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Spectroscopy of one-dimensionally inhomogeneous media with quadratic nonlinearity

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Abstract. **We present a brief review of the results of fifty years of development efforts in spectroscopy of one-dimensionally inhomogeneous media with quadratic nonlinearity. The recent original results obtained by the authors show the fundamental possibility of determining, from experimental data, the coordinate dependences of complex quadratic susceptibility tensor components of a one-dimensionally inhomogeneous (along the z axis) medium with an arbitrary frequency dispersion, if the linear dielectric properties of the medium also vary along the z axis and are described by a diagonal tensor of the linear dielectric constant. It is assumed that the medium in question has the form of a plane-parallel plate, whose surfaces are perpendicular to the direction of the inhomogeneity. Using the example of several components of the tensors $\hat{\chi}^{(2)}(z, \omega_1 \pm \omega_2; \omega_1, \pm \omega_2)$, we describe two methods for finding their spatial profiles, which differ in the interaction geometry of plane monochromatic fundamental waves with frequencies ω_1 and ω_2. The both methods are based on assessing the intensity of the waves propagating from the plate at the sum or difference frequency and require measurements over a range of angles of incidence of the fundamental waves. Such measurements include two series of additional estimates of the intensities of the waves generated under special conditions by using the test and additional reference plates, which eliminates the need for complicated phase measurements of the complex amplitudes of the waves at the sum (difference) frequency.**

Keywords: *one-dimensionally inhomogeneous medium, quadratic susceptibility, inverse problem, second harmonic generation, sum-frequency generation, difference-frequency generation.*

1. Introduction

Nonlinear optical phenomena in media with quadratic susceptibility have been the subject of study since creation of the first lasers [1–7]. In particular, Maker et al. [5] investigated experimentally the dependence of the intensity $I_{2\omega}$ of the second harmonic generated in an optically thick ($L = 0.782$ mm $\approx 1000\lambda$) plane-parallel quartz plate on the angle α of incidence of a plane fundamental wave. It was found that the measured dependence is well described by a simple formula:

$$I_{2\omega} = 2\pi c P^2 (k_{2\omega}/\Delta k)^2 \sin^2(\Delta k L), \qquad (1)$$

where $\Delta k = |\boldsymbol{k}_{2\omega} - 2\boldsymbol{k}_\omega|$; $\boldsymbol{k}_{2\omega}$ and $\boldsymbol{k}_\omega$ are the wave vectors of the second harmonic and fundamental radiation in the plate, respectively; P is the modulus of the amplitude of the nonlinear polarisation at the doubled frequency, induced by the wave with frequency ω; c is the speed of light in vacuum.

Intensity oscillations of the second harmonic generated in a plane-parallel plate upon changing the angle of incidence of the fundamental radiation wave [5] formed the basis of one of the most versatile methods for measuring the quadratic susceptibility tensor components, i.e., the Maker-fringe analysis [8–10]. Soon, however, it was found that the use of the approximate formula (1), which does not take into account, in particular, multiple reflections of the fundamental waves and waves generated from the surfaces of the plane-parallel plate, leads to a significant scatter in the results of measurements obtained by different authors [11]. It turned out that the approximation is poorly applied, in particular, to thin plates, as well as in the case of media with a high dielectric constant. Papers [11–13] were devoted to improvement of the formulas describing the SHG process in a homogeneous plane-parallel plate, the process being first considered theoretically in detail in [7]. This process of improvement continues to this day [14], which allows one to achieve accurate reconstruction of the quadratic optical susceptibility of different homogeneous crystals, comparable with the accuracy provided by other measurement methods [15–20].

The methods for finding the coordinate dependences of the quadratic nonlinearity tensor components in one-dimensionally inhomogeneous media whose properties vary only in one direction are far less developed. And this despite the fact that interest (which emerged at the beginning of the era of nonlinear optics [21–23]) in structures with periodic modulation of the quadratic dielectric susceptibility only continues to increase [24–30]. The interest is associated with the possibility of implementing so-called quasi-phase matching conditions in media with periodic modulation of the quadratic susceptibility, which allows highly efficient conversion of the optical radiation frequency. A field of application for such media expanded dramatically after development (at the end of the last century) of effective methods for forming sufficiently sophisticated periodic domain structures in some ferroelectrics. At the same time, researchers showed much interest in quasi-periodic structures [31], suitable for conversion of radiation with a broad spectrum, as well as in the possibility of using strongly inhomogeneous, localised in a small region

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of thickness of about 5 μm, quadratic nonlinearity, which arises under certain conditions in silica glass [32]. This control of the domain structure quality and study of the mechanisms of nonlinearity formation in different types of glasses gave, apparently, an impetus to intensive development of both destructive [32–34] and nondestructive [35–39] methods for finding the spatial profile of the quadratic nonlinearity in one-dimensionally inhomogeneous media. The task was greatly simplified by the fact that, as a rule, linear optical properties of the systems under study were assumed homogeneous, and the media producing them – nonabsorbing. However, recent research shows that this approximation is not always fully justified [40].

Destructive methods usually include measurement of the SHG intensity as a function of sample thickness [32, 41, 42], as well as the study of the sample cut in the plane along the axis of inhomogeneity using different techniques [33, 34, 43–45]. The resolution of these methods achieves one micron.

