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Nonlinear diffraction of light on near-surface microdomain structures

A.V. Kondratov, M.V. Gorkunov

Abstract. **The nonlinear diffraction on periodic microdomain structures with different orientations of the spontaneous polarisation axis relative to the surface is theoretically considered. The possibility of reliable non-destructive diagnostics of the period, duty cycle and depth of domain structures, as well as of the shape of the domain walls under the crystalline surface is shown.**

Keywords: *nonlinear light diffraction, periodic microdomain structures, domain walls.*

1. Introduction

Studies of microscopic domain structures are important both to clarify the peculiarities of the occurence and stabilisation of spontaneous polarisation in ferroelectric crystals and to understand the prospects for the use of these structures in photonics and nanoelectronics. Historically, structures with an alternating polar axis direction were considered primarily as nonlinear photonic crystals, in which the one-dimensional periodicity compensates for the phase mismatch of the interacting waves, the so-called effect of quasi-phase matching, and can significantly improve the efficiency of nonlinear frequency conversion [1]. 2D structures open up additional possibilities for matching nonlinear interaction of waves propagating in different directions [2]. Much effort was made to increase the conversion efficiency [3], including taking into account the nonideality of the periodic structure [4] and related nonlinear processes, such as the formation of rapidly oscillating solitons [5].

In recent years, domain structures in ferroelectrics have turned out to be interesting for nanoelectronics due to the special electrical properties of domain walls, which make these structures promising for high-density nonvolatile memory [6]. Much attention is paid to various methods of controlling walls: both by point irradiation with a focused electron beam [7] and by optical methods [8]. In addition to domain walls, it is also proposed to use the form of the domains themselves for encoding information with a sequence of triangular electric pulses [9].

Domain structures with a characteristic scale of about ten micrometers can be fabricated by applying an electrical voltage to structured electrodes deposited on the surface of a ferroelectric crystal [10]. The formation of submicron-scale structures (so-called microdomain structures) requires qualitatively different approaches and can be carried out with an atomic-force microscope (AFM) by applying a high voltage to the nanoscale tip of the probe [11, 12]. By irradiating small areas of the crystal surface with a