

example of this is shown in table 4.3 where the uncertainty of Δ becomes larger at a large angle of incidence. The optimum choice of angles for measurement is a compromise between the need for increased sensitivity of the DEHM values, the absence of cross correlation, and the uncertainty of Δ caused by inaccuracy of measurement of the angle of incidence.

The above computational considerations of MAI measurements have been successfully applied [175] to resolve numerous questions concerning this technique in earlier investigations particularly those of McCrackin and Colson [167], Burge and Bennet [156], Oldham [172], and Johnson and Bashara [179].

4.6. Reflection and transmission by isotropic stratified planar structures

The method of addition of multiple reflections becomes impractical when considering the reflection and transmission of polarized light at oblique incidence by a multilayer film between semi-infinite ambient and substrate media. A more elegant approach that employs 2×2 matrices will now be discussed, and is based on the fact that the equations that govern the propagation of light are linear and that the continuity of the tangential fields across an interface between two isotropic media can be regarded as a 2×2 -linear-matrix transformation.²⁴

Consider a stratified structure that consists of a stack of $1, 2, 3, \dots, j, \dots, m$ parallel layers (strata) sandwiched between two semi-infinite ambient (0) and substrate ($m+1$) media, fig. 4.38. Let all media be linear homogeneous and isotropic, and let the complex index of refraction of the j th layer be N_j and its thickness d_j . N_0 and N_{m+1} represent the complex indices of refraction of the ambient and substrate media, respectively. An incident monochromatic plane wave in medium 0 (the ambient) generates a resultant reflected plane wave in the same medium and a resultant transmitted plane wave in medium $m+1$ (the substrate). The total field inside the j th layer, which is excited by the incident plane wave, consists of two plane waves: a forward-travelling plane wave denoted by (+), and a backward-travelling plane wave denoted by (-). The wave-vectors of all plane waves lie in the same plane (the plane of incidence), and the wave-vectors of the two plane waves in the j th layer make equal angles with the z -axis which is perpendicular to the plane boundaries directed toward the substrate. When the incident wave in the ambient is linearly polarized with its electric vector

²⁴Work on the use of 2×2 -matrix methods to study the reflection and transmission of light by multilayered structures was pioneered by Abelès. (For an account of Abelès work see, for example, ref. 23, pp. 51ff.) The present development is similar to that used by Hayfield and White [146].

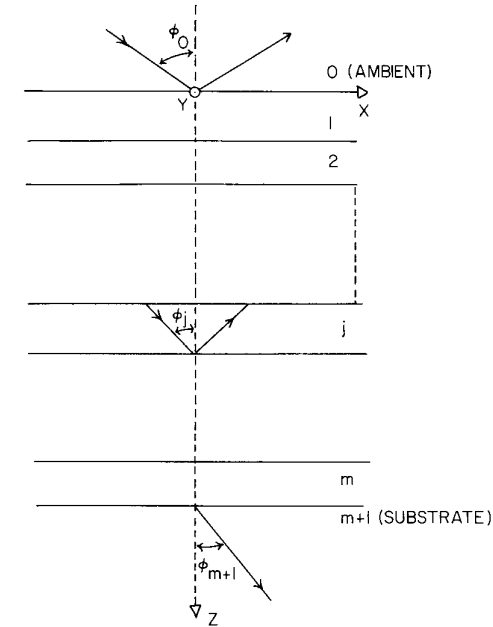


Fig. 4.38. Reflection and transmission of a plane wave by a multi-film structure (films $1, 2, \dots, m$) sandwiched between semi-infinite ambient (0) and substrate ($m+1$) media. ϕ_0 is the angle of incidence; ϕ_j and ϕ_{m+1} are the angles of refraction in the j th film and substrate, respectively.

vibrating parallel (p) or perpendicular (s) to the plane of incidence, all plane waves excited by that incident wave in the various layers of the stratified structure will be similarly polarized, parallel or perpendicular to the plane of incidence, respectively. In the following, it will be assumed that all waves are either p- or s-polarized.

