Reciprocity in reflection and transmission of light

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The manifestation of the time-reversal symmetry in the optical response is considered. Using the formulation of the reciprocity law for unpolarized light, the symmetry in the coherent reflection and/or transmission matrix R and the Stokes matrix Q is deduced by analyzing a gedanken measurement for the R or Q matrix. Implications of the combined time-reversal and spatial symmetry are studied for oblique specular reflection, forward transmission and, more extensively, for the normal-incidence reflection. The case of chiral gyrotropic media is discussed, and it is shown that a circular dichroism is symmetry allowed if the normal-incidence reflection leads to a partial depolarization of the beam. Nonreciprocity in the optical response is discussed. Phenomenological symmetry analysis of the experiments searching for the time-reversal-symmetry violation in high-T_c superconductors is presented.

I. INTRODUCTION

It is well known that time-reversal symmetry (T symmetry) of microscopic processes manifests itself also on the macroscopic level. Microscopic T symmetry underlies the Onsager relations for kinetic coefficients, which linearly relate the response to the forces in a macroscopic system.[1] In an optical experiment, one measures the electric field $\mathbf{E}_{\rm out}$ in the reflected or transmitted beam generated by the incoming beam with a given electric field $\mathbf{E}_{\rm in}$. The fields in the (weak) beams are linearly related by the reflectivity or transmission matrix $\hat{R}$, so that the "response" $\mathbf{E}_{\rm out}$ to the "force" $\mathbf{E}_{\rm in}$ is given by $\mathbf{E}_{\rm out}=\hat{R}\,\mathbf{E}_{\rm in}$. Partially polarized light is conveniently characterized by Stokes parameters or the Stokes four-vector $\mathbf{S}$.[2] The Stokes vectors $\mathbf{S}_{\rm in}$ and $\mathbf{S}_{\rm out}$ of incoming and outgoing beams are related by a matrix $\hat{Q}$, $\mathbf{S}_{\rm out}=\hat{Q}\mathbf{S}_{\rm in}$.

In the present paper, we address the problem of the constraints imposed by time-reversal symmetry on the optical kinetic coefficients, the reflection and/or transmission matrix R, or the Stokes matrix Q. These constraints can be considered as a consequence of the Onsager principle or the reciprocity law of optics.[2,3] Being well known as a general statement, its particular implications for reflection or transmission do not seem to have been analyzed in a complete manner.

The main motivation for the present study is given by recent optical experiments searching for a spontaneous T-symmetry violation in high-temperature superconductors (for a review, see, e.g., Ref. 4). Knowledge of the time-reversal transformation properties of the optical kinetic coefficients is needed for the analysis of experimental data in order to discriminate between a manifestation of T-symmetry violation and the effects of a different origin.

It should be noted, that there is no rigorous way of deriving the discussed properties from Onsager symmetry in the dielectric $\hat{\epsilon}$ and the magnetic permeability $\hat{\mu}$ tensors even when the macroscopic description is meaningful. For example, reflectivity, calculated with spatial dispersion taken into account, is known to be dependent on the boundary conditions for the electric and magnetic fields at the interface.[5,6] Generally speaking, the boundary conditions cannot be found without going into details of the microscopic origin of the dispersion. In case of gyrotropic or, in other terminology, chiral crystals, the real problem of boundary conditions sometimes is disguised by a certain freedom in choice of $\hat{\epsilon}$ and $\hat{\mu}$: Gyrotropy can be included only in $\hat{\epsilon}$ or in both $\hat{\epsilon}$ and $\hat{\mu}$.[7] Then observable quantities, such as partial reflectivities, turn out to be dependent on the choice of $\hat{\epsilon}$ and $\hat{\mu}$.[7,8] In this situation it is useful to know general symmetry properties, independent of any microscopic theory, as a guide and test for selecting the structure of a phenomenological theory.

The properties of the Stokes matrix, brought about by the reciprocity law, have been investigated by Perrin[9] in the context of elastic scattering of light by nonmagnetic liquids or gases. Figueiredo and Raab[10] and Graham[11] applied the results of Ref. 9 to the study of electro-optical and magneto-optical effects in Rayleigh scattering. Reciprocity of the optical response was demonstrated by Chandrasekhar[12] in the framework of the kinetic theory of radiative transfer. Onsager symmetry in impedance closely related to the R matrix was derived in Ref. 3. Recently, normal-incidence reflection has been analyzed by Halperin.[13]

A straightforward way to derive time-reversal symmetry in the optical response of a macroscopic body could consist in the application of microscopic T symmetry, which is well known in quantum electrodynamics.[14] However, we choose an alternative method that is an application of the reciprocity law of classical optics. The classical approach appears to be the most natural one for the description of a standard reflectivity or transmission measurement.

Perrin[9] postulated the reciprocity law of optics by comparing the results of two measurements: (i) The incoming wave has linear polarization $\mathbf{e}_1$ and wave vector $\mathbf{q}_1$, and the detector measures the intensity of emerging light with linear polarization $\mathbf{e}_2$ and wave vector $\mathbf{q}_2$; (ii) the beams are reversed, and their polarizations are interchanged, so that the incoming wave is characterized by $\mathbf{e}_2$ and $-\mathbf{q}_2$, and the wave $(\mathbf{e}_1, -\mathbf{q}_1)$ is measured. The reciprocity means the identity of the two intensities.

In the present paper, we will use a slightly different formulation of the reciprocity law. We consider unpolarized light, i.e., an incoherent mixture of all possible polarization states on equal footing. In this formulation (Sec. II), the source produces unpolarized light and the detector is insensitive to polarization. (Absence of a definite polarization in the source may be thought to be a property of the statistical ensemble comprising monochromatic, polarized sources of light or, simply, as a property of a quasimonochromatic source.) The reciprocity law is then formulated as an invariance of the readings of the detector when the source and detector are interchanged simultaneously with time reversal of the state of the system illuminated by light. If the system is invariant relative to time reversal, this formulation is equivalent to the one put forward by Perrin.

A plane monochromatic wave, having been specularly reflected by a crystal, remains essentially space and time coherent and the description in terms of the reflectivity and/or transmission matrix (Sec. III A) is an adequate approximation. The Stokes description (Sec. III B) is more appropriate when the outgoing wave loses its complete coherence as a result of, e.g., the multidomain structure of the crystal or quasielastic processes. In Sec. III C we present a derivation of time-reversal symmetry in the Stokes and reflectivity and/or transmission matrices. In essence, our derivation is a modified version of Perrin's arguments.

In Sec. IV a combination of T and spatial symmetries is considered. For illustration, we show a symmetry-allowed structure of the reflectivity and Stokes matrices for an oblique incidence reflection from crystals, with different spatial symmetries, placed in a magnetic field. Transmission through a centrosymmetric crystal is also briefly discussed. Application of the T and spatial symmetries to the reflection under normal incidence is considered in Sec. V with reference to (i) a crystal in a magnetic field (or the "anyon state," which possesses the same symmetry) and (ii) chiral media.

In the last section, we summarize the result and discuss the condition of their applicability. Possibilities of optical detection of a broken time-reversal symmetry are discussed together with the experiments searching for T-symmetry violation in high-temperature superconductors.

II. RECIPROCITY LAW

We consider a setup comprising a physical system in an equilibrium state, a source, and detector of light, situated far enough from the system and each other. The system includes the sample under study and auxiliary elements to control the light beams. It may also include sources of magnetic fields, e.g., permanent magnets, to create a certain configuration of the field in the sample and auxiliary elements.

The total Hamiltonian of the system illuminated by light is supposed to be T invariant, and therefore the time-reversal transformation applied to a macroscopic state $\{t\}$ generates the state $\{-t\}$, which is also a possible equilibrium state of the system. If the conjugated states $\{t\}$ and $\{-t\}$ are macroscopically distinctive, the T symmetry is (spontaneously) broken. The symmetry may be broken by the sample being spin polarized or current carrying. Another example is given by a hypothetical anyon state, where the quasiparticles possess fractional statics and a broken T symmetry is an intrinsic property (see, e.g., references in Ref. 4). Besides, the magnetizations and magnetic fields of the magnets included in the system have opposite directions in the conjugated states, so that their presence makes the states $\{t\}$ and $\{-t\}$ distinctive.

