Perturbation approach to the reflection and transmission of light

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A perturbation approach to the calculation of the optical response of a crystalline slab within the framework of phenomenological electrodynamics is presented. Analytical expressions for the change in the reflection and transmission coefficients of the slab due to a small perturbation are derived. The main advantage of the approach is that it takes into account both surface and bulk contributions to optical effects. The theory is well suited to the analysis of experiments in which the optical effects due to the spatial dispersion of a dielectric tensor are measured. We apply the derived formulas to the analysis of the nonreciprocal reflection of light from magnetoelectric Cr_2O_3.

I. INTRODUCTION

In investigations of solids by optical methods one often deals with reflection and transmission effects of a small magnitude. Such effects may be considered as originated from a small addition $\delta\hat{\epsilon}$ to some background optical dielectric tensor $\hat{\epsilon}^0$. This small addition may be due to both external perturbations and/or some internal interactions. Despite the smallness of $\delta\hat{\epsilon}$, it often considerably complicates calculations of corresponding optical effects. If $\delta\hat{\epsilon}$ is a local function of coordinates, then this difficulty is purely algebraic in its nature and can be overcome by making certain approximations. If, however, $\delta\hat{\epsilon}$ is nonlocal we face the well-known difficulty of principle. The point is that the boundary conditions for electric and magnetic fields of light waves needed for the calculation of an optical response cannot be found without going into details of light interaction with matter inside a thin surface layer (see, for example, Refs. 1 and 2). When $\delta\hat{\epsilon}$ is nonlocal, then the relative surface contribution to corresponding optical effects may be appreciable and should be taken into account in a proper way. This difficulty has been known for a long time and has manifested itself again in the recent search for a breakdown of time-reversal symmetry in high-T_c superconductors,[3–8] where theoretical calculations of the optical effects must be consistent with the Onsager symmetry principle. To avoid this difficulty a symmetry approach to the reflection and transmissiom of light was proposed.[9,10] Being absolutely rigorous, this approach has proved to be very useful in analyzing experimental data. A limitation of this approach is an inability to get analytical expressions for reflection and transmission coefficients in terms of a few physical parameters characterizing the optical properties of a medium.

The purpose of this paper is to obtain analytical expressions for the reflection and transmission coefficients for a spatially dispersive medium with arbitrary symmetry within the framework of phenomenological electrodynamics. More precisely, we shall get expressions for variations of these coefficients caused by a small but otherwise arbitrary variation $\delta\hat{\epsilon}$ of the dielectric tensor of a medium which are valid up to the first order in $\delta\hat{\epsilon}$. Thus our theory is a perturbation theory. The derived expressions for the optical response will automatically satisfy the Onsager symmetry principle.

The problem we consider here is closely related with calculations of surface corrections to the reflection and transmission coefficients. This last problem was studied in many papers, and among them we note those where the Green's function method was used.[11–16] The Green's function formalism is a general basis for a perturbation treatment and can be applied to our problem. However, we choose another approach, which is more simple and directly leads to the sought-after result.

The formal derivation of expressions for the reflection and transmission coefficients is presented in Sec. II. The derived formulas are especially suitable for an analysis of optical experiments where effects of the spatial dispersion are studied. In Sec. III we demonstrate this by analyzing the nonreciprocal reflection of light from magnetoelectric Cr_2O_3.

II. THEORY

We consider monochromatic light incident from the left on a crystalline slab of thickness d. We take the z axis to be perpendicular to the slab and the incident wave vector $\mathbf{k}$ to be in the xz plane [see Fig. 1(a)]. The slab occupies the space $0 < z < d$. We assume the slab obeys macroscopic parallel translational symmetry, so that after the cell-averaging along the surface the x dependence of the fields has the form $e^{ik_x x}$, where k_x is the projection of the wave vector of the incident wave onto the x axis. The electric field of the light wave may be represented in the form

$$\mathbf{E}(x,z) = e^{ik_x x}\begin{cases}\mathbf{I}e^{ik_z z} + \mathbf{R}e^{-ik_z z}, & z < 0\\ \mathbf{T}(z), & z > 0,\end{cases} \tag{1}$$

where $\mathbf{I}$, $\mathbf{R}$, and $\mathbf{T}$ represent the incident, reflected, and transmitted waves, respectively. Here and in the following we omit the factor $e^{-i\omega t}$ and the frequency argument.

