Boundary conditions in the simplest model of linear and second harmonic magneto-optical effects

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This paper is concerned with linear and nonlinear magneto-optical effects in multilayered magnetic systems when treated by the simplest phenomenological model that allows their response to be represented in terms of electric polarization. The problem is addressed by formulating a set of boundary conditions at infinitely thin interfaces, taking into account the existence of surface polarizations. Essential details are given that describe how the formalism of distributions (generalized functions) allows these conditions to be derived directly from the differential form of Maxwell's equations. Using the same formalism we show the origin of alternative boundary conditions that exist in the literature. The boundary value problem for the wave equation is formulated, with an emphasis on the analysis of second harmonic magneto-optical effects in ferromagnetically ordered multilayers. An associated problem of conventions in setting up relationships between the nonlinear surface polarization and the fundamental electric field at the interfaces separating anisotropic layers through surface susceptibility tensors is discussed. A problem of self-consistency of the model is highlighted, relating to the existence of rescaling procedures connecting the different conventions. The linear approximation with respect to magnetization is pursued, allowing rotational anisotropy of magneto-optical effects to be easily analyzed owing to the invariance of the corresponding polar and axial tensors under ordinary point groups. Required representations of the tensors are given for the groups ∞m, $4mm$, $mm2$, and $3m$. With regard to centrosymmetric multilayers, nonlinear volume polarization is also considered. A concise expression is given for its magnetic part, governed by an axial fifth-rank susceptibility tensor being invariant under the Curie group $\infty\infty m$.

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I. INTRODUCTION

The boundary conditions to be discussed in this paper are essential to the development of the simplest theoretical models of surface linear and second harmonic magneto-optical effects in ferromagnetically ordered multilayers. Any multilayer can be visualized as a set of alternating layers composed of different materials [Fig. 1(a)]. Each homogeneous region or layer transforms into the neighboring one within an interface that is very narrow and inhomogeneous. When excited by monochromatic light of high intensity, the multilayer may exhibit magneto-optical Kerr effects at both the fundamental frequency ω and second harmonic frequency $\omega_S = 2\omega$. If the multilayer is transparent enough, the Faraday effect, at both frequencies, may also be observed. Normally, the response of the multilayer generating the effects is completely described by means of electric (effective) polarization.[1–4] Since optical nonlinearity is assumed to be small, a consideration of the effects is carried out iteratively, i.e., within a simple perturbation procedure. Consequently, there are two separate parts of the phenomenological solution to the problem.[5]

In the first part, the influence of the nonlinearity on the response is neglected. Volume and surface linear polarizations, induced by incident light in the layers and interfaces (transition regions), are related to the primary electric field through linear susceptibility tensors. The tensors may depend respectively on volume and surface magnetization, thereby introducing linear volume- and surface-sensitive magneto-optical effects. Since the influence of surface polarizations is small, they can be considered as perturbations. Moreover, the interfaces themselves are thought of as ideal surfaces (infinitely thin) on the basis that their thicknesses are much smaller than the wavelength of the radiation. The linear problem is solved in an iterative way. In the first instance, any surface polarizations are neglected. The electromagnetic field in all space is obtained from Maxwell's equations subject to the conventional boundary conditions. The magnitude and state of polarization (the Kerr angle and elllipticity) of these fields in the transparent medium [Fig. 1(a)] contain information about volume-sensitive magneto-optical effects. The next procedure concerns the surface susceptibility tensors that determine the surface polarizations driven by the unperturbed field. It should be noted that the normal component of

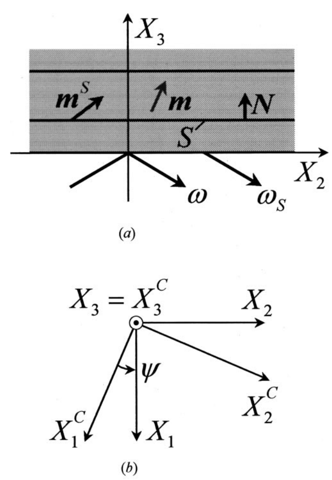

FIG. 1. (a) Multilayer configuration for the model of magneto-optical effects. (b) Orientation of the physical and crystallographic coordinate systems.

this field, across ideal interfaces, is discontinuous, and the consequences of this requires careful consideration. The surface polarizations become sources (instead of incident light) radiating waves at the fundamental frequency. The corresponding electric field, carrying information about surface contributions to linear magneto-optical effects, can be obtained from Maxwell's equations, which now need to be accompanied by the unconventional boundary conditions that take into account singular polarizations that are assumed to be localized exactly at the ideal interfaces.

The second part of the solution is similar to the first. Primarily this concerns the state of the polarization and intensity of second harmonic waves in the transparent (incident) medium. Instead of incident light, nonlinear surface and volume polarizations play the role of sources. The boundary conditions for the second harmonic fields obviously remain the same as those for the linear perturbation fields, involving a nonlinear surface polarization in place of a linear surface polarization. The wave equation for the electric field in each layer may be inhomogeneous because of the nonlinear volume polarization. How both kinds of polarizations are related to the fundamental unperturbed field is a matter of approximation. If each layer possesses a center of inversion, then, within the electric-dipole approximation, there exist no volume polarizations because a polar third-rank tensor is identically zero. For this reason, volume-sensitive second harmonic magneto-optical effects are referred to as forbidden in centrosymmetric media. However, they are not forbidden within the electric-quadrupole approximation that takes into account the nonlocality of the response (spatial dispersion) through a polar fourth-rank tensor.[1,6] While considering surfaces and interfaces, it is always sufficient to take the electric-dipole approximation into account. Since inversion symmetry is broken at surfaces and interfaces, surface-sensitive second harmonic magneto-optical effects are never forbidden by symmetry. Irrespective of these points the above-mentioned problem concerning the convention related to the driving field remains. For noncentosymmetric multilayers the electric-dipole approximation does allow volume-sensitive second harmonic magneto-optical effects. If the volume polarizations dominate over the surface ones, the latter are disregarded, and the conventional boundary conditions may be used.[7]

Following this approach, an interesting treatment of the surface-sensitive transverse Kerr effect was given for a semi-infinite medium showing that it is possible to use linear magneto-optics for probing surface magnetism.[8] Similar ideas also apply to the development of simple models of the second harmonic magneto-optical effects in centrosymmetric, ferromagnetically ordered multilayers.[5,9]

Although such phenomenological solutions are common, our return to the basic principles has been influenced by subtleties that often cause ambiguities in the literature. These are due to the fact that boundary conditions involving surface polarization at an ideal interface exist in two versions,[10–14] as well as due to a variety of conventions for the surface polarization itself in terms of its driving field. A relevant question emerges as to whether or not it is always true that a particular convention, which should include both a preferred set of boundary conditions and a driving field, has relationships with the others. This problem is considered in Sec. III, to provide general rescaling procedures linking the tensors related to often-encountered conventions. Unfortunately, such a rescaling is not always possible in the case of anisotropic adjacent layers. To illustrate this feature of the phenomenological model, an example will be given (Sec. V). In Sec. II, we give a rather elegant and quick method for deriving either version of the boundary conditions directly from the differential form of Maxwell's equations, valid in the space of distributions (generalized functions). Although the validity may be intuitively clear, we outline why this is so, for to accept it merely as a postulate, as suggested before,[10] seems insufficient. The version of boundary conditions we prefer is considered thoroughly. For the case of normal incidence this version was also given elsewhere, but without details of its derivation.[11] If it had been shown how the boundary conditions relevant to magneto-optics came about, there would have been no need to seek an analogy with electrostatics.[12] In Sec. IV, the same method is used to describe the origin of an alternative and less simple set of boundary conditions.[13,14] In view of the similarities in considering linear and second harmonic magneto-optical effects, we confine ourselves to the formulation of a boundary value problem for looking at the latter. Section V is devoted to this problem, with an emphasis on the effects in centrosymmetric, ferromagnetically ordered, multilayers. An auxiliary analysis revealing the symmetry of the associated susceptibility tensors will also be carried out, pursuing the linear approximation with respect to magnetization.[15] In this respect it is advantageous to examine the invariance of the relevant polar and axial tensors under ordinary point groups. In the case of surface-sensitive magneto-optical effects, this will be done for the often encountered groups $4mm$, $mm2$, and $3m$, and the Curie group ∞m governing the symmetry of an isotropic surface. The tensors will be given in a 3×6 matrix form, and with reference to a physical rather than a crystallographic coordinate system [Fig. 1(b)], so that it will be most convenient to use the results while looking at the rotational anisotropy[16] of magneto-optical effects. To observe quadrupole contributions to surface magneto-optical effects, along with the known result for the nonmagnetic part of volume polarizations (suitable for cubic and isotropic layers), a representation for the magnetic part will be given, provided that the corresponding axial fifth-rank susceptibility tensor is invariant under the Curie group $\infty\infty m$. Finally, we shall summarize the essential features of the model in an attempt to clarify the arguments.