Let $E^+(z)$ and $E^-(z)$ denote the complex amplitudes of the forward- and backward-travelling plane waves at an arbitrary plane z . The total field at z can be described by a 2×1 -column vector

$$\mathbf{E}(z) = \begin{bmatrix} E^+(z) \\ E^-(z) \end{bmatrix}. \quad (4.145)$$

If we consider the fields at two different planes z' and z'' parallel to the layer boundaries then, by virtue of system linearity, $\mathbf{E}(z'')$ and $\mathbf{E}(z')$ must be

related by a 2×2 -matrix transformation

$$\begin{bmatrix} E^+(z') \\ E^-(z') \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix} \begin{bmatrix} E^+(z'') \\ E^-(z'') \end{bmatrix}. \quad (4.146)$$

More concisely, eq. (4.146) can be written as

$$E(z') = SE(z''), \quad (4.147)$$

where

$$S = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix}. \quad (4.148)$$

Note that S must characterize that part of the stratified structure confined between the two parallel planes at z' and z'' .

By choosing z' and z'' to lie immediately on opposite sides of the $(j-1)j$ interface, located at z_j between layers $j-1$ and j , eq. (4.147) becomes

$$E(z_j - 0) = I_{(j-1)j} E(z_j + 0), \quad (4.149)$$

where $I_{(j-1)j}$ is a 2×2 matrix characteristic of the $(j-1)j$ interface alone. On the other hand, if z' and z'' are chosen inside the j th layer at its boundaries, eq. (4.147) becomes

$$E(z_j + 0) = L_j E(z_j + d_j - 0), \quad (4.150)$$

where L_j is a 2×2 matrix characteristic of the j th layer alone whose thickness is d_j . Only the reflected wave in the ambient medium and the transmitted wave in the substrate are accessible for measurement, so that it is necessary to relate their fields to those of the incident wave. By taking the planes z' and z'' to lie in the ambient and substrate media, immediately adjacent to the 01 and $m(m+1)$ interfaces respectively, eq. (4.147) will read

$$E(z_1 - 0) = SE(z_{m+1} + 0). \quad (4.151)$$

Equation (4.151) defines a *scattering matrix* S which represents the overall reflection and transmission properties of the stratified structure. S can be expressed as a product of the interface and layer matrices I and L that describe the effects of the individual interfaces and layers of the entire stratified structure, taken in proper order, as follows:

$$S = I_{01} L_1 I_{12} L_2 \dots I_{(j-1)j} L_j \dots L_m I_{m(m+1)} \quad (4.152)$$

Equation (4.152) may be proved readily by repeated application of eq. (4.147) to the successive interfaces and layers of the stratified structure, starting with the ambient-first film (01) interface and ending by the last film-substrate interface $[m(m+1)]$.

From eq. (4.152) it is evident that to determine the stratified structure scattering matrix S , the individual interface and layer matrices I and L have to be calculated.

The matrix I of an interface between two media a and b relates the fields on both its sides as

$$\begin{bmatrix} E_a^+ \\ E_a^- \end{bmatrix} = \begin{bmatrix} I_{11} & I_{12} \\ I_{21} & I_{22} \end{bmatrix} \begin{bmatrix} E_b^+ \\ E_b^- \end{bmatrix}. \quad (4.153)$$

Consider the special cases when one plane wave is incident on the ab interface. In terms of the complex amplitude E_a^+ of an incident plane wave in medium a , the complex amplitudes of the transmitted and reflected plane waves in media b and a , respectively (fig. 4.39-left), are given by

$$E_b^+ = t_{ab} E_a^+, \quad (4.154a)$$

$$E_a^- = r_{ab} E_a^+, \quad (4.154b)$$

and $E_b^- = 0$, where r_{ab} and t_{ab} are the Fresnel reflection and transmission coefficients of the ab interface. However, in accordance with eq. (4.153) we have

$$\begin{bmatrix} E_a^+ \\ E_a^- \end{bmatrix} = \begin{bmatrix} I_{11} & I_{12} \\ I_{21} & I_{22} \end{bmatrix} \begin{bmatrix} E_b^+ \\ 0 \end{bmatrix},$$

whose expansion gives

$$E_a^+ = I_{11} E_b^+, \quad (4.155a)$$

$$E_a^- = I_{21} E_b^+. \quad (4.155b)$$

Comparison between eqs. (4.154) and (4.155) leads to

$$I_{11} = 1/t_{ab}, \quad (4.156)$$

$$I_{21} = r_{ab}/t_{ab}. \quad (4.157)$$

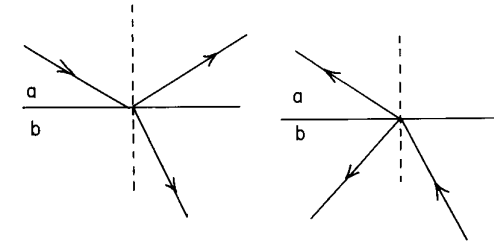


Fig. 4.39. Left: reflection and transmission (or refraction) of a plane wave at a two-media (ab) interface. Right: the direction of propagation of the transmitted (refracted) wave is assumed to have been reversed.