The source of light is assumed to be pointlike and to produce monochromatic unpolarized isotropic radiation. The detector, tuned on the frequency of the source, mea-

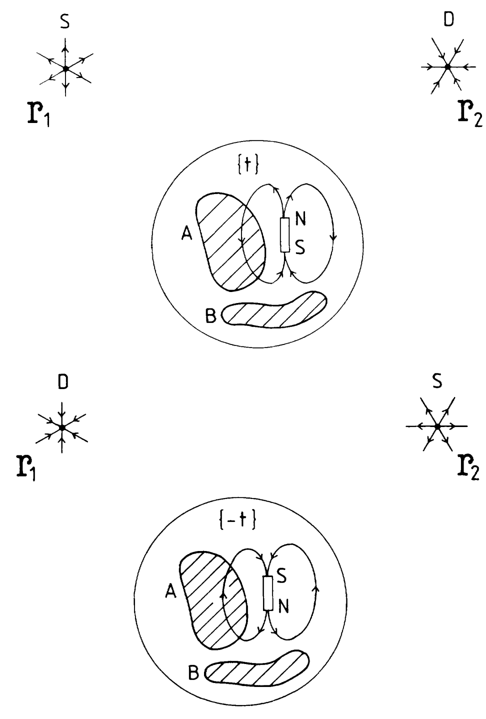

FIG. 1. Formulation of the reciprocity law. Unpolarized light from the source S illuminates a physical system shown surrounded by an imaginary sphere. The system is comprised of "bodies" A and B and, optionally, a (permanent) magnet with the poles N and S. The detector D registers the total intensity of the radiation falling on it. (a) In the first measurement, the source is at the point $\mathbf{r}_1$, the system is in an equilibrium state $\{t\}$, and the detector is at the point $\mathbf{r}_2$. The reading of D is $I_{21}(\{t\})$. (b) In the second measurement, S and D are interchanged and the system is in the state $\{-t\}$ generated from $\{t\}$ by time reversal. The reversal of the spins in the permanent magnet leads to the reversal of its magnetic fields, as shown in (b). The intensity measured in the second experiment $I_{12}(\{-t\})$ is equal to $I_{21}(\{t\})$ by virtue of the reciprocity law.

sures the intensity of radiation integrated over the direction of propagation and polarization. A set of three independent identical pointlike dipoles, closely spaced and oriented along x, y, and z axes, gives a realization for the source as well as the detector.

To express reciprocity quantitatively, we compare the results of two measurements (Fig. 1): (i) the system is in a state $\{t\}$, and the source of light is at a point $\mathbf{r}_1$; the detector registers an intensity $I_{12}(\{t\})$ at a point $\mathbf{r}_2$; (ii) if we substitute the positions of the source and detector as well as reverse the state of the system from $\{t\}$ to $\{-t\}$, the detector will then display an intensity $I_{21}(\{-t\})$. The reciprocity law can be expressed as

$$I_{12}(\{t\})=I_{21}(\{-t\}) . \tag{1}$$

When the states $\{t\}$ and $\{-t\}$ are identical, Eq. (1) follows from the reciprocity theorem of Helmholtz.[2] This theorem, which was formulated for scalar waves, is valid here as soon as the source generates unpolarized light and the detector is insensitive to polarization.[2] Equation (1) can also be derived by applying the Onsager principle to the kinetic coefficients, relating the response of the dipoles in the detector to the forces acting on the dipoles in the source of light. The quantum derivation of the Onsager relations[1] explicitly exploits the time-reversal transformation of the wave functions of the system, and from the latter point of view, the reciprocity law [Eq. (1)] is a direct consequence of microscopic time-reversal symmetry.

III. RECIPROCITY IN REFLECTIVITY AND STOKES MATRICES

In this section we consider the implications of the reciprocity law for the symmetry properties of the optical kinetic coefficients. We first give a formal definition of the coefficients.

A. Reflectivity matrix

In the case of specular reflection of polarized monochromatic light by a sample with uniform surface, incoherent effects can often be neglected, and the electric field outside of the sample,

$$\mathbf{E}(r,t)=\mathrm{Re}[(\mathbf{E}_{\mathrm{in}}e^{i\mathbf{q}_{\mathrm{in}}\cdot\mathbf{r}}+\mathbf{E}_{\mathrm{out}}e^{i\mathbf{q}_{\mathrm{out}}\cdot\mathbf{r}})e^{-i\omega t}] , \tag{2}$$

is a superposition of incoming and outgoing plane waves $\mathbf{q}_{\mathrm{in}}$ and $\mathbf{q}_{\mathrm{out}}$ with frequency $\omega>0$. The complex amplitudes of the waves, $\mathbf{E}_{\mathrm{in}}$ and $\mathbf{E}_{\mathrm{out}}$, have two components perpendicular to the corresponding wave vectors. The reflectivity (2×2) matrix $\hat{R}$ couples the independent Cartesian components of $\mathbf{E}_{\mathrm{in}}$ and $\mathbf{E}_{\mathrm{out}}$,

$$\begin{bmatrix} E_{x2} \\ E_{y2} \end{bmatrix}_{\mathbf{q}_{2,\ \mathrm{out}}}=\hat{R}(\mathbf{q}_2,\mathbf{q}_1)\begin{bmatrix} E_{x1} \\ E_{y1} \end{bmatrix}_{\mathbf{q}_{1,\ \mathrm{in}}} , \tag{3}$$

$$\begin{bmatrix} E_{x1} \\ E_{y1} \end{bmatrix}_{-\mathbf{q}_{1,\ \mathrm{out}}}=\hat{R}(-\mathbf{q}_1,-\mathbf{q}_2)\begin{bmatrix} E_{x2} \\ E_{y2} \end{bmatrix}_{-\mathbf{q}_{2,\ \mathrm{in}}} , \tag{4}$$

for the reflections $\mathbf{q}_1\to\mathbf{q}_2$ and $-\mathbf{q}_2\to-\mathbf{q}_1$, respectively. Here and below the same coordinate system is used for the description of the polarization of the waves $\mathbf{q}_1$ $(\mathbf{q}_2)$ and $-\mathbf{q}_1$ $(-\mathbf{q}_2)$. In the case of forward transmission through a plate, the incoming and transmitted waves are given by

$$\mathbf{E}_{\mathrm{in,out}}(\mathbf{r},t)=\mathrm{Re}(\mathbf{E}_{\mathrm{in,out}}e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)}) . \tag{5}$$

The transmission (2×2) matrix $\hat{R}(\mathbf{q},\mathbf{q})$ relates the transverse components of $\mathbf{E}_{\mathrm{out}}$ to those of $\mathbf{E}_{\mathrm{in}}$,

$$\begin{bmatrix} E_x \\ E_y \end{bmatrix}_{\mathrm{out}}=\hat{R}(\mathbf{q},\mathbf{q})\begin{bmatrix} E_x \\ E_y \end{bmatrix}_{\mathrm{in}} , \tag{6}$$

in a coordinate system with x, y, and z axes $\|\mathbf{q}$. We use the same axes for the description of the waves $\mathbf{q}$ and $-\mathbf{q}$.

B. Stokes matrix

Reflection of a monochromatic plane wave from, or transmission of it through, a crystalline sample may produce a wave which is only partially coherent. For example, the light emerging after reflection of a fully polarized light from a multidomain anisotropic crystal is perceived by an analyzing system as being partially polarized. The Stokes description of a light beam is then the most convenient one. The description is adequate for an analysis of any phase-insensitive optical experiment with (quasi)monochromatic beams.