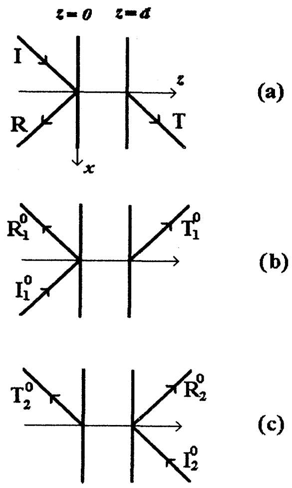

FIG. 1. Geometry of reflection-transmission from a slab. (a) Original problem with the perturbed dielectric tensor $\hat{\epsilon} = \hat{\epsilon}^0 + \delta\hat{\epsilon}$. Corresponding solution of Maxwell's equations is denoted by $\mathbf{E}(x, z)$ in the text. (b) and (c) illustrate the origin of the two auxiliary solutions $\mathbf{E}_1^0$ and $\mathbf{E}_2^0$, respectively, for the unperturbed problem.

For $z > d$ $\mathbf{T}(z) = \mathbf{T}\exp(ik_z z)$. Note that inside the slab the field (1) contains a microscopic part, i.e., a part rapidly varying with z through a crystal cell. Due to the presence of the boundaries the macroscopic field cannot be unambiguously defined.[15]

The starting point of our analytical calculations is the well-known[1] bilinear relation between two arbitrary free-of-source solutions $\mathbf{E}^0$ and $\mathbf{E}$ of Maxwell equations

$$\operatorname{div}\mathbf{N} = i\,\frac{\omega}{c}\,(\mathbf{D}\mathbf{E}^0 - \mathbf{D}^0\mathbf{E}), \tag{2}$$

where $\mathbf{N} = (\mathbf{E}^0 \times \mathbf{B} - \mathbf{B}^0 \times \mathbf{E})$; $\mathbf{B}$ and $\mathbf{D}$ are magnetic induction and displacement vectors, respectively. We assume that $\mathbf{H} = \mathbf{B}$ and all magnetic effects are incorporated in $\mathbf{D}$, i.e., all induced currents are included in the definition of the displacement vector through the material relation $\mathbf{D}(\mathbf{r}) = \int d\mathbf{r}'\,\hat{\epsilon}(\mathbf{r}, \mathbf{r}')\mathbf{E}(\mathbf{r}')$, where $\hat{\epsilon}(\mathbf{r}, \mathbf{r}')$ is an optical dielectric tensor. We assume that the solutions $\mathbf{E}^0$ and $\mathbf{E}$ in Eq. (2) correspond to the dielectric tensors of the slab $\hat{\epsilon}^0$ and $\hat{\epsilon} = \hat{\epsilon}^0 + \delta\hat{\epsilon}$, respectively. Here, $\delta\hat{\epsilon}$ is a dielectric perturbation.

To be more concrete we assume the tensor $\hat{\epsilon}^0$ is time-odd (i.e., all optical effects with the unperturbed medium have to be reciprocal) and its dependence on x and y is local, i.e., $\epsilon^0_{ik}(\mathbf{r}, \mathbf{r}') = \epsilon^0_{ik}(z, z')\delta(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel})$, where $\mathbf{r}_{\parallel} = (x, y)$. Note, that our consideration can be easily extended to the tensor ϵ^0_{ik} of a quite general form. An arbitrary dependence of ϵ^0_{ik} on z is allowed, in particular, the slab may have a multilayer structure. The tensor $\delta\hat{\epsilon}$ may be nonlocal and in general has a coordinate dependence $\delta\hat{\epsilon}_{ik}(\mathbf{r}, \mathbf{r}') = \delta\hat{\epsilon}(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}; z, z')$.