Now consider a plane wave incident on the ba interface from medium b , fig. 4.39–*right*, at an angle of incidence equal to the angle of refraction in medium b in the above case described in fig. 4.39–*left*. In this case, the fields immediately adjacent to the ba interface are related by

$$E_b^+ = r_{ba} E_b^-, \quad (4.158a)$$

$$E_a^- = t_{ba} E_b^-, \quad (4.158b)$$

and $E_a^+ = 0$, where r_{ba} and t_{ba} are the Fresnel reflection coefficients of the ba interface, respectively. On the other hand, eq. (4.153) tells us that, in this case,

$$\begin{bmatrix} 0 \\ E_a^- \end{bmatrix} = \begin{bmatrix} I_{11} & I_{12} \\ I_{21} & I_{22} \end{bmatrix} \begin{bmatrix} E_b^+ \\ E_b^- \end{bmatrix},$$

$$0 = I_{11} E_b^+ + I_{12} E_b^-, \quad (4.159a)$$

$$E_a^- = I_{21} E_b^+ + I_{22} E_b^-. \quad (4.159b)$$

Substitution of I_{11} and I_{21} from eqs. (4.156) and (4.157) into eqs. (4.159) transforms the latter equations into

$$E_b^+ = -I_{12} t_{ab} E_b^-, \quad (4.160a)$$

$$E_a^- = \left[\frac{r_{ab} r_{ba}}{t_{ab}} + I_{22} \right] E_b^-. \quad (4.160b)$$

Identification of the results shown by eqs. (4.158) and (4.160) leads to

$$I_{12} = -r_{ba}/t_{ab} \quad (4.161)$$

$$I_{22} = (t_{ab} t_{ba} - r_{ab} r_{ba})/t_{ab}. \quad (4.162)$$

Finally, use of the relationships between the interface Fresnel coefficients for both directions of propagation, namely $r_{ba} = -r_{ab}$ and $t_{ba} = (1 - r_{ab}^2)/t_{ab}$ [eqs. (4.29) and (4.30)], produces a final interface matrix of the form

$$\begin{aligned} \mathbf{I}_{ab} &= \begin{bmatrix} 1/t_{ab} & r_{ab}/t_{ab} \\ r_{ab}/t_{ab} & 1/t_{ab} \end{bmatrix} \\ &= (1/t_{ab}) \begin{bmatrix} 1 & r_{ab} \\ r_{ab} & 1 \end{bmatrix}. \end{aligned} \quad (4.163)$$

The interface Fresnel reflection and transmission coefficients that appear in eq. (4.163) must be evaluated using the complex indices of refraction of the two media that define the interface and the *local* angle of incidence. The latter can be found by the repeated application of Snell's law

$$N_0 \sin \phi_0 = N_1 \sin \phi_1 = \dots = N_j \sin \phi_j = \dots = N_{m+1} \sin \phi_{m+1}. \quad (4.164)$$

Now that we have determined the interface matrix \mathbf{I} , we turn our attention to the effect of propagation through a homogenous layer of index of refraction N and thickness d on the relationship between the fields inside that layer at both ends. This can be simply described by

$$\begin{bmatrix} E_0^+ \\ E_0^- \end{bmatrix} = \begin{bmatrix} e^{j\beta} & 0 \\ 0 & e^{-j\beta} \end{bmatrix} \begin{bmatrix} E_d^+ \\ E_d^- \end{bmatrix}, \quad (4.165)$$

where the subscripts 0 and d identify the beginning and end of the layer (along the direction of the forward-travelling wave) and the phase shift (layer phase thickness) β is given by

$$\beta = \frac{2\pi d N}{\lambda} \cos \phi, \quad (4.166)$$

with ϕ representing the angle between the direction of propagation in the layer and the perpendicular to its boundaries (the z -axis). The layer matrix \mathbf{L} can therefore be written as

$$\mathbf{L} = \begin{bmatrix} e^{j\beta} & 0 \\ 0 & e^{-j\beta} \end{bmatrix}. \quad (4.167)$$