The beam is described by the Stokes "vector" $\mathbf{S}$ with four components S_i $(i=0,1,2,3)$, expressed via the time or ensemble average of the complex amplitude $\mathbf{E}$ in the beam[2] as

$$\begin{aligned} S_0&=\langle(|E_x|^2+|E_y|^2)\rangle , \\ S_1&=\langle(|E_x|^2-|E_y|^2)\rangle , \\ S_2&=2\,\mathrm{Re}\langle E_x^*E_y\rangle,\quad S_3=2\,\mathrm{Im}\langle E_x^*E_y\rangle \end{aligned} \tag{7}$$

or, in a more compact form, as

$$S_i=\langle\hat{E}^{\dagger}\hat{\sigma}_i\hat{E}\rangle , \tag{8}$$

where $\hat{E}$ is a column built of E_x and E_y, $\hat{E}^{\dagger}$ stands for the $\hat{E}$ Hermitian conjugate, and $\hat{\sigma}_0=\hat{1}$, $\hat{\sigma}_1$, $\hat{\sigma}_2$, and $\hat{\sigma}_3$ are the Pauli matrices $\hat{\sigma}_z$, $\hat{\sigma}_x$, and $\hat{\sigma}_y$, respectively.

When the incoming beam with the wave vector $\mathbf{q}$ and Stokes parameters $\mathbf{S}_{\mathrm{in},\mathbf{q}_1}$ is reflected by the sample into the wave $\mathbf{q}_2$, the Stokes vector $\mathbf{S}_{\mathrm{out},\mathbf{q}_2}$ of the outgoing beam is linearly coupled to $\mathbf{S}_{\mathrm{in},\mathbf{q}_1}$ via the Stokes matrix Q,

$$\mathbf{S}_{\mathrm{out},\mathbf{q}_2}=\hat{Q}(\mathbf{q}_2,\mathbf{q}_1)\mathbf{S}_{\mathrm{in},\mathbf{q}_1} , \tag{9}$$

where $\hat{Q}$ is a (4×4) real matrix.

If reflection, as such, leads only to a negligible loss of coherence, the matrix $\hat{Q}(\mathbf{q}_2,\mathbf{q}_1)$ can be expressed via the corresponding reflectivity matrix $\hat{R}(\mathbf{q}_2,\mathbf{q}_1)$,

$$(\hat{Q})_{ij}=\tfrac{1}{2}\mathrm{Tr}(\hat{\sigma}_i\hat{R}\hat{\sigma}_j\hat{R}^{\dagger}) , \tag{10}$$

as follows from Eqs. (3), (8), and (9). In this case the reflectivity and Stokes matrix give equivalent descriptions of a phase-insensitive experiment.

C. Reciprocity relation

Apart from the requirement of energy conservation, which leads to certain inequalities for the matrix elements, the most general constraint imposed on the structure, the R and Q matrices, is brought about by time-reversal symmetry or, in other words, the reciprocity law [Eq. (1)].

To deduce the constraints, we consider a system built of the sample under study and some idealized auxiliary elements which imitate the measurement of the R or Q matrix. (The system may include sources of magnetic fields to create a certain field environment in the sample and auxiliary elements.) Analysis of the gedanken measurement allows one to derive the symmetry properties in the R or Q matrix from the reciprocity law [Eq. (1)]. The arguments in the case of R and Q matrices are similar, and we show the derivation with reference to the Stokes description.

Consider the measurement of $\hat{Q}_t \equiv \hat{Q}(\mathbf{q}_2,\mathbf{q}_1;\{t\})$, the Stokes matrix corresponding the sample in a state $\{t\}$. The system includes two absorbing screens with apertures (Fig. 2), so that the sample is illuminated by the light with the wave vector $\mathbf{q}_1$, and the detector registers the light with the wave vector $\mathbf{q}_2$. The incoming and outgoing beams pass through the ideal polarizers P_1 and P_2, respectively. The polarizer P_1 (P_2) is described by the Stokes matrix $\hat{\Pi}_{1,t}$ ($\hat{\Pi}_{2,t}$), which, as in Eq. (9), gives the Stokes vector of the wave $\mathbf{q}_1$ ($\mathbf{q}_2$) emerging from it. The Stokes matrix for the combined sample and polarizer system is given by an ordered product of the corresponding matrices, with the order determined by the path of the beam. The incoming light is produced by an unpolarized source, and its Stokes vector reduces to the zeroth component proportional to the intensity of the source, I_0. A polarization-insensitive detector measures the total intensity given by the zeroth component of the Stokes vector of the outgoing beam. Therefore the signal from the detector is

$$I_{12}(\{t\}) = I_0 A(\hat{\Pi}_{2,t}\hat{Q}_t\hat{\Pi}_{1,t})_{00} , \tag{11}$$

where A is a purely geometrical factor accounting for the propagation of the light in free space from the source to the sample and then to the detector.

In the "time-reversed" experiment, where the whole system is in the state $\{-t\}$ and the source and detector are interchanged, the matrix $\hat{Q}_{-t} \equiv \hat{Q}(-\mathbf{q}_1,-\mathbf{q}_2;\{-t\})$ is measured. The directions of the beams are reversed, and the same polarizers P_1 and P_2 are now described by new matrices $\hat{\Pi}_{1,-t}$ and $\hat{\Pi}_{2,-t}$. The detector displays an intensity $I_{21}(\{-t\})$ given by Eq. (11), with t substituted for $-t$ and also with interchanged indexes 1 and 2. By equating the two intensities by virtue of the reciprocity law Eq. (1); we obtain

$$(\hat{\Pi}_{2,t}\hat{Q}_t\hat{\Pi}_{1,t})_{00} = (\hat{\Pi}_{1,-t}\hat{Q}_{-t}\hat{\Pi}_{2,-t})_{00} , \tag{12}$$

for arbitrary polarizers and any pair $\{t\}$ and $\{-t\}$ states of the system coupled by time reversal.

The Stokes matrix $\hat{\Pi}$ of an ideal polarizer can be presented as a direct product

$$(\hat{\Pi})_{ij} = \tfrac{1}{2}(\boldsymbol{L})_i(\mathbf{M})_j \tag{13}$$

of two four-component vectors which represent the Stokes vectors of a fully polarized light, i.e., of the form $(1,n_1,n_2,n_3)$ with $\sum n_i^2 = 1$. The vector $\boldsymbol{L}$ gives the Stokes vector of the fully polarized light emerging from the polarizer, and $\mathbf{M}$ determines the polarization state of the incoming light where no loss of the intensity in the polarizer take place. A general ideal polarizer can be built of (i) a linear polarizer and (ii) a set of birefrigent transparent plates sandwiching the linear polarizer. By varying the optical properties of the birefrigent plates, the three components of $\boldsymbol{L}$ and $\mathbf{M}$ can be arbitrarily altered.

For two time-conjugated configurations (where the directions of the propagation of the beam are opposite[15]), the Stokes matrices of an ideal polarizer, $\hat{\Pi}_t$ and $\hat{\Pi}_{-t}$, relate to each other as

$$\hat{\Pi}_t = \hat{T}\tilde{\hat{\Pi}}_{-t}\hat{T}^{-1} , \tag{14}$$

where the overhead tilde on the matrix, here and thereafter, denotes transposition of the matrix, and the diagonal matrix $\hat{T}$ is

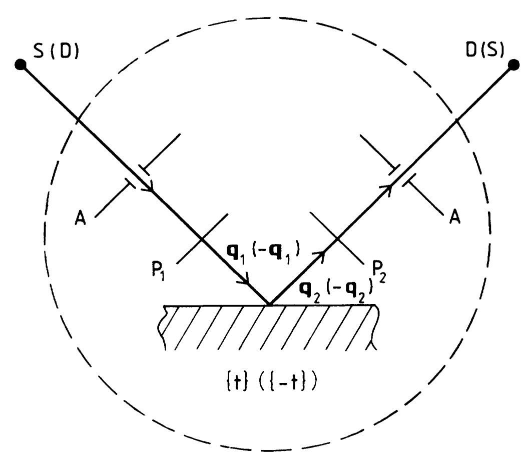

FIG. 2. Gedanken measurement of the reflectivity or Stokes matrices. As in Fig. 1, the physical system (shown encircled) is illuminated by light from the source (S). The system includes two screens with apertures (A), the polarizer and/or analyzer P_1 and P_2, and the sample under study. The system may include sources (not shown) of magnetic or other fields acting on the sample and/or P_1 and P_2. In the first measurement, the system is in a state $\{t\}$. The wave $\mathbf{q}_1$, polarized by P_1, falls on the sample. The outgoing wave $\mathbf{q}_2$ traverses P_2, which, in this configuration, plays role of an analyzer selecting a certain polarization from the reflected beam. D measures $I_{12}(\{t\})$, the intensity of light emerging from the system. The symbols in brackets denote the time-conjugated arrangement where the source and detector are interchanged, and the state of the whole system (the sources of field included) is changed from $\{t\}$ to $\{-t\}$. Now the incoming wave $-\mathbf{q}_2$ is polarized by P_2 and the polarization state of the reflected wave $-\mathbf{q}_1$ is analyzed by P_1. The detector displays $I_{21}(\{-t\})$, which is equal to $I_{12}(\{t\})$ by virtue of the reciprocity law.

$$\hat{T}=\mathrm{diag}(1,1,1,-1)\ . \tag{15}$$

The relation in Eq. (14) can be checked for any given realization of an ideal polarizer.