Our key assumption is that the problem with $\hat{\epsilon}^0$ can be solved exactly for the fields $\mathbf{E}^0$ and $\mathbf{B}^0$, and now we use the relation (2) to obtain the solutions of the reflection and transmission problems for the case where $\hat{\epsilon} = \hat{\epsilon}^0 + \delta\hat{\epsilon}$. To this end we introduce two auxiliary solutions for the unperturbed problem. In the first of them, $\mathbf{E}_1^0$, the incident and reflected beams are interchanged as compared with Eq. (1) [see Fig. 1(b)]. This solution has the same form as (1), except k_x must be replaced by $-k_x$:

$$\mathbf{E}_1^0(x, z) = e^{-ik_x x}\begin{cases}\mathbf{I}_1^0 e^{ik_z z} + \mathbf{R}_1^0 e^{-ik_z z}, & z < 0\\ \mathbf{T}_1^0(z), & z > 0.\end{cases} \tag{3}$$

The second auxiliary solution, $\mathbf{E}_2^0$, corresponds to the light incident on the slab from the right and having the direction opposite to the direction of the incident light in Eq. (1) [see Fig. 1(c)]. This solution may be represented as

$$\mathbf{E}_2^0(x, z) = e^{-ik_x x}\begin{cases}\mathbf{I}_2^0 e^{-ik_z z} + \mathbf{R}_2^0 e^{ik_z z}, & z > d\\ \mathbf{T}_2^0(z), & z < d.\end{cases} \tag{4}$$

We can make the physical meaning of the solutions $\mathbf{E}_1^0$ and $\mathbf{E}_2^0$ more clear if we notice that in an experimental setup corresponding to our problem there are one source and two detectors: the first of them measures the intensity of the reflected wave and the second measures the intensity of the transmitted wave. If we put the source onto the place of the detector 1 or 2 (and set $\delta\hat{\epsilon} = 0$) we obtain the solution $\mathbf{E}_1^0$ or $\mathbf{E}_2^0$, respectively.

Now we substitute $\mathbf{E}_1^0$ into Eq. (2) for $\mathbf{E}^0$ and integrate this relation over z from 0 to d. Due to specific choice of $\mathbf{E}$ and $\mathbf{E}^0$ the relation (2) becomes independent of x and the values $N_z(0)$ and $N_z(d)$ can be expressed via the reflectivity (2×2) matrix $r_{\alpha\beta}$, which couples the amplitudes of the incident and reflected beams: $R_\alpha = r_{\alpha\beta} I_\beta$, where $\alpha(\beta)$ denotes the s or p component of the reflected (incident) wave. After simple but somewhat lengthy algebra we obtain $N_z(d) = 0$ and

$$2k_z(r_{\alpha\beta} - r^0_{\alpha\beta})I^0_{1\alpha}I_\beta = i\,\frac{\omega}{c}\int_0^d dz\,d\mathbf{r}'\,\mathbf{E}_1^0(\mathbf{r})\delta\hat{\epsilon}(\mathbf{r}, \mathbf{r}')\mathbf{E}(\mathbf{r}'), \tag{5}$$

where $r^0_{\alpha\beta}$ is the reflectivity matrix for the unperturbed problem. In deriving relation (5) we have taken into account that $r^0_{\alpha\beta} = r^0_{1\beta\alpha}$ (see Ref. 9), where $r^0_{1\alpha\beta}$ connects the reflected and incident waves in the solution $\mathbf{E}_1^0$, Eq. (3). We note that the integrand in Eq. (5), in fact, does not depend on x and it could be transformed to a form in which this independence of x would be apparent. However, for further applications it is more convenient to retain the dependence of all the functions in the integrand of Eq. (5) on x (see Sec. III).