However, of greatest practical interest are, of course, nondestructive methods for finding the coordinate dependence of the quadratic nonlinearity. One of the most developed among them is the Maker-fringe analysis, generalised to the case of a plane-parallel plate formed by a one-dimensionally inhomogeneous nonlinear medium with homogeneous linear properties [35, 37, 46–54]. The direction in which the medium is inhomogeneous, is perpendicular to the plate surface. Apparently, formula (1) was first generalised to this case in [35]:

$$I_{2\omega} = AI^2\left|\int_0^d \chi^{(2)}_{\mathrm{eff}}(z)\exp(\mathrm{i}\Delta k z)\,\mathrm{d}z\right|^2. \tag{2}$$

Here, d is the thickness of the plate or of the nonlinearity region inside it; $\chi^{(2)}_{\mathrm{eff}}$ is expressed in terms of the components of the quadratic susceptibility tensor $\hat{\chi}^{(2)}$ and the Fresnel coefficients, the form of this dependence being determined by the geometry of measurements ; I is the intensity of the fundamental wave; A is the normalisation factor. The integral in (2) has the form of the Fourier transform, which is known to be reversible. However, even in the case of a commonly used approximation of absence of linear and nonlinear absorption, where $\chi^{(2)}_{\mathrm{eff}}(z)$ is a real function, for its unambiguous determination it is needed to find from the experimental data not only the modulus, but also the complex-integral argument entering into (2) [52]. The latter is quite a challenge. Therefore, many researchers use the relations of the form (2) in combination with different *a priori* assumptions about the form of functions that define the shape of the spatial profiles of the components of the tensor $\hat{\chi}^{(2)}$, and then find the values of several fitting parameters of these functions that give the best agreement with the experiment [36, 46–50]. Naturally, this approach cannot guarantee the uniqueness of the reconstruction of the coordinate dependence of the quadratic susceptibility tensor components, and at best gives only an assessment of its basic parameters, and under certain circumstances can even lead to erroneous results. The authors of [49, 51–54] suggested several methods for solving this problem; the basic idea of the methods is to use an auxiliary plate or a mirror. However, all of them are only applicable to nonabsorbing media. In addition, papers [49, 51–54] as well as all other papers that use the formula of the form (2) do not take into account multiple reflections of waves from the interfaces of the main and auxiliary plates. As mentioned above, even in the case of homogeneous media, neglect of multiple reflections can significantly reduce the accuracy of finding the quadratic susceptibility tensor components [11].

In [55–59], formula (1) was generalised to the media consisting of homogeneous layers with different linear properties. The results obtained by solving the direct problem take into account all possible multiple reflections from surfaces in such a multilayer system. Nevertheless, the problem of finding the quadratic susceptibility tensor components describing each of the structure layers was studied much less than in the above case of a linear homogeneous medium [56, 58]. Another promising method for diagnosing the form of a coordinate dependence of the quadratic optical susceptibility is the analysis of $\omega - k$ spectra of parametric scattering of light [39, 60–66]. However, despite the construction of a sufficiently complete theory of this phenomenon [67], the development of a technique used to solve the inverse problem is still far from complete.

In this paper, using the example of the component $\chi^{(2)}_{yxy}$ and $\chi^{(2)}_{yyy}$ of the complex tensors $\hat{\chi}^{(2)}(z,\omega_1+\omega_2;\omega_1,\omega_2)$ and $\hat{\chi}^{(2)}(z,\omega_1-\omega_2;\omega_1,-\omega_2)$, we demonstrated for the first time the possibility of finding the coordinate dependence of the quadratic optical susceptibility tensor of the one-dimensionally inhomogeneous medium along the z axis without any *a priori* assumptions about the form of the functions $\hat{\chi}^{(2)}(z,\omega_1+\omega_2;\omega_1,\omega_2)$ and $\hat{\chi}^{(2)}(z,\omega_1-\omega_2;\omega_1,-\omega_2)$ if its linear dielectric properties also vary along the z axis and are described by the diagonal tensor of the linear dielectric constant $\hat{\varepsilon}(z,\omega)$, which depends on the frequency in an arbitrary manner.

In this paper we propose and justify the two methods for reconstructing the coordinate dependence of the quadratic susceptibility tensor. In the first method, use is made of the wave with frequency ω_1, normally incident on the plate, and of the wave with frequency ω_2, incident on the plate at a certain angle. In this case, the components of the tensor $\hat{\chi}^{(2)}(z,\omega_1+\omega_2;\omega_1,\omega_2)$ can be reconstructed by measuring the complex amplitude of the wave propagating from the plate at the sum frequency in a certain range of angles α of incidence of the plane wave with frequency ω_2. Similarly, we can reconstruct the profiles of the components of the quadratic susceptibility tensor $\hat{\chi}^{(2)}(z,\omega_1-\omega_2;\omega_1,-\omega_2)$, which describes the difference-frequency generation.

Unfortunately, in finding the spatial dependence of the components of $\hat{\chi}^{(2)}(z,\omega_1-\omega_2;\omega_1,-\omega_2)$, the first method is ineffective if $|\omega_1-\omega_2|\ll\omega_2$. The wave at the difference frequency in this case will propagate from the plate in the form of a homogeneous wave only at small angles α (if the condition $\omega_2\sin\alpha \leqslant |\omega_1-\omega_2|$ is fulfilled). Because of the small range of angles of incidence at which the amplitude of the reflected wave can be measured at the difference frequency, it is almost impossible to provide any reasonable accuracy of the quadratic susceptibility profile reconstruction. On the other hand, this frequency relation arises in many important practical applications, such as generation of terahertz waves by nonlinear optics methods.

In this case, the second suggested method is more effective for finding the coordinate dependence of the quadratic optical susceptibility. It involves the use of a biharmonic fundamental wave (formed by two collinearly propagating monochromatic waves with frequencies ω_1 and ω_2) which is incident at an angle α to the plane-parallel plate. In this scheme, the angle of reflection or transmission of the wave at the difference (sum) frequency through the plate is also equal to α. To implement the method, it is sufficient to measure the complex amplitude of the (sum-) difference-frequency wave propagating to one of the sides of the plate in some range of angles of incidence.

