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NONLINEAR OPTICAL PHENOMENA

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Determining the coordinate dependence of some components of the cubic susceptibility tensor $\hat{\chi}^{(3)}(z,\omega,-\omega,\omega,\omega)$ of a one-dimensionally inhomogeneous absorbing plate at an arbitrary frequency dispersion

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Abstract. **The possibility of unique reconstruction of the spatial profile of the cubic nonlinear susceptibility tensor component $\hat{\chi}^{(3)}_{yyyy}(z,\omega,-\omega,\omega,\omega)$ of a one-dimensionally inhomogeneous plate whose medium has a symmetry plane m_y perpendicular to its surface is proved for the first time and the unique reconstruction algorithm is proposed. The amplitude complex coefficients of reflection and transmission (measured in some range of angles of incidence) as well as of conversion of an *s*-polarised plane signal monochromatic wave into two waves propagating on both sides of the plate make it possible to reconstruct the profile. These two waves result from nonlinear interaction of a signal wave with an intense plane wave incident normally on the plate. All the waves under consideration have the same frequency ω, and so its variation helps study the frequency dispersion of the cubic nonlinear susceptibility tensor component $\hat{\chi}^{(3)}_{yyyy}(z,\omega,-\omega,\omega,\omega)$. For media with additional symmetry axes 2_z, 4_z, 6_z, or ∞_z that are perpendicular to the plate surface, the proposed method can be used to reconstruct the profile and to examine the frequency dispersion of about one third of all independent complex components of the tensor $\hat{\chi}^{(3)}$.**

Keywords: *dielectric constant, reflection coefficient, transmission coefficient, cubic susceptibility, inverse problem, one-dimensionally inhomogeneous medium.*

Identification and control of the dielectric properties of one-dimensionally inhomogeneous structures, including multilayer systems, are becoming a popular practical problem [1]. Different methods exist to solve this problem in linear media [1–8]. However, most these methods are either inapplicable in optics due to some reasons (neglect of absorption [2] or frequency dispersion in a broad frequency range [3], use of extremely simple models of this dispersion [4], etc.) or can be used only for weakly inhomogeneous media [1, 5]. For nonlinear media, the solution of these problems is at the initial stage [9–14].

In papers [6, 7] we proposed and tested a method for determining the coordinate dependences of the complex tensor components of the dielectric constant of a one-dimensionally inhomogeneous plate in linear media, the method being free from the above drawbacks. In this paper, we first generalise this method to nonlinear media. As a result, we pay main attention to the method of unique reconstruction of the coordinate dependence of some complex cubic susceptibility tensor components $\hat{\chi}^{(3)}(z,\omega,-\omega,\omega,\omega)$ of a nonlinear medium whose dielectric properties change along the z axis that is perpendicular to its two parallel flat surfaces, and arbitrary depend on the frequency.

Let the layer of this medium adjoin the homogeneous isotropic linear nonabsorbing media with a real dielectric constant $\varepsilon_0(\omega)$ along the planes $z = z_1$ and $z = z_2$ ($z_2 > z_1$). We assume that the point group symmetry of the medium producing the plate is such that one of its symmetry elements is the symmetry plane perpendicular to the plate surface. We will direct the axis $x \perp z$ along this plane and assume that an s-polarised plane signal low-intensity wave propagating in the direction positive or negative to the z axis is incident on this plate at a nonzero angle α. In the first case, the electric field strength of this wave is equal to $E_+\boldsymbol{e}_y\exp\{\mathrm{i}[\omega t - k_x x - k_z(z - z_1)]\}$ + c.c. (at $z < z_1$), while in the second case, $E_-\boldsymbol{e}_y\exp\{\mathrm{i}[\omega t - k_x x + k_z(z - z_2)]\}$ + c.c. (at $z > z_2$). Here, e_y is the unit vector perpendicular to the symmetry plane of the medium; ω is the wave frequency; $k_x = k_0\sin\alpha$; $k_z = k_0\cos\alpha$; $k_0 = \omega\sqrt{\varepsilon_0}/c$; c is the speed of light in vacuum. In addition, let an intense fundamental-radiation plane wave with the electric field strength equal to $E_0\boldsymbol{e}_y\exp\{\mathrm{i}[\omega t - k_0(z - z_1)]\}$ + c.c. at $z < z_1$ be incident on a plate perpendicular to its surface in the positive direction of the z axis. In other words, we simultaneously consider in this paper two independent problems. In the first problem, the intense and signal waves fall onto the same side of the plate under study (the subscript ‘plus’), while in the second – onto the opposite sides (the subscript ‘minus’).

We assume that as the phase-matching condition is violated, harmonics are not generated in the plane. As a result, only three waves [one intense ($E_{\rm f}(z)\boldsymbol{e}_y\exp(\mathrm{i}\omega t)$ + c.c.) wave and two weak waves: the initial signal ($E_{\rm s\pm}(z)\boldsymbol{e}_y\exp[\mathrm{i}(\omega t - k_x x)]$ + c.c.) wave and a new ($E_{\rm g\pm}(z)\boldsymbol{e}_y\exp[\mathrm{i}(\omega t + k_x x)]$ + c.c.) wave produced in the nonlinear medium] with the frequency ω can efficiently interact. Because the medium is inhomogeneous, each of these waves in the plate is a superposition of two counter-propagating variable-amplitude travelling waves. To describe their propagation, we can use both a system of six exact first-order equations for travelling-wave amplitudes and a system of three second-order wave equations for the total electric field of each wave. For the problem under study, using the second-order equations is more preferable, because it allows one to simplify the presentation.

