mathbf{k}_0\mathbf{R})]}{1 - \alpha^2 F^2 \exp(\mathrm{i}2k_0 R)} E_{0I}^\beta,$$

$$\tag{11.16}$$

$$E_{02}^y = \frac{\exp(\mathrm{i}\mathbf{k}_0\mathbf{R}) + 2\alpha G \exp(\mathrm{i}k_0 R)}{1 - 4\alpha^2 G^2 \exp(\mathrm{i}2k_0 R)} E_{0I}^y,$$

$$E_{02}^\beta = \frac{\exp(\mathrm{i}\mathbf{k}_0\mathbf{R}) - \alpha F \exp(\mathrm{i}k_0 R)}{1 - \alpha^2 F^2 \exp(\mathrm{i}2k_0 R)} E_{0I}^\beta.$$

Thus, it can be seen from formulas (11.16) that the field at the position of an atom is generally different from the external field. The situation is determined by the values of the factors αF and αG, which depend on the frequency of the external field and the interatomic separation. The fields (11.16) are equal to the external field only if each of these factors is much less than unity, and this condition can be fulfilled when either the interatomic separation is sufficiently large or the frequency of the external field differs considerably from the resonant frequency.

The complex amplitudes of the fields at each atom can be related to each other by writing

$$\mathbf{E}_{02} = (E_{01}^x \mathbf{x}_0 + E_{01}^z \mathbf{z}_0) \exp(\mathrm{i}k_r R) + E_{01}^y \exp(\mathrm{i}k_l R)\mathbf{y}_0, \tag{11.17}$$

where $\mathbf{x}_0, \mathbf{y}_0, \mathbf{z}_0$ are the unit vectors of the coordinate system, and

$$k_r = \frac{\mathbf{k}_0\mathbf{R}}{R} - \frac{\mathrm{i}}{R} \ln \frac{1 - \alpha F \exp[\mathrm{i}(k_0 R - \mathbf{k}_0\mathbf{R})]}{1 - \alpha F \exp[\mathrm{i}(k_0 R + \mathbf{k}_0\mathbf{R})]},$$

$$k_l = \frac{\mathbf{k}_0\mathbf{R}}{R} - \frac{\mathrm{i}}{R} \ln \frac{1 + 2\alpha G \exp[\mathrm{i}(k_0 R - \mathbf{k}_0\mathbf{R})]}{1 + 2\alpha G \exp[\mathrm{i}(k_0 R + \mathbf{k}_0\mathbf{R})]}. \tag{11.18}$$

The expression (11.17), together with Eqn (11.6), imply that the field in the system is a superposition of two waves, a transverse wave with the wave vector $\mathbf{k}_r$, and a longitudinal wave with the wave vector $\mathbf{k}_l$, either vector being directed along the y axis.

We are now, finally, in a position to determine the light wave field outside the small object by inserting Eqn (11.6) into Eqn (11.1). In this case the Green function $\exp(\mathrm{i}k_0 R_j)/R_j$ links the jth dipole and the observation point $\mathbf{r}$ outside the small object. Below, the wave zone field due to a small object is examined numerically.

11.2 Two atoms in the field of an intense quasi-resonant plane wave

We apply the field equation (11.1) to describing the optical properties of a small object made up of two resonance atoms, in an intense light wave field

$$\mathbf{E}_I(\mathbf{r}, t) = \mathbf{e}_I E_{0I} \exp\left(-\mathrm{i}[\mathbf{k}_0\mathbf{r} - \omega t]\right) + \text{c.c.},$$

where $\mathbf{e}_I$ is the real unit polarization vector, E_{0I} is the wave amplitude, and ω and $\mathbf{k}_0$ are the frequency and wave vector of the plane wave, respectively ($k_0 = \omega/c$). Then the dipole moment of the jth atom can be written as

$$\mathbf{p}_j = \frac{1}{2} \mathbf{d}(u_j + \mathrm{i}v_j) \exp(\mathrm{i}\omega t) + \text{c.c.}, \tag{11.19}$$

where $\mathbf{d}$ is the matrix element of the dipole moment for the transition between two chosen states. The quantities u_j and v_j in Eqn (11.9) satisfy the following equations [24]:

$$\dot{u}_j = -(\omega_0 - \omega)v_j - \frac{2}{\hbar}(\mathbf{d}\mathbf{e}_j)E_{0j}'' w_j - \frac{u_j}{T_2'},$$

$$\dot{v}_j = (\omega_0 - \omega)u_j + \frac{2}{\hbar}(\mathbf{d}\mathbf{e}_j)E_{0j}' w_j - \frac{v_j}{T_2'}, \tag{11.20}$$

$$\dot{w}_j = \frac{2}{\hbar}(\mathbf{d}\mathbf{e}_j)(E_{0j}'' u_j - E_{0j}' v_j) - \frac{(w_j - w_0)}{T_1},$$

where ω_0 is the frequency of the resonant transition chosen in the spectrum of the atom, w_j is the inversion of the corresponding quantum states on the jth atom, w_0 is the initial value of the inversion, and T_1 and T_2' are the relaxation times [24]. The quantities E_{0j}' and E_{0j}'' are the real and imaginary parts of the electric field amplitude at the jth atom:

$$\mathbf{E}(\mathbf{r}_j, t) = \mathbf{e}_j(E_{0j}' + iE_{0j}'')\exp(i\omega t) + \text{c.c.},\qquad (11.21)$$

$\mathbf{e}_j$ being the real polarization vector of the field at the jth atom.

Thus, we have established a self-consistent system of equations for the field and atomic variables.