The both proposed method for reconstructing the coordinate dependence of the components of the tensors $\hat{\chi}^{(2)}(z, \omega_1 \pm \omega_2; \omega_1, \pm\omega_2)$, in contrast to all existing methods, are applicable to media with any, including piecewise-continuous, dependence of linear and nonlinear optical properties of the medium. They are based on the solution of inhomogeneous Fredholm equations of the first kind, take into account all multiple reflections of the waves, and consist of three series of measurements of the intensities of the waves generated under special conditions with the use of the test and additional reference plates. This helps to avoid complicated phase measurements of the complex amplitudes of the waves at the difference (sum) frequency.

In all the cases below, we assume that the amplitudes and frequencies of the incident waves, as well as the quantities $\chi^{(2)}_{jlm}(z, \omega_1 \pm \omega_2; \omega_1, \pm\omega_2)$, where $j, l, m = x, y, z$, are such that the waves are strongly generated at sum and (or) the difference frequencies in the medium, which is sufficient for reliable measurements. However, these waves and the waves at the doubled frequency are not involved in the generation of the waves with other frequencies and do not affect significantly the propagation of the initial waves and each other.

2. Reconstruction of spatial profiles of the components of the tensors $\hat{\chi}^{(2)}(z, \omega_1 \pm \omega_2; \omega_1, \pm\omega_2)$ with the help of noncollinear interaction of the waves

Consider a plane-parallel plate bounded by the planes $z = z_1$ and $z = z_2$ ($z_2 > z_1$) and placed in a homogeneous isotropic linear medium with real dielectric constant ε_0. The plate medium is nonmagnetic, one-dimensionally inhomogeneous along the z axis, and its linear dielectric properties, when the directions of the Cartesian axes x, y are properly chosen, are described by the diagonal tensor of the dielectric constant $\varepsilon(z, \omega)$.

Let a plane monochromatic wave with frequency ω_1, whose electric field strength vector is $E^{(1)}_{01}\boldsymbol{e}_x \exp[\mathrm{i}(\omega_1 t + k_1(z - z_2))]$ + c.c. at $z > z_2$, be incident perpendicularly to the surface of the plates in the negative direction of the z axis. We also assume that in addition to this wave, an s-polarised wave with frequency ω_2 is incident on the plate at an angle α. The electric field strength vector of the latter at $z > z_2$ is $E^{(1)}_{02}\boldsymbol{e}_y \exp[\mathrm{i}(\omega_2 t - k_{2x}x + k_{2z}(z - z_2))]$ + c.c. Here, $k_{1,2} = \omega_{1,2} \times \sqrt{\varepsilon_0}/c$; $\boldsymbol{e}_x$ and $\boldsymbol{e}_y$ are the unit vectors directed, respectively, along the axes x and y; $k_{2x} = k_2 \sin\alpha$; $k_{2z} = k_2 \cos\alpha$.

As a result, the wave $E^{(1)}_{01}E^{(1)}_1(z)\boldsymbol{e}_x \exp(\mathrm{i}\omega_1 t)$ + c.c with frequency ω_1 and the wave $E^{(1)}_{02}E_2(z)\boldsymbol{e}_y \exp[\mathrm{i}(\omega_2 t - k_{2x}x)]$ + c.c with frequency ω_2 will propagate in the plate. Changes in their dimensionless amplitudes $E^{(1)}_1(z)$ and $E_2(z)$ are described by the equations

$$\frac{\mathrm{d}^2 E^{(1)}_1}{\mathrm{d}z^2} + \frac{\omega_1^2}{c^2}\varepsilon_{xx}(z, \omega_1)E^{(1)}_1 = 0, \quad (3)$$

$$\frac{\mathrm{d}^2 E_2}{\mathrm{d}z^2} + \left[\frac{\omega_2^2}{c^2}\varepsilon_{yy}(z, \omega_2) - k^2_{2x}\right]E_2 = 0, \quad (4)$$

whose solutions at $z = z_{1,2}$ satisfy the boundary conditions

$$\left.\frac{\mathrm{d}E^{(1)}_1}{\mathrm{d}z}\right|_{z=z_1} - \mathrm{i}k_1 E^{(1)}_1(z_1) = 0,$$
$$\left.\frac{\mathrm{d}E^{(1)}_1}{\mathrm{d}z}\right|_{z=z_2} + \mathrm{i}k_1 E^{(1)}_1(z_2) = 2\mathrm{i}k_1, \quad (5)$$

$$\left.\frac{\mathrm{d}E_2}{\mathrm{d}z}\right|_{z=z_1} - \mathrm{i}k_{2z} E_2(z_1) = 0,$$
$$\left.\frac{\mathrm{d}E_2}{\mathrm{d}z}\right|_{z=z_2} + \mathrm{i}k_{2z} E_2(z_2) = 2\mathrm{i}k_{2z}, \quad (6)$$

which follow directly from Maxwell's boundary conditions.