The new wave $E_{\rm g\pm}(z)\boldsymbol{e}_y\exp[\mathrm{i}(\omega t + k_x x)]$ + c.c. appears during the nonlinear interaction of intense and signal waves

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and, in turn, can affect propagation of the latter. This nonlinear interaction takes place because the plate medium has a nonlinearity that is characterised by a symmetric (with respect to permutation of the last two subscripts) cubic susceptibility tensor $\hat{\chi}^{(3)}(z,\omega,-\omega,\omega,\omega)$. Because the medium has a plane of local symmetry m_y perpendicular to the y axis, in the case of the chosen polarisation of incident waves, the expression for the electric induction vector in the plate at the frequency ω can be written in the form [15]

$$\boldsymbol{D}_{\pm}(x,z,t) = \varepsilon_{yy}(z)\boldsymbol{E}_{0\pm} + 4\pi\chi^{(3)}_{yyyy}(z)(\boldsymbol{E}^*_{0\pm}\boldsymbol{E}_{0\pm})\boldsymbol{E}_{0\pm} + \text{c.c.} \quad (1)$$

Here, $\boldsymbol{E}_{0\pm}(x,z,t) = [E_{\rm f}(z) + E_{{\rm s}\pm}(z)\exp(-{\rm i}k_x x) + E_{{\rm g}\pm}(z)\exp({\rm i}k_x x)] \times \boldsymbol{e}_y\exp({\rm i}\omega t)$ is the spectral component of the electric field strength in a nonlinear medium at the frequency ω; $\varepsilon_{yy}(z)$ is the tensor component of the linear dielectric constant of the medium under consideration. In (1) and some subsequent expressions, the frequency arguments at the tensor components $\hat{\varepsilon}(z,\omega)$ and $\hat{\chi}^{(3)}(z,\omega,-\omega,\omega,\omega)$ are omitted for brevity.

In the case of a relatively weak signal wave, expression (1) can use only the terms that are linear in $E_{{\rm s}\pm}(z)$ and $E_{{\rm g}\pm}(z)$. Then, we have

$$\begin{aligned}\boldsymbol{D}_{\pm} = \{&\varepsilon_{yy}(z)[E_{\rm f} + E_{{\rm s}\pm}\exp(-{\rm i}k_x x) + E_{{\rm g}\pm}\exp({\rm i}k_x x)] \\ &+ 4\pi\chi^{(3)}_{yyyy}(z)[|E_{\rm f}|^2E_{\rm f} + (2|E_{\rm f}|^2E_{{\rm s}\pm} + E_{\rm f}^2E^*_{{\rm g}\pm}) \\ &\times\exp(-{\rm i}k_x x) + (2|E_{\rm f}|^2E_{{\rm g}\pm} + E_{\rm f}^2E^*_{{\rm s}\pm})\exp({\rm i}k_x x)]\} \\ &\times\boldsymbol{e}_y\exp({\rm i}\omega t) + \text{c.c.}\end{aligned} \quad (2)$$

It follows from (1) that in the used geometry, all the waves under study are transverse due to the presence of the m_y symmetry in the medium plane. Using (2), we can easily derive the equations for wave propagation:

$$\mathrm{d}^2E_{\rm f}/\mathrm{d}z^2 + 0.5\omega^2[\varepsilon_{yy}(z) + \varepsilon_{\rm n}(z)]E_{\rm f}/c^2 = 0, \quad (3)$$

$$\begin{aligned}&\mathrm{d}^2E_{{\rm s}\pm}/\mathrm{d}z^2 + [\omega^2\varepsilon_{\rm n}(z)/c^2 - \lambda]E_{{\rm s}\pm} + r(z)E^*_{{\rm g}\pm} = 0, \\ &\mathrm{d}^2E^*_{{\rm g}\pm}/\mathrm{d}z^2 + [\omega^2\varepsilon^*_{\rm n}(z)/c^2 - \lambda]E^*_{{\rm g}\pm} + r^*(z)E_{{\rm s}\pm} = 0,\end{aligned} \quad (4)$$

where $\lambda = k_x^2$; $\varepsilon_{\rm n}(z) = \varepsilon_{yy}(z) + 8\pi\chi^{(3)}_{yyyy}(z)|E_f(z)|^2$; $r(z) = 4\pi\omega^2\chi^{(3)}_{yyyy}(z)E_{\rm f}^2(z)/c^2$.

Because of Maxwell's boundary conditions on the layer surface, we have

$$\begin{aligned}&E_{\rm f}(z_1) = (1+R)E_0,\ \mathrm{d}E_{\rm f}/\mathrm{d}z|_{z=z_1} = -{\rm i}k_0(1-R)E_0, \\ &E_{\rm f}(z_2) = TE_0,\ \mathrm{d}E_{\rm f}/\mathrm{d}z|_{z=z_2} = -{\rm i}k_0TE_0,\end{aligned} \quad (5)$$

where R and T are the amplitude complex coefficients of intense light wave reflection by the plate and transmission through the plate, respectively. In addition,

$$\begin{aligned}&E_{{\rm s}+}(z_1) = (1+R_+)E_+,\ \mathrm{d}E_{{\rm s}+}/\mathrm{d}z|_{z=z_1} = -{\rm i}k_z(1-R_+)E_+, \\ &E_{{\rm s}+}(z_2) = T_+E_+,\ \mathrm{d}E_{{\rm s}+}/\mathrm{d}z|_{z=z_2} = -{\rm i}k_zT_+E_+, \\ &E_{{\rm s}-}(z_2) = (1+R_-)E_-,\ \mathrm{d}E_{{\rm s}-}/\mathrm{d}z|_{z=z_2} = {\rm i}k_z(1-R_-)E_-, \\ &E_{{\rm s}-}(z_1) = T_-E_-,\ \mathrm{d}E_{{\rm s}-}/\mathrm{d}z|_{z=z_1} = {\rm i}k_zT_-E_-.\end{aligned} \quad (6)$$

Here, $R_\pm$ are the amplitude complex reflectivities of signal waves by the plate, the waves propagating respectively in the positive and negative directions to the z axis; and $T_\pm$ are the amplitude complex transmittivities of the corresponding waves through the plate. Recall that the subscript 'plus' is used when an intense and signal waves are incident on one side of the plate under study, while the subscript 'minus' is used when these waves are incident on the opposite sides of the plate. In the experiment we can use any of these measurement schemes or, if necessary, both these schemes to obtain more accurate data.