II. WAY OF DERIVING THE BOUNDARY CONDITIONS

The boundary conditions involving a surface polarization have the same form in all situations where the idealizations, described in Sec. I, are applicable. However, since second harmonic magneto-optical effects are of particular significance, we shall derive the boundary conditions relevant to this case. Obviously, it is sufficient to consider a plane surface S between semi-infinite, anisotropic, not necessarily magnetic, media [Fig. 2(a)].

The assumed idealization implies that Maxwell's equa-

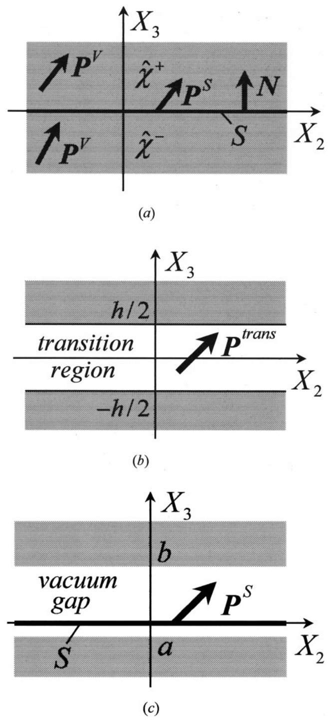

FIG. 2. (a) Ideal interface between semi-infinite homogeneously magnetized media. (b) Corresponding transition region used for deriving the boundary conditions in the classical way. (c) Illustration concerned with the convention defining the surface polarization through Eq. (9).

tions hold in the space of distributions $\mathfrak{D}'(\mathbb{R}^3)$. This assertion unequivocally follows from a procedure that introduces the idealization correctly. The procedure begins with the polarization being continuous in all space including a transition region of finite width h. A test function φ, belonging to $\mathfrak{D}(\mathbb{R}^3)$, is then taken in order to obtain the purely regular distributions from the corresponding classical Maxwell equations in the usual way.[17,18] Let f_h be a certain component of any of the fields, which obviously depends on h. For different values of h there is a sequence of actions $\langle f_h,\varphi\rangle$. In $\mathfrak{D}'(\mathbb{R}^3)$, the weak convergence $\langle f_h,\varphi\rangle \to \langle f,\varphi\rangle$ as $h\to 0$ may result in a singular distribution along with the regular one, both having a physical sense. A remarkable property, asserted by the theorem,[17] is that for any multi-index α of the differential operator D^α the following is always true: $\langle D^\alpha f_h,\varphi\rangle \to \langle D^\alpha f,\varphi\rangle$ as $h\to 0$. This means that, on undergoing the weak limit procedure, the classical Maxwell equations apply to the space $\mathfrak{D}'(\mathbb{R}^3)$, where they have exactly the same form, except that the densities of sources are defined as distributions. With regard to the corresponding differential operators the following result will be often used below. If a regular distribution f has a discontinuity of the first kind at a smooth surface S, then

$$\partial f/\partial x_i = \{\partial f/\partial x_i\} + [f]_S N_i \delta_S . \tag{1}$$

Here the derivative in the curly brackets is a function defined almost everywhere (in the classical sense). The next term on the right-hand side (RHS) of Eq. (1) is a singular distribution referred to as a single layer on S, where δ_S is the surface delta function, and $[f]_S = f^+ - f^-$ stands for the jump of f across S. The signs $\pm$ are linked to a chosen positive direction of the unit normal $\mathbf{N}$ to S.[17,18]

The optical and magneto-optical response of the whole medium is governed by the constitutive relation (in *SI* units)

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}^V + \mathbf{P}^S \delta_S , \tag{2}$$

where all the fields are at the frequency ω_S and belong to $\mathfrak{D}'(\mathbb{R}^3)$. The relation $\mathbf{B} = \mu_0 \mathbf{H}$ assumes that the medium does not respond to the magnetic field $\mathbf{H}$ at optical frequencies. The field $\mathbf{H}$ is a regular distribution, and so are the magnetic induction $\mathbf{B}$ and the bulk polarization $\mathbf{P}^V$. The latter is unambiguously defined through the linear susceptibility tensors as

$$\mathbf{P}^V = \begin{cases} \varepsilon_0 \hat{\chi}^{V+} \mathbf{E} + \mathbf{P}^{NV}, & x_3 > 0, \\ \varepsilon_0 \hat{\chi}^{V-} \mathbf{E} + \mathbf{P}^{NV}, & x_3 < 0, \end{cases}$$

and comprises the already known nonlinear volume contribution $\mathbf{P}^{NV}$ originating from the quadrupole or/and dipole approximation. We do not define $\mathbf{P}^V$ at the surface itself, for doing so would be meaningless. The polarizations $\mathbf{P}^S$ and $\mathbf{P}^{NV}$ can be related to the fundamental electric field through a surface and volume susceptibility tensor respectively. The RHS of Eq. (2) includes a singular distribution $\mathbf{P}^S \delta_S$ (single layer). Consequently, the electric displacement $\mathbf{D}$ is a linear combination of the regular and singular distributions.

In $\mathfrak{D}'(\mathbb{R}^3)$, Maxwell's equations relevant to the problem are

$$\mathrm{curl}\mathbf{H} = -i\omega_S \mathbf{D}, \tag{3a}$$

$$\mathrm{curl}\mathbf{E} = i\omega_S \mu_0 \mathbf{H}, \tag{3b}$$

$$\mathrm{div}\,\mathbf{H} = 0, \tag{3c}$$

$$\mathrm{div}\,\mathbf{D} = 0. \tag{3d}$$

Their forms are exactly the same as we understand them in the classical sense, i.e., for regions where all the fields are continuous. However, a significant difference is that Eqs. (3) are valid in all space $\mathbb{R}^3$, provided they are understood in the sense of distributions. Consequently, the differential operators in Eqs. (3) lead to classical Maxwell equations and corresponding boundary conditions. Indeed, taking into account Eq. (1), a calculation of $\mathrm{div}\,\mathbf{H}$ in Eq. (3c) leads to $\mathrm{div}\,\mathbf{H} = \{\mathrm{div}\,\mathbf{H}\} + ([\mathbf{H}]_S, \mathbf{N})\delta_S$. Here $\{\mathrm{div}\,\mathbf{H}\}$ is a regular distribution corresponding to the classical divergence (where it exists). The next term is defined as a simple layer of surface density $([\mathbf{H}]_S, \mathbf{N})$, where $[\mathbf{H}]_S = \mathbf{H}^+ - \mathbf{H}^-$ is the hypothetical jump of $\mathbf{H}$ across S. The sign + corresponds to a chosen positive direction of the unit normal $\mathbf{N}$ to S [Fig. 2(a)]. On the other hand, the equation $\mathrm{div}\,\mathbf{H} = 0$ should be valid everywhere in $\mathbb{R}^3$. Therefore, we have $\{\mathrm{div}\,\mathbf{H}\} = 0$, which is the classical Maxwell equation, plus the conventional boundary condition $([\mathbf{H}]_S, \mathbf{N}) = 0$ revealing the continuity of the normal component of the magnetic field across S:

$$H_3^+ - H_3^- = 0. \tag{4}$$

Equation (3d), where the displacement defined by Eq. (2), is more subtle. To cope with this the identity $\langle \mathrm{div}(\mathbf{P}^S\delta_S),\varphi\rangle = -\int_S(\mathbf{P}^S,\nabla\varphi)dS$, involving a test function $\varphi\in\mathfrak{D}(\mathbb{R}^3)$, has to be used. Leaving the calculation of $\mathrm{div}\,\mathbf{E}$ until later, we arrive at the equation

$$\varepsilon_0\,\mathrm{div}\mathbf{E}+\{\mathrm{div}\,\mathbf{P}^V\}+[P_3^V]_S\delta_S+(\partial P_1^S/\partial x_1+\partial P_2^S/\partial x_2)\delta_S + \delta(P_3^S\delta_S)/\partial x_3=0, \tag{5}$$

where, in addition to a linear combination of the simple layers, a different kind of singularity—a double layer [the last term on the left-hand side (LHS)] has appeared. Consequently, Eq. (5) can be satisfied only if the electric field $\mathbf{E}$ comprises the singular term $-\varepsilon_0^{-1}(0,0,P_3^S\delta_S)$. Hence the constitutive relation [Eq. (2)] suggests that the normal component of $\mathbf{D}$ must possess no singularity. This singularity must therefore be attached to the normal component of $\mathbf{E}$. On substituting the representation $\mathrm{div}\,\mathbf{E}=\{\mathrm{div}\,\mathbf{E}\}+[E_3]_S\delta_S-\varepsilon_0^{-1}\partial(P_3^S\delta_S)/\partial x_3$ into Eq. (5), there follows the classical Maxwell equation $\{\mathrm{div}\,\mathbf{D}\}=0$, plus the boundary condition

$$D_3^+-D_3^-=-\partial P_1^S/\partial x_1-\partial P_2^S/\partial x_2. \tag{6}$$