Now we take the simplest form of the perturbation theory and replace the electric field $\mathbf{E}$ in the integrand in Eq. (5) by the field $\mathbf{E}^0$. Thus we obtain a simple expression for the change of the reflectivity matrix $\delta r_{\alpha\beta} = r_{\alpha\beta} - r^0_{\alpha\beta}$ due to $\delta\hat{\epsilon}$ which is valid up to the first order in $\delta\hat{\epsilon}$:

$$2k_z\delta r_{\alpha\beta}I^0_{1\alpha}I_\beta = i\,\frac{\omega}{c}\int_0^d dz\,d\mathbf{r}'\,\mathbf{E}_1^0(\mathbf{r})\delta\hat{\epsilon}(\mathbf{r}, \mathbf{r}')\mathbf{E}^0(\mathbf{r}'). \tag{6}$$

The expression for the change in the transmission matrix

$t_{\alpha\beta}$, defined through the relation $T_\alpha = t_{\alpha\beta} I_\beta$, can be obtained quite analogously if we substitute $\mathbf{E}_2^0$ into Eq. (2) for $\mathbf{E}^0$ and perform operations similar to those used in deriving Eq. (6). The result is

$$2k_z \delta t_{\alpha\beta} I_{1\alpha}^0 I_\beta = i\,\frac{\omega}{c}\int_0^d dz\, d\mathbf{r}'\, \mathbf{E}_2^0(\mathbf{r})\delta\hat{\epsilon}(\mathbf{r},\mathbf{r}')\mathbf{E}^0(\mathbf{r}'). \quad (7)$$

In deriving Eqs. (6) and (7) we have avoided the problem with the boundary conditions. Instead, however, we need to know the tensor $\delta\hat{\epsilon}(\mathbf{r},\mathbf{r}')$ within a thin surface layer.

Of course, the first-order perturbation theory and, consequently, Eqs. (6) and (7) are not always applicable, even if $\delta\hat{\epsilon}$ is small as compared with $\hat{\epsilon}^0$. Though we cannot give a rigorous mathematical formulation for the applicability of Eqs. (6) and (7), usually it is not too difficult to do this for every concrete problem. An example of such a problem is the total internal reflection from a chiral medium.[17] Despite existing limitations, there is a wide class of problems such as investigations of symmetry changes at phase transitions by optical methods, which may be solved by using the presented approach.

A comment concerning the above-mentioned possibility of applying the Green's function method to our problem is in order here. The expressions for the reflected and transmitted waves explicitly containing Green's functions[11–16] are inconvenient for calculations of the optical response, especially for an anisotropic medium and oblique incidence. The advantage of the presented approach is that it avoids from the beginning the explicit use of the Green's functions and directly leads to the relatively simple expressions (6) and (7) for the optical response. Though the difference between both approaches is not a difference of principle, it is essential for a practical usage.

As we have already mentioned, all the fields entering Eqs. (5)–(7) contain microscopic, i.e., short-wavelength parts. For a practical usage of Eqs. (6) and (7) it is necessary to introduce the macroscopic description of light propagation in the slab. The presence of the boundaries complicates the issue. The point is that the components E_z, D_x, and D_y rapidly vary across the boundary. For this reason the separation of these components into short- and long-wavelength parts has no physical meaning. However, the components D_z, E_x, and E_y, slowly varying across the boundary (if the first-order perturbation theory is applicable), can be partitioned into the microscopic and macroscopic parts in the unique way. Consequently, for one of the two factors in the products of the type $D_i^0 E_i$ [see Eq. (2)] the macroscopic part can be unambiguously defined. Due to this fact, the macroscopic description is still possible. Indeed, the displacement vector $\mathbf{D}$ is transverse and this is also true about its short-wavelength part, $\mathbf{D}_{\text{mic}}$, but the short-wavelength part of the electric field, $\mathbf{E}_{\text{mic}}$, is almost longitudinal.[1] Hence, *after the integration over* z, the scalar products of the type $\mathbf{D}_{\text{mic}}^0\mathbf{E}_{\text{mic}}$ contribute negligibly small to the right-hand sides of Eqs. (5)–(7). [Remember that the fields (1), (3), and (4) have been already cell-averaged along the surface.] The terms linear in these microscopic parts disappear after the integration over z. Consequently, we may consider all the fields as macroscopic and use the macroscopic representation for the tensor $\delta\hat{\epsilon}$ to express the reflection and transmission coefficients in terms of a few phenomenological parameters.