In what follows we consider only steady processes, i.e., we assume the time that elapsed since the external field is on to be $t \gg T_1, T_2'$, so that $\dot{u}_j = \dot{v}_j = \dot{w}_j = 0$. With this restriction and using Eqns (11.20), the atomic variables can be expressed in terms of the field variables as follows:

$$u_j = -\frac{w_0 \varkappa_j T_2'\left(E_{0j}'(\omega - \omega_0)T_2' + E_{0j}''\right)}{\Delta_j},$$

$$v_j = \frac{w_0 \varkappa_j T_2'\left(E_{0j}' - E_{0j}''(\omega_0 - \omega)T_2'\right)}{\Delta_j},\qquad (11.22)$$

$$w_j = \frac{w_0\left(1 + (\omega_0 - \omega)^2(T_2')^2\right)}{\Delta_j},$$

where $\Delta_j = 1 + (\omega_0 - \omega)^2(T_2')^2 + T_1 T_2' \varkappa_j^2((E_{0j}')^2 + (E_{0j}'')^2)$, $\varkappa_j = 2\mathbf{d}\mathbf{e}_j/\hbar$.

We choose the origin of the coordinate system to be at point $\mathbf{r}_1$, so that we have $\mathbf{r}_1(0,0,0)$ and $\mathbf{r}_2(0, R, 0)$. We also take the wave vector $\mathbf{k}_0$ of the external wave to be along $\mathbf{r}_2$ and the vectors $\mathbf{e}_I$ and $\mathbf{e}_j$, to be parallel to the vector $\mathbf{d}$. This yields the following equations for the fields at the positions of the atoms:

$$E_{0j}' + iE_{0j}'' = E_{0I}\exp(-ik_0 r_j) + Ad(u_j + iv_j)\exp(-ik_0 R),$$

$$(11.23)$$

where

$$A = \frac{k_0^2}{R} + i\frac{k_0}{R^2} - \frac{1}{R^3},$$

with d being the magnitude of the dipole moment for the transition.

Thus, because the quantities u_j and v_j can be expressed in terms of field variables, we have a system of two complex, nonlinear algebraic equations to work with.

For weak fields satisfying the inequality

$$T_1 T_2' \varkappa_j^2 |E_{0I}|^2 \ll 1,\qquad (11.24)$$

the system (11.23) reduces to a linear algebraic system. The solutions of this system have been examined by us in detail in Ref. [69]. For strong fields, the inequality (11.24) is reversed, and the corresponding system has only one solution — which, however, is of no interest here.

Suppose now the fields to be such that

$$T_1 T_2' \varkappa^2 |E_{0I}|^2 \sim 1.$$

Also assume that the resonance frequency is

$$\omega_0 = 2.9 \times 10^{15} \text{ s}^{-1},$$

and the relaxation times are

$$T_1 = 10^{-3} \text{ s}, \quad T_2' = 10^{-8} \text{ s}, \quad d = 1.3 \times 10^{-18} \text{ CGSE}.$$

The numerical solution of the system (11.23) yields the complex amplitude of the electric field at each atom as a function of the wave vector k_0 for both the external field amplitude and the interatomic atomic separation held fixed. For certain values of E_{0I} and R, this function is multi-valued, i.e., several amplitudes of the electric field at atoms, or several field states, correspond to a given value of k_0. This phenomenon is known as optical multistability. Precisely how the system makes the transition to a particular state depends on which states the system has passed through before the transition — i.e., the phenomenon of hysteresis takes place. Let $R = 1$; then the numerical analysis of the solutions of the system shows that multistability occurs for external fields satisfying the inequality

$$10^{-2} < |E_{0I}| < 1 \text{ CGSE}.$$

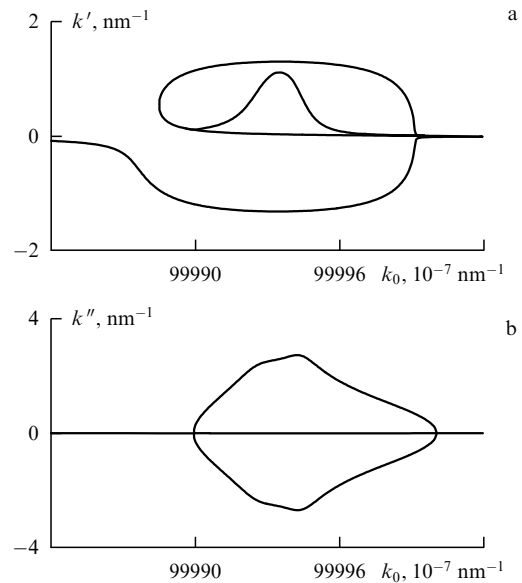Dispersion laws for waves excited in a two-atom small object are shown in Fig. 12a.

Figure 12. Dispersion of (a) the real and (b) the imaginary part of the wave vector **k**. The atomic separation is $R = 1.6$ nm and the external field strength is $E_{0I} = 0.5$ CGSE.

11.3 Optical recording of quantum information in a two-qubit quantum computer

It has been proven that only two elements (gates), one-qubit and two-qubit, are needed to construct a quantum computer [70]. The qubit — the unit of quantum information — is known to be a superposition of the quantum states of a certain system $|\psi\rangle = \alpha|0\rangle + \beta|1\rangle$, where $|0\rangle$, $|1\rangle$ are the wave functions of two states of the system, and α, β are quantum numbers, $|\alpha|^2 + |\beta|^2 = 1$ [71]. Gadomskiĭ [68] proposed a method by which quantum information can be recorded optically in a system of two atoms situated a short distance apart ($R \ll \lambda$, λ the light wavelength, R the atomic separation) using the inversions w_1 and w_2 of the first and second atoms of

the system to measure quantum information. The system was described using Eqns (11.22) and (11.1) for the atoms interacting in a steady-state manner with the field of an intense quasi-resonant radiation.