The new wave $E_{{\rm g}\pm}(z)$ appearing in the plate during the nonlinear interaction propagates in homogeneous linear media adjacent to nonlinear medium in the form of a wave $E_{{\rm g}1\pm}\boldsymbol{e}_y\exp\{{\rm i}[\omega t + k_x x + k_z(z-z_1)]\}$ + c.c. at $z < z_1$ and in the form $E_{{\rm g}2\pm}\boldsymbol{e}_y\exp\{{\rm i}[\omega t + k_x x - k_z(z-z_2)]\}$ + c.c. at $z > z_2$. In this case, $E_{{\rm g}1\pm}$, $E_{{\rm g}2\pm}$ and $E_{{\rm g}\pm}(z)$ meet Maxwell's boundary conditions

$$\begin{aligned}&E_{{\rm g}\pm}(z_1) = E_{{\rm g}1\pm},\ \mathrm{d}E_{{\rm g}\pm}/dz|_{z=z_1} = {\rm i}k_zE_{{\rm g}1\pm}, \\ &E_{{\rm g}\pm}(z_2) = E_{{\rm g}2\pm},\ \mathrm{d}E_{{\rm g}\pm}/dz|_{z=z_2} = -{\rm i}k_zE_{{\rm g}2\pm}.\end{aligned} \quad (7)$$

It follows from equations (4), (6), and (7) that $E^*_{{\rm g}1\pm}$ and $E^*_{{\rm g}2\pm}$ are proportional to $E_\pm$ for these $\varepsilon_{\rm n}(z)$ and $r(z)$. It means that we can introduce $E_\pm$-independent signal-wave conversion coefficients $G^{(1)}_\pm = E^*_{{\rm g}1\pm}/E_\pm$ and $G^{(2)}_\pm = E^*_{{\rm g}2\pm}/E_\pm$ characterising the conversion efficiency of the signal wave incident on the plate into the waves of the same frequency, propagating away from the plate. In this case, the propagation direction of the latter ones differs from the propagation direction of reflected and transmitted signal waves. Using these coefficients, boundary conditions (7) can be conveniently rewritten in the form

$$\begin{aligned}&E^*_{{\rm g}\pm}(z_1) = G^{(1)}_\pm E_\pm,\ \mathrm{d}E^*_{{\rm g}\pm}/dz|_{z=z_1} = -{\rm i}k_zG^{(1)}_\pm E_\pm, \\ &E^*_{{\rm g}\pm}(z_2) = G^{(2)}_\pm E_\pm,\ \mathrm{d}E^*_{{\rm g}\pm}/dz|_{z=z_2} = {\rm i}k_zG^{(2)}_\pm E_\pm.\end{aligned} \quad (8)$$

If the dependences $\varepsilon_{\rm n}(z)$ and $r(z)$ are known, having solved equations (4), (6), and (8), we can unambiguously calculate $R_\pm$, $T_\pm$, $G^{(1)}_\pm$, $G^{(2)}_\pm$ for any angles of incidence of a plane signal wave.

In Appendix 1 we show for the first time that the converse is also true. If in some interval of angles of the plane signal wave incidence we have measured the amplitude complex transmission, reflection, and conversion coefficients in the presence of an intense fundamental-radiation wave [T_+, R_+, $G^{(1),(2)}_+$ or T_-, R_-, $G^{(1),(2)}_-$], the dependences $\varepsilon_{\rm n}(z)$ and $r(z)$ for a layer of the given thickness can be unambiguously found with the help of the obtained data. The linear dielectric constant profile $\varepsilon_{yy}(z)$ can be also uniquely reconstructed, as follows from papers [6, 7], by the amplitude reflectivities and transmittivities of the signal wave in the absence of an intense fundamental-radiation wave.

If the dependences $\varepsilon_{\rm n}(z)$, $r(z)$ and $\varepsilon_{yy}(z)$ are found, $\chi^{(3)}_{yyyy}(z,\omega,-\omega,\omega,\omega)$ can be uniquely reconstructed with the help of one of two equivalent expressions:

$$\begin{aligned}\chi^{(3)}_{yyyy}(z,\omega,-\omega,\omega,\omega) &= [\varepsilon_{\rm n}(z) - \varepsilon_{yy}(z)]/[8\pi|E_{\rm f}(z)|^2] \\ &= c^2r(z)/[4\pi\omega^2E_{\rm f}^2(z)],\end{aligned} \quad (9)$$

which follow from the definitions of the quantities $\varepsilon_{\rm n}(z)$ and $r(z)$ presented after the system of equations (4). The strength $E_{\rm f}(z)$ entering into (9) is unambiguously found from the linear differential equation (3) with the known coefficients and boundary conditions

$$\begin{aligned} &{\rm d}E_{\rm f}/{\rm d}z\,|_{\,z=z_1} - {\rm i}k_0E_{\rm f}(z_1) = -2{\rm i}k_0E_0, \\ &{\rm d}E_{\rm f}/{\rm d}z\,|_{\,z=z_2} + {\rm i}k_0E_{\rm f}(z_2) = 0, \end{aligned} \tag{10}$$

that follow directly from (5).

Thus, we have obtained the following result. Let the profile of the linear dielectric constant of the plate, $\varepsilon_{yy}(z,\omega)$, be known and, the amplitude complex reflection, transmission, and conversion coefficients [T_+, R_+, $G_+^{(1),(2)}$ or T_-, R_-, $G_-^{(1),(2)}$] of the signal wave in some interval of angles of the plane signal wave incidence be measured in the presence of the intense fundamental-radiation wave with the known amplitude E_0. These data will be sufficient to find unambiguously the cubic nonlinear susceptibility $\chi^{(3)}_{yyyy}(z,\omega,-\omega,\omega,\omega)$ of the plate under consideration.

In practice, $\chi^{(3)}_{yyyy}(z,\omega,-\omega,\omega,\omega)$ can be reconstructed, for example, with algorithm generalising the method used to find the coordinate dependence of the tensor components of the dielectric constant of a linear medium to the nonlinear medium, the method being proposed in [6] and realised in a numerical experiment [7]. This method is based on determination of the unique zero minimum of a specially constructed functional from the probe functions describing the coordinate dependence of the dielectric properties of the plate under study [6, 7]. This functional can be constructed differently but the main requirement to it is that it vanishes only for the probe profile of the linear dielectric constant to which there correspond signal-wave reflection and transmission coefficients known from the experiments and calculated on its basis in some specified interval of angles of incidence.