It is now clear that the three singular components of $\mathbf{P}^S\delta_S$ belong, respectively, to D_1, D_2, and E_3. This simplifies the procedure for dealing with the rest of the equations. It follows from Eq. (1) that the LHS of Eq. (3a) is represented as $\mathrm{curl}\mathbf{H}=\{\mathrm{curl}\mathbf{H}\}-[[\mathbf{H}]_S,\mathbf{N}]\delta_S$. Since the singular term of $\mathbf{D}$ is $(P_1^S,P_2^S,0)\delta_S$, Eq. (3a) becomes

$$\{\mathrm{curl}\mathbf{H}\}+(-[H_2]_S,[H_1]_S,0)\delta_S = -i\omega_S\{\mathbf{D}\}-i\omega_S(P_1^S,P_2^S,0)\delta_S,$$

which instantly yields the classical Maxwell equation $\{\mathrm{curl}\mathbf{H}\}=-i\omega_S\{\mathbf{D}\}$, being valid everywhere in $\mathbb{R}^3$ except the surface S, plus the linked boundary conditions for the two tangential components of the magnetic field:

$$H_1^+-H_1^-=-i\omega_S P_2^S,\quad H_2^+-H_2^-=i\omega_S P_1^S. \tag{7}$$

Since the field $\mathbf{E}$ possesses the above-mentioned singularity and the field $\mathbf{H}$ does not possess any, Eq. (3b) becomes

$$\{\mathrm{curl}\mathbf{E}\}-[[\mathbf{E}]_S,\mathbf{N}]\delta_S-\varepsilon_0^{-1}\mathrm{curl}(0,0,P_3^S\delta_S)=i\omega_S\mu_0\{\mathbf{H}\},$$

and leads immediately to the classical Maxwell equation $\{\mathrm{curl}\mathbf{E}\}=i\omega_S\mu_0\{\mathbf{H}\}$ and the boundary conditions for the two tangential components of the electric field:

$$E_1^+-E_1^-=-\varepsilon_0^{-1}\partial P_3^S/\partial x_1,\quad E_2^+-E_2^-=-\varepsilon_0^{-1}\partial P_3^S/\partial x_2. \tag{8}$$

It is worth noting that if the surface polarization vanishes, boundary conditions (6)–(8) turn into conventional ones. Moreover, if certain components of $\mathbf{P}^S$ vanish or do not depend on either x_1 or x_2, then some of the tangential components of $\mathbf{H}$ and $\mathbf{E}$ may be continuous as in the conventional case.

The unconventional boundary conditions (6)–(8) can also be derived from the integral form of Maxwell's equations. However, if this classical method is pursued, the ideal surface has to be replaced with a transition region of width h with continuous polarization $\mathbf{P}^{\mathrm{trans}}$ within it [Fig. 2(b)]. The surface polarization appears then almost in the sense of the weak limit: $\int_{-h/2}^{h/2}\mathbf{P}^{\mathrm{trans}}dx_3\to\mathbf{P}^S$ as $h\to 0$. In contrast to the method described above, it necessary however to assume that $\int_{-h/2}^{h/2}D_3dx_3\to 0$ as $h\to 0$, when applying Stoke's theorem and integrating the component $E_3=\varepsilon_0^{-1}D_3-P_3^{\mathrm{trans}}$ across the transition region.

Whichever way of deriving the boundary conditions is used, the assumption of ideal interfaces is only justified by the condition that thickness of the real interface is much less than the wavelength. The corresponding surface polarization $\mathbf{P}^S$, in fact, has to be defined from a nonlocal polarization within the ''smooth'' interface, and this can only be done correctly using microscopic theory.[3,20] How $\mathbf{P}^S$ is related to the fundamental field $\boldsymbol{\mathcal{E}}$ at the ideal interface is, however, a matter of convention.

III. CONVENTIONS ON THE SURFACE POLARIZATION

In the ideal model of second harmonic magneto-optical effects, the surface polarization $\mathbf{P}^S$ is induced by the fundamental electric field $\boldsymbol{\mathcal{E}}$ at the surface, and is related to the latter through a nonlinear surface susceptibility tensor.[1–4,19,21] However, an ambiguity occurs in setting up such a relationship, since the normal component $\mathcal{E}_3$ of the field is discontinuous across the ideal surface. This inevitably leads to an uncertainty in choosing the most suitable field to drive the polarization. Actually the driving field may be chosen according to a clearly defined convention, and such a solution does not ultimately lead to any logical inconsistencies.[22]

To visualize how the most convenient convention comes about, the following expedient is pertinent. The anisotropic media can imaginatively be offset by pushing the layers slightly off the ideal interface at $x_3=0$ [Fig. 2(c)]. As a consequence, a vacuum gap appears, while the position of the surface carrying $\mathbf{P}^S$ remains unchanged. To return to the initial configuration, it is necessary to collapse the vacuum gap, i.e., the coordinates of a and b of the surfaces between the anisotropic media and vacuum must go to zero. In the first instance, the conventional boundary conditions for the displacement $\boldsymbol{\mathcal{D}}$ (at the fundamental frequency) have to be written down. They are $\mathcal{D}_3(a)=\varepsilon_0\mathcal{E}_3^+(a)$ and $\varepsilon_0\mathcal{E}_3^-(b)=\mathcal{D}_3(b)$, where a dependence on only the essential coordinate is shown explicitly. At the surface, where $\mathbf{P}^S$ is localized, all the components of the electric field $\boldsymbol{\mathcal{E}}$ are continuous, and, in particular, $\mathcal{E}_3^-(0)=\mathcal{E}_3^+(0)=\mathcal{E}_3(0)$. It is the field $\boldsymbol{\mathcal{E}}(0)$ that is related to the surface polarization through a surface susceptibility tensor. If the vacuum gap is collapsed, both a and b tend to zero, and we have $\varepsilon_0^{-1}\mathcal{D}_3(a)=\mathcal{E}_3^+(a)\to\mathcal{E}_3(0)$ and $\varepsilon_0^{-1}\mathcal{D}_3(b)=\mathcal{E}_3^-(b)\to\mathcal{E}_3(0)$. Hence the normal component of the driving field can be defined exactly at the surface as $\mathcal{E}_3(0)=\varepsilon_0^{-1}\mathcal{D}_3(0)$. This justifies the particular conventions[21] avoiding the discontinuity of the driving field across the ideal surface:

$$P_i^S=\varepsilon_0\chi_{ijk}^S(\mathbf{m}^S)F_jF_k, \tag{9}$$

where $F_1=\mathcal{E}_1$, $F_2=\mathcal{E}_2$, and $F_3=\varepsilon_0^{-1}\mathcal{D}_3$. The surface susceptibility tensor χ^S_{ijk} in Eq. (9) depends on the surface magnetization whose direction is indicated by the unit vector $\mathbf{m}^S$. Alternatively, for the same surface polarization two other conventions are possible and equally acceptable:

$$P_i^S=\varepsilon_0\chi^{S+}_{ijk}(\mathbf{m}^S)\mathcal{E}_j^+\mathcal{E}_k^+ , \tag{10a}$$

$$P_i^S=\varepsilon_0\chi^{S-}_{ijk}(\mathbf{m}^S)\mathcal{E}_j^-\mathcal{E}_k^- . \tag{10b}$$

Here $\mathcal{E}_i^+$ and $\mathcal{E}_i^-$ are components of the field $\boldsymbol{\mathcal{E}}$ at the positive and negative sides of the interface, respectively, as defined by a positive direction of the earlier introduced unit normal $\mathbf{N}$ [Fig. 2(a)].

It is wholly immaterial which convention is preferred. The form (symmetry) of the corresponding susceptibility tensor remains the same, i.e., all the relevant tensors have coinciding indices for their zero components. However, a difference occurs but is solely in the values of the nonzero components that are linked to the third component of the driving field. The form of the tensors is determined by their intrinsic symmetry as well as by specific magnetic symmetry of the interface. The former is universal and apparently consists in invariance of the tensor under the permutation of its last two indices. Since $\mathbf{P}^S$ must not depend on a choice of any possible conventions, the three quadratic equations, concerning the relationship between Eqs. (10a) and (10b),

$$\begin{aligned}&\chi^{S+}_{i11}\mathcal{E}_1^2+\chi^{S+}_{i22}\mathcal{E}_2^2+\chi^{S+}_{i33}(\mathcal{E}_3^+)^2+2\chi^{S+}_{i23}\mathcal{E}_2\mathcal{E}_3^+\\&\quad+2\chi^{S+}_{i13}\mathcal{E}_1\mathcal{E}_3^++2\chi^{S+}_{i12}\mathcal{E}_1\mathcal{E}_2\\&\quad=\chi^{S-}_{i11}\mathcal{E}_1^2+\chi^{S-}_{i22}\mathcal{E}_2^2+\chi^{S-}_{i33}(\mathcal{E}_3^-)^2+2\chi^{S-}_{i23}\mathcal{E}_2\mathcal{E}_3^-\\&\qquad+2\chi^{S-}_{i13}\mathcal{E}_1\mathcal{E}_3^-+2\chi^{S-}_{i12}\mathcal{E}_1\mathcal{E}_2\end{aligned}\tag{11}$$