This problem was solved [68] numerically for various small-object excitation conditions. It was shown that for equal atomic frequencies $\omega_{01} = \omega_{02}$ the atomic inversions w_1 and w_2 were also equal for different angles φ between the vector $\mathbf{k}_0$ and the axis y passing through the two atoms. It was also shown that for $\omega_{01} = \omega_{02}$ and certain values of detuning, multipolarity appears due to the lack of uniqueness in the determination of the atomic inversions. Figure 13 shows the numerical results for the atomic inversions w_1 and w_2 for the case in which the first and second atoms differ somewhat in frequency. The atomic inversions may differ significantly in this case, and this fact can be used for recording quantum information on individual atoms in a quantum computer (which is a small object).
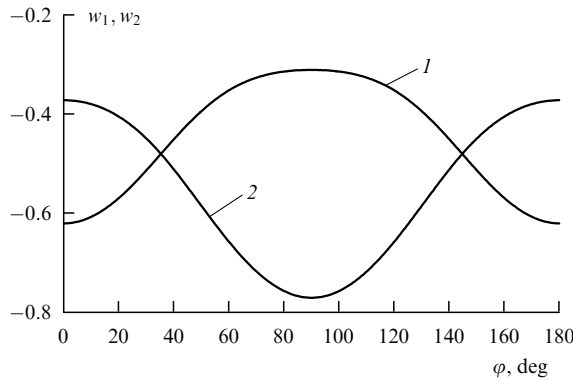
Figure 13. Dependence of the inversion of the atoms in a small object on the angle between the wave vector $\mathbf{k}_0$ of the external wave and the axis passing through the atoms. Curves *1* and *2* represent the inversions w_1 and w_2 of the first and second atoms, respectively.

11.4 Optical holography of small objects

Let us consider the interference of polarizing fields, Eqn (11.1), produced by two atoms of a small object, with a reference field with wave vector $\mathbf{k}_0'$ (Fig. 14) [72].

We start by calculating the field at observation points in the wave zone, where

$$k_0|\mathbf{r} - \mathbf{r}_1| \gg 1, \qquad k_0|\mathbf{r} - \mathbf{r}_2| \gg 1.$$

For such observation points, the terms proportional to $1/R_j$ are dominant in Eqn (11.7), and the electric and magnetic field strengths at $\mathbf{r}$ assume the respective forms [8]

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_I'(\mathbf{r}, t) - \frac{[\mathbf{n}_1 \times [\mathbf{n}_1 \times \ddot{\mathbf{p}}_1]]}{c^2 R_1} - \frac{[\mathbf{n}_2 \times [\mathbf{n}_2 \times \ddot{\mathbf{p}}_2]]}{c^2 R_2},$$

$$\tag{11.25}$$

$$\mathbf{H}(\mathbf{r}, t) = \mathbf{H}_I'(\mathbf{r}, t) + \frac{1}{c^2 R_1}[\ddot{\mathbf{p}}_1 \times \mathbf{n}_1] + \frac{1}{c^2 R_2}[\ddot{\mathbf{p}}_2 \times \mathbf{n}_2],$$

where $\mathbf{E}_I'$ and $\mathbf{H}_I'$ are, respectively, the electric and magnetic fields of the reference plane wave with wave vector $\mathbf{k}_0'$. For a plane wave we have [18]

$$\mathbf{H}_I' = \frac{1}{k_0'}[\mathbf{k}_0' \times \mathbf{E}_I'],$$
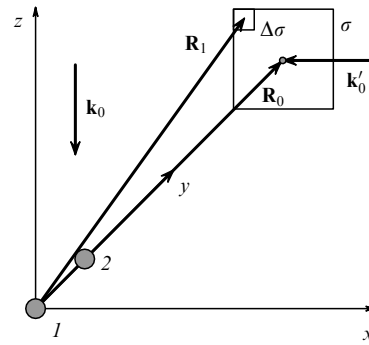
Figure 14. Optical scheme of the holographic recording of a small object consisting of two closely spaced atoms. The plane of the hologram is at a distance R_0 from the first atom and coincides with the xz plane; $\mathbf{k}_0$ is the wave vector of the object beam, $\mathbf{k}_0 \parallel \mathbf{z}$; $\mathbf{k}_0'$ is the wave vector of the reference beam, $\mathbf{k}_0' \perp \mathbf{z}$; $\Delta\sigma$ is an element of the plane hologram, located at a distance R_1 from the first atom, $R \ll \lambda$, $R_0 \gg \lambda$, and λ is the wavelength of the reference and the object radiation, both from one source.

where $k_0' = k_0$. Since the induced dipole moments for both atoms in a small object are given by Eqns (11.19) for the times $t - R_1/c$ and $t - R_2/c$, using Eqn (11.15) we obtain

$$\ddot{\mathbf{p}}_j^{\gamma} = -\omega^2 p_{0j}^{\gamma} \exp(ik_0 R_j) \exp(-i\omega t) + \text{c.c.}, \tag{11.26}$$

where the index γ refers to the x-, y- or z-components of the induced dipole moment of the jth atom, $R_j = |\mathbf{r} - \mathbf{r}_j|$.