The basic principles of construction of the functional which we suggest using to uniquely determine $\chi^{(3)}_{yyyy}(z,\omega,-\omega,\omega,\omega)$ and for more symmetric media considered below and other tensor components of the cubic nonlinear susceptibility of the tested plate, do not undergo any noticeable changes as compared to those in [6, 7] and consist in the following. Having replaced $\varepsilon_{\rm n}(z)$ by an arbitrary probe profile $q(z)$ in (3) and solved the resultant equation with boundary conditions (10), we will find the electric field distribution of an intense wave, $E_{{\rm f}q}(z)$, corresponding to the selected probe profile. With the known amplitude reflection, transmission, and conversion coefficients of the signal wave, we will find, by virtue of Maxwell's boundary conditions (6) and (8), the electric field strengths and the first signal-wave derivative on both surfaces of the plate for two weak waves propagating inside it. We will use these values on one of the plate surfaces as boundary conditions and solve system (4) describing a change in the electric field of weak waves inside the plate, by replacing in it $\varepsilon_{\rm n}(z)$ by $q(z)$ and $r(z)$ by $q_1(z) = \omega^2[q(z) - \varepsilon_{yy}(z)]\,E^2_{{\rm f}q}(z)/[2c^2|\,E_{{\rm f}q}(z)\,|^2]$.

As a result, we will find the electric field strengths of weak waves and their first derivatives on the opposite surface of the plate. In the general case, they will naturally differ from those that are known to us from the measured signal-wave transmission, reflection, and conversion coefficients. The functional $G_{\rm n}[q(z)]$ is constructed in such a way that it is a measure of the difference between these calculated values of the electric field strength and its derivative from those we know from the measured signal-wave transmission, reflection, and conversion coefficients. In Appendix 2 we present a specific example of such a functional. Finding its unique zero minimum allows us to reconstruct unambiguously not only $\chi^{(3)}_{yyyy}(z,\omega,-\omega,\omega,\omega)$ but also the corresponding distribution of the electric field strength of an intense wave, $E_{\rm f0}(z)$, inside the plate.

So far we have assumed that the medium forming the layer has only the symmetry plane m_y perpendicular to its surface. We will consider now briefly the media additionally having the symmetry axis 2_z, 4_z, 6_z, or ∞_z. Without changing the polarisation of the intense fundamental-radiation wave, we will rotate the plane of incidence and the signal-wave polarisation vector by 90°. Then, the latter will be specified by one of two formulae: $E_+\boldsymbol{e}_x\exp\{{\rm i}[\omega t - k_y y - k_z(z - z_1)]\}$+c.c. when the intense and signal waves are incident on the same side of the studied plate in the positive direction to the z axis or $E_-\boldsymbol{e}_x\exp\{{\rm i}[\omega t - k_y y + k_z(z - z_2)]\}$ + c.c. when these waves are incident on opposite sides of the plate. Here, $k_y = k_0\sin\alpha$ and e_x is the unit vector directed along the x axis. In this case, the expressions for the nonzero components of the electric induction vector in the above approximations will have the form [15]:

$$\begin{aligned} D_{\pm x} = {}&\{\{[\varepsilon_{xx}(z) + 8\pi\chi^{(3)}_{xyxy}(z)\,|E_{\rm f}|^2]\,E_{{\rm s}\pm} + 4\pi\chi^{(3)}_{xxyy}(z)E^2_{\rm f}E^*_{{\rm g}\pm}\} \\ &\times\exp(-{\rm i}k_y y) + \{[\varepsilon_{xx}(z) + 8\pi\chi^{(3)}_{xyxy}(z)\,|E_{\rm f}|^2]\,E_{{\rm g}\pm} \\ &+ 4\pi\chi^{(3)}_{xxyy}(z)E^2_{\rm f}E^*_{{\rm s}\pm}\}\exp({\rm i}k_y y)\}\exp({\rm i}\omega t) + {\rm c.c.} \\ D_{\pm y} = {}&[\varepsilon_{yy}(z) + 4\pi\chi^{(3)}_{yyyy}(z)\,|E_{\rm f}|^2]\,E_{\rm f}\exp({\rm i}\omega t) + {\rm c.c.} \end{aligned}$$

Recall that the used tensor $\hat{\chi}^{(3)}$ is symmetric to permutation of the last two subscripts. With the fundamental-radiation wave as powerful as before, equation (3), boundary conditions (10), and the dependence $E_{\rm f0}(z)$ will not change and the system of equations for $E_{{\rm s}\pm}$ and $E^*_{{\rm g}\pm}$ will retain the form (4) with an accuracy of substitution of the parameter λ by $\bar{\lambda} = k_y^2$ and the functions $\varepsilon_{\rm n}(z)$ and $r(z)$ by

$$\begin{aligned} &\bar{\varepsilon}_{\rm n}(z) = \varepsilon_{xx}(z) + 8\pi\chi^{(3)}_{xyxy}(z)\,|E_{\rm f0}(z)|^2, \\ &\bar{r}(z) = 4\pi\omega^2\chi^{(3)}_{xxyy}(z)E^2_{\rm f0}(z)/c^2. \end{aligned} \tag{11}$$

Because the system of equations for $E_{{\rm s}\pm}$ and $E^*_{{\rm g}\pm}$ retains the form (4), all the results obtained in Appendices 1 and 2 can be applied to it. In particular, knowing the four new coefficients of signal-wave transmission, reflection, and conversion in some interval of angles of incidence, we can unambiguously reconstruct the profiles $\bar{\varepsilon}_{\rm n}(z)$ and $\bar{r}(z)$ in the medium under study. To this end, we should find the only zero minimum of the functional $G[q(z), q_1(z)]$ constructed similarly to the functional $G_{\rm n}[q(z)]$ described in Appendix 2. The only difference consists in the fact that now the functions $q(z)$ and $q_1(z)$, which are probe for the profiles $\bar{\varepsilon}_{\rm n}(z)$ and $\bar{r}(z)$, respectively, and enter into auxiliary equation (A2.1), are not related in any way. We already know the function $E_{\rm f0}(z)$ and the dependence $\varepsilon_{xx}(z,\omega)$ is either known ($\varepsilon_{xx} = \varepsilon_{yy}$ for all the studied media except those with the mm2 symmetry) or can be reconstructed [6, 7]. Thus, having found the functions $\bar{q}(z)$ and $\bar{q}_1(z)$ corresponding to the only zero minimum of the functional $G[q(z), q_1(z)]$ and, consequently, coinciding with the desired functions $\bar{\varepsilon}_{\rm n}(z)$ and $\bar{r}(z)$, we can calculate the spatial profiles of two new components of the cubic nonlinear tensor with the help of the expressions

$$\chi^{(3)}_{xyxy}(z) = [\bar{q}(z) - \varepsilon_{xx}(z)]/[8\pi\,|E_{f0}(z)|^2],$$

$$\chi^{(3)}_{xxyy}(z) = c^2\bar{q}_1(z)/[4\pi\omega^2 E^2_{f0}(z)],$$

which follow from (11).