Using Eq. (14), one can rewrite Eq. (12) as

$$\sum_{i,j}(\boldsymbol{L}_2)_i(\hat{Q}_t-\hat{T}\tilde{\hat{Q}}_{-t}\hat{T}^{-1})_{ij}(\boldsymbol{L}_1)_j=0\ , \tag{16}$$

where the representation in Eq. (13) has been used, and $\boldsymbol{L}_1$ ($\boldsymbol{L}_2$) is the four-vector corresponding to $\hat{\Pi}_{1,t}$ ($\hat{\Pi}_{2,t}$). The unit three-vectors in $\boldsymbol{L}_1$ and $\boldsymbol{L}_2$ are arbitrary and independent of each other, and Eq. (16) is satisfied only if the matrix, sandwiched by $\boldsymbol{L}_2$ and $\boldsymbol{L}_1$, is zero. Therefore the reciprocity law [Eq. (1)] results in the relation

$$\hat{Q}(\mathbf{q}_2,\mathbf{q}_1;\{t\})=\hat{T}\tilde{\hat{Q}}(-\mathbf{q}_1,-\mathbf{q}_2;\{-t\})\hat{T}^{-1}\ , \tag{17}$$

for the Stokes matrices which describe reflection (or transmission) from the sample in two time-conjugated equilibrium states $\{t\}$ and $\{-t\}$.

Symmetry in the reflectivity (or transmission) matrix,

$$\hat{R}(\mathbf{q}_2;\mathbf{q}_1;\{t\})=\tilde{\hat{R}}(-\mathbf{q}_1,-\mathbf{q}_2;\{-t\})\ , \tag{18}$$

can be proved by similar arguments.[16]

Equations (17) and (18) express time-reversal symmetry in the optical response and give an optical analog to Onsager symmetry of the kinetic coefficients. These relations are quite general, and such factors as absorption, gyrotropy, etc., do not affect their validity.

In the case when the states $\{t\}$ and $\{-t\}$ are microscopically identical, Eq. (17) gives a convenient matrix representation for the relations between the elements of the Stokes matrix first obtained by Perrin.[9] Equation (17) is also in agreement with the symmetry derived by Chandrasekhar[12] from analysis of the transport equation for radiation propagating through a nonmagnetic random media.

General relations [Eqs. (17) and (18)] can be written in a more detailed form in a situation when the distinction between states $\{t\}$ and $\{-t\}$ is specified. In the simplest case, T symmetry is preserved or optionally broken only by the presence of a magnet. Then Eq. (18) reads as

$$\hat{R}(\mathbf{q}_2,\mathbf{q}_1;\mathbf{H})=\tilde{\hat{R}}(-\mathbf{q}_1,-\mathbf{q}_2;-\mathbf{H})\ , \tag{19}$$

where $\mathbf{H}$ is the magnetic field of the magnet experienced by the sample. If the sample is characterized by a T-odd order parameter, the states $\{t\}$ and $\{-t\}$ differ also in the sign of the order parameter: For example, for a (anti)ferromagnet with spontaneous (sublattice) magnetization $\mathbf{M}$, Eq. (17) gives

$$\hat{Q}(\mathbf{q}_2,\mathbf{q}_1;\mathbf{M})=\hat{T}\tilde{\hat{Q}}(-\mathbf{q}_1,-\mathbf{q}_2;-\mathbf{M})\hat{T}^{-1}\ . \tag{20}$$

Similar relations can be written, e.g., for superfluids with nondissipative currents or unconventional superconductors.

As we see in the following sections, more information can be extracted from Eqs. (17) and (18) (i) when the T and spatial symmetries are combined and (ii) for a special case of the normal-incidence reflection.

IV. COMBINATION OF TIME-REVERSAL AND SPATIAL SYMMETRIES

T symmetry couples the reversed processes $\mathbf{q}_1\to\mathbf{q}_2$ and $-\mathbf{q}_2\to-\mathbf{q}_1$. There are also certain spatial transformations $\mathcal{P}$ which interchange the incoming and outgoing beams. Consideration of the combined T and spatial symmetries $\mathcal{P}$ allows one to derive the properties of the R or Q matrix with fixed $\mathbf{q}_1$ and $\mathbf{q}_2$.

For reflection, there are two spatial transformations of interest: (i) $\mathcal{P}=C_2$, i.e., a 180° rotation around the axis perpendicular to the reflection plane; (ii) $\mathcal{P}=\sigma_v$, i.e., a mirror transformation in the plane perpendicular to the plane of incidence. In the case of transmission, the beam can be reversed by the space inversion $\mathcal{P}=I$.

Invariance relative to two simultaneous transformations, (i) the time reverse [Eqs. (17) and (18)] and (ii) the above spatial transformation of the coordinate system, is expressed as

$$\hat{R}(\mathbf{q}_2,\mathbf{q}_1;\{t\})=\hat{\mathcal{P}}_E\tilde{\hat{R}}(\mathbf{q}_2,\mathbf{q}_1;\{-t\}_{\mathcal{P}})\hat{\mathcal{P}}_E^{-1} \tag{21}$$

and

$$\hat{Q}(\mathbf{q}_2,\mathbf{q}_1;\{t\})=\hat{\mathcal{P}}_S\hat{T}\tilde{\hat{Q}}(\mathbf{q}_2,\mathbf{q}_1;\{-t\}_{\mathcal{P}})\hat{T}^{-1}\hat{\mathcal{P}}_S^{-1}\ , \tag{22}$$

for the R and Q matrices, respectively. Here $\{-t\}_{\mathcal{P}}$ stands for the state produced from $\{t\}$ by time reversal combined with $\mathcal{P}$. The spatial transformation generally changes the electric field in the beam, $\mathcal{P}\mathbf{E}=\hat{\mathcal{P}}_E\mathbf{E}$, or the Stokes vector, $\mathcal{P}\mathbf{S}=\hat{\mathcal{P}}_S\mathbf{S}$. The corresponding matrices $\hat{\mathcal{P}}_E$ and $\hat{\mathcal{P}}_S$ enter Eqs. (21) and (22). In the coordinate system shown in Fig. 3, they are ($\mathcal{P}=C_2$, σ_v, and I)

$$(\hat{C}_2)_E=(\hat{I})_E=-\hat{1},\quad (\hat{C}_2)_S=(\hat{I})_S=\hat{1}\ , \tag{23}$$

$$(\hat{\sigma}_v)_E=\begin{bmatrix}-1 & 0\\ 0 & 1\end{bmatrix},\quad (\hat{\sigma}_v)_S=\mathrm{diag}(1,-1,1,-1)\ . \tag{24}$$

If symmetry elements of the sample include the above transformations so that the states $\{t\}$ and $\{-t\}_{\mathcal{P}}$ are identical, Eq. (21) or (22) couples the (symmetric) off-diagonal elements of the R or Q matrices. When the spatial symmetry is broken, e.g., by an applied field, Eqs. (21)

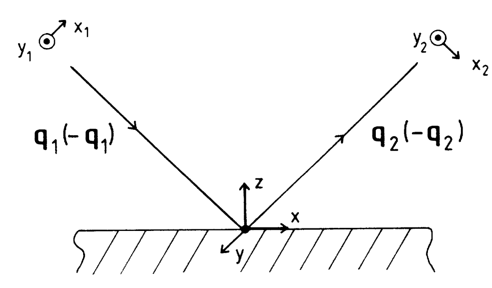

FIG. 3. Oblique-incidence reflection. The wave $\mathbf{q}_1$ ($-\mathbf{q}_2$) is reflected into the wave $\mathbf{q}_2$ ($-\mathbf{q}_1$). The polarization of the waves is defined in the coordinate systems with the x_1 and y_1 axes for the waves $\pm\mathbf{q}_1$ and the x_2 and y_2 axes for the waves $\pm\mathbf{q}_2$. The x_1 and x_2 axes are in the incidence plane, and $y_{1,2}$ is directed to the eye. The applied magnetic field is expanded in the coordinate system with the x, y, and z axes.