Propagation of the waves $E^{(1)}_{01}E^{(1)}_1(z)\boldsymbol{e}_x \times \exp(\mathrm{i}\omega_1 t)$ + c.c and $E^{(1)}_{02}E_2(z)\boldsymbol{e}_y \exp[\mathrm{i}(\omega_2 t - k_{2x}x)]$ + c.c. in the plate leads, in particular, to the emergence of nonlinear polarisation of the medium at the sum frequency:

$$P_{1j}(z, \omega_s) = \chi^{(2)}_{jxy}(z, \omega_s; \omega_1, \omega_2)$$
$$\times I_1 E^{(1)}_1(z)E_2(z)\exp[\mathrm{i}(\omega_s t - k_{2x}x)] + \text{c.c.} \quad (7)$$

Here, $I_1 = 2E^{(1)}_{01}E^{(1)}_{02}$; $\omega_s = \omega_1 + \omega_2$; $j = x, y, z$. In writing (7), we take into account that $\chi^{(2)}_{jlm}(z, \omega_s; \omega_2, \omega_1) = \chi^{(2)}_{jml}(z, \omega_s; \omega_1, \omega_2)$ for all, including absorbing, media. As a result, s- and p-polarised waves with frequency ω_s are generated in the plate. In this case, the electric field strength vector of the s-polarised sum-frequency waves in the plate can be written as $I_1 E_s(z)\boldsymbol{e}_y \times \exp[\mathrm{i}(\omega_s t - k_{2x}x)]$ + c.c. The change in the dimensionless amplitude $E_s(z)$ is described by the equation

$$\frac{\mathrm{d}^2 E_s}{\mathrm{d}z^2} + \left[\frac{\omega_s^2}{c^2}\varepsilon_{yy}(z, \omega_s) - k^2_{2x}\right]E_s =$$
$$-\frac{4\pi\omega_s^2}{c^2}\chi^{(s)}_{yxy}(z)E^{(1)}_1(z)E_2(z), \quad (8)$$

where $\hat{\chi}^{(s)}(z) \equiv \hat{\chi}^{(2)}(z, \omega_s; \omega_1, \omega_2)$.

We consider the linear dielectric constant of the medium of the one-dimensionally inhomogeneous plate in question to be known. Recall that we examine media in which the tensors $\hat{\varepsilon}(z, \omega_1)$, $\hat{\varepsilon}(z, \omega_2)$ and $\hat{\varepsilon}(z, \omega_s)$ are diagonal. Their components can be found using the method proposed in [68–70] and experimentally implemented for homogeneous media in [71]. Therefore, the dependences $E^{(1)}_1(z)$ and $E_2(z)$, uniquely defined by (3)–(6) can be also regarded as known.

The s-polarised wave appearing in the plate at the sum frequency $I_1 E_s(z)\boldsymbol{e}_y \exp[\mathrm{i}(\omega_s t - k_{2x}x)]$ + c.c. continues to propagate in homogeneous linear media bordering the nonlinear medium: in the region $z < z_1$ (behind the plate) – in the form of a wave with electric field strength $S^{(t)}_s I_1 \boldsymbol{e}_y \times \exp\{\mathrm{i}[\omega_s t - k_{2x}x + k_{sz}(z - z_1)]\}$ + c.c. and in the region $z > z_2$ (in front of the plate) – in the form of a wave with $S^{(r)}_s I_1 \boldsymbol{e}_y \exp\{\mathrm{i}[\omega_s t - k_{2x}x - k_{sz}(z - z_2)]\}$ + c.c. [here, $k_{sz} = (\omega_s^2\varepsilon_0/c^2 - k^2_{2x})^{1/2}$].

Note that $k_{2x} = (\omega_2/c)\varepsilon_0^{1/2}\sin\alpha < (\omega_s/c)\varepsilon_0^{1/2}$ and, therefore, k_{sz} is a positive real value at all angles of incidence of the wave with frequency ω_2. In these formulas, the coefficients $S^{(t)}_s(\alpha)$ and $S^{(r)}_s(\alpha)$ characterise the conversion efficiency. The incident waves with frequencies ω_1, ω_2 and orthogonal linear polarisations are converted by the plate into the s-polarised sum-frequency waves propagating behind and in front of the plate. In what follows we will call $S^{(r)}_s$ and $S^{(t)}_s$ the coefficients of conversion into the sum-frequency waves upon reflection and transmission, respectively. On flat surfaces of the plate, the functions $E_{1s}(z)$ satisfy Maxwell's boundary conditions, which, with the above notations taken into account, can be written in the form:

$$E_s(z_1) = S_s^{(t)}, \quad dE_s/dz\,|_{z=z_1} = ik_{sz}S_s^{(t)},$$
$$E_s(z_2) = S_s^{(r)}, \quad dE_s/dz\,|_{z=z_2} = -ik_{sz}S_s^{(r)}. \tag{9}$$

Let $R_s(z,\alpha)$ be any continuously differentiable solution to the homogeneous equation (8):

$$\frac{d^2R_s}{dz^2} + \left[\frac{\omega_s^2}{c^2}\varepsilon_{yy}(z,\omega_s) - k_{2x}^2\right]R_s = 0. \tag{10}$$

Multiplying equations (8) and (10) by $R_s(z)$ and $E_s(z)$, respectively, and subtracting the second product from the first, we obtain

$$-\frac{4\pi\omega_s^2}{c^2}\chi_{yxy}^{(s)}(z)E_1^{(1)}(z)E_2(z,\alpha)R_s(z,\alpha)$$
$$= \frac{d^2E_s}{dz^2}R_s - \frac{d^2R_s}{dz^2}E_s. \tag{11}$$