For media with the symmetry axis 2_z, we can also obtain the dependences of the components $\chi^{(3)}_{xxxx}$, $\chi^{(3)}_{yxyx}$ and $\chi^{(3)}_{yyxx}$ of the tensor $\hat{\chi}^{(3)}(z,\omega,-\omega,\omega,\omega)$ on the coordinate z except the above-mentioned three components. To do this, we should fully repeat all the measurements described in this paper, by rotating the plate by 90° around the z axis, and then, perform all the calculations again. For media with the symmetry axis 4_z, 6_z, or ∞_z, these additional measurements and calculations are not necessary because in these media $\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy}$, $\chi^{(3)}_{yxyx} = \chi^{(3)}_{xyxy}$ and $\chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx}$ [15]. In addition, in media with the symmetry axis 6_z and ∞_z, the equality $\chi^{(3)}_{yyyy} = \chi^{(3)}_{xxyy} + 2\chi^{(3)}_{xyxy}$ is valid [15].

As is known, the surface layer properties of any medium can noticeably differ from the layer properties in the bulk. The surface layer, in particular, cannot have a three-dimensional inversion centre as well as symmetry plane parallel to the surface, i.e., cannot be three-dimensionally isotropic. However, the flat boundary between the inhomogeneous media is a special case of a one-dimensionally inhomogeneous system. As a result, the one-dimensionally inhomogeneous medium, strictly speaking, cannot be isotropic. It does not have a three-dimensional inversion centre and the symmetry plane perpendicular to the direction where the nonuniformity of the properties takes place. As a result, out of 32 classes and 7 limiting symmetry groups, that are possible for homogeneous media [15], in the case of one-dimensionally inhomogeneous media, only 10 classes (1, 2, m, mm2, 3, 4, 6, 3m, 4mm, 6mm) and 2 limiting groups (∞, ∞m) where the above-mentioned inversion center and symmetry plane perpendicular to the inhomogeneity direction are absent.

The symmetry of the given one-dimensionally inhomogeneous plate depends both on the medium the plate is made of and on orientation of its surfaces perpendicular to the direction of the inhomogeneity with respect to the crystallographic axes X_1, X_2, X_3 [15] of this medium. Unfortunately, our method does not allow one to determine and control the cubic nonlinearity of one-dimensionally inhomogeneous media with the class symmetry 1, 2, 3, 4, 6, and ∞. For the remaining 6 symmetry classes of one-dimensionally inhomogeneous media, Table 1 illustrates the specific features of the proposed method. In particular, in studying inhomogeneous mm2-symmetry plates, it is possible to reconstruct six of fifteen independent tensor components $\hat{\chi}^{(3)}(z,\omega,-\omega,\omega,\omega)$ and all the three tensor components of the dielectric constant $\hat{\varepsilon}(z,\omega)$ {the profile of the component $\varepsilon_{zz}(z,\omega)$ for classes mm2, 3, 3m, 4, 4mm, 6, ∞, 6mm, ∞m can be reconstructed by using the results of paper [8]}.

Note also that varying the frequency ω of the intense and signal waves, we can reconstruct the profiles of the cubic nonlinear susceptibility tensor components listed in Table 1 at different frequencies and study the frequency dispersion of the medium nonlinearity.

Table 1. Number of independent components of the $\hat{\chi}^{(3)}(z,\omega,-\omega,\omega,\omega)$ and $\hat{\varepsilon}(z,\omega)$ tensors of one-dimensional inhomogeneous media of different symmetry classes and their components, whose spatial profiles can be reconstructed using the suggested procedure.

Symmetry class	$\hat{\varepsilon}$ tensor		$\hat{\chi}^{(3)}$ tensor	
	Number of independent components	Reconstructed components	Number of independent components	Reconstructed components
m (m_y)	4	ε_{yy}	28	$\chi^{(3)}_{yyyy}$
mm2	3	$\varepsilon_{xx}, \varepsilon_{yy}, \varepsilon_{zz}$	15	$\chi^{(3)}_{yyyy}, \chi^{(3)}_{xyxy}, \chi^{(3)}_{xxyy}, \chi^{(3)}_{xxxx}, \chi^{(3)}_{yxyx}, \chi^{(3)}_{yyxx}$
3m			10	$\chi^{(3)}_{yyyy}$
4mm	2	$\varepsilon_{yy} = \varepsilon_{xx}, \varepsilon_{zz}$	8	$\chi^{(3)}_{yyyy}, \chi^{(3)}_{xyxy}, \chi^{(3)}_{xxyy}$
6mm, ∞m			7	$\chi^{(3)}_{yyyy}, \chi^{(3)}_{xxyy}$

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Appendix 1

The system of equations (4) and boundary conditions (6), (8) have the physical sense when the values of $k_z \in (0, k_0)$ are real (the value $k_z = k_0$ corresponds to the normal incidence of the signal wave for which the signal and intense waves are physically indiscernible). However, equations (4), (6), (8) and the quantities entering into them can be formally considered at any (including complex) values k_z and $\lambda \equiv k_x^2 = k_0^2 - k_z^2$, which is done in this Appendix.