TABLE I. Symmetry-allowed structure of the reflectivity matrix for oblique-incidence reflection with different orientations of the applied magnetic field **H** along the axes x, y, and z (Fig. 3). The first row shows the structure of the R matrix for reflection from a crystal with a twofold rotation axis $C_2 \| z$. The same for a crystal with a mirror plane (σ_v), perpendicular to the plane of incidence, is shown in the second row. The third row refers to the case when both C_2 and σ_v are symmetry elements. The elements labeled by g or u are even or odd functions, respectively, of the corresponding component of the field. The unlabeled elements do not have definite parity with respect to the field.

Symmetry \ Field	$\mathbf{H}\|x$	$\mathbf{H}\|y$	$\mathbf{H}\|z$
C_2	$\begin{bmatrix} a & c \\ c & b \end{bmatrix}$	$\begin{bmatrix} a & c \\ c & b \end{bmatrix}$	$\begin{bmatrix} a_g & c_g+d_u \\ c_g-d_u & b_g \end{bmatrix}$
σ_v	$\begin{bmatrix} a_g & c_u+d_g \\ c_u-d_g & b_g \end{bmatrix}$	$\begin{bmatrix} a & d \\ -d & b \end{bmatrix}$	$\begin{bmatrix} a & d \\ -d & b \end{bmatrix}$
$C_2 \times \sigma_v$	$\begin{bmatrix} a_g & c_u \\ c_u & b_g \end{bmatrix}$	$\begin{bmatrix} a & 0 \\ 0 & b \end{bmatrix}$	$\begin{bmatrix} a_g & d_u \\ -d_u & b_g \end{bmatrix}$

and (22) couple the reflections for different orientations of the field.

For an illustration, we apply Eqs. (21) and (22) to a situation when a crystalline sample is placed in a magnetic field **H** (or it possesses an order parameter with the same symmetry properties, e.g., magnetization) and has (i) C_2, (ii) σ_v, and (iii) $C_2 \times \sigma_v$ as symmetry elements. The vector $\mathbf{H}(H_x,H_y,H_z)$ is expanded in the coordinate system shown in Fig. 3. A symmetry-allowed structure of the reflectivity matrix is presented in Table I for different symmetries of the crystal and orientations of the magnetic field. In Table I the matrix elements labeled by g (or u) are even (or odd) functions of the corresponding component of the field.

As for the Stokes matrix, if the sample is symmetric relative to C_2, it follows from Eqs. (22) and (23) that

$$Q_{ij}(H_x,H_y,H_z)=\alpha_i\alpha_j Q_{ji}(H_x,H_y,-H_z)\ , \tag{25}$$

where α_i and α_j are equal to (-1) for $i=j=3$ and to unity otherwise. For the σ_v symmetry,

$$Q_{ij}(H_x,H_y,H_z)=\beta_i\beta_j Q_{ji}(-H_x,H_y,H_z)\ , \tag{26}$$

where β_i and β_j are equal to (-1), for $i,j=2$ or 3 and to unity otherwise.

In the case of the $\sigma_v \times C_2$ symmetry, Eqs. (25) and (26) are valid simultaneously and the Q matrix has the following form for a field oriented along x, y, or z axis:

$$\mathbf{H}(H_x,0,0):\ \hat{Q}=\begin{bmatrix} g_1 & g_2 & u_1 & u_2 \\ g_2 & g_3 & u_3 & u_4 \\ u_1 & u_3 & g_4 & g_5 \\ -u_2 & -u_4 & -g_5 & g_6 \end{bmatrix}, \tag{27}$$

$$\mathbf{H}(0,\mathbf{H}_y,0):\ \hat{Q}=\begin{bmatrix} a_1 & a_2 & 0 & 0 \\ a_2 & a_3 & 0 & 0 \\ 0 & 0 & a_4 & a_5 \\ 0 & 0 & -a & a \end{bmatrix}, \tag{28}$$

$$\mathbf{H}(0,0,H_z):\ \hat{Q}=\begin{bmatrix} g_1 & g_2 & u_1 & u_2 \\ g_2 & g_3 & u_3 & u_4 \\ -u_1 & -u_3 & g_4 & g_5 \\ u_2 & -u_4 & g_5 & g_6 \end{bmatrix}. \tag{29}$$

In Eqs. (27) and (29), as in Table I, the matrix elements denoted by g (u) are even (odd) functions of the field, whereas the a's in Eq. (28) are allowed to be arbitrary functions.

If light passes through a plate with inversion symmetry, Eq. (21) or (22) with $\mathcal{P}=I$ reads as

$$\hat{R}(\mathbf{q},\mathbf{q};\mathbf{H})=\tilde{\hat{R}}(\mathbf{q},\mathbf{q};-\mathbf{H}) \tag{30}$$

or

$$\hat{Q}(\mathbf{q},\mathbf{q};\mathbf{H})=\hat{T}\tilde{\hat{Q}}(\mathbf{q},\mathbf{q};-\mathbf{H})\hat{T}^{-1}\ . \tag{31}$$

Note that the inversion symmetry of the plate, and therefore Eqs. (30) and (31), may be violated even in case of a centrosymmetric crystal if the crystal is placed on a substrate or if the interfaces of the plate are not identical.

V. NORMAL-INCIDENCE REFLECTION

The reflection under normal incidence is a special case as the reflections $\mathbf{q}_1 \to \mathbf{q}_2$ $(=-\mathbf{q}_1)$ and $-\mathbf{q}_2 \to -\mathbf{q}_1$ are the same processes. Therefore, for any spatial symmetry of the sample, the reciprocity law [Eqs. (17) and (18)] applied to the normal-incidence reflection reads as

$$\hat{R}(\{t\})=\tilde{\hat{R}}(\{-t\})\ , \tag{32}$$

$$\hat{Q}(\{t\})=\hat{T}\tilde{\hat{Q}}(\{-t\})\hat{T}^{-1}\ , \tag{33}$$

for the reflectivity and Stokes matrices, respectively.

In the case of normal incidence, the R matrix can always be expressed via the impedance matrix $\hat{\zeta}$, which enters the effective boundary condition on the surface of the crystal and couples the electric **E** and magnetic **H** fields: $\mathbf{E}=\hat{\zeta}[\mathbf{n}\times\mathbf{H}]$, $\mathbf{n}$ being a unit vector orthogonal to the surface. From this point of view, Eq. (32) follows from the symmetry of the impedance matrix derived in Ref. 3 from the Onsager principle. An equivalent [to Eq. (32)] relation for the reflectivity matrix in the representation of the circularly polarized waves has been recently obtained by Halperin.[13]

Normal incidence is also a special case for the spatial symmetry elements: The mirror transformation σ_v in any plane orthogonal to the reflection plane, as well as any rotation in the reflection plane, does not change the direction of the incoming and outgoing beams. Therefore the R and Q matrices transform through themselves under these operations. Below we consider the restrictions imposed by the combined T and spatial symmetries on the R and Q matrices.

The R matrix is conveniently presented as the Pauli matrix expansion of the form

$$\hat{R}=R_0\hat{1}+R_1[\hat{\sigma}_z\cos(2\varphi_1)-\hat{\sigma}_x\sin(2\varphi_1)]+iR_2[\hat{\sigma}_z\cos(2\varphi_2)-\hat{\sigma}_x\sin(2\varphi_2)]+R_3\hat{\sigma}_y\ , \tag{34}$$

where R_1 and R_2 are real, whereas R_0 and R_3 are generally complex.