(where $i=1$, 2, or 3) that involve the four field components must be met along with the essential requirement for the tensors χ^{S+}_{ijk} and χ^{S-}_{ijk} to have the same form. The boundary condition for $\mathcal{D}_3$,

$$\varepsilon_{31}^+\mathcal{E}_1+\varepsilon_{32}^+\mathcal{E}_2+\varepsilon_{33}^+\mathcal{E}_3^+=\varepsilon_{31}^{-1}\mathcal{E}_1+\varepsilon_{32}^-\mathcal{E}_2+\varepsilon_{33}^-\mathcal{E}_3^- ,$$

where the signs labelling the components of the permittivity tensor refer to the layers above and below the interface according to the direction of $\mathbf{N}$, allows Eqs. (11) to become a system of three homogeneous equations with respect to three field components. Since the system must be satisfied for arbitrary field components, the following relationships between the two conventions come about:

$$\begin{aligned}\chi^{S-}_{i11}&=\chi^{S+}_{i11}-2\chi^{S+}_{i13}(\varepsilon_{31}^+-\varepsilon_{31}^-)/\varepsilon_{33}^++\chi^{S+}_{i33}[(\varepsilon_{31}^+-\varepsilon_{31}^-)/\varepsilon_{33}^+]^2,\\ \chi^{S-}_{i22}&=\chi^{S+}_{i22}-2\chi^{S+}_{i23}(\varepsilon_{32}^+-\varepsilon_{32}^-)/\varepsilon_{33}^++\chi^{S+}_{i33}[(\varepsilon_{32}^+-\varepsilon_{32}^-)/\varepsilon_{33}^+]^2,\\ \chi^{S-}_{i33}&=\chi^{S+}_{i33}(\varepsilon_{33}^-/\varepsilon_{33}^+)^2,\\ \chi^{S-}_{i23}&=\chi^{S+}_{i23}\varepsilon_{33}^-/\varepsilon_{33}^+-\chi^{S+}_{i33}\varepsilon_{33}^-(\varepsilon_{32}^+-\varepsilon_{32}^-)/(\varepsilon_{33}^+)^2,\\ \chi^{S-}_{i13}&=\chi^{S+}_{i13}\varepsilon_{33}^-/\varepsilon_{33}^+-\chi^{S+}_{i33}\varepsilon_{33}^-(\varepsilon_{31}^+-\varepsilon_{31}^-)/(\varepsilon_{33}^+)^2,\\ \chi^{S-}_{i12}&=\chi^{S+}_{i12}-\chi^{S+}_{i23}(\varepsilon_{31}^+-\varepsilon_{31}^-)/\varepsilon_{33}^+-\chi^{S+}_{i13}(\varepsilon_{32}^+-\varepsilon_{32}^-)/\varepsilon_{33}^+\\&\quad+\chi^{S+}_{i33}(\varepsilon_{31}^+-\varepsilon_{31}^-)(\varepsilon_{32}^+-\varepsilon_{32}^-)/(\varepsilon_{33}^+)^2.\end{aligned}\tag{12a}$$

Alternatively, to invert these expressions (to have χ^{S+} in terms of χ^{S-}), it is necessary merely to change all the superscripts "+" for "−" and vice versa. A similar rescaling procedure can be derived for the conventions defined by Eqs. (9) and (10a):

$$\begin{aligned}\chi^{S+}_{i11}&=\chi^{S}_{i11}+2\chi^{S}_{i13}\varepsilon_{31}^++\chi^{S}_{i33}(\varepsilon_{31}^+)^2,\\ \chi^{S+}_{i22}&=\chi^{S}_{i22}+2\chi^{S}_{i23}\varepsilon_{32}^++\chi^{S}_{i33}(\varepsilon_{32}^+)^2,\\ \chi^{S+}_{i33}&=\chi^{S}_{i33}(\varepsilon_{33}^+)^2,\\ \chi^{S+}_{i23}&=\chi^{S}_{i23}\varepsilon_{33}^++\chi^{S}_{i33}\varepsilon_{32}^+\varepsilon_{33}^+,\\ \chi^{S+}_{i13}&=\chi^{S}_{i13}\varepsilon_{33}^++\chi^{S}_{i33}\varepsilon_{31}^+\varepsilon_{33}^+,\\ \chi^{S+}_{i12}&=\chi^{S}_{i12}+\chi^{S}_{i23}\varepsilon_{31}^++\chi^{S}_{i13}\varepsilon_{32}^++\chi^{S}_{i33}\varepsilon_{31}^+\varepsilon_{32}^+.\end{aligned}\tag{12b}$$

Equations (12) constitute the most general rescaling procedures, which are undoubtedly correct if all the 18 components of the surface susceptibility tensor, related to a particular convention, are different. However, if there exist zero components along with independent ones, serious doubts arise about the validity of such rescaling. In other words, the inevitable arbitrariness in choosing the driving field not always maintains the identity of symmetry properties of χ^S_{ijk}, χ^{S+}_{ijk}, and χ^{S-}_{ijk}. After these properties have been considered in Sec. V, an example of such an inconsistency will be given. This rather bewildering feature of the ideal phenomenological model vanishes if all the off-diagonal components ε_{31} and ε_{32} in Eqs. (12) are zero. For instance, an optically isotropic multilayer in the polar configuration (magnetization is normal to the interface) obeys this restriction. Thus, the three conventions become equivalent in the sense that, as follows from Eqs. (12), the tensors χ^S_{ijk}, χ^{S+}_{ijk}, and χ^{S-}_{ijk} are simply related to one another:

$$\begin{aligned}\chi^S_{ijk}&=\chi^{S+}_{ijk}=\chi^{S-}_{ijk},\quad j,k\neq 3,\\ \chi^S_{ij3}&=\chi^{S+}_{ij3}/\varepsilon_{33}^+=\chi^{S-}_{ij3}/\varepsilon_{33}^-,\quad j\neq 3,\\ \chi^S_{i33}&=\chi^{S+}_{i33}/(\varepsilon_{33}^+)^2=\chi^{S-}_{i33}/(\varepsilon_{33}^-)^2.\end{aligned}\tag{13}$$

The rescaling procedure defined by Eqs. (13) is also legitimate when the off-diagonal components are so small that the contributions to the driving field they cause can be neglected. If they are related exclusively to magnetization (as normally occurs in linear magneto-optics), then that is the case.

Whether or not any possible convention, associated with the choice of the driving field, is related to the others through rescaling procedures, the components of the associated surface susceptibility tensor are entirely material parameters. Their values (normally unknown) can only be obtained from experimental data on the state of polarization of second harmonic light.[4] To avoid ambiguity, it is absolutely essential to state clearly the convention preferred when obtaining and presenting such data.

IV. ALTERNATIVE BOUNDARY CONDITIONS

Another set of boundary conditions can arise if the response of the whole medium [Fig. 1(a)] is interpreted differently to that outlined in Sec. II. This certainly occurs when the displacement, in contrast with Eq. (2), is represented through the permittivity tensor as

$$\mathbf{D}=\varepsilon_0\hat{\varepsilon}\mathbf{E}+\mathbf{P}^{NV}+\mathbf{P}^S\delta_S, \tag{14}$$

and, in addition to this, the tensor itself is artificially defined at the surface to make this representation meaningful. For instance, let $\varepsilon_{ij}^+=\delta_{ij}+\chi_{ij}^{V+}$ if $x_3\geqslant 0$, and $\varepsilon_{ij}^-=\delta_{ij}+\chi_{ij}^{V-}$ if $x_3<0$. Clearly, the incorporation of the permittivity tensor in this way is actually equivalent to imposing some singularity on the bulk polarization, which should realistically be unrelated to any singularity at all. Nevertheless, it is necessary to find out the consequences of such incorporation, and the method described in Sec. II can readily be applied for this purpose.