Integrating equality (11) from z_1 to z_2 and using the method of integration by parts and boundary conditions (9) to calculate the integral in the right-hand side of (11), after some transformations we obtain for the function $\chi_{yxy}^{(s)}(z)$ the Fredholm equation of the first kind:

$$\int_{z_1}^{z_2}\chi_{yxy}^{(s)}(u)K_s(u,\alpha)du = [R_s'(z_1) - ik_{sz}R_s(z_1)]S_s^{(t)}(\alpha)$$
$$-[R_s'(z_2) + ik_{sz}R_s(z_2)]S_s^{(r)}(\alpha) \tag{12}$$

with a known normalised kernel $K_s(z,\alpha) = -4\pi\omega_s^2E_1^{(1)}(z)\times E_2(z,\alpha)R_s(z,\alpha)/c^2$. Note that the right-hand side of equation (12) becomes independent of $S_s^{(t)}$ if $R_s(z,\alpha)$ satisfies the boundary conditions

$$R_s(z_1) = 1, \quad (dR_s/dz)\big|_{z=z_1} = ik_{sz}, \tag{13}$$

and is independent of $S_s^{(r)}$, if

$$R_s(z_2) = 1, \quad (dR_s/dz)\big|_{z=z_2} = -ik_{sz}. \tag{14}$$

Suppose that for a given layer thickness in some range of angles of incidence of the wave with frequency ω_2, we know from the experiment the values of the conversion coefficients $S_s^{(r)}(\alpha)$ and (or) $S_s^{(t)}(\alpha)$. Then, with the appropriate choice of boundary conditions for the auxiliary function $R_s(z,\alpha)$, the right-hand side of equation (12) becomes known. Thus, using the standard methods of solution of the Fredholm equations of the first kind [72, 73], we can find the coordinate dependence of the component $\chi_{yxy}^{(s)}(z,\omega_s;\omega_1,\omega_2)$.

If the plate is rotated by 90° around the z axis without changing the incidence plane and polarisation of incident waves, then after measuring a new coefficient of conversion into the s-polarised wave at the sum frequency in some range of incidence angles and acting as in the previous of case, we can reconstruct the profile of the component $\chi_{xyx}^{(s)}(z)$.

Similarly, we can reconstruct the components $\chi_{xyx}^{(d)}$ and $\chi_{yxy}^{(d)}$ of the tensor $\hat{\chi}^{(d)}(z) \equiv \hat{\chi}^{(2)}(z,\omega_d;\omega_1,-\omega_2)$, $\omega_d = \omega_1 - \omega_2$, describing the difference-frequency generation in the plate. However, the difference-frequency waves with the wave vector projections $\pm\tilde{k}_{dz}$ on the z axis propagate in the medium surrounding the plate; here,

$$\tilde{k}_{dz} \equiv (\omega_d^2\varepsilon_0/c^2 - k_{2x}^2)^{1/2} = [\varepsilon_0(\omega_d^2 - \omega_2^2\sin^2\alpha)]^{1/2}/c.$$

Obviously, these waves will be homogeneous only if $\omega_2\sin\alpha \leqslant |\omega_d|$. This fact makes it almost impossible to use this technique to reconstruct the components of the tensor $\hat{\chi}^{(d)}(z)$ when $|\omega_1-\omega_2| \ll \omega_2$. In the practically important case, the collinear geometry of interaction of the waves, discussed below, proves more efficient.

3. Reconstruction of the components of the tensors $\hat{\chi}^{(2)}(z,\omega_1\pm\omega_2;\omega_1,\pm\omega_2)$ with the help of a biharmonic fundamental wave

Let the s-polarised plane fundamental wave with two monochromatic components, propagating in the negative direction of the z axis, be incident on the investigated plate at an angle α; the electric field strength vector at $z > z_2$ has the form

$$E_{01}^{(2)}\boldsymbol{e}_y\exp[i(\omega_1t - k_{1x}x + k_{1z}(z-z_2))]$$
$$+E_{02}^{(2)}\boldsymbol{e}_y\exp[i(\omega_2t - k_{2x}x + k_{2z}(z-z_2))] + \text{c.c.}$$

Here, $k_{nx} = k_n\sin\alpha$ and $k_{nz} = k_n\cos\alpha$ are the wave-vector projections $k_n = \omega_n\sqrt{\varepsilon_0}/c$ of the wave with frequency ω_n on the x and z axes; $n = 1, 2$.

The change in the dimensionless amplitude of each of the two waves propagating in the plate, $E_{0n}^{(2)}E_n(z)\boldsymbol{e}_y\times\exp[i(\omega_nt - k_{nx}x)]$ + c.c., with frequency ω_n is described by the equations and boundary conditions that are similar to relations (4) and (6):

$$\frac{d^2E_n}{dz^2} + \left[\frac{\omega_n^2}{c^2}\varepsilon_{yy}(z,\omega_n) - k_{nx}^2\right]E_n = 0, \tag{15}$$

$$\left.\frac{dE_n}{dz}\right|_{z=z_1} - ik_{nz}E_n(z_1) = 0,$$
$$\left.\frac{dE_n}{dz}\right|_{z=z_2} + ik_{nz}E_n(z_2) = 2ik_{nz}. \tag{16}$$

Propagation of the waves in the plate, $E_{0n}^{(2)}E_n(z)\boldsymbol{e}_y\times\exp[i(\omega_nt - k_{nx}x)]$ + c.c., leads, in particular, to the emergence of nonlinear polarisation of the medium at the difference frequency:

$$P_{2j}(z,\omega_d) = I_2\chi_{jyy}^{(d)}(z)E_1(z)[E_2(z)]^*$$
$$\times\exp[i(\omega_dt - k_{dx}x)] + \text{c.c.}, \tag{17}$$

where $j = x, y, z$; $I_2 = 2E_{01}^{(2)}(E_{02}^{(2)})^*$; $\omega_d = \omega_1 - \omega_2$; $k_{dx} = k_{1x} - k_{2x} = (\omega_d\varepsilon_0^{1/2}/c)\sin\alpha$; the asterisk denotes complex conjugation. In deriving (17), we have taken into account that

$\chi^{(2)}_{jlm}(z,\omega_d;\omega_1,-\omega_2)=\chi^{(2)}_{jml}(z,\omega_d;-\omega_2,\omega_1)$ for all, including the absorbing, media.