The coordinate system, where the electric field $\mathbf{E}$ is expanded in the components E_x and E_y, is determined by the axes of the optical setup measuring the R matrix. Given the representation in Eq. (34), a rotation of the axes through an angle φ or, equivalently, a rotation of the sample through the angle $-\varphi$ does not change the R's in Eq. (34) and shifts the φ's: $\varphi_{1,2}\rightarrow\varphi_{1,2}+\varphi$. The mirror transformation σ_v in the plane $x0z$ (or $y0z$) reduces to $\varphi_{1,2}\rightarrow-\varphi_{1,2}$ and $R_3\rightarrow-R_3$. The symmetry properties relative to time reversal (T), i.e., $\{t\}\rightarrow\{-t\}$, can be found from Eq. (32). Summarizing, the parameters in the representation Eq. (34) have the following transformation properties:

$$\text{rotation}(\varphi):\quad R_3\rightarrow R_3,\quad \varphi_{1,2}\rightarrow\varphi_{1,2}+\varphi; \tag{35}$$

$$\sigma_v:\quad R_3\rightarrow -R_3,\quad \varphi_{1,2}\rightarrow-\varphi_{1,2}; \tag{36}$$

$$T:\quad R_3\rightarrow -R_3,\quad \varphi_{1,2}\rightarrow\varphi_{1,2}, \tag{37}$$

while R_0, R_1, and R_2 are invariant relative to time reversal and all the spatial transformations. The simplicity of the above symmetry properties is the reason for the choice of the representation in Eq. (34).

Analogously, the Q matrix is conveniently written as a sum of T-even ($\hat{Q}_g$) and T-odd ($\hat{Q}_u$) parts:

$$\hat{Q}=\hat{Q}_g+\hat{Q}_u\ , \tag{38}$$

$$\hat{Q}_g=\begin{bmatrix} G_1 & g_1\cos2\varphi_1 & g_1\sin2\varphi_1 & J \\ g_1\cos2\varphi_1 & G_2+g_3\cos4\varphi_3 & g_3\sin4\varphi_3 & -g_2\sin2\varphi_2 \\ g_1\sin2\varphi_1 & g_3\sin4\varphi_3 & G_2-g_3\cos4\varphi_3 & g_2\cos2\varphi_2 \\ -J & g_2\sin2\varphi_2 & -g_2\cos2\varphi_2 & G_3 \end{bmatrix}, \tag{39}$$

$$\hat{Q}_u=\begin{bmatrix} 0 & -u_1\sin2\psi_1 & u_1\cos2\psi_1 & U_1 \\ u_1\sin2\psi_1 & 0 & U_2 & u_2\cos2\psi_2 \\ -u_1\cos2\psi_1 & -U_2 & 0 & u_2\sin2\psi_2 \\ U_1 & u_2\cos2\psi_2 & u_1\sin2\psi_2 & 0 \end{bmatrix}, \tag{40}$$

where $\hat{Q}_{g(u)}$ is defined as $2\hat{Q}_{g(u)}=\hat{Q}+(-)\hat{T}\tilde{\hat{Q}}\hat{T}^{-1}$. All the G's and g's in Eq. (39) are invariant relative to time reversal and all the spatial transformations. The rest of the parameters in Eqs. (39) and (40) have the following transformation properties:

$$\text{rotation}(\varphi):\quad J\rightarrow J,\quad (U_i,u_k)\rightarrow(U_i,u_k),(\varphi_i,\psi_k)\rightarrow(\varphi_i+\varphi,\psi_i+\varphi); \tag{41}$$

$$\sigma_v:\quad J\rightarrow -J,\quad (U_i,u_k)\rightarrow(-U_i,-u_k),(\varphi_i,\psi_k)\rightarrow(-\varphi_i,-\psi_k); \tag{42}$$

$$T:\quad J\rightarrow J,\quad (U_i,u_k)\rightarrow(-U_i,-u_k),(\varphi_i,\psi_k)\rightarrow(\varphi_i,\psi_k), \tag{43}$$

where i and k numerate the corresponding parameters in Eqs. (39) and (40).

Note also, that the Q matrix [Eqs. (38)–(40)] as well as the R matrix [Eq. (34)] are not changed by the 180° rotation of the coordinate system, and therefore all the Q- and R-matrix elements are functions only of C_2 invariants regardless of the crystal symmetry.

With the properties in Eqs. (35)–(37) and (41)–(43) and the requirement of C_2 invariance, a symmetry-allowed structure of the R and Q matrices and its dependence on external fields can be found in any given situation when the symmetries of the crystal and field environment are specified.

The general form of the matrices is simplified when T and σ_v are symmetry elements: For a T-symmetric state, R_3 in Eq. (34) and $\hat{Q}_u$ must be zero as T-odd quantities.

In the coordinate system with the x or y axis passing through the mirror plane, all the φ's in Eq. (34) and (39) are equal to zero (or $\pi/2$, depending on convention). A rotation of the coordinates (or the sample) makes them finite, but keeps them equal. The parameter J in Eq. (39), being σ_v odd, must be zero identically. Rotational symmetry brings further simplification.

As examples of analysis of the rotational invariance, we consider two situations where σ_v is not a symmetry element: (i) Both σ_v and T are broken, but the combination $\sigma_v T$ is preserved. This is the symmetry of a crystal with a relevant mirror plane, placed in a magnetic field H_z orthogonal to the surface. The role of H_z can also be played by the magnetization of a ferromagnet or the order parameter of the anyon state.[4] (ii) Light is reflected by a nonmagnetic crystal, the point group of which does not contain σ_v. This is the symmetry of a chiral crystal, exhibiting natural optical activity (gyrotropy).

In case (i), by virtue of the time-reversal transformation properties in Eqs. (37) and (43), R_3 and $\hat{Q}_u$ are proportional to H_z, whereas $R_{0,1,2}$ and $\hat{Q}_g$ are functions of H_z^2 only. The combined $\sigma_v T$ symmetry allows one to conclude from Eqs. (36) and (42) that (a) $J=0$ and (b) $\varphi_{1,2}$ in Eq. (34) or $\varphi_{1,2,3}$ and $\psi_{1,2}$ in Eqs. (39) and (40) can be simultaneously put to zero if the axes of the measuring setup are adjusted to the mirror plane of the sample. These conclusions are valid for arbitrary rotational symmetry of the sample. If the sample has an n-fold rotation axis C_n, $n \geq 3$, perpendicular to the surface, the rotational invariance necessitates $R_1=R_2=0$ and $g_1=g_2=g_3=u_1=u_2=0$, with the exception that the C_4 symmetry is compatible with a finite g_3.

In case (ii) the reflectivity matrix [Eq. (34)] must be proportional to the unit matrix when the sample has a rotation axis C_n, $n \geq 3$: Time-reversal symmetry requires $R_3=0$, as follows from Eq. (37), and $R_1=R_2=0$ because of rotational invariance.[17] Therefore, in the R-matrix approximation, normal-incidence reflection by optically isotropic gyrotropic and/or chiral media has the properties of reflection by a simple isotropic medium.

This interesting manifestation of the combined rotation and time-reversal symmetry has recently been stressed by Halperin.[13] A negative result of the measurement of the circular dichroism in normal-incidence reflection from a gyrotropic cubic crystal α-LiO_3 (Ref. 8) is consistent with Halperin's observation.

The Stokes matrix, corresponding to case (ii), must be T even, so that $\hat{Q}_u=0$ and its structure is given by $\hat{Q}_g$ in Eq. (39). Again, the rotation symmetry C_n, $n \geq 3$, leads to simplifications in the general form and necessitates $g_1=g_2=g_3=0$, with the only exception that C_4 symmetry allows g_3 to be finite.