In this case boundary condition (4) for the normal component of $\mathbf{H}$ obviously remains the same. By virtue of Eq. (14), Eq. (3c) is transformed into

$$\varepsilon_0\,\mathrm{div}(\hat{\varepsilon}\mathbf{E}+\mathbf{P}^{NV})+(\partial P_1^S/\partial x_1+\partial P_2^S/\partial x_2)\,\delta_S+\partial(P_3^S\delta_S)/\partial x_3 \\ =0, \tag{15}$$

where the double layer can be annihilated if the field $\hat{\varepsilon}^+\mathbf{E}$ comprises the simple layer $-\varepsilon_0^{-1}(0,0,P_3^S\delta_S)$. Since the medium in the upper half-space [Fig. 2(a)] may be optically isotropic, i.e., $\varepsilon_{ij}^+=\varepsilon_{33}^+\delta_{ij}$, the singularity $-(\varepsilon_0\varepsilon_{33}^+)^{-1}P_3^S\delta_S$ must be associated with E_3. Therefore, the expression $\mathrm{div}\,\hat{\varepsilon}\mathbf{E}=\{\mathrm{div}(\hat{\varepsilon}\mathbf{E}+\mathbf{P}^{NV})\}+([\hat{\varepsilon}\mathbf{E}]_S,\mathbf{N})\,\delta_S-\varepsilon_0^{-1}\partial(P_3^S\delta_S)/\partial x_3$ on being substituted into Eq. (15), yields boundary condition (6). Since D_1 and D_2 are related to E_3 through the tensor, the extra singularities $-\varepsilon_{13}^+/\varepsilon_{33}^+P_3^S\delta_S$ and $-\varepsilon_{23}^+/\varepsilon_{33}^+P_3^S\delta_S$ are, respectively, inherited by them. This feature inevitably results in essential differences in the rest of the boundary conditions, compared to Eqs. (7) and (8). Indeed, taking into account the regular and singular terms of Eq. (3a), we arrive at

$$\{\mathrm{curl}\mathbf{H}\}-[[\mathbf{H}]_S,\mathbf{N}]\delta_S=-i\omega_S\{\mathbf{D}\}-i\omega_S(P_1^S,P_2^S,0)\,\delta_S \\ +i\omega_S(\varepsilon_{13}^+/\varepsilon_{33}^+,\varepsilon_{23}^+/\varepsilon_{33}^+,1)P_3^S\delta_S,$$

which leads to the boundary conditions for the tangential components of $\mathbf{H}$:

$$H_1^+-H_1^-=-i\omega_S(P_2^S-\varepsilon_{23}^+/\varepsilon_{33}^+P_3^S),$$

$$H_2^+-H_2^-=i\omega_S(P_1^S-\varepsilon_{13}^+/\varepsilon_{33}^+P_3^S). \tag{16}$$

This is clearly different from the previous condition [Eq. (7)]. Likewise, Eq. (3b) transforms into

$$\{\mathrm{curl}\mathbf{E}\}-[[\mathbf{E}]_S,\mathbf{N}]\delta_S-(\varepsilon_0\varepsilon_{33}^+)^{-1}\mathrm{curl}(0,0,P_3^S\delta_S) \\ =i\omega_S\mu_0\{\mathbf{H}\}$$

yielding the boundary conditions for the tangential components of $\mathbf{E}$:

$$E_1^+-E_1^-=-(\varepsilon_0\varepsilon_{33}^+)^{-1}\partial P_3^S/\partial x_1,$$

$$E_2^+-E_2^-=-(\varepsilon_0\varepsilon_{33}^+)^{-1}\partial P_3^S/\partial x_2. \tag{17}$$

Again, this is different from Eq. (8). On the basis of the assumption, stated in the beginning of this section, boundary conditions (4), (6), (16), and (17) have been derived (in Gaussian units) in the classical way.[13] Furthermore, they have been used to develop a phenomenological model of second harmonic magneto-optical Kerr effects for semi-infinite media.[23]

The surface polarization $\mathbf{P}^S$, naturally going together with definition (14), is $P_i^S=\varepsilon_0\eta_{ijk}^{S+}(\mathbf{m}^S)\mathcal{E}_j^+\mathcal{E}_k^+$, where the susceptibility tensor η_{ijk}^{S+} is different from χ_{ijk}^{S+} in Eq. (10). If the tensor $\hat{\varepsilon}$ were defined at the surface as $\hat{\varepsilon}^-$, instead of $\hat{\varepsilon}^+$, the components ε_{ij}^+ would need to be replaced by ε_{ij}^- in Eqs. (16) and (17). Consequently, we have another set of boundary conditions,

$$H_1^+-H_1^-=-i\omega_S(P_2^S-\varepsilon_{23}^-/\varepsilon_{33}^-P_3^S),$$

$$H_2^+-H_2^-=i\omega_S(P_1^S-\varepsilon_{13}^-/\varepsilon_{33}^-P_3^S),$$

$$E_1^+-E_1^-=-(\varepsilon_0\varepsilon_{33}^-)^{-1}\partial P_3^S/\partial x_1,$$

$$E_2^+-E_2^-=-(\varepsilon_0\varepsilon_{33}^-)^{-1}\partial P_3^S/\partial x_2,$$

that involves the surface polarization defined through the surface susceptibility tensor η_{ijk}^{S-} as $P_i^S=\varepsilon_0\eta_{ijk}^{S-}(\mathbf{m}^S)\mathcal{E}_j^-\mathcal{E}_k^-$. In contrast with Eq. (2), definition (14) results in a dichotomy which might have caused an ambiguity about the boundary conditions in question. The rescaling procedure for η_{ijk}^{S-} and η_{ijk}^{S+} is similar to that considered in Sec. III, for χ_{ijk}^{S-} and χ_{ijk}^{S+}. Moreover, the relationship, for instance, between χ_{ijk}^{S+} and χ_{ijk}^{S+},

$$\chi_{1jk}^{S+}=\eta_{1jk}^{S+}-\eta_{3jk}^{S+}\varepsilon_{13}^+/\varepsilon_{33}^+,$$

$$\chi_{2jk}^{S+}=\eta_{2jk}^{S+}-\eta_{3jk}^{S+}\varepsilon_{23}^+/\varepsilon_{33}^+,$$

$$\chi_{3jk}^{S+}=\eta_{3jk}^{S+}/\varepsilon_{33}^+,$$

coming from the two sets of boundary conditions makes these sets totally equivalent.

V. BOUNDARY VALUE PROBLEM AND SYMMETRY OF THE MULTILAYERS

The two versions of boundary conditions and associated conventions on the involved surface polarization are a foundation allowing us to set up, unambiguously, a boundary value problem for analyzing second harmonic magneto-optical effects in ferromagnetically ordered multilayers [Fig. 1(a)].

It follows from Eqs. (3) that in each layer characterized by its own permittivity tensor, as well as in the transparent medium, the electric field **E** must obey the wave equation

$$\nabla\,\mathrm{div}\,\mathbf{E}-\nabla^2\mathbf{E}=k_{0S}^2(\hat{\varepsilon}\mathbf{E}+\varepsilon_0^{-1}\mathbf{P}^{NV}), \tag{18}$$

where all the quantities, including the wave number $k_{0S}=\omega_S/c$, are defined at the frequency ω_S. The corresponding magnetic fields in the layers are available through Eq. (3b). At the interfaces between adjacent layers, or between the outer layers and transparent medium, the fields **E** and **H** must also obey boundary conditions (7) and (8). The rest of boundary conditions, i.e., Eqs. (4) and (6), are then met automatically. A solution of this linear boundary value problem, allowing us to obtain the field **E** in the transparent medium, is quite straightforward, and most simple within the plane-wave approximation. This faciliates a modified formalism of characteristic matrices to be used, which ultimately leads to a required expression for **E** involving all the permittivity tensors and volume polarizations of the layers as well as surface polarizations of the interfaces. An adequate consideration of these quantities is only possible within microscopic theory,[3,20] which is beyond the scope of his paper. However, the symmetry of the tensors is macroscopic,[1–3,25,26] and will be considered concisely below.

Let the multilayer exhibit, exclusively, ferromagnetic ordering and each of its magnetic layers be homogeneously magnetized. The internal magnetization is determined by the arbitrarily directed unit vector **m**. Normally, the magneto-optical effects, being linear in magnetization, are of significance. In this approximation, the susceptibility tensor of any layer can be represented as $\chi_{ij}^V(\mathbf{m})=\tilde{\chi}_{ij}+\tilde{\chi}_{ijk}m_k$, where $\tilde{\chi}_{ij}$ and $\tilde{\chi}_{ijk}$ are *i* tensors, the former being polar and the latter axial, since m_i is an axial *c* tensor.[24] The notations we use are as follows. The tilde ($\sim$), or a letter (V,S) above the χ tensors, means their definition in the physical coordinate system [axes X_1, X_2, and X_3; see Fig. 1(b)], which is transformed into the crystallographic coordinate system (axes X_1^C, X_2^C, and $X_3^C=X_3$) by a clockwise rotation about X_3 (in viewing against this axis) through an angle ψ, the axis X_1 being chosen as a reference for the angle. A lack of these symbols refers to a definition of the tensors in the crystallographic coordinate system. A simplification of $\tilde{\chi}_{ij}$ and $\tilde{\chi}_{ijk}$ comes from their intrinsic symmetry: $\tilde{\chi}_{ij}=\tilde{\chi}_{ji}$, $\tilde{\chi}_{ijk}=-\tilde{\chi}_{jik}$.[2,24,25] In accordance with Neumann's principle, they must also be invariant under the ordinary point group that describes the crystallographic symmetry of the layer.[24,26] The invariance implies a matrix representation of a particular point group—an isomorphic group of 3×3 matrices. For any matrix $\hat{C}$ belonging to the group, the equations $\chi_{ij}=C_{ik}C_{jl}\chi_{kl}$ and $\chi_{ijk}=(\det\hat{C})C_{il}C_{jm}C_{kn}\chi_{lmn}$ are held and signify transformations of the tensors into themselves. Such equations constitute the direct inspection method for simplifying tensors, i.e., for revealing their zero and independent components. To ensure maximum simplification it is sufficient to engage consecutively all the generating matrices of the group.[24] If the layers are optically isotropic and are described by one of the groups $m3m$, $m3$, or the Curie group $\infty\infty m$, each possessing inversion symmetry, then it is easy to show that $\chi_{ij}=\chi_{11}\delta_{ij}$ and $\chi_{ijk}=\chi_{123}e_{ijk}$, where e_{ijk} is the Levi-Civita tensor (permutation symbol). Since δ_{ij} and e_{ijk} are invariant under rotation, we have $\chi_{ij}^V(\mathbf{m})=\chi_{11}\delta_{ij}+\chi_{123}e_{ijk}m_k$. Hence, for any optically isotropic layer with magnetization along the unit vector **m**, the permittivity tensor is

$$\varepsilon_{ij}=n^2(\delta_{ij}-iQe_{ijk}m_k), \tag{19}$$

where n is the complex refractive index [$n^2=1+\chi_{11}$; $\mathrm{Im}\,n>0$, since the time-dependent factor $\exp(-i\omega_S t)$ is used], and $Q=in^{-2}\chi_{123}$ is the complex magneto-optical parameter, which is small, $|Q|\ll1$.