Let $\Delta\sigma$ be a surface element in the neighborhood of the observation point $\mathbf{r}$ at the surface of the hologram. Then the intensity of optical radiation at this point is given by

$$I(\mathbf{r}) = \Delta\sigma \cdot \frac{c}{4\pi} \cdot I_{\sigma}(\mathbf{r}), \qquad I_{\sigma}(\mathbf{r}) = \left| [\mathbf{E} \times \mathbf{H}]_y \right|, \tag{11.27}$$

where the quantities $\mathbf{E}$ and $\mathbf{H}$ at $\mathbf{r}$ are given by Eqn (11.25). By changing the point of observation, we find the intensity distribution for the interfering fields in the plane of the hologram. Below we will present characteristic holographic pictures obtained from Eqn (11.27) for various conditions.

Formulas (11.15) contain denominators whose real parts can vanish as the external frequency ω or the separation R are varied. In this case the amplitudes of the induced dipole moments of the first and second atoms reach their maximum values. From Eqn (11.15), the conditions for this to occur are

$$\text{Re}\left\{\alpha_1 \alpha_2 G^2 \exp(2ik_0 R)\right\} = \frac{1}{4},$$

$$\text{Re}\left\{\alpha_1 \alpha_2 F^2 \exp(2ik_0 R)\right\} = 1, \tag{11.28}$$

depending on the orientation of the dipole moments relative to the coordinate axes.

Let us separate out resonances with wave vectors k_{01} and k_{02}, respectively, in the spectra of the first and second atoms in the small object. The polarizabilities α_1 and α_2 of the atoms are different according to Eqn (11.11). Note that $T_j = \gamma_{0j}^{-1}$, where $\gamma_{0j} = 2e^2 k_{0j}^2/(3mc)$ ($j = 1, 2$). Let

$$k_{01} = 89000 \text{ cm}^{-1}, \qquad k_{02} = 92000 \text{ cm}^{-1};$$

then

$$\gamma_{01} = 0.45 \times 10^8 \text{ s}^{-1}, \qquad \gamma_{02} = 0.48 \times 10^8 \text{ s}^{-1}.$$

In the case of exact resonance

$$\mathrm{Re}\,(\alpha_1) = 1.8 \times 10^{-16}\,\mathrm{s}^3\,, \quad \mathrm{Re}\,(\alpha_2) = 1.6 \times 10^{-16}\,\mathrm{s}^3\,,$$

$$\mathrm{Im}\,(\alpha_1) = 3.6 \times 10^{-16}\,\mathrm{cm}^3\,, \quad \mathrm{Im}\,(\alpha_2) = 3.2 \times 10^{-16}\,\mathrm{cm}^3\,.$$

Thus, in the absence of interaction between the atoms we have two isolated resonances, which correspond to two isotropic atoms.

The self-consistent atom-atom interaction determined by the solution (11.15) considerably modifies the spectroscopic properties of the atoms that constitute the small object. Numerical analysis shows that a small object of size $R = 1$ nm has four dimensional resonances at the following wave vectors:

$$k'_{01} = 86992.9\,\mathrm{cm}^{-1}\,, \qquad k'_{02} = 93900.2\,\mathrm{cm}^{-1}\,,$$

$$k'_{03} = 88326.3\,\mathrm{cm}^{-1}\,, \qquad k'_{04} = 92646.9\,\mathrm{cm}^{-1}\,.$$

The positions of the dimensional resonances on the axis of wave vectors depend strongly on the separation and polarizabilities α_1 and α_2 of the isolated atoms the small object is made of. The first and the second atom behave as anisotropic particles. The effective polarizabilities of the atoms, in the case of exact dimensional resonances, differ considerably from their isolated-atom resonance values α_1 and α_2. Furthermore, near the dimensional resonances

$$k'_{02} = 93900.2\,\mathrm{cm}^{-1}\,, \qquad k'_{03} = 88326.3\,\mathrm{cm}^{-1}$$

negative dispersion is exhibited by the first and the second atom.

Let us now examine some properties of the optical holograms which form when a two-atom small object is irradiated by a coherent object wave.

Figure 15 illustrates numerical calculations of the intensity (11.27) for a plane hologram at a distance $R_0 = 100$ cm from a small object. The frequencies of the object and reference waves are equal and match the dimensional resonances. The separation between the atoms in the small object is $R = 1$ nm. The phase plane of the reference wave is at an angle $\alpha_0 = 0$ to the plane of the hologram. Both the object and reference waves are linearly polarized, and the vectors $\mathbf{E}_{0I}$ and $\mathbf{E}'_{0I}$ have the following components:

$$\mathbf{E}_{0I}(E_{0I}, 0, 0)\,, \qquad \mathbf{E}'_{0I}(0, 0, E'_{0I})\,.$$

The angle θ between the vector $\mathbf{k}_0$ and $\mathbf{R}$ is $90°$. The field intensity in the plane of the hologram is expressed in units of $\Delta\sigma(c/(4\pi))$, where $\Delta\sigma = \Delta x \cdot \Delta z$. The area of the hologram is $L_x L_y = 1$ cm^2. Numerical calculations show the dimensional resonances to be narrow, with a width of about 0.1 cm^{-1}.