Note that neither time-reversal nor rotational symmetries are in contradiction with a finite J in Eq. (39). It is generally finite if σ_v is not a symmetry element, and therefore the properties of normal-incidence reflection from a chiral media differ from that of a nonchiral one. For instance, $(S_0)_{\text{out}}$, the intensity of light emerging after reflection of a circularly polarized wave of unit intensity ($S_0=1$, $S_3=\pm 1$) from a chiral crystal with symmetry C_n, $n \geq 3$, is given by

$$(S_0)_{\text{out}}=G_1+JS_3 \ , \tag{44}$$

in accordance with Eqs. (9) and (39) and the above remarks. The intensity depends on the handedness of the incoming wave S_3, that is, a circular dichroism. Therefore a chiral isotropic media, e.g., sugar, may reveal its gyrotropy in normal-incidence reflection.

The contradiction with the conclusion from the analysis of the R matrix can be resolved as follows. In the description of reflection in terms of the R matrix, the outgoing beam is believed to be a coherent plane wave with a definite amplitude $\mathbf{E}_{\text{out}}$. An incoherent component, i.e., fluctuating part of $\mathbf{E}_{\text{out}}$, arising, e.g., as a result of static or dynamic optical roughness of the interface, is completely neglected. On the other hand, the Stokes vector is an average of a bilinear form built of $\mathbf{E}_{\text{out}}$ and $\mathbf{E}^*_{\text{out}}$, and it includes both the coherent and incoherent parts of light on equal footing. Thus the reflectivity and Stokes matrix represent different descriptions of reflection. Only when the incoherent part of light is neglected can the Stokes matrix be expressed via the R matrix with the help of Eq. (10). The two descriptions do not have to give identical predictions when the reflected wave is only partially coherent.

A signature of incoherent effects is the loss of a complete polarization, as a result of reflection. For a fully polarized wave, the combination

$$\Delta^2=S_0^2-S_1^2-S_2^2-S_3^2 \geq 0 \tag{45}$$

is zero and, generally, Δ/S_0 shows the weight of the incoherent (fully depolarized) component in the beam.[2] When the incoming beam is completely and circularly polarized, the Stokes vector of the outgoing beam has $(S_0)_{\text{out}}$ given by Eq. (51), $(S_3)_{\text{out}}=-J+G_3$, and $(S_{1,2})_{\text{out}}=0$. The depolarization parameter of the outgoing wave can be written as

$$\Delta^2=(G_1+G_3)(G_1-G_3-2JS_3) \ . \tag{46}$$

A finite J is incompatible with $\Delta=0$, as G_1+G_3 can be shown to be always finite (and positive). The R-matrix approximation in Eq. (10), applied to the present situation, automatically gives $G_1-G_3=J=0$, and both the depolarization and dichroism are zero. Generally, the representation in Eq. (10) is not valid and J as well as $G_1-G_3 \geq 2|J|$ is finite. Then the dichroism is possible, but with depolarization as an inevitable attribute.

Therefore the circular dichroism and all other effects related to chirality or gyrotropy are seen in normal-incidence reflection only in the incoherent part of light and they are present to the extent that incoherent effects are pronounced. Similarly, the aforementioned anisotropy in case of relfection from a crystal with C_4 rotation symmetry can also be shown to arise solely due to the incoherent processes. Incoherent reflection is not described by the R matrix, and this resolves the above contradiction.

We note that the Stokes matrix has more independent parameters, generally 16, than the R matrix, which has only 7 (apart from the global phase). The $16-7=9$ parameters are reserved for the contributions of incoherent processes. As for a physical mechanism of the processes, we note only that, generally speaking, any violation of translational invariance in the reflection plane, such as roughness or contamination of the surface, multidomain structure, thermal fluctuations, etc., leads to the loss of a complete coherence. A more detailed discussion of this and related questions is beyond the scope of this paper.

VI. DISCUSSION AND CONCLUSIONS

In this paper we have considered the constraints imposed on the optical kinetic coefficients by time-reversal symmetry. In the course of the derivation, light has been regarded as a classical field. In the quantum theory, T symmetry leads to constraints imposed on the scattering matrix (S matrix).[14] When only the coherent interaction of light with a thermally equilibrium media is taken into account, T symmetry in the S matrix can be shown to lead to identities between the amplitudes of photon scattering, which are equivalent to Eq. (18). Symmetry in the Stokes description [Eq. (17)] holds for both coherent and incoherent elastic processes. For quasielastic processes Eq. (17) is in agreement with quantum theory if $\Delta\omega$, that is, the broadening of the emerging beam, is small enough, i.e., $\Delta\omega \ll k_B T/\hbar$, where T is the temperature of the sample. We note in passing that the quantum approach allows one to generalize Eq. (17) to the case of Raman scattering.

In Secs. IV and V we have considered the combined time-reversal and spatial symmetries. If constraints imposed by the reciprocity law are quite universal, the discussed spatial symmetry elements must be understood as invariance of the *sample* relative to spatial transformations. The sample is normally less symmetric than the material it is made of. For example, a structure, made of crystalline but disoriented layers, does not have any σ_v mirror plane even if the crystal has one. Also, the screw axis or a glide mirror plane is not equivalent to the ordinary one, although the inequivalence leads to small effects when the wavelength of the light is large in comparison to the lattice constant. The effective spatial symmetry of the sample with a contaminated surface or multidomain structure depends on the wavelength and size of the incoming beam.

As was mentioned in the Introduction, there exist contradictory results in the theory of gyrotropic media.[5–8] Based on the microscopic theory of exciton resonances, calculation of the reflectivity matrix of a gyrotropic crystal was done by Ivchenko[5] and Ivchenko and Selkin.[6] Their results for general oblique incidence are in full agreement with the reciprocity law as expressed by Table I (in the limit $\mathbf{H}\to 0$). In particular, no rotation of polarization or circular dichroism is predicted for normal incidence. Silberman[7] and Luk'anov and Novikov[8] calculated the R matrix in a phenomenological theory of gyrotropic crystals. A variant of their theory, where standard boundary conditions for the fields and inductions as well as the Born constitutive relations for $\hat{\epsilon}$ and $\hat{\mu}$ were adopted, predicts circular dichroism for normal incidence. In accordance with Halperin[13] and Sec. V, this contradicts T symmetry, and by this argument the above version appears theoretically unsatisfactory. We note that agreement with the symmetry properties in Eq. (18) can be used as a test for a phenomenological theory of gyrotropic crystals.

As discussed in Sec. V, the Stokes vector of the beam back reflected by a chiral medium does contain information about the gyrotropy of the medium. The symmetry arguments do not forbid a circular dichroism with the sign controlled by the handedness of the chiral media. However, chirality reveals itself only in the incoherent part of the reflected light. Similarly, symmetry does not forbid an anisotropy in reflection from cubic crystals (Sec. V), but again, only to the extent depolarization is present.

The reciprocity relations in Eqs. (17) and (18) make it possible to introduce classification by time-reversal symmetry, i.e., symmetry of the R and Q response matrices relative to the time reversal of the probed system, $\{t\}\to\{-t\}$. Experimental detection of a nonreciprocal response, i.e., a quantity which is T odd in this classification, unequivocally proves that T symmetry of the state of the system is broken.

The elements of neither the R nor Q matrix for given $\mathbf{q}_1$ and $\mathbf{q}_2$ do not have definite T symmetry. Only certain combinations of them measured in the geometries $\mathbf{q}_1\to\mathbf{q}_2$ (I) and $-\mathbf{q}_2\to-\mathbf{q}_1$ (II) do. For example, the combination $\hat{R}_{\mathrm{I}}-\hat{R}_{\mathrm{II}}$, built of the reflectivity matrices $\hat{R}_{\mathrm{I}}$ and $\hat{R}_{\mathrm{II}}$ measured in the geometries I and II, is T odd: By virtue of Eq. (14), it changes its sign under $\{t\}\to\{-t\}$. The other example is given by the split in Eq. (38), where $\hat{Q}_g$ is T even and $\hat{Q}_u$ is T odd.

Therefore the detection of a T-odd quantity necessarily requires two measurements differing in their geometry, as discussed. Measurements in transmission or oblique-incidence reflection modes have to be done with the reverse of the beam. In the case of normal-incidence reflection, the 90° rotation of the sample may play the role of the reversal: If the incident polarization is along the x axis and the outgoing y polarization is detected, i.e., $x\to y$ process, then the rotation interchanges the axes and the reversed process $y\to x$ is measured.