Often, if the linear approximation with respect to magnetization is ignored, a symmetry analysis of the tensors has to be carried out on the basis of their invariance under magnetic point groups (identical to black-and-white point groups because time inversion and color changing are equivalent operations).[19,24] Such an analysis shows that the number of independent tensor components depends dramatically on the magnetization direction. This complexity is obviously excessive unless magneto-optical effects of higher order in magnetization have to be considered.

Since we have been pursuing the linear approximation, for any interface, which is magnetized in an arbitrary direction of the unit vector $\mathbf{m}^S$, the surface susceptibility tensor introduced in accordance with convention (9) can be decomposed as $\chi_{ijk}^S(\mathbf{m}^S)=\tilde{\chi}_{ijk}+\tilde{\chi}_{ijkl}m_l^S$. The polar and axial *i* tensor in this expression possesses an apparent intrinsic symmetry: $\tilde{\chi}_{ijk}=\tilde{\chi}_{ikj}$, $\tilde{\chi}_{ijkl}=\tilde{\chi}_{ikjl}$. Hence the number of independent components is reduced, respectively, to 18 and 54. Further simplification may be due to the invariance of χ_{ijk} and χ_{ijkl} under an ordinary point group of the interface.[15,24–26] The direct inspection method implies a matrix representation of the group and a consecutive involvement of associated matrices in the equations $\chi_{ijk}=C_{il}C_{jm}C_{kn}\chi_{lmn}$, $\chi_{ijkl}=(\det\hat{C})C_{im}C_{jn}C_{kp}C_{lq}\chi_{mnpq}$. The generating matrices should necessarily be involved for revealing utmost simplification bestowed by symmetry.[24] It is convenient to rewrite Eq. (9) in a matrix form,

$$\begin{bmatrix} P_1^S\\ P_2^S\\ P_3^S \end{bmatrix}=\varepsilon_0(\hat{A}+m_1^S\hat{B}_1-m_2^S\hat{B}_2+m_3^S\hat{B}_3)\begin{bmatrix} F_1^2\\ F_2^2\\ F_3^2\\ 2F_2F_3\\ 2F_1F_3\\ 2F_1F_2 \end{bmatrix}, \tag{20}$$

where the four 3×6 matrices on the RHS are to be defined in the physical coordinate system, i.e., they are sensitive to the angle ψ, unless the interface is isotropic. Matrix $\hat{A}$, corresponding to $\tilde{\chi}_{ijk}$ and governing second harmonic optical effects, is well known for all point groups.[27] This is not so for the other three matrices, which are associated, respectively, with the transverse, longitudinal, and polar surface-sensitive second harmonic magneto-optical effects. It is important to see that the numbered configurations are defined with reference to the plane of incidence $x_1=0$ [Fig. 1(a)].

The matrices will be given below for often encountered point groups, and are particularly relevant, as an example, to an interface that separates fcc layers.

Group $4mm$. The generating matrices are:

$$\begin{bmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad \begin{bmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (001) \text{ interface},$$

$$\hat{A}=\begin{bmatrix} 0 & 0 & 0 & 0 & \chi_{113} & 0 \\ 0 & 0 & 0 & \chi_{113} & 0 & 0 \\ \chi_{311} & \chi_{311} & \chi_{333} & 0 & 0 & 0 \end{bmatrix},$$

$$\hat{B}_1=\begin{bmatrix} \tilde{\chi}_{1111} & -\tilde{\chi}_{1111} & 0 & 0 & 0 & \tilde{\chi}_{1121} \\ \tilde{\chi}_{2111} & \tilde{\chi}_{2221} & \chi_{2331} & 0 & 0 & -\tilde{\chi}_{1111} \\ 0 & 0 & 0 & \chi_{3231} & 0 & 0 \end{bmatrix},$$

$$\hat{B}_2=\begin{bmatrix} \tilde{\chi}_{2221} & \tilde{\chi}_{2111} & \chi_{2331} & 0 & 0 & \tilde{\chi}_{1111} \\ \tilde{\chi}_{1111} & -\tilde{\chi}_{1111} & 0 & 0 & 0 & \tilde{\chi}_{1121} \\ 0 & 0 & 0 & 0 & \chi_{3231} & 0 \end{bmatrix},$$

$$\hat{B}_3=\begin{bmatrix} 0 & 0 & 0 & \chi_{1233} & 0 & 0 \\ 0 & 0 & 0 & 0 & -\chi_{1233} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix},$$

where $\tilde{\chi}_{1111}=\frac{1}{4}\Delta\sin 4\psi$, $\tilde{\chi}_{1121}=\chi_{1121}-\frac{1}{2}\Delta\sin^2 2\psi$, $\tilde{\chi}_{2111}=\chi_{2111}-\frac{1}{2}\Delta\sin^2 2\psi$, $\tilde{\chi}_{2221}=\chi_{2221}+\frac{1}{2}\Delta\sin^2 2\psi$, and $\Delta=\chi_{2111}-\chi_{2221}+2\chi_{1121}$. There are nine independent parameters: three optical and six magneto-optical.[28] The interface is optically isotropic,[29] for the optical parameters do not depend on ψ, but magneto-optically anisotropic. The state of polarization and intensity of second harmonic light would exhibit a fourfold rotational anisotropy. The effect observed in Bi-substituted iron-garnet films[16] is an excellent experimental confirmation. If the linear approximation with respect to magnetization is not used, then particularly for the in-plane magnetization $\mathbf{m}^S \| [110]$ invariance of the tensor $\chi^S_{ijk}(\mathbf{m}^S)$ under the magnetic point group $mm2$ has to be considered. This leaves ten independent parameters to be dealt with.[19]

The obtained tensor can readily be used now to show, as stated in Sec. III, that the rescaling procedures [Eq. (12)] may not always be carried out. Let both adjacent layers be characterized by tensor (19), so that, for arbitrary directions of their magnetizations, $\varepsilon^{\pm}_{31}=-im^{\pm}_2(n^{\pm})^2Q^{\pm}$, $\varepsilon^{\pm}_{32}=im^{\pm}_1(n^{\pm})^2Q^{\pm}$, and $\varepsilon^{\pm}_{33}=(n^{\pm})^2$. The vector $\mathbf{m}^S$ may also have any direction. In the linear approximation, with respect to magnetization, procedure (12a) particularly leads to the equations

$$m^S_1\tilde{\chi}^-_{2111}-m^S_2\tilde{\chi}^-_{1111}=m^S_1\tilde{\chi}^+_{2111}-m^S_2\tilde{\chi}^+_{1111},$$

$$m^S_1\tilde{\chi}^-_{1111}+m^S_2\tilde{\chi}^-_{2111}=m^S_1\tilde{\chi}^+_{1111}+m^S_2\tilde{\chi}^+_{2111},$$

$$m^S_1\tilde{\chi}^-_{1111}-m^S_2\tilde{\chi}^-_{2221}=m^S_1\tilde{\chi}^+_{1111}-m^S_2\tilde{\chi}^+_{2221}+2i\chi^+_{113}[m^+_2Q^+ - m^-_2(n^-/n^+)^2Q^-],$$

$$m^S_1\tilde{\chi}^-_{2221}+m^S_2\tilde{\chi}^-_{1111}=m^S_1\tilde{\chi}^+_{2221}+m^S_2\tilde{\chi}^+_{1111}-2i\chi^+_{113}[m^+_1Q^+ - m^-_1(n^-/n^+)^2Q^-],$$

$$m^S_1\chi^-_{3231}=\{m^S_1\chi^+_{3231}-i\chi^+_{333}[m^+_1Q^+-m^-_1(n^-/n^+)^2Q^-)\}\times(n^-/n^+)^2,$$

$$m^S_2\chi^-_{3231}=\{m^S_2\chi^+_{3231}-i\chi^+_{333}[m^+_2Q^+-m^-_2(n^-/n^+)^2Q^-]\}\times(n^-/n^+)^2,$$

which are seen to become contradictory unless direction of magnetization both in the layers and at the interface is the same ($\mathbf{m}^+=\mathbf{m}^-=\mathbf{m}^S$). This is even more obvious for the tensors defined in the crystallographic coordinate system, since $\psi=0$ entails the disappearance of $\tilde{\chi}_{1111}$.