From the interface and layer matrices \mathbf{I} and \mathbf{L} of eqs. (4.163) and (4.167) the overall scattering matrix \mathbf{S} of the stratified structure can be found by straightforward matrix multiplication as indicated by eq. (4.152). Equation (4.151) can be expanded as

$$\begin{bmatrix} E_a^+ \\ E_a^- \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix} \begin{bmatrix} E_s^+ \\ E_s^- \end{bmatrix}, \quad (4.168)$$

where, for simplicity, the subscripts a and s refer to the ambient and substrate media respectively, and $E_s^- = 0$. Further expansion of eq. (4.168) yields the overall reflection and transmission coefficients of the stratified structures as

$$R = \frac{E_a^-}{E_a^+} = \frac{S_{21}}{S_{11}}, \quad (4.169)$$

$$T = \frac{E_s^+}{E_a^+} = \frac{1}{S_{21}}, \quad (4.170)$$

respectively. From eq. (4.169) and (4.170) it is clear that only the elements of the first column of scattering matrix \mathbf{S} determine the overall reflection and transmission coefficients of the stratified structure.

For the purpose of ellipsometry, the stratified-structure scattering matrix \mathbf{S} (or its elements S_{11} and S_{21}) have to be calculated for both linear polarizations parallel (p) and perpendicular (s) to the plane of incidence. Such calculations

are readily programmable for execution on a digital computer. Let S_p and S_s represent the scattering matrices for the p and s polarizations. The p and s reflection and transmission coefficients become

$$R_p = \frac{S_{21p}}{S_{11p}}, \quad (4.171)$$

$$R_s = \frac{S_{21s}}{S_{11s}}, \quad (4.172)$$

$$T_p = \frac{1}{S_{21p}}, \quad (4.173)$$

$$T_s = \frac{1}{S_{21s}}. \quad (4.174)$$

The quantities that are measurable by the ellipsometer are the ratio of the complex reflection coefficients for the p and s polarizations

$$\rho_r = R_p/R_s = \frac{S_{21p}}{S_{11p}} \times \frac{S_{11s}}{S_{21s}}, \quad (4.175)$$

and the ratio of the complex transmission coefficients for the same polarizations

$$\rho_t = T_p/T_s = \frac{S_{21s}}{S_{21p}}. \quad (4.176)$$

Note that S_p and S_s are different because each of the interface matrices I that appear in eq. (4.152) is different for the p and s polarizations. The layer matrices L appearing in eq. (4.152) are the same for these two polarizations.

As an example of the application of the above procedure, consider the case of a single film (1) sandwiched between semi-infinite ambient (0) and substrate (2) media, fig. 4.14. From eq. (4.152) the scattering matrix S in this case is given by

$$S = I_{01}L_1I_{12}, \quad (4.177)$$

which upon substitution from eqs. (4.163) and (4.167) becomes

$$S = \left(\frac{1}{t_{01}t_{12}} \right) \begin{bmatrix} 1 & r_{01} \\ r_{01} & 1 \end{bmatrix} \begin{bmatrix} e^{i\beta} & 0 \\ 0 & e^{-i\beta} \end{bmatrix} \begin{bmatrix} 1 & r_{12} \\ r_{12} & 1 \end{bmatrix}, \quad (4.178)$$

or

$$S = \left(\frac{e^{i\beta}}{t_{01}t_{12}} \right) \begin{bmatrix} (1 + r_{01}r_{12}e^{-j2\beta}) & (r_{12} + r_{01}e^{-j2\beta}) \\ (r_{01} + r_{12}e^{-j2\beta}) & (r_{01}r_{12} + e^{-j2\beta}) \end{bmatrix}, \quad (4.179)$$

when matrix multiplication is carried out. From eq. (4.179) we have

$$S_{11} = \left(\frac{e^{i\beta}}{t_{01}t_{12}} \right) (1 + r_{01}r_{12}e^{-j2\beta}), \quad (4.180a)$$

$$S_{21} = \left(\frac{e^{i\beta}}{t_{01}t_{12}} \right) (r_{01} + r_{12}e^{-j2\beta}). \quad (4.180b)$$

Substitution of these values of S_{11} and S_{21} into eqs. (4.169) and (4.170) yields

$$R = \frac{r_{01} + r_{12}e^{-j2\beta}}{1 + r_{01}r_{12}e^{-j2\beta}},$$

$$T = \frac{t_{01}t_{12}e^{-i\beta}}{r_{01} + r_{12}e^{-j2\beta}},$$

which are the same expressions as previously found in §4.3 [eqs. (4.34) and (4.36)] by use of the method of adding multiply-reflected and transmitted partial waves.