Because nonlinear polarisation (17) is present in the medium, s-and p-polarised waves are generated at frequency ω_d in the plate. The electric field strength vector of the s-polarised difference-frequency wave in the plate can be written as $I_2E_d(z)\boldsymbol{e}_y\exp[i(\omega_d t-k_{dx}x)]$ + c.c. Changes in its dimensionless amplitude are described by the equation

$$\frac{d^2E_d}{dz^2}+\left[\frac{\omega_d^2}{c^2}\varepsilon_{yy}(z,\omega_d)-k_{dx}^2\right]E_d$$

$$=-\frac{4\pi\omega_d^2}{c^2}\chi^{(d)}_{yyy}(z)E_1(z)[E_2(z)]^*. \quad (18)$$

We still consider the linear dielectric constant of the medium of the plate under study and, consequently, the dependences $E_1(z)$ and $E_2(z)$, uniquely defined by relations (15) and (16), to be known [68–70]. At points $z=z_{1,2}$ the dimensionless amplitude $E_d(z)$ satisfies the boundary conditions that are similar to (9):

$$E_d(z_1)=S_d^{(t)},\quad \left.\frac{dE_d}{dz}\right|_{z=z_1}=ik_{dz}S_d^{(t)},$$

$$E_d(z_2)=S_d^{(r)},\quad \left.\frac{dE_d}{dz}\right|_{z=z_2}=-ik_{dz}S_d^{(r)}, \quad (19)$$

where $k_{dz}=(\omega_d^2\varepsilon_0/c^2-k_{dx}^2)^{1/2}=(\omega_d\varepsilon_0^{1/2}/c)\cos\alpha$; $S_d^{(t)}(\alpha)$ and $S_d^{(r)}(\alpha)$ are the coefficients of conversion of the fundamental waves with two equally s-polarised monochromatic components into the difference-frequency waves with the same polarisation, which propagate from the plate in the negative and positive directions along the z axis, respectively. If the values of the coefficients $S_d^{(r)}(\alpha)$ or $S_d^{(t)}(\alpha)$ are known within a certain range of angles of incidence of the biharmonic wave, then we can reconstruct the spatial dependence of the component $\chi^{(d)}_{yyy}(z)$. The procedure of $\chi^{(d)}_{yyy}(z)$ reconstruction, as in the previous case, is reduced to solving the Fredholm integral equation of the first kind with normalised kernel and known right-hand side. The latter is obtained from equation (18) and boundary conditions (19) in the same way as equation (12) was derived, and has the form:

$$\int_{z_1}^{z_2}\chi^{(d)}_{yyy}(u)K_d(u,\alpha)du$$

$$=[R'_d(z_1)-ik_{dz}R_d(z_1)]S_d^{(t)}(\alpha)$$

$$-[R'_d(z_2)+ik_{dz}R_d(z_2)]S_d^{(r)}(\alpha), \quad (20)$$

where $K_d(z,\alpha)=-4\pi\omega_d^2E_1(z,\alpha)[E_2(z,\alpha)]^*R_d(z,\alpha)/c^2$; $R_d(z,\alpha)$ is any continuously differentiable solution to the homogeneous equation (18):

$$\frac{d^2R_d}{dz^2}+\left[\frac{\omega_d^2}{c^2}\varepsilon_{yy}(z,\omega_d)-k_{dx}^2\right]R_d=0. \quad (21)$$

As in the previous case, the right-hand side of equation (20) ceases to depend on the conversion coefficient $S_d^{(t)}(\alpha)$ of the propagating fundamental wave, if the auxiliary function $R_d(z,\alpha)$ satisfies the boundary conditions

$$R_d(z_1)=1,\quad \left.\frac{dR_d}{dz}\right|_{z=z_1}=ik_{dz}, \quad (22)$$

and does not depend on the conversion coefficient $S_d^{(r)}$upon reflection, if

$$R_d(z_2)=1,\quad \left.\frac{dR_d}{dz}\right|_{z=z_2}=-ik_{dz}. \quad (23)$$

If the plate is rotated by 90° around the z axis, without changing the incidence plane and polarisation of the incident biharmonic wave, then we can reconstruct the profile of the component $\chi^{(d)}_{xxx}(z)$. To do this, it is needed to measure a new coefficient of conversion into the s-polarised wave at the difference frequency in some range of incidence angles and to act as in the previous of case. Similarly, we can reconstruct the components $\chi^{(s)}_{yyy}(z)$ and $\chi^{(s)}_{xxx}(z)$ of the tensor $\hat{\chi}^{(2)}(z,\omega_s;\omega_1,\omega_2)$, responsible for generating the sum frequency.

Note that one-dimensionally inhomogeneous media can have a local symmetry, described by one of ten crystal classes (1, 2, m, mm2, 3, 4, 6, 3m, 4mm, 6mm) or one of two limiting symmetry groups (∞, ∞m) [74, 75]. Three classes of symmetry (1, 2, and m) are not considered in this paper, because the linear dielectric properties of the corresponding media are described in the general case by a nondiagonal second-rank tensor. The reduction of possible classes and limiting symmetry groups in comparison with homogeneous media is due to the fact that a one-dimensionally inhomogeneous system, strictly speaking, can only have a symmetry axis, whose direction coincides with the direction of inhomogeneity, and a symmetry plane containing the direction. Let the axes x, y and z coincide, respectively, with the axes X_1, X_2 and X_3 of the crystallophysical coordinate system [75] of the medium forming the plate. Then, in the media under study, the following relations between the components of the complex tensors $\hat{\chi}^{(s),(d)}(z)$ are fulfilled [75]:

$$\chi^{(s),(d)}_{yyx}(z)=\chi^{(s),(d)}_{xyy}(z)=-\chi^{(s),(d)}_{xxx}(z)=\chi^{(s),(d)}_{yxy}(z)=\sigma_1^{(s),(d)}(z),$$

$$\chi^{(s),(d)}_{xxy}(z)=\chi^{(s),(d)}_{yxx}(z)=-\chi^{(s),(d)}_{yyy}(z)=\chi^{(s),(d)}_{xyx}(z)=\sigma_2^{(s),(d)}(z). \quad (24)$$

In this case, the functions $\sigma_1^{(s),(d)}(z)$ are not identically equal to zero only in class 3, whereas $\sigma_2^{(s),(d)}(z)$ – only in class 3m (with the symmetry plane perpendicular to the x axis) and in class 3 [75].