Group ∞m. The interface described by this Curie group is isotropic in its plane. The matrices derived for the group $4mm$ are applicable, provided that $\Delta=0$. Therefore, eight independent parameters (three optical and five magneto-optical) characterize such an interface. It obviously exhibits no rotational anisotropy. According to Eq. (20), the nonlinear surface polarization can now be written

$$\begin{aligned}\varepsilon_0^{-1}\mathbf{P}^S=&2\chi_{113}(\mathbf{N},\mathbf{F})\mathbf{F}+\chi_{311}(\mathbf{F},\mathbf{F})\mathbf{N}\\ &+(\chi_{333}-\chi_{311}-2\chi_{113})(\mathbf{N},\mathbf{F})^2\mathbf{N}\\ &+2\chi_{1121}([\mathbf{N},\mathbf{m}^S],\mathbf{F})\mathbf{F}+\chi_{2111}(\mathbf{F},\mathbf{F})[\mathbf{N},\mathbf{m}^S]\\ &-2\chi_{1233}(\mathbf{N},\mathbf{F})[\mathbf{m}^S,\mathbf{F}]\\ &-(\chi_{2111}-\chi_{2331}+2\chi_{1233})(\mathbf{N},\mathbf{F})^2[\mathbf{N},\mathbf{m}^S]\\ &+2(\chi_{3231}+\chi_{1233}-\chi_{1121})(\mathbf{N},\mathbf{F})([\mathbf{N},\mathbf{m}^S],\mathbf{F})\mathbf{N}.\end{aligned}$$

The axial vector $\mathbf{m}^S$ is seen to form only those combinations with the polar vectors $\mathbf{N}=(0,0,1)$ and $\mathbf{F}$, which insure that $\mathbf{P}^S$ remains a polar vector. A similar, though slightly different, expression was already given.[11,23] However, it may only be valid in the linear approximation with a small parameter associated with $\mathbf{N}$.

Group $mm2$. The generating matrices are

$$\begin{bmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad \begin{bmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (110) \text{ interface}.$$

To avoid cumbersome expressions we give tensors χ_{ijk} and χ_{ijkl} in the crystallographic coordinate system, i.e., for $\psi=0$. It is then fairly straightforward to carry out their transformations $\tilde{\chi}_{ijk}=C_{il}C_{jm}C_{kn}\chi_{lmn}$ and $\tilde{\chi}_{ijkl}=C_{im}C_{jn}C_{kp}C_{lq}\chi_{mnpq}$, with the matrix

$$\hat{C}=\begin{bmatrix} \cos\psi & \sin\psi & 0 \\ -\sin\psi & \cos\psi & 0 \\ 0 & 0 & 1 \end{bmatrix}.$$

Concerning the fcc layers, the directions $[\bar{1}10]$, [001], and [110] are now along the axes X_1, X_2, and X_3. The matrices comprising Eq. (20) are

$$\hat{A}=\begin{bmatrix} 0 & 0 & 0 & 0 & \chi_{113} & 0 \\ 0 & 0 & 0 & \chi_{223} & 0 & 0 \\ \chi_{311} & \chi_{322} & \chi_{333} & 0 & 0 & 0 \end{bmatrix},$$

$$\hat{B}_1=\begin{bmatrix} 0 & 0 & 0 & 0 & 0 & \chi_{1121} \\ \chi_{2111} & \chi_{2221} & \chi_{2331} & 0 & 0 & 0 \\ 0 & 0 & 0 & \chi_{3231} & 0 & 0 \end{bmatrix},$$

$$\hat{B}_2=-\begin{bmatrix} \chi_{1112} & \chi_{1222} & \chi_{1332} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & \chi_{2122} \\ 0 & 0 & 0 & 0 & \chi_{3132} & 0 \end{bmatrix},$$

$$\hat{B}_3=\begin{bmatrix} 0 & 0 & 0 & \chi_{1233} & 0 & 0 \\ 0 & 0 & 0 & 0 & \chi_{2133} & 0 \\ 0 & 0 & 0 & 0 & 0 & \chi_{3123} \end{bmatrix}.$$

The number of independent parameters is 18 (five optical and 13 magneto-optical).[28,30] The in-plane magnetization $\mathbf{m}^S \| X_1$ allows ten of them to survive. The same configuration is described by the magnetic point group $\underline{2}$, which yields 18 independent parameters[31] of the tensor $\chi^S_{ijk}(\mathbf{m}^S)$. Magneto-optical effects coming from such an interface can be shown to exhibit a twofold rotational anisotropy.[16]

Group $3m$. The generating matrices are

$$\begin{bmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad \begin{bmatrix} -1/2 & \sqrt{3}/2 & 0 \\ -\sqrt{3}/2 & -1/2 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (111)\ \text{interface}.$$

The directions $[2\bar{1}\bar{1}]$, $[01\bar{1}]$, and [111] are along the axes X^C_1, X^C_2, and X_3, a vertical plane of symmetry being normal to X^C_2. In this case there are 12 independent parameters (four optical and eight magneto-optical)[28] in the matrices

$$\hat{A}=\begin{bmatrix} \tilde{\chi}_{111} & -\tilde{\chi}_{111} & 0 & 0 & \chi_{113} & \tilde{\chi}_{112} \\ \tilde{\chi}_{112} & -\tilde{\chi}_{112} & 0 & \chi_{113} & 0 & -\tilde{\chi}_{111} \\ \chi_{311} & \chi_{311} & \chi_{333} & 0 & 0 & 0 \end{bmatrix}, \quad \hat{B}_1=\begin{bmatrix} 0 & 0 & 0 & \tilde{\chi}_{1231} & \tilde{\chi}_{1131} & \chi_{1121} \\ \chi_{2111} & \chi_{2221} & \chi_{2331} & -\tilde{\chi}_{1131} & \tilde{\chi}_{1231} & 0 \\ \tilde{\chi}_{3111} & -\tilde{\chi}_{3111} & 0 & \chi_{3231} & 0 & \tilde{\chi}_{3121} \end{bmatrix},$$

$$\hat{B}_2=\begin{bmatrix} \chi_{2221} & \chi_{2111} & \chi_{2331} & \tilde{\chi}_{1131} & -\tilde{\chi}_{1231} & 0 \\ 0 & 0 & 0 & \tilde{\chi}_{1231} & \tilde{\chi}_{1131} & \chi_{1121} \\ -\tilde{\chi}_{3121} & \tilde{\chi}_{3121} & 0 & 0 & \chi_{3231} & \tilde{\chi}_{3111} \end{bmatrix}, \quad \hat{B}_3=\begin{bmatrix} \tilde{\chi}_{1113} & -\tilde{\chi}_{1113} & 0 & \chi_{1233} & 0 & \tilde{\chi}_{1123} \\ \tilde{\chi}_{1123} & -\tilde{\chi}_{1123} & 0 & 0 & -\chi_{1233} & -\tilde{\chi}_{1113} \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix},$$

where $\tilde{\chi}_{111}=\chi_{111}\cos 3\psi$, $\tilde{\chi}_{112}=-\chi_{111}\sin 3\psi$, $\tilde{\chi}_{1231}=\chi_{1231}\cos 3\psi$, $\tilde{\chi}_{1131}=\chi_{1231}\sin 3\psi$, $\tilde{\chi}_{3121}=\chi_{3121}\cos 3\psi$, $\tilde{\chi}_{3111}=\chi_{3121}\sin 3\psi$, $\tilde{\chi}_{1123}=\chi_{1123}\cos 3\psi$, and $\tilde{\chi}_{1113}=\chi_{1123}\sin 3\psi$, and the relation $\chi_{2111}-\chi_{2221}+2\chi_{1121}=0$ occurs. This result suggests a threefold rotational anisotropy of surface-sensitive magneto-optical effects, and this has experimental evidence.[16] The importance of the approximation used can be illustrated for $\mathbf{m}^S$ being normal to the interface. Hence two of the magneto-optical parameters have to be taken into consideration. The same result[19] also follows from invariance of the tensor $\chi^S_{ijk}(\mathbf{m}^S)$ under the magnetic point group $3\underline{m}$. However, this group turns into the trivial one if a direction of $\mathbf{m}^S$ becomes arbitrary. Such a reduction in symmetry inevitably entails 18 independent magneto-optical parameters, in contrast to the eight we have owing to the approximation.