Let us next perform four numerical experiments taking the object and reference frequencies to match the exact dimensional resonances with the wave numbers

$$k'_{01} = 86992.9\,\mathrm{cm}^{-1}\,, \qquad k'_{02} = 93900.2\,\mathrm{cm}^{-1}\,,$$

$$k'_{03} = 88326.3\,\mathrm{cm}^{-1}\,, \qquad k'_{04} = 92646.9\,\mathrm{cm}^{-1}\,.$$

In this case the dimensional resonances

$$k'_{01} = 86992.9\,\mathrm{cm}^{-1}\,, \qquad k'_{02} = 93900.2\,\mathrm{cm}^{-1}$$
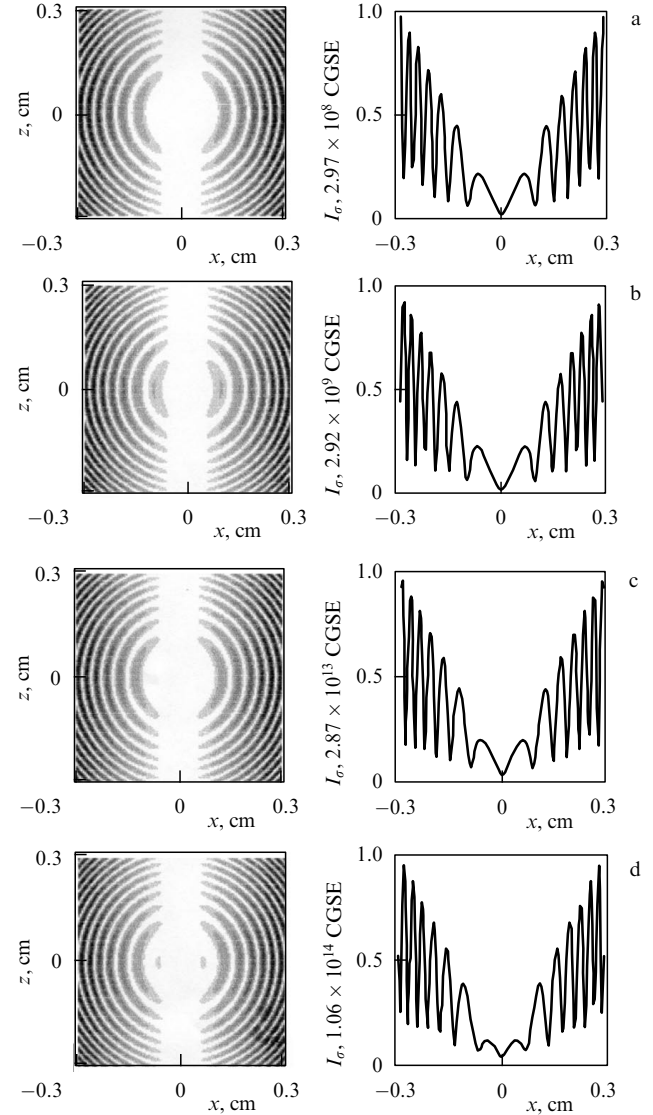

Figure 15. Distribution of the intensity I_σ in the plane xz of a hologram of a two-atom nanostructural object, for a normally incident object wave. $\bar{E}_{0I}$ has the coordinates $(0, E^y_{0I}, 0)$, $E^y_{0I} = 1$ CGSE unit, the distance R_0 from the object to the center of the hologram is 100 cm, and the coordinates of the wave vector $\bar{\mathbf{k}}'_0$ of the reference wave are $(0, 0, -k_0)$; $I_{\max} = (I_\sigma)_{\max}$ in CGSE units. The object and reference waves have equal frequencies, corresponding to four resonances with wave numbers (a) 86992.9 cm^{-1}, (b) 93900.2 cm^{-1}, (c) 88326.3 cm^{-1}, and (d) 92646.9 cm^{-1}. The atoms that constitute the object are different and have eigenfrequencies with wave numbers 89000 cm^{-1} and 92000 cm^{-1}. The interatomic separation in the object is 1 nm. The hologram measures 0.6×0.6 cm^2 in size.

separate out the y-components of the induced dipole moments of the first and second atoms in the small object, because the x- and z-components vanish for these values. Conversely, the dimensional resonances

$$k'_{03} = 88326.3\,\mathrm{cm}^{-1}\,, \qquad k'_{04} = 92646.9\,\mathrm{cm}^{-1}$$

separate out the x- and z-components of the induced dipole moments of the first and second atoms. Thus, using various dimensional resonances, it is possible to investigate the induced anisotropy of a small object.

12. Conclusion

Thus, this review presents equations, nonlocal in both space and time, that describe the propagation of photons [Eqns (5.11), (5.18)] and of classical fields [Eqns (6.1)–(6.3), (6.8), (6.16), (6.17), (6.30), (6.31)] taking into account the influence of the surrounding charges (electrons and atomic nuclei) on the process of the interaction between external radiation and an electron at a certain point of observation within the system.

These nonlocal equations are derived based on the quantum electrodynamical and semiclassical approaches. The former approach separates out those third-order effects which are accompanied by the exchange of virtual photons between atomic electrons and which involve the emission (absorption) of a real photon. The Feynman diagrams for these effects are shown in Fig. 1. It is shown that these are precisely the effects that control the refractive index, i.e., are responsible for the formation of a wave propagating in the optical medium with a wave vector different from that of the wave in vacuum. This fundamental statement is proved within the framework of the electric dipole approximation by going over from Eqn (5.11) to the integral equation for electromagnetic waves in an optical medium, the equation which is well known in classical optics and which, in turn, allows the Lorentz–Lorentz formula for the refractive index of the medium to be rigorously derived using the Ewald–Oseen procedure.

The third-order effects (see Fig. 1) can be represented, as shown in Section 3, in terms of polarizing fields which depend on the types of intermediate states involved and on the types of quantum transitions present in the spectrum of the interacting fields. Based on the concepts of polarizing fields, the following conclusions are made.