Spontaneous violation of T symmetry along with the loss of mirror symmetry is an intrinsic property of a so-called anyon model of high-T_c superconductivity (for a review, see Ref. 4). Wen and Zee[18] suggested to search for the violation by observation of circular effects in optical properties. We briefly discuss the reported experimental data[19–23] in the nomenclature adopted in this paper. We believe that a phenomenological language of the R or Q matrix is the most appropriate for analysis of the data. The choice between R and Q is dictated by the type of experiment.

Lyons *et al.*[19] have reported a circular dichroism in normal-incidence reflection from high-T_c films. In their apparature the beam traverses a set of optically active plates and the reflected light transformed by the plates is registered by a diode detector. An experiment of this type can be completely described in the Stokes approach. One can show that the output signal η_o (Refs. 19 and 22) from the null detector is expressed by the elements of the

Q matrix as

$$2\eta_0 = \frac{Q_{30}}{Q'_{11}} , \tag{47}$$

where Q'_{11} is the rotationally invariant part of Q_{11} [equal to G_2 in Eq. (39)]. Q_{30} is odd relative to a mirror transformation P (here and below we use the notation from the anyon literature, $P \equiv \sigma_v$). However, Q_{30} alone does not have a definite T symmetry: Only a T-odd combination $Q_{30}+Q_{03}$ does. Therefore the observation of a finite Q_{30} proves only that the part of the sample under the light spot does not have any mirror plane P as a symmetry element. No conclusions follow about the T-invariance violation. (In different terms this problem was discussed in Refs. 19 and 22.)

Ellipsometric data for normal-incidence reflection have been also reported by Weber *et al.*[21] They use an optical setup, where the output signals are ζ_A (Ref. 24)

$$\zeta_A = \zeta_u + \zeta_g ,$$
$$4\zeta_u = \frac{Q_{30}+Q_{03}}{Q_{32}-Q_{23}}, \quad 4\zeta_g = \frac{Q_{31}-Q_{13}}{Q_{32}-Q_{23}} , \tag{48}$$

and ζ_B, which can also be expressed via elements of the Q matrix [Eq. (38)] similarly to ζ_A. The parameter ζ_A (as well as ζ_B) is P odd, but does not have definite T symmetry: It is built of T-odd (ζ_u), and T-even (ζ_g) parts (see Sec. V). As in the experiment of Ref. 19, detection of a finite ζ_A only shows that P symmetry is broken without any firm conclusions about T symmetry. The transmission experiment,[21] where only one direction of propagation is measured, suffers from the same uncertainty in the interpretation.

It has been attempted in Ref. 21 to discriminate between ζ_u and ζ_g by making use of the difference in their symmetry properties: A linear coupling to the external magnetic field is allowed only for ζ_u. A rigid correlation between the sign of $\zeta_{A,B}$ and the direction of the magnetic field applied in the process of cooling has been reported for single-crystal samples[21] and later for thin high-T_c films.[23] A Curie-Weiss-type behavior of the Faraday effect in transmission has also been reported.[23] Taken as published, the above magnetic-field data hardly can be understood unless T symmetry is indeed broken below certain transition temperatures. However, these findings have not been confirmed so far by other groups, and the situation remains controversial.

A qualitatively different experiment has been performed by Spielman *et al.*[20] They use an optical gyroscope setup and measure the interference of two waves having traversed a high-T_c film in two opposite directions. The setup is sensitive only to coherent processes: The waves reach the sample with a delay $\tau = L/c$, $L = 1.5$ km being the length of the gyroscope fiber. The long delay guarantees that only the emerging light within a very narrow frequency window, $\Delta f \sim 1/2\pi\tau \sim 50$ kHz, contributes to the interference signal. Besides this frequency filtering, the light eventually transforms into a wave propagating in a single-mode fiber, so that a spatial filtering takes place. An experiment of this type can be completely described in the R-matrix formalism. For the experimental arrangement of Ref. 20, the output signal ϕ can be written in our notations as[25]

$$\phi = \mathrm{Im} \rightarrow \mathrm{Arg}[(R_0^+ + R_3^+)(R_0^- - R_3^-)^*] , \tag{49}$$

where $R_{0,3}^+$ and $R_{0,3}^-$ are the coefficients in the Pauli matrix expansion of $\hat{R}(\mathbf{q},\mathbf{q},\{t\})$ and $\hat{R}(-\mathbf{q},-\mathbf{q},\{t\})$, respectively, and the indices 0 and 3 numerate the coefficients as in Eq. (34). By virtue of Eq. (37), ϕ is a T-odd quantity, and therefore the signal is controlled exclusively by an nonreciprocal response of the sample. The part of the nonreciprocal response, which is given by Eq. (49), has been accurately measured with a negative result.

As we see, phenomenologically, the three experiments measure three different quantities. The same object, i.e., the Stokes matrix, is measured in the ellipsometric experiments of Refs. 19, 21, and 23. However, the "rotation angles" η_o and ζ_A are given by different combinations of the elements of the Q matrix. This and differences in the sample structures make a quantitative comparison of the two ellipsometric experiments difficult. A qualitatively different physical quantity, i.e., the amplitude of a certain coherent process, is measured in the gyroscope setup.[20] The reported "angles" η_o, ζ_A, and ϕ are, in the phenomenological approach, independent parameters and can be in any relation. The negative result of the interference experiment may turn out to be compatible with the ellipsometric data, when the latter are interpreted in favor of a broken T symmetry. As an example of a situation where different measuring techniques may give seemingly incompatible results, we recall again the back reflection from a chiral media: The P-symmetry violation (gyrotropy) would not be seen in the coherent part of the reflected light, but might reveal itself in the incoherent part. We do not see any general argument forbidding a microscopic model which has a similar property with respect to T-symmetry violation. Another possibility has been suggested by Dzyaloshinskii:[26] If the space inversion combined with time reversal is preserved, ϕ, the rotation in transmission, must be zero [see Eq. (21)], but a nonreciprocal reflection is not forbidden. At present, when the experimental situation is still controversial and no convincing theory of the anyon state in high-T_c cuprates has been put forward, room for various speculations remains wide.

In conclusion, we have considered the derivation of time-reversal symmetry in the optical kinetic coefficients describing reflection or transmission of polarized light. The derived T symmetry in the coherent reflection and/or transmission R matrix and Stokes Q matrix is a general property that is valid in spite of absorption, gyrotropy, etc. The symmetry in the Stokes matrix allows also for quasielastic processes. We have considered implications of the combined time-reversal and spatial symmetries as well. The derived time-reversal transformation properties allow one to discriminate between the reciprocal and nonreciprocal parts of the optical response in experiments searching for a spontaneously broken T symmetry.

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[16]With an indefinite phase factor on the right-hand side of it, Eq. (18) follows from Eqs. (10) and (17). The choice of the phase factor can be justified by the analysis of a phase-sensitive measurement, when the sample is complemented with a semitransparent mirror, so that the sample and mirror comprise a Fabry-Pérot-type resonator. In this case the intensity of light emerging from the combined sample and mirror system is controlled by multiple reflection and is sensitive to the global phase of the reflectivity matrix of the sample. Applied to the combined system, Eq. (17) leads to Eq. (18).

[17]For a low-symmetry crystal without the axes C_n, $n \geq 3$, the R matrix has its general form given by Eq. (34) apart from $R_3=0$. The role of gyrotropy, i.e., the presence of the antisymmetric part of the dielectric tensor ϵ proportional to the wave vector, cannot be established by general arguments. One of the possibilities is that the gyrotropic transition takes place in an orthorhombic crystal and the symmetric part of ϵ remains to be diagonal in the same axes as in the high-symmetry phase. Then φ_1 and φ_2 in Eq. (34) are proportional to the gyrotropy (and $\epsilon_{xx}-\epsilon_{yy}$). However, the low symmetry does not guarantee the absence of a monoclinic distortion of the lattice, and it does not fix the directions of the axes even when, by chance, the distortion is absent. Moreover, the real and imaginary symmetric parts of ϵ are generally diagonal in different axes. Therefore φ_1, φ_2, and $\varphi_1-\varphi_2$ may be finite without gyrotropy taken into account.

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