For another example, consider the case of two films (1 and 2) between semi-infinite ambient (0) and substrate (3) media. The method of adding multiply-reflected and transmitted waves becomes quite awkward in this case, although such calculation has been made [180]. In contrast, by use of eq. (4.152) the scattering matrix for this system can be easily obtained as

$$S = I_{01}L_1I_{12}L_2I_{23}. \quad (4.181)$$

Upon substitution from eqs. (4.163) and (4.167) eq. (4.181) becomes

$$S = \left(\frac{1}{t_{01}t_{12}t_{23}} \right) \begin{bmatrix} 1 & r_{01} \\ r_{01} & 1 \end{bmatrix} \begin{bmatrix} e^{i\beta_1} & 0 \\ 0 & e^{-j\beta_1} \end{bmatrix} \begin{bmatrix} 1 & r_{12} \\ r_{12} & 1 \end{bmatrix} \begin{bmatrix} e^{i\beta_2} & 0 \\ 0 & e^{-j\beta_2} \end{bmatrix} \begin{bmatrix} 1 & r_{23} \\ r_{23} & 1 \end{bmatrix}, \quad (4.182)$$

or

$$S = \left(\frac{e^{j(\beta_1+\beta_2)}}{t_{01}t_{12}t_{23}} \right) \begin{bmatrix} [(1 + r_{01}r_{12}e^{-j2\beta_1}) + (r_{12} + r_{01}e^{-j2\beta_1})r_{23}e^{-j2\beta_2}] & [(1 + r_{01}r_{12}e^{-j2\beta_1})r_{23} + (r_{12} + r_{01}e^{-j2\beta_1})e^{-j2\beta_2}] \\ [(r_{01} + r_{12}e^{-j2\beta_1}) + (r_{01}r_{12} + e^{-j2\beta_1})r_{23}e^{-j2\beta_2}] & [(r_{01} + r_{12}e^{-j2\beta_1})r_{23} + (r_{01}r_{12} + e^{-j2\beta_1})e^{-j2\beta_2}] \end{bmatrix} \quad (4.183)$$

after matrix multiplications have been carried out explicitly. From eq. (4.183) we have

$$S_{11} = \left(\frac{e^{j(\beta_1+\beta_2)}}{t_{01}t_{12}t_{23}} \right) [(1 + r_{01}r_{12}e^{-j2\beta_1}) + (r_{12} + r_{01}e^{-j2\beta_1})r_{23}e^{-j2\beta_2}], \quad (4.184a)$$

$$S_{21} = \left(\frac{e^{j(\beta_1 + \beta_2)}}{t_{01}t_{12}t_{23}} \right) [(r_{01} + r_{12} e^{-j2\beta_1}) + (r_{01}r_{12} + e^{-j2\beta_1})r_{23} e^{-j2\beta_2}]. \quad (4.184b)$$

Substitution of eqs. (4.184a) and (4.184b) into eqs. (4.169) and (4.170) yields

$$R = \frac{(r_{01} + r_{12} e^{-j2\beta_1}) + (r_{01}r_{12} + e^{-j2\beta_1})r_{23} e^{-j2\beta_2}}{(1 + r_{01}r_{12} e^{-j2\beta_1}) + (r_{12} + r_{01} e^{-j2\beta_1})r_{23} e^{-j2\beta_2}}, \quad (4.185)$$

and

$$T = \frac{t_{01}t_{12}t_{23} e^{-j(\beta_1 + \beta_2)}}{(r_{01} + r_{12} e^{-j2\beta_1}) + (r_{01}r_{12} + e^{-j2\beta_1})r_{23} e^{-j2\beta_2}}. \quad (4.186)$$

Equations (4.185) and (4.186) apply to the p and s polarizations by simple attachment of a subscript p or s to R , T and the individual interface Fresnel reflection (r_{01} , r_{12} , r_{23}) and transmission (t_{01} , t_{12} , t_{23}) coefficients. The ratios of overall reflection and transmission coefficients that can be measured ellipsometrically are given by eqs. (4.175) and (4.176).