First, in the electric dipole approximation the polarizing field is produced by a dipole and is treated as a field of virtual photons. In a system of two atoms this field is of a nonlocal nature. The dipole of one of the atoms is excited by the external field at the position of this particular atom, and the transition with the emission (absorption) of a real photon occurs at the position of the other atom.

Second, depending on the types of quantum transitions occurring in the spectrum of interacting atoms, different photon propagation equations can be obtained, which can be represented as nonlocal integral equations for a continuous medium.

In the present review, some boundary value problems were solved to demonstrate the advantages of the nonlocal equations over Maxwell's local equations. In doing this, the existence of the near-field effect in the optics of dielectrics was proved theoretically, and from this fact the concept of a discrete-continuous dielectric with a structural factor as a necessary element was developed. This factor is most important in the surface layers of dielectrics, in superthin films, and also in small objects with linear dimensions much less than the light wavelength involved. It can also be argued that the near-field effect is a more general concept than the local field effect [26]. The latter is predicted theoretically to occur in nonlinear resonant optical phenomena as one goes over from the microscopic to the macroscopic field using the relations (8.2) in the modified Bloch equations [24]. The near field effect introduces the structural factor and is also taken into account by the relation (8.12) if an optical effect is, for example, considered in a superthin film consisting of resonant atoms [73].

Of particular interest from the point of view of the application of the nonlocal equations are so-called small objects, with linear dimensions much less than the wavelength of the light. This review considers two problems concerning two-atom small objects in the field of quasi-resonant radiation, for weak and for strong radiation. The solutions of these problems suggest important conclusions about the use of optical radiation for the study of small objects, for obtaining the holograms of such objects, depending on the processes inside them, and for using small objects as quantum computer components.

13. Appendix

A. The tensor $\widehat{\Pi}$ is described by the following, rather cumbersome, equations:

$$\widehat{\Pi}(x, y, z) = a_0\left[\widehat{\mathcal{L}}(z) + \widehat{T}(z)\right] + \widehat{S}(x, y, z),$$

$$\widehat{\Pi}(0) = a_0\widehat{J} + \widehat{S}(0),$$

$$\widehat{\mathcal{L}}(z) = \widehat{\Phi}^{-1}\widehat{\mathcal{L}}'(z)\widehat{\Phi}, \quad z \neq 0,$$

where the rotation matrix $\widehat{\Phi}$ has the form

$$\widehat{\Phi} = \begin{pmatrix} -\cos\Phi_I & -\sin\Phi_I & 0 \\ \sin\Phi_I & -\cos\Phi_I & 0 \\ 0 & 0 & 1 \end{pmatrix}.$$

The inverse matrix $\widehat{\Phi}^{-1}$ is obtained by the replacement $\Phi_I \to -\Phi_I$. The symmetric third-rank tensor $\widehat{\mathcal{L}}'(z)$ has the following components:

$$\mathcal{L}'_{xy}(z) = \mathcal{L}'_{zy}(z) = 0,$$

$$\mathcal{L}'_{xx}(z) = -2\pi\exp\left(ik_0\sqrt{\varepsilon^2 + z^2}\right)\varepsilon^2\frac{1 - ik_0\sqrt{\varepsilon^2 + z^2}}{(\varepsilon^2 + z^2)^{3/2}}\frac{J_1(\xi)}{\xi},$$

$$\mathcal{L}'_{yy}(z) = 2\pi\exp\left(ik_0\sqrt{\varepsilon^2 + z^2}\right)$$
$$\times \left\{\frac{\xi J_1(\xi)}{\sqrt{\varepsilon^2 + z^2}} - \varepsilon^2\frac{1 - ik_0\sqrt{\varepsilon^2 + z^2}}{(\varepsilon^2 + z^2)^{3/2}}\left[J_0(\xi) - \frac{J_1(\xi)}{\xi}\right]\right\},$$

$$\mathcal{L}'_{zz}(z) = \mathcal{L}'_{xx}(z) + \mathcal{L}'_{yy}(z),$$

$$\mathcal{L}'_{zx}(z) = 2\pi\exp\left(ik_0\sqrt{\varepsilon^2 + z^2}\right)i\varepsilon z\frac{1 - ik_0\sqrt{\varepsilon^2 + z^2}}{(\varepsilon^2 + z^2)^{3/2}}J_1(\xi),$$

where $\xi = k_0\varepsilon\sin\theta_I$ and J_n is the nth-order Bessel function.

The tensor $T(z)$ is defined by

$$\widehat{T}(z) = \widehat{\Phi}^{-1}\widehat{T}'(z)\widehat{\Phi},$$

where $\widehat{T}'(z)$ is a symmetric third-rank tensor having the following components:

$$T'_{xy}(z) = T'_{xy}(z) = 0,$$

$$T'_{xx}(z) = 2\pi\int_0^\varepsilon J_0\left(k_0\rho\sin\theta_I\right)G(R)$$
$$\times\left[\left(\frac{z}{R}\right)^2\left(-k_0^2 - i\frac{3k_0}{R} + \frac{3}{R^2}\right) + \left(ik_0 - \frac{1}{R}\right)\frac{1}{R}\right]\rho\,d\rho,$$

$$T'_{zz}(z) = -2\pi k_0^2 \sin^2\theta_I \int_0^\varepsilon J_0(k_0\rho\sin\theta_I)G(R)\rho\,\mathrm{d}\rho\,,$$

$$T'_{yy}(z) = T'_{xx}(z) + T'_{zz}(z)\,,$$

$$T'_{xz}(z) = 2\pi\mathrm{i}k_0 z\sin\theta_I$$
$$\times \int_0^\varepsilon J_0(k_0\rho\sin\theta_I)G(R)\left(\mathrm{i}k_0 - \frac{1}{R}\right)\frac{\rho}{R}\,\mathrm{d}\rho\,,$$