The both proposed methods can be also used to reconstruct the profiles of other components of the complex quadratic susceptibility tensors $\hat{\chi}^{(s)}(z)$ and $\hat{\chi}^{(d)}(z)$ of one-dimensionally inhomogeneous media belonging to classes mm2, 3, 4, 6, 3m, 4mm and 6mm or limiting groups ∞ and ∞m. To this end, it is necessary to investigate not only generation of s- but also of p-polarised waves with frequencies ω_s and ω_d, using the fundamental waves with specially selected polarisations. In this case, the reconstruction problem is reduced to solving the Fredholm integral equations [that are similar to (12)] with the known right-hand side. The capabilities of the both methods used to reconstruct the profiles of various components of the tensors $\hat{\chi}^{(2)}(z,2\omega;\omega,\omega)$, $\hat{\chi}^{(2)}(z,\omega_s;\omega_1,\omega_2)$ and $\hat{\chi}^{(2)}(z,\omega_d;\omega_1,-\omega_2)$, including the conditions of uniqueness of reconstruction, have been investigated in detail in [76–78].

4. Replacement of phase measurements by additional measurements of the intensity

Until recently the complex conversion coefficients of the fundamental wave into the sum- or difference-frequency wave have been assumed experimentally known. However, their determination requires rather complicated phase measurements. Let us prove that the phase measurements can be avoided if three series of measurements of the modulus of the conversion coefficients are performed for each value of the angle. The first series of measurements is carried out only with the plate in question, the two other series – with the test and additional plates. Linear and nonlinear properties of the additional plate should be known, the symmetry class of its medium can be any other than classes 1, 2, and m, and the axes X_1, X_2 and X_3 of the crystallophysical coordinate system should be oriented parallel to the axes x, y and z, respectively.

Let us measure the intensity of the sum frequency wave in the case of reflection. We will use the boundary conditions (13) and (22) for the functions $R_s(z,\alpha)$ and $R_d(z,\alpha)$ entering into the integral equation (12) and (20), respectively. Then, equations (12) and (20) can be written in the general form:

$$\int_{z_1}^{z_2} q(u)K(u,\alpha)\,du = I_0(\alpha), \quad (25)$$

where $I_0(\alpha) = -[R'(z_2) + ik_z R(z_2)]S(\alpha)$. Equation (25) transforms into equation (12), if we put $q(z) = \chi^{(s)}_{yxy}(z)$, $K(z,\alpha) = K_s(z,\alpha)$, $R(z,\alpha) = R_s(z,\alpha)$, $k_z = k_{sz}$ and $S(\alpha) = S^{(r)}_s(\alpha)$. If $q(z) = \chi^{(d)}_{yyy}(z)$, $K(z,\alpha) = K_d(z,\alpha)$, $R(z,\alpha) = R_d(z,\alpha)$, $k_z = k_{dz}$ and $S(\alpha) = S^{(r)}_d(\alpha)$ equation (25) transforms into equation (20). Because the complex functions $K(z,\alpha)$ and $R(z,\alpha)$ are determined only by the linear properties of the investigated plate, they are assumed known. Therefore, the most important step to finding $q(z)$ is to determine the real and imaginary parts of the complex function $I_0(\alpha)$ from the experiment.

We place an additional plate with well-known linear and nonlinear properties in front of the test plate (not necessarily close to it) in the region $z_2 < z < z_{a1}$. For the new one-dimensionally inhomogeneous structure we can write an equation, similar to (25):

$$\int_{z_1}^{z_{a1}} q_{a1}(u)K_{a1}(u,\alpha)\,du = -[R'_{a1}(z_{a1}) + ik_z R_{a1}(z_{a1})]S_{a1}(\alpha). \quad (26)$$

Here, $S_{a1}(\alpha)$ is the coefficient characterising the efficiency of conversion of the fundamental waves into the reflected sum- or difference-frequency wave, the wave being converted by the test and reference plates. Functions $q_{a1}(z)$, $K_{a1}(z,\alpha)$, $R_{a1}(z,\alpha)$ entering into (26) are defined by the relations that are similar to those given after equation (25).Their values in the region $z_2 < z_1 < z_{a1}$ are known for the known linear properties of the investigated plate, as well as for linear and nonlinear dielectric parameters of the reference plate. Therefore, the function

$$J_1(\alpha) \equiv \int_{z_2}^{z_{a1}} q_{a1}(u)K_{a1}(u,\alpha)\,du \quad (27)$$

is also assumed known.

Note that when $z \in [z_1, z_2]$, the equalities $q_{a1}(z) = q(z)$ and $R_{a1}(z,\alpha) = R(z,\alpha)$ are fulfilled. The first equality is obvious, and the validity of the latter relation follows from the fact that the boundary conditions (13) and (22) for the auxiliary functions $R_s(z,\alpha)$ and $R_d(z,\alpha)$ are set at point $z = z_1$ and, therefore, the solutions of equations (10) and (21) for these functions at $z \in [z_1, z_2]$ do not depend on what is in the region $z > z_2$.