Recall that for a broad class of the functions $\varepsilon_{\mathrm{n}}(z)$ and $r(z)$ (piecewise continuous and bounded or even only integrated [16] functions), system (4) has continuously differentiable solutions which we will write for brevity as a column:

$$\overleftrightarrow{\varphi}(z) = \begin{pmatrix}\varphi_{\mathrm{s}}(z)\\ \varphi_{\mathrm{g}}(z)\end{pmatrix} = \begin{pmatrix}E_{\mathrm{s}}(z)\\ E^*_{\mathrm{g}}(z)\end{pmatrix}.$$

Let the columns $\overleftrightarrow{\varphi}_m(z,\lambda)$ where $m = 1,2,3,4$, be the solutions of system (4) with the boundary conditions

$$\begin{aligned}
&\overleftrightarrow{\varphi}_1(z_1,\lambda) = \begin{pmatrix}1\\0\end{pmatrix},\ \mathrm{d}\overleftrightarrow{\varphi}_1(z,\lambda)/\mathrm{d}z\,|_{z=z_1} = \begin{pmatrix}0\\0\end{pmatrix},\\
&\overleftrightarrow{\varphi}_2(z_1,\lambda) = \begin{pmatrix}0\\1\end{pmatrix},\ \mathrm{d}\overleftrightarrow{\varphi}_2(z,\lambda)/\mathrm{d}z\,|_{z=z_1} = \begin{pmatrix}0\\0\end{pmatrix},\\
&\overleftrightarrow{\varphi}_3(z_1,\lambda) = \begin{pmatrix}0\\0\end{pmatrix},\ \mathrm{d}\overleftrightarrow{\varphi}_3(z,\lambda)/\mathrm{d}z\,|_{z=z_1} = \begin{pmatrix}1\\0\end{pmatrix},\\
&\overleftrightarrow{\varphi}_4(z_1,\lambda) = \begin{pmatrix}0\\0\end{pmatrix},\ \mathrm{d}\overleftrightarrow{\varphi}_4(z,\lambda)/\mathrm{d}z\,|_{z=z_1} = \begin{pmatrix}0\\1\end{pmatrix}.
\end{aligned} \tag{A1.1}$$

Then, $\overleftrightarrow{\varphi}_m(z,\lambda)$ at any, including complex, value of λ produce a fundamental system of solutions of system (4), and the general solution of problem (4), (6), (8) can be written in the form

$$\begin{aligned}
\overleftrightarrow{\varphi}_\pm(z,\lambda) = [&C_{1\pm}\overleftrightarrow{\varphi}_1(z,\lambda) + C_{2\pm}\overleftrightarrow{\varphi}_2(z,\lambda)\\
&+ C_{3\pm}\overleftrightarrow{\varphi}_3(z,\lambda) + C_{4\pm}\overleftrightarrow{\varphi}_4(z,\lambda)]E_\pm.
\end{aligned} \tag{A1.2}$$

Note that due to the symmetry of system (4) and boundary conditions (A1.1), the elements of the columns $\overleftrightarrow{\varphi}_m(z,\lambda)$ at real values of λ are related by the expressions

$$\begin{aligned}
&\varphi_{\mathrm{s}2}(z,\lambda) = \varphi^*_{\mathrm{g}1}(z,\lambda), \quad \varphi_{\mathrm{g}2}(z,\lambda) = \varphi^*_{\mathrm{s}1}(z,\lambda),\\
&\varphi_{\mathrm{s}4}(z,\lambda) = \varphi^*_{\mathrm{g}3}(z,\lambda), \quad \varphi_{\mathrm{g}4}(z,\lambda) = \varphi^*_{\mathrm{s}3}(z,\lambda).
\end{aligned} \tag{A1.3}$$

Substituting the relations for the functions $\overleftrightarrow{\varphi}_{\pm}(z,\lambda)$ from (A1.2) into (6), (8) at real λ and taking into account boundary conditions (A1.1) and equalities (A1.3), we obtain the expressions for the coefficients $C_{m\pm}$:

$$C_{1+} = 1 + R_+,\ C_{1-} = T_-,\ C_{2\pm} = G_\pm^{(1)},$$

$$C_{3+} = -\mathrm{i}k_z(1 - R_+),\ C_{3-} = \mathrm{i}k_zT_-,\ C_{4\pm} = -\mathrm{i}k_zG_\pm^{(1)},$$

and eight linear equations relating the elements of the columns $\overleftrightarrow{\varphi}_m(z,\lambda)$ and their derivatives at point $z = z_2$ with the coefficients $R_\pm$, $T_\pm$, $G_\pm^{(1)}$ and $G_\pm^{(2)}$:

$$T_-f_1(k_z) + G_-^{(1)}f_2(k_z) = 1 + R_-,$$
$$G_-^{(1)*}f_1(k_z) + T_-^*f_2(k_z) = G_-^{(2)*}, \tag{A1.4}$$

$$T_-f_3(k_z) + G_-^{(1)}f_4(k_z) = \mathrm{i}k_z(1 - R_-),$$
$$G_-^{(1)*}f_3(k_z) + T_-^*f_4(k_z) = -\mathrm{i}k_zG_-^{(2)*}, \tag{A1.5}$$

$$R_+f_1(k_z) + G_+^{(1)}f_2(k_z) + f_5(k_z) = T_+,$$
$$G_+^{(1)*}f_1(k_z) + R_+^*f_2(k_z) + f_6(k_z) = G_+^{(2)*},$$
$$R_+f_3(k_z) + G_+^{(1)}f_4(k_z) + f_7(k_z) = -\mathrm{i}k_zT_+,$$
$$G_+^{(1)*}f_3(k_z) + R_+^*f_4(k_z) + f_8(k_z) = -\mathrm{i}k_zG_+^{(2)*}. \tag{A1.6}$$

In (A1.4)–(A1.6) we used the notations

$$f_{1,2}(k_z) \equiv \Psi_{\mathrm{s1,s2}}(\lambda) \pm \mathrm{i}k_z\Psi_{\mathrm{s3,s4}}(\lambda),$$
$$f_{3,4}(k_z) \equiv \Psi_{\mathrm{s1}z,\mathrm{s2}z}(\lambda) \pm \mathrm{i}k_z\Psi_{\mathrm{s3}z,\mathrm{s4}z}(\lambda),$$
$$f_{5,6}(k_z) \equiv \Psi_{\mathrm{s1,s2}}(\lambda) \mp \mathrm{i}k_z\Psi_{\mathrm{s3,s4}}(\lambda),$$
$$f_{7,8}(k_z) \equiv \Psi_{\mathrm{s1}z,\mathrm{s2}z}(\lambda) \mp \mathrm{i}k_z\Psi_{\mathrm{s3}z,\mathrm{s4}z}(\lambda), \tag{A1.7}$$

where $\Psi_{\mathrm{s}m}(\lambda) = \varphi_{\mathrm{s}m}(z_2,\lambda)$; $\Psi_{\mathrm{s}mz}(\lambda) = \mathrm{d}\varphi_{\mathrm{s}m}(z,\lambda)/\mathrm{d}z\,|_{z=z_2}$. Recall that m takes the values from one to four. Equations (A1.4)–(A1.6) and the Wronskian constancy of system (4) allow one to obtain, in particular, that $|T_+|^2 - |G_+^{(2)}|^2 = |T_-|^2 - |G_-^{(1)}|^2$.