The above arguments are only qualitative, but, in principle, Eq. (5) may serve as a starting point for a definition of macroscopic parameters describing the optical response of the slab in terms of a microscopic dielectric function. It must be emphasized that small variations of D_z, E_x, and E_y across the boundary are the key condition for a macroscopic description of light reflection and, as a consequence, are a common feature of the other approaches.[12–16]

III. APPLICATION TO NONRECIPROCAL REFLECTION FROM Cr_2O_3

The nonreciprocal (NR) reflection from antiferromagnetic magnetoelectric Cr_2O_3 was predicted theoretically[18] and observed experimentally.[19] The essence of the effect is that the ellipticity and rotation of the reflected light change their signs when so do all the spins of Cr^{3+} ions, that is, nonreciprocal effects explicitly probe a violation of the time-reversal symmetry. As it has been already pointed out,[19] the surface plays an important role in these effects and should be properly taken into account. In this section we calculate the NR optical response of semi-infinite Cr_2O_3 and demonstrate the advantage of our approach in analyzing such types of problems.

In order to calculate $r_{\alpha\beta}$ by using Eq. (6) we need an explicit form for the tensor $\delta\hat{\epsilon}$. In media with zero net magnetic moment, the optical nonreciprocal effects, if any, may be due only to the spatial dispersion of an optical dielectric tensor, i.e., spatial derivatives must be included in material relations. A special feature of magnetoelectrics is that three-dimensional parity symmetry is broken. Therefore, the lowest, i.e., the first-order derivatives of the fields are sufficient to describe the magnetoelectric (ME) effect in optics. Extending the Onsager symmetry principle to an inhomogeneous medium[20,21] with broken time-reversal symmetry and zero net magnetic moment we can write[19]

$$\int d\mathbf{r}'\,\delta\hat{\epsilon}_{ij}(\mathbf{r},\mathbf{r}')\mathbf{E}_j(\mathbf{r}') = \frac{1}{2}\frac{\partial[\gamma_{ikl}^s(\mathbf{r})+\gamma_{ikl}^a(\mathbf{r})]}{\partial r_l}E_k(\mathbf{r}) + \gamma_{ikl}^s(\mathbf{r})\frac{\partial E_k(\mathbf{r})}{\partial r_l}, \quad (8)$$

where $\gamma_{ikl}(\mathbf{r})$ is a time-odd tensor of rank three. Superscripts s and a denote, respectively, symmetrical and antisymmetrical, in permutations of the indices i and k, parts of the tensor γ_{ikl}. The arguments given at the end of Sec. II allow us to neglect other than the first-order derivatives in Eq. (8). As seen from Eq. (8), transmission optical effects in a homogeneous medium do not depend on γ_{ikl}^a. Consequently, this tensor describes the influence of the boundary on the optical response.

Below $T_N = 307\,\text{K}$, where the tensor γ_{ikl} is nonzero, Cr_2O_3 has the magnetic point group $\bar{3}'m'$ with spins of

Cr^{3+} ions pointing along the threefold axis C_3 in an alternating manner. We consider the normal incidence reflection and take the orientation of the crystal axes such that $z \parallel U_2$ (twofold axis) and $x \parallel C_3$.

Mathematically the nonreciprocity in reflection manifests itself as a linear dependence of the r-matrix elements on the tensor γ_{ikl}. For normal incidence only the off-diagonal elements r_{xy} and r_{yx} may be time-odd;[9] therefore, we calculate $\delta r_{xy} = -\delta r_{yx}$.