$$R = \sqrt{\rho^2 + z^2}\,.$$

Unlike the tensors $\widehat{\mathcal{L}}$ and $\widehat{T}$, which are functions of the coordinate z only, the tensor $\widehat{S}$ also depends on the coordinates of the point of observation in the (x, y) plane:

$$\widehat{S}(x,y,z) = \begin{pmatrix} S_{xx}(x,y,z) & S_{yx}(x,y,z) & S_{zx}(x,y,z) \\ S_{yx}(x,y,z) & S_{yy}(x,y,z) & S_{zy}(x,y,z) \\ S_{zx}(x,y,z) & S_{zy}(x,y,z) & S_{zz}(x,y,z) \end{pmatrix},$$

$$S_{kl}(x,y,z) = a_0^3 \sum_{a_j} G(R_{a_j})$$
$$\times \exp\left[-\mathrm{i}k_0 R_{a_j}\sin\theta_I(\cos\Phi_I n_{a_jx} + \sin\Phi_I n_{a_jy})\right]$$
$$\times \left[k_0^2(\delta_{kl} - n_{a_jk}n_{a_jl}) - \mathrm{i}k_0\frac{3n_{a_jk}n_{a_jl} - \delta_{kl}}{R_{a_j}} + \frac{3n_{a_jk}n_{a_jl} - \delta_{kl}}{R_{a_j}^2}\right],$$

$$R_{a_j} = \sqrt{(x - x_{a_j})^2 + (y - y_{a_j})^2 + z^2}\,,$$

$$n_{a_jx} = \frac{x - x_{a_j}}{R_{a_j}}\,, \qquad n_{a_jy} = \frac{y - y_{a_j}}{R_{a_j}}\,, \qquad n_{a_jz} = \frac{z}{R_{a_j}}\,,$$

where δ_{kl} is the Kronecker delta. The summation runs over the atoms of the jth layer lying within the sphere of radius ε centered at the observation point.

The tensor $\widehat{S}$ has a translational symmetry:

$$\widehat{S}(x + a_{0x}, y, z) = \widehat{S}(x + a_{0x}, y + a_{0y}, z)$$
$$= \widehat{S}(x, y + a_{0y}, z) = \widehat{S}(x, y, z)\,,$$

where a_{0x} and a_{0y} are the lattice constants in the directions x and y, respectively. In the calculations of this paper, $a_{0x} = a_{0y} = a_0$.

The tensor $\widehat{J}$ has the form
$$\widehat{J} = \widehat{\Phi}^{-1}\widehat{J}'\widehat{\Phi}\,;$$

$\widehat{J}'$ is a diagonal tensor with the components

$$J'_{xx} = 2\pi\frac{\exp(\mathrm{i}k_0\varepsilon)}{\varepsilon}\left[J_0(\xi) + \frac{J_1(\xi)}{\xi}(\mathrm{i}k_0\varepsilon - 1)\right]$$
$$+ 2\pi k_0\sin^2\theta_I\left[k_0\int_0^\varepsilon \exp(\mathrm{i}k_0\rho)\frac{J_1(a)}{a}\,\mathrm{d}\rho - \frac{\mathrm{i}}{1 + \cos\theta_I}\right],$$

$$J'_{yy} = 2\pi\frac{\exp(\mathrm{i}k_0\varepsilon)}{\varepsilon}(1 - \mathrm{i}k_0\varepsilon)\frac{J_1(\xi)}{\xi}$$
$$+ 2\pi k_0\left[\frac{\mathrm{i}}{\cos\theta_I} - k_0\int_0^\varepsilon \exp(\mathrm{i}k_0\rho)J_0(a)\,\mathrm{d}\rho\right],$$

$$J'_{zz} = -2\pi\frac{\exp(\mathrm{i}k_0\varepsilon)}{\varepsilon}J_0(\xi)$$
$$+ 2\pi k_0\left[\frac{\mathrm{i}}{\cos\theta_I} - k_0\int_0^\varepsilon \exp(\mathrm{i}k_0\rho)J_0(a)\,\mathrm{d}\rho\right]$$
$$- 2\pi k_0\sin^2\theta_I\left[k_0\int_0^\varepsilon \exp(\mathrm{i}k_0\rho)\frac{J_1(a)}{a}\,\mathrm{d}\rho - \frac{\mathrm{i}}{1 + \cos\theta_I}\right],$$

where $\xi = k_0\varepsilon\sin\theta_I$

B. The tensor $\widehat{\Pi}$ in Eqns (9.5) and (9.10) is given by

$$\widehat{\Pi}(z) = (z_{j-1} - z_j)[\widehat{\mathcal{L}}(z) + \widehat{T}(z)] + \widehat{S}(z)\,, \qquad z \neq 0\,,$$

$$\widehat{\Pi}(0) = (z_{j-1} - z_j)\widehat{J} + \widehat{S}(0)\,, \qquad z = 0\,.$$