The presence of the additional reference plate will, of course, change the complex field amplitude of the fundamental wave in the test plate. In this case, the functions $E^{(1)}_{1,1a}(z)$ and $E_{n,1a}(z)$, where $n = 1, 2$, should at $z = z_1$ satisfy the same homogeneous boundary conditions as functions $E^{(1)}_1(z)$ and $E_n(z)$ [see (5) and (16)]. Because any homogeneous boundary condition determines the solution of the second-order differential equation with an accuracy to a constant factor, then

$$E^{(1)}_{1,1a}(z) = C^{(1)}E^{(1)}_1(z),\ E_{n,1a}(z,\alpha) = C^{(1)}_n(\alpha)E_n(z,\alpha) \quad (28)$$

for $z \in [z_1, z_2]$. Here, $C^{(1)}$, $C^{(1)}_n$ are the known constant complex factors which depend only on the frequency and angle of incidence of the fundamental waves onto the plates, as well as on the linear properties and mutual arrangement of the plates. It follows from (28) and definition of the function $K(z,\alpha)$ that at $z \in [z_1, z_2]$ the function $K_{a1}(z,\alpha) = C_1(\alpha)K(z,\alpha)$, where the complex constant C_1 is equal to $C^{(1)}C^{(1)}_2$ or $C^{(1)}_1\left(C^{(1)}_2\right)^*$, depending on the used method of measurements. Therefore, dividing the integration domain in the left-hand side of equation (26) into two subdomains ($[z_1, z_2]$ and $[z_2, z_{a1}]$) and using (25) and (27), equation (26) for a composite plate can be written as

$$J_1(\alpha) + C_1(\alpha)I_0(\alpha) = -[R'_{a1}(z_{a1}) + ik_z R_{a1}(z_{a1})]S_{a1}(\alpha). \quad (29)$$

If we now shift the additional plate for some distance along the z axis or replace it with another reference plate such that it be in the region $z_2 < z < z_{a2}$, then the value of the right-hand side of (27) changes to $J_2(\alpha)$. Using the same line of reasoning as in the first case, we obtain

$$J_2(\alpha) + C_2(\alpha)I_0(\alpha) = -[R'_{a2}(z_{a2}) + ik_z R_{a2}(z_{a2})]S_{a2}(\alpha). \quad (30)$$

After measuring the moduli of the conversion coefficients $S(\alpha)$, $S_{a1}(\alpha)$ and $S_{a2}(\alpha)$, the moduli of the right-hand sides of equations (25), (29) and (30), which we denote, respectively, A_0, A_1 and A_2, prove to be the known quantities, and it is possible to write the relations

$$|I_0(\alpha)| = A_0, \quad |I_0(\alpha) + \tilde{J}_1(\alpha)| = \tilde{A}_1,$$

$$|I_0(\alpha) + \tilde{J}_2(\alpha)| = \tilde{A}_2, \quad (31)$$

where $\tilde{J}_n(\alpha) = J_n(\alpha)/C_n(\alpha)$; $\tilde{A}_n = A_n/|C_n(\alpha)|$; $n = 1, 2$.

After some transformations, we obtain from (31) a system of linear equations

$$\mathrm{Re}\{\tilde{J}_{1,2}\}\,\mathrm{Re}\{I_0\} + \mathrm{Im}\{\tilde{J}_{1,2}\}\,\mathrm{Im}\{I_0\} = (\tilde{A}^2_{1,2} - A^2_0 - |\tilde{J}_{1,2}|^2)/2, \quad (32)$$

allowing one to find unambiguously $\mathrm{Re}\{I_0\}$ and $\mathrm{Im}\{I_0\}$, if the determinant $\mathrm{Re}\{\tilde{J}_1\}\,\mathrm{Im}\{\tilde{J}_2\} - \mathrm{Im}\{\tilde{J}_1\}\,\mathrm{Re}\{\tilde{J}_2\} = \mathrm{Im}\{\tilde{J}^*_1\tilde{J}_2\}$ is not zero. Note that the latter is the decisive factor when choosing reference plates and their location relative to the test plate. Fulfilment of this condition should be checked before the measurements. It is easy to see that the proposed method of replacement of phase measurements by intensity measurements is based on the interference of the waves at the sum (difference) frequency, and also takes into account the interference of the fundamental waves in the system of two parallel plates. Therefore, it is necessary to measure the mutual position of the plates with an error much smaller than the wavelength at the sum (difference) frequency.

5. Conclusions

Thus, significant progress, made in recent years in the field of spectroscopy of one-dimensionally inhomogeneous media with quadratic nonlinearity, indicates the fundamental possibility of unambiguous determination (by the experimental data) of the coordinate dependences of the components of complex quadratic susceptibility tensors, if the medium under study has the form of a plane-parallel plate, the surfaces of which are perpendicular to the direction of the inhomogeneity of its linear and nonlinear dielectric properties. The proposed methods for unique reconstruction of the profile of the tensor components, $\hat{\chi}^{(2)}(z, \omega_1 \pm \omega_2; \omega_1, \pm\omega_2)$, are applicable for a medium with an arbitrary frequency dispersion, if there exists a coordinate system in which the tensor of its linear dielectric constant is diagonal. They include three series of measurements of the wave intensities at the sum (difference) frequency, generated under special conditions with the test and additional reference plates, which eliminates the need for complicated phase measurements. By varying the frequencies ω_1 and (or) ω_2 of the incident waves, we can reconstruct the profiles of the tensor components, $\hat{\chi}^{(2)}(z, \omega_1 \pm \omega_2; \omega_1, \pm\omega_2)$ at different frequency arguments and, therefore, investigate the frequency dispersion of the quadratic susceptibility of different parts of the medium. The latter, in particular, can be used for the problems of nondestructive testing of the internal structure of the various devices.

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