Substituting expression (8) in Eq. (6) and using the properties of the zero-order solution for the semi-infinite crystal,[18] we easily obtain

$$\delta r_{xy} = \frac{i}{(1+n_x)(1+n_y)} \left(\gamma^a_{xyz} + \gamma^s_{xyz} \frac{n_x - n_y}{n_x + n_y} \right), \quad (9)$$

where n_x and n_y are indices of refraction for light waves polarized along the x and y axes, respectively. In deriving Eq. (9) we have used simple relations[18] between the amplitudes of the incident I^0_x and transmitted E^0_x waves at $z = 0$: $E^0_x = 2I^0_x/(1+n_x)$ and $E^0_y = 2I^0_y/(1+n_y)$. In addition, we suppose that the spatial width of the surface transition layer [where $\gamma^s_{ikl}(\mathbf{r})$ and $\gamma^a_{ikl}(\mathbf{r})$ vary rapidly] is much less than the wavelength of light. The components γ^s_{xyz} and γ^a_{xyz} are responsible for the NR rotation and ellipticity of the reflected light. The former quantity may be measured in transmission experiments, but the latter describes the influence of the surface on reflection. It is important to emphasize that this surface term is not a small correction to the bulk one. These two terms are, in general, of the same order of magnitude.[19]

It is interesting to compare the reflection coefficient (9) with the result obtained in Ref. 18 by solving Maxwell's equations together with boundary conditions. These boundary conditions have the tangential components of $\mathbf{E}$ and $\mathbf{H} = \mathbf{B} - \hat{\alpha}\mathbf{E}$ continuous across the boundary. Here $\hat{\alpha}$ is the magnetoelectric tensor at optical frequencies. If we set

$$\gamma^s_{xyz} = \frac{1}{2}(\alpha_{xx} - \alpha_{yy}), \quad (10a)$$

$$\gamma^a_{xyz} = \frac{1}{2}(\alpha_{xx} + \alpha_{yy}), \quad (10b)$$

and substitute these expressions in Eq. (9), we obtain for δr_{xy} exactly the same result as in Ref. 18. However, as it follows from Eqs. (10), the optical ME tensor $\hat{\alpha}$ defined through the usual material relations[18,22]

$$\mathbf{D} = \hat{\epsilon}^0\mathbf{E} + \hat{\alpha}\mathbf{H}, \quad (11)$$

$$\mathbf{B} = \mathbf{H} + \hat{\alpha}^T\mathbf{E} \quad (12)$$

$(\alpha^T_{ij} = \alpha_{ji})$ may not be considered as a purely bulk quantity, since the component γ^a_{xyz} is influenced by the surface. The representation (9) [and the general formulas (6) and (7)] allow one to get information about the surface contribution to the optical response if the bulk parameters are known from transmission experiments.

The calculations in the considered example are rather simple due to the absence of the normal component E_z of the electric field. At oblique incidence the E_z is nonzero and the integration over z in Eq. (6) becomes somewhat more complex. Fortunately, there is a simple way to resolve the difficulty,[12,13] expressing E_z in terms of the components D_z, E_x, and E_y which are slowly varying across the boundary.

IV. CONCLUSION

We have derived the expressions for the optical response of a media with a nonlocal optical dielectric tensor. The only assumption made in our derivation was that the nonlocal part $\delta\hat{\epsilon}$ of the total dielectric tensor $\hat{\epsilon} = \hat{\epsilon}^0 + \delta\hat{\epsilon}$ can be treated within the framework of the first-order perturbation theory. There is a wide variety of real systems for which such separation is justified.

The distinctive property of systems with a nonlocal optical dielectric tensor is the relatively large influence of the surface on an optical response. The approach we presented in this paper is especially suited for such systems, since it permits one to obtain a symmetry-allowed parametrization of the optical response in the form in which the surface contribution is separated from the bulk one. In particular, the expressions (6) and (7) can be applied to the cases where the bulk contribution to optical effects equals zero. For example, the bulk contribution to the NR reflection from antiferromagnetic La_2CuO_4 equals zero,[22] but it is not the case for the surface contribution, since antiferromagnetic ordering in this crystal does not break translational invariance along the surface (assuming surface normal to be perpendicular to CuO_2 layers).

Our results can also be applied to the study of the optical effects which are due to the spatial dispersion in macroscopically inhomogeneous media. An example of such a medium is a current-carrying superconductor. In Ref. 23 we have obtained the symmetry-allowed forms of the r matrices for this system. However, the explicit calculation of the reflection coefficients for an oblique incidence and anisotropic crystal by using the traditional method is rather difficult. Our approach permits one to carry out the calculations without any essential difficulties.

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