The symmetric third-rank tensor $\widehat{\mathcal{L}}(z)$ has the following components:

$$\mathcal{L}_{xy}(z) = \mathcal{L}_{zy}(z) = 0\,,$$

$$\mathcal{L}_{xx}(z) = -2\pi\exp(\mathrm{i}k_0 L_0)(L_0^2 - z^2)\frac{1 - \mathrm{i}k_0 L_0}{L_0^3}\frac{J_1(\xi)}{\xi}\,,$$

$$\mathcal{L}_{yy}(z) = 2\pi\exp(\mathrm{i}k_0 L_0)\left\{\frac{\xi J_1(\xi)}{L_0} - (L_0^2 - z^2)\frac{1 - \mathrm{i}k_0 L_0}{L_0^3}\right.$$
$$\left.\times\left[J_0(\xi) - \frac{J_1(\xi)}{\xi}\right]\right\},$$

$$\mathcal{L}_{zz}(z) = \mathcal{L}_{xx}(z) + \mathcal{L}_{yy}(z)\,,$$

$$\mathcal{L}_{zx}(z) = 2\pi\exp(\mathrm{i}k_0 L_0)\,\mathrm{i}z\sqrt{L_0^2 - z^2}\,\frac{1 - \mathrm{i}k_0 L_0}{L_0^3}J_1(\xi)\,,$$

where $\xi = k_0\sqrt{L_0^2 - z^2}\sin\theta_I$, and J_n is the nth-order Bessel function.

$\widehat{T}(z)$ is a symmetric third-rank tensor:

$$T_{xy}(z) = T_{xy}(z) = 0\,,$$

$$T_{xx}(z) = 2\pi\int_0^{\sqrt{L_0^2 - z^2}} J_0(k_0\rho\sin\theta_I)G(R)$$
$$\times \left[(zR)^2\left(-k_0^2 - \mathrm{i}\frac{3k_0}{R} + \frac{3}{R^2}\right) + \left(\mathrm{i}k_0 - \frac{1}{R}\right)\frac{1}{R}\right]\rho\,\mathrm{d}\rho\,,$$

$$T_{zz}(z) = -2\pi k_0^2\sin^2\theta_I\int_0^{\sqrt{L_0^2 - z^2}} J_0(k_0\rho\sin\theta_I)G(R)\rho\,\mathrm{d}\rho\,,$$

$$T_{yy}(z) = T_{xx}(z) + T_{zz}(z)\,,$$

$$T_{xz}(z) = 2\pi\mathrm{i}k_0 z\sin\theta_I\int_0^{\sqrt{L_0^2 - z^2}} J_0(k_0\rho\sin\theta_I)G(R)$$
$$\times \left(\mathrm{i}k_0 - \frac{1}{R}\right)\frac{\rho}{R}\,\mathrm{d}\rho\,,$$

where $R = \sqrt{\rho^2 + z^2}$.

$\widehat{S}$ is a symmetric tensor with the components

$$S_{kl}(z) = \frac{1}{N}\sum_{a_j} G(R_{a_j})\exp\left[-\mathrm{i}k_{Ix}(x - x_{a_j})\right]$$
$$\times \left[k_0^2(\delta_{kl} - n_{a_jk}n_{a_jl}) - \mathrm{i}k_0\frac{3n_{a_jk}n_{a_jl} - \delta_{kl}}{R_{a_j}}\right.$$
$$\left.+ \frac{3n_{a_jk}n_{a_jl} - \delta_{kl}}{R_{a_j}^2}\right],$$

$$R_{a_j} = \sqrt{(x - x_{a_j})^2 + (y - y_{a_j})^2 + z^2}\,,$$

where δ_{kl} is the Kronecker delta. The summation runs over the atoms of the jth layer lying within the sphere of radius $(L_0^2 - z^2)^{1/2}$.

$\widehat{J}$ is a diagonal tensor with the components

$$J_{xx} = 2\pi\frac{\exp\left(\mathrm{i}k_0\sqrt{L_0^2 - z^2}\right)}{\sqrt{L_0^2 - z^2}}$$
$$\times \left[J_0(\xi) + \frac{J_1(\xi)}{\xi}\left(\mathrm{i}k_0\sqrt{L_0^2 - z^2} - 1\right)\right] + 2\pi k_0\sin^2\theta_I$$
$$\times \left[k_0\int_0^{\sqrt{L_0^2 - z^2}} \exp(\mathrm{i}k_0\rho)\frac{J_1(a)}{a}\,\mathrm{d}\rho - \frac{\mathrm{i}}{1 + \cos\theta_I}\right],$$

$$J_{yy} = 2\pi \frac{\exp\left(\mathrm{i}k_0\sqrt{L_0^2 - z^2}\right)}{\sqrt{L_0^2 - z^2}} \left(1 - \mathrm{i}k_0\sqrt{L_0^2 - z^2}\right) \frac{J_1(\xi)}{\xi}$$

$$+ 2\pi k_0 \left[\frac{i}{\cos\theta_I} - k_0 \int_0^{\sqrt{L_0^2 - z^2}} \exp(\mathrm{i}k_0\rho) J_0(a)\,\mathrm{d}\rho\right],$$

$$J_{zz} = -2\pi \frac{\exp\left(\mathrm{i}k_0\sqrt{L_0^2 - z^2}\right)}{\sqrt{L_0^2 - z^2}} J_0(\xi)$$

$$+ 2\pi k_0 \left[\frac{i}{\cos\theta_I} - k_0 \int_0^{\sqrt{L_0^2 - z^2}} \exp(\mathrm{i}k_0\rho) J_0(a)\,\mathrm{d}\rho\right]$$

$$- 2\pi k_0 \sin^2\theta_I \left[k_0 \int_0^{\sqrt{L_0^2 - z^2}} \exp(\mathrm{i}k_0\rho) \frac{J_1(a)}{a}\,\mathrm{d}\rho - \frac{i}{1 + \cos\theta_I}\right],$$

where $\xi = k_0\sqrt{L_0^2 - z^2}\sin\theta_I$, $a = k_0\rho\sin\theta_I$.

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