In addition, it follows from (A1.4), (A1.5) that at $k_z \neq 0$ the inequality $|T_-| \neq |G_-^{(1)}|$ is valid. Thus, the functions $f_1(k_z)$ and $f_2(k_z)$ can be found from the equations (A1.4), at least for the parameters $k_z \in (0, k_0)$ most interesting from the physical point of view:

$$f_1(k_z) = \frac{T_-^*(1 + R_-) - G_-^{(1)}G_-^{(2)*}}{|T_-|^2 - |G_-^{(1)}|^2},$$
$$f_2(k_z) = \frac{T_-G_-^{(2)*} - (1 + R_-)G_-^{(1)*}}{|T_-|^2 - |G_-^{(1)}|^2}. \tag{A1.8}$$

Besides, at each fixed $z \in [z_1, z_2]$, the quantities $\varphi_{\mathrm{s}m}(z,\lambda)$ and $\varphi_{\mathrm{g}m}(z,\lambda)$ are known to be single-valued analytic functions λ without singularities in the final part of the plane, i.e., integer functions [16,17]. Thus, $\Psi_{\mathrm{s}m}(\lambda)$ and $\Psi_{\mathrm{g}m}(\lambda) = \varphi_{\mathrm{g}m}(z_2,\lambda)$ are also integer functions of λ and, hence, $k_z^2 = k_0^2 - \lambda$. The latter equality means that $\Psi_{\mathrm{s}m}$ and $\Psi_{\mathrm{g}m}$ are the even integer functions of k_z, while f_1 and f_2, by definition of (A1.7), are the integer functions of k_z. Using the parity of the functions $\Psi_{\mathrm{s}m}$ with respect to k_z, we can obtain from (A1.7) the relations:

$$\Psi_{\mathrm{s1,s2}}(\lambda) = [f_{1,2}(k_z) + f_{1,2}(-k_z)]/2,$$
$$\Psi_{\mathrm{s3,s4}}(\lambda) = \pm[f_{1,2}(k_z) - f_{1,2}(-k_z)]/(2\mathrm{i}k_z). \tag{A1.9}$$

Applying the results of [18] to system (4), we immediately obtain that it is sufficient to know $\Psi_{\mathrm{s}m}(\lambda)$ and $\Psi_{\mathrm{g}m}(\lambda)$ on the entire complex plane λ to determine uniquely $\varepsilon_{\mathrm{n}}(z)$ and $r(z)$. Let the coefficients T_-, R_- and $G_-^{(1),(2)}$ be known at some interval of angles of incidence $0 < \alpha_1 \leqslant \alpha \leqslant \alpha_2 < \pi/2$. Then, using (A1.8), for real values of $k_z \in [k_0\cos\alpha_2, k_0\cos\alpha_1] \subset (0, k_0)$ we can find $f_1(k_z)$ and $f_2(k_z)$ that are the integer functions which is sufficient for their unambiguous analytic continuation to the entire complex plane k_z [17]. Knowing $f_1(k_z)$ and $f_2(k_z)$, we can find $\Psi_{\mathrm{s}m}(\lambda)$ with the help of (A1.9). Then, using (A1.3) to find $\Psi_{\mathrm{g}m}(\lambda)$ at real λ and holding an unambiguous analytic continuation, we can also find $\Psi_{\mathrm{g}m}(\lambda)$ for any complex values of λ and thus determine the dependences $\varepsilon_{\mathrm{n}}(z)$ and $r(z)$. A similar result can be obtained by using the known coefficients T_+, R_+ and $G_+^{(1),(2)}$.

Appendix 2

Let the signal-wave transmission, reflection, and conversion coefficients $T_+(k_x)$, $R_+(k_x)$, and $G_+^{(1),(2)}(k_x)$ and (or) $T_-(k_x)$, $R_-(k_x)$, and $G_-^{(1),(2)}(k_x)$ be known for some interval K of values k_z for a layer with the known profile of the linear dielectric constant $\varepsilon_{yy}(z)$ whose boundaries have the coordinates $z = z_1$ and $z = z_2$ in the presence of an intense fundamental-radiation wave with the known amplitude. In other words, it is known that there exist the functions $\chi_{yyyy}^{(3)}(z)$, $\varepsilon_{\mathrm{n}}(z)$, and $r(z)$ for which the problem (3), (4), (6), (8), (10) at these $T_+(k_x)$, $R_+(k_x)$, $G_+^{(1),(2)}(k_x)$ and (or) $T_-(k_x)$, $R_-(k_x)$, $G_-^{(1),(2)}(k_x)$ as well as at $\varepsilon_{yy}(z)$ and E_0 has a nontrivial solution at any $k_x \in K$ and $E_+ \neq 0$ and (or) $E_- \neq 0$. To reconstruct $\varepsilon_{\mathrm{n}}(z)$, $E_{\mathrm{f}}(z)$ and consequently $\chi_{yyyy}^{(3)}(z)$ by virtue of (9), we will find the solution $E_{\mathrm{f}q}(z)$ of problem (3), (10) with the probe function $q(z)$ instead of the function $\varepsilon_{\mathrm{n}}(z)$ and four solutions $E_{\mathrm{s}m}$, $E_{\mathrm{g}m}^*$ ($m = 1$–4) of the auxiliary system of equations, coinciding with (4) at $q(z) = \varepsilon_{\mathrm{n}}(z)$:

$$\mathrm{d}^2E_{\mathrm{s}}/\mathrm{d}z^2 + [\omega^2q(z)/c^2 - \lambda]E_{\mathrm{s}} + q_1(z)E_{\mathrm{g}}^* = 0,$$
$$\mathrm{d}^2E_{\mathrm{g}}^*/\mathrm{d}z^2 + [\omega^2q^*(z)/c^2 - \lambda]E_{\mathrm{g}}^* + q_1^*(z)E_{\mathrm{s}} = 0, \tag{A2.1}$$