Along with the surface polarization $\mathbf{P}^S$ comprising the boundary conditions, it is necessary to consider the volume polarization $\mathbf{P}^{NV}$ in wave equation (18). We confine this consideration to a layer possessing inversion symmetry. Therefore, $\mathbf{P}^{NV}$ has to be taken into account in the electric-quadrupole approximation[1,6,7,21,29,32]

$$P^{NV}_i=\varepsilon_0\chi^V_{ijkl}(\mathbf{m})\mathcal{E}_j\frac{\partial}{\partial x_k}\mathcal{E}_l, \tag{21}$$

where $\boldsymbol{\mathcal{E}}$ is the electric field (at the fundamental frequency) in the layer. In the decomposition of the nonlinear volume susceptibility tensor only terms linear in magnetization are to be retained: $\chi^V_{ijkl}(\mathbf{m})=\tilde{\chi}_{ijkl}+\tilde{\chi}_{ijkln}m_n$, where no intrinsic symmetry is assumed for the polar and axial i tensor.

Let symmetry of the layer be described by the point group $m3m$. Symmetry of the tensors χ_{ijkl} and χ_{ijkln} can be revealed in a very similar way as we have outlined above. The generating matrices are

$$\begin{bmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad \begin{bmatrix} 0 & 1 & 0 \\ 0 & 0 & 1 \\ 1 & 0 & 0 \end{bmatrix}, \quad \text{and} \quad \begin{bmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{bmatrix}.$$

The last matrix represents inversion and does not lead to any simplification of either tensor. There are 21 surviving components of χ_{ijkl}, four of them being independent.[27,32] In the physical coordinate system the nonzero components are

$$\tilde{\chi}_{1111}=\tilde{\chi}_{2222}=\chi_{1111}-\tfrac{1}{2}\delta\sin^2 2\psi,\quad \chi_{3333}=\chi_{1111},\tag{22}$$

$$\tilde{\chi}_{2212}=\tilde{\chi}_{1222}=\tilde{\chi}_{2221}=\tilde{\chi}_{2122}=-\tilde{\chi}_{1112}=-\tilde{\chi}_{1211}=-\tilde{\chi}_{2111}$$
$$=-\tilde{\chi}_{1121}=\tfrac{1}{4}\delta\sin 4\psi,$$
$$\tilde{\chi}_{1212}=\tilde{\chi}_{2121}=\chi_{1212}+\tfrac{1}{2}\delta\sin^2 2\psi,$$
$$\tilde{\chi}_{2112}=\tilde{\psi}_{1221}=\chi_{2112}+\tfrac{1}{2}\delta\sin^2 2\psi,$$
$$\tilde{\chi}_{2211}=\tilde{\chi}_{1122}=\chi_{1122}+\tfrac{1}{2}\delta\sin^2 2\psi,$$
$$\chi_{1313}=\chi_{2323}=\chi_{3232}=\chi_{3131}=\chi_{1212},$$
$$\chi_{3113}=\chi_{3223}=\chi_{1331}=\chi_{2332}=\chi_{2112},$$
$$\chi_{3311}=\chi_{3322}=\chi_{2233}=\chi_{1133}=\chi_{1122},$$

where $\delta=\chi_{1111}-\chi_{1122}-\chi_{1212}-\chi_{2112}$. This result allows us to observe how volume anisotropic optical contributions influence rotational anisotropy of magneto-optical effects.

If the layer is isotropic, the Curie group $\infty\infty m$ describes its symmetry. Relations (22) hold valid, provided that $\delta=0$. Consequently, there are three independent optical parameters, and the nonmagnetic part of $\mathbf{P}^{NV}$, as follows from Eq. (21), is

$$\varepsilon_0^{-1}\mathbf{P}^{NV}_{\mathrm{nonmagn}}=\chi_{2112}(\boldsymbol{\mathcal{E}},\nabla)\boldsymbol{\mathcal{E}}+\chi_{1122}\boldsymbol{\mathcal{E}}\,\mathrm{div}\,\boldsymbol{\mathcal{E}}+\tfrac{1}{2}\chi_{1212}\nabla(\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{E}}),\tag{23}$$

which is known,[31] although for a different combination of the involved parameters.[7,21,29,33]

Contributions due to the axial fifth-rank tensor must be fairly small. Nevertheless, to see their symmetry, the invariance of χ_{ijkln} under $\infty\infty m$ is worth considering as the simplest example. Only 60 of the 243 components are nonzero, six of them being independent.[28] On carrying out a symmetry analysis, the magnetic part of $\mathbf{P}^{NV}$ can be written down in vector form:

$$\begin{aligned}\varepsilon_0^{-1}\mathbf{P}^{NV}_{\mathrm{magn}}=&\chi_{12131}[[\mathbf{q},\nabla],\boldsymbol{\mathcal{E}}]+\chi_{31121}(\boldsymbol{\mathcal{E}},\nabla)\mathbf{q}+(\chi_{12131}\\&+\chi_{32111})\mathbf{q}\,\mathrm{div}\,\boldsymbol{\mathcal{E}}+\chi_{11321}\boldsymbol{\mathcal{E}}\,\mathrm{div}\,\mathbf{q}+\chi_{12311}(\mathbf{q},\nabla)\boldsymbol{\mathcal{E}}\\&+\tfrac{1}{2}\chi_{31211}[\mathbf{m},\nabla](\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{E}}),\end{aligned}\tag{24}$$

where $\mathbf{q}=[\mathbf{m},\boldsymbol{\mathcal{E}}]$. The field $\boldsymbol{\mathcal{E}}$ in Eq. (24), which we believe to be a new result, must be taken as unperturbed, i.e., independent of magneto-optical parameters.

At this stage the boundary value problem to look at second harmonic magneto-optical effects is unambiguously set up. It should also be clear that the simplest phenomenological model for dealing with linear surface magneto-optical effects follows a very similar formulation to the boundary value problem that has been given for the second harmonic case.

VI. SUMMARY

An analysis has been given of fundamental aspects related exclusively to a formulation of the boundary value problem for looking at optical and magneto-optical effects that originate within surfaces and interfaces of multilayered magnetic media. A particular and currently important example of such effects arises in the generation of second harmonic magneto-optical effects in ferromagnetically ordered multilayers that possess inversion symmetry. Another example is the possibility of specific surface contributions to the first-order linear magneto-optical effects.

Concerning second harmonic magneto-optical effects, it has been assumed that the response of the medium is adequately described in terms of surface and volume electric polarizations and that all the interfaces are ideal (infinitely thin). The polarizations are related, respectively (within electric-dipole and -quadrupole approximations), to the fundamental field through the surface and volume susceptibility tensors, which might be available from adequate microscopic theory or experiment. A cursory perusal of the literature related to this topic reveals considerable confusion in the mathematical treatment of surface-sensitive magneto-optical effects, particularly with respect to fundamental issues.

Here we have attempted to highlight some of the problems and provide a reasonably self-consistent analysis. A central issue concerns an elegant way of deriving two versions of the relevant boundary conditions: Eqs. (4) and (6)–(8), and alternatively a less simple set of equations (4), (6), (16), and (17). We have shown how this derivation can be carried out through validity of the differential form of Maxwell's equations in the space of generalized functions. Either version forms a crucial part of the boundary value problem for the wave equation (18). Its solution ultimately allows the state of polarization and intensity of light (generated solely from the surfaces and interfaces) to become known in the transparent medium, usually air, where these quantities determine the magneto-optical effects. We purposefully leave out quite straightforward solution of the boundary value problem.

For completeness, the relevant problem of revealing the symmetry of the form of the susceptibility tensors has been outlined in the linear approximation with respect to magnetization. This approximation, apart from being naturally reasonable (magneto-optical effects are normally small), has been shown to be advantageous, because it is sufficient to analyze the invariance of the corresponding tensors merely under ordinary crystallographic point groups. This leads to a significantly lower number of nonzero tensor components than would be delivered by the often-used invariance under magnetic point groups. To illustrate this, we have given corresponding expressions of the surface polarization [Eq. (20)] for the four typical point groups: ∞m, $4mm$, $mm2$, and $3m$. The volume polarization has been considered thoroughly for the Curie group $\infty\infty m$, its nonmagnetic part also for $m3m$. Expression (24), believed to be a new result, illustrates the role of the magnetic part of the volume polarization in an isotropic layer. The tensors have been defined in the physical coordinate system, and this allows the results to be used for analysing rotational anisotropy of second harmonic magneto-optical effects.

An additional and particularly important issue has also been discussed in relation to uncertainty in the definition of the surface polarization in terms of susceptibility tensor and fundamental electric field. An ambiguity arises because of the discontinuity of its normal component across an ideal interface. We have considered the implications of different conventions on such a definition and shown that, in the general case of anisotropic adjacent layers, unless certain components of their dielectric tensors are zero, there may be no relationship between the conventions. An example has been given to illustrate why this occurs within the linear approximation with respect to magnetization. If the condition is met, the relationships are simple and imply a rescaling procedure of some tensor components in accordance with Eqs. (13). Since the issue of convention is an inevitable feature of the simplest model described in this paper, we emphasize the need to state clearly the choice, which must be common to both theoretical and experimental treatments of magneto-optical effects.

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