Although the 2×2 -matrix method can be easily extended to derive explicit expressions for R and T in cases that involve three or more films, such explicit expressions are of little practical value because all calculations can be handled by a computer without them.

Throughout this section, we assumed that the optical properties are uniform within each layer of the stratified structure, and that they change abruptly at the sharp interfaces between layers. When the optical properties change continuously (and not in a discontinuous step-like fashion) within an inhomogeneous layer, the method of this section can still be applied. This requires, however, that we divide the inhomogeneous layer into an adequately large number of sublayers, each of which is approximately homogeneous. In fact, this is the most general approach for a problem of this kind, because closed-form solutions are only possible for a few simple cases as, for example, when the refractive index of the layer changes linearly or exponentially along the direction of stratification [181].

4.7. Reflection and transmission by anisotropic stratified planar structures

A 4×4 -matrix method has recently been developed by Billard [182], Teitler and Henvis [183], and Berreman and Scheffer [184] to study the reflection and transmission of obliquely incident polarized light by stratified anisotropic planar structures. The method is a generalization of the Abelès 2×2 -matrix method [185] applicable to stratified isotropic media and previously discussed in §4.6. The following presentation of the 4×4 -matrix method is based on the general exposition given by Berreman [186]. Notice, however, that we use Maxwell's equations in rationalized units, that we

adhere to the $e^{j\omega t}$ time dependence, and that we use different notation in some places.

With the $e^{j\omega t}$ time dependence assumed, the two Maxwell's "curl" equations take the form

$$-\text{curl } \mathbf{E} = j\omega \mathbf{B}, \quad \text{curl } \mathbf{H} = j\omega \mathbf{D} \quad (4.187)$$

where \mathbf{E} , \mathbf{H} , \mathbf{D} , and \mathbf{B} are the electromagnetic-field vectors, as usual. In Cartesian coordinates, eqs. (4.187) can be combined in matrix form as

$$\begin{bmatrix} 0 & 0 & 0 & 0 & -\partial/\partial z & \partial/\partial y \\ 0 & 0 & 0 & \partial/\partial z & 0 & -\partial/\partial x \\ 0 & 0 & 0 & -\partial/\partial y & \partial/\partial x & 0 \\ 0 & \partial/\partial z & -\partial/\partial y & 0 & 0 & 0 \\ -\partial/\partial z & 0 & \partial/\partial x & 0 & 0 & 0 \\ \partial/\partial y & -\partial/\partial x & 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \\ H_x \\ H_y \\ H_z \end{bmatrix} = j\omega \begin{bmatrix} D_x \\ D_y \\ D_z \\ B_x \\ B_y \\ B_z \end{bmatrix}, \quad (4.188)$$

or shortly,

$$\mathbf{O}\mathbf{G} = j\omega \mathbf{C}. \quad (4.189)$$

In eq. (4.189), \mathbf{O} is a 6×6 symmetric matrix operator which can be partitioned into four 3×3 submatrices to take the form

$$\mathbf{O} = \begin{bmatrix} \mathbf{0} & \text{curl} \\ -\text{curl} & \mathbf{0} \end{bmatrix}. \quad (4.190)$$

$\mathbf{0}$ denotes the 3×3 zero matrix, and **curl** is the curl operator

$$\text{curl} = \begin{bmatrix} 0 & -\partial/\partial z & \partial/\partial y \\ \partial/\partial z & 0 & -\partial/\partial x \\ -\partial/\partial y & \partial/\partial x & 0 \end{bmatrix}. \quad (4.191)$$

\mathbf{G} is a 6×1 column vector whose elements are the Cartesian components of \mathbf{E} followed by those of \mathbf{H} , and \mathbf{C} is a 6×1 column vector whose elements are the Cartesian components of \mathbf{D} followed by those of \mathbf{B} .

In the absence of nonlinear optical effects and spatial dispersion, the constitutive relation between \mathbf{C} and \mathbf{G} can be generally put as

$$\mathbf{C} = \mathbf{M}\mathbf{G}, \quad (4.192)$$

where the 6×6 matrix \mathbf{M} carries all the information about the anisotropic optical properties of the medium that supports the electromagnetic fields. \mathbf{M} ,