where $q_1(z) = \omega^2[q(z) - \varepsilon_{yy}(z)]E_{\mathrm{f}q}^2(z)/\left[2c^2\left|E_{\mathrm{f}q}(z)\right|^2\right]$. The four solutions (A2.1) we are interested in meet the boundary conditions:

$$E_{\mathrm{s1}}(z_1) = (1 + R_+),\ \mathrm{d}E_{\mathrm{s1}}/\mathrm{d}z\,|_{z=z_1} = -\mathrm{i}k_z(1 - R_+),$$
$$E_{\mathrm{s2}}(z_2) = T_+,\ \mathrm{d}E_{\mathrm{s2}}/\mathrm{d}z\,|_{z=z_2} = -\mathrm{i}k_zT_+,$$
$$E_{\mathrm{s3}}(z_1) = T_-,\ \mathrm{d}E_{\mathrm{s3}}/\mathrm{d}z\,|_{z=z_1} = \mathrm{i}k_zT_-,$$
$$E_{\mathrm{s4}}(z_2) = (1 + R_-),\ \mathrm{d}E_{\mathrm{s4}}/dz\,|_{z=z_2} = \mathrm{i}k_z(1 - R_-),$$
$$E_{\mathrm{g1}}^*(z_1) = G_+^{(1)},\ \mathrm{d}E_{\mathrm{g1}}^*/\mathrm{d}z\,|_{z=z_1} = -\mathrm{i}k_zG_+^{(1)},$$
$$E_{\mathrm{g2}}^*(z_2) = G_+^{(2)},\ \mathrm{d}E_{\mathrm{g2}}^*/\mathrm{d}z\,|_{z=z_2} = \mathrm{i}k_zG_+^{(2)}, \tag{A2.2}$$

$$E^*_{g3}(z_1) = G^{(1)}_-, \ \ dE^*_{g3}/dz\,|_{z=z_1} = -ik_z G^{(1)}_-,$$

$$E^*_{g4}(z_2) = G^{(2)}_-, \ \ dE^*_{g4}/dz\,|_{z=z_2} = ik_z G^{(2)}_-.$$

Consider now a nonnegative functional from the probe profile $q(z)$

$$G_n[q(z)] = \int_K dk_x \sum_{m=1}^{4} \Big[\mu_{sm}\big|E_{sm}(\bar{d}_m) - a_{sm}\big|^2 + \mu_{gm}\big|E^*_{gm}(\bar{d}_m) - a_{gm}\big|^2 + \beta_{sm}\big|dE_{sm}/dz\,|_{z=\bar{d}_m} - b_{sm}\big|^2 + \beta_{gm}\big|dE^*_{gm}/dz\,|_{z=\bar{d}_m} - b_{gm}\big|^2\Big] \quad \text{(A2.3)}$$

constructed in accordance with the principles described in the main part of this paper. Here, $\bar{d}_{1,3} = z_2$; $\bar{d}_{2,4} = z_1$; $a_{s1,s4} = T_\pm$; $a_{s2,s3} = 1 + R_\pm$; $b_{s1,s4} = \mp ik_z T_\pm$; $b_{s2,s3} = \mp ik_z(1 - R_\pm)$; $a_{g1,g2} = G^{(2),(1)}_+$; $a_{g3,g4} = G^{(2),(1)}_-$; $b_{g1,g2} = \pm ik_z G^{(2),(1)}_+$; $b_{g3,g4} = \pm ik_z G^{(2),(1)}_-$. In addition, the weight functions $\mu_{sm}, \mu_{gm}, \beta_{sm}$ and β_{gm} in (A2.3) are any fixed nonnegative numbers, which are simultaneously nonzero. In this case, if we know only $T_+(k_x)$, $R_+(k_x)$ and $G^{(1),(2)}_+(k_x)$, we have $\mu_{s1,s2} \neq 0, \mu_{g1,g2} \neq 0, \beta_{s1,s2} \neq 0$ и $\beta_{g1,g2} \neq 0$, while the other weight coefficients are equal to zero. If we know only $T_-(k_x)$, $R_-(k_x)$ and $G^{(1),(2)}_-(k_x)$, we have $\mu_{s3,s4} \neq 0$, $\mu_{g3,g4} \neq 0$, $\beta_{s3,s4} \neq 0$ and $\beta_{g3,g4} \neq 0$, while the other weight functions are equal to zero.

The functional $G_n[q(z)]$, on the one hand, is a measure of difference of the values of the electric field strength and its derivatives calculated with formulae (A2.1), (A2.2) and corresponding to the probe profile $q(z)$ on one of the plate surfaces from those we know from the measured transmission, reflection, and conversion coefficients of a signal wave with a unit amplitude. On the other hand, $G_n[q(z)]$ is a measure of difference of the transmission, reflection, and conversion coefficients $T_{q\pm}(k_x)$, $R_{q\pm}(k_x)$ and $G^{(1),(2)}_{q\pm}(k_x)$ for the layer with the profile $q(z)$ from the coefficients known from the measurements. Indeed, comparison of expressions (6), (8) at $E_\pm = 1$ with (A2.2), (A2.3) shows that $G_n[q(z)] = 0$ only when the coefficients $T_{q+}(k_x)$, $R_{q+}(k_x)$, and $G^{(1),(2)}_{q+}(k_x)$ and (or) $T_{q-}(k_x)$, $R_{q-}(k_x)$, and $G^{(1),(2)}_{q-}(k_x)$ coincide with the coefficients $T_+(k_x)$, $R_+(k_x)$, $G^{(1),(2)}_+(k_x)$ and (or) $T_-(k_x)$, $R_-(k_x)$, $G^{(1),(2)}_-(k_x)$ in the interval K. We showed however in Appendix 1 that this coincidence is possible only in one case. Therefore, reconstruction of $\chi^{(3)}_{yyyy}(z)$ is reduced to the search for the function $q_0(z)$ corresponding to the only zero minimum of functional (A2.3) and to the subsequent use of expression

$$\chi^{(3)}_{yyyy}(z) = [q_0(z) - \varepsilon_{yy}(z)]/[8\pi|E_{f0}(z)|^2]$$

from (9). Here, $E_{f0}(z)$ is the solution of problem (3), (10) with the function $q_0(z)$ instead of the function $\varepsilon_n(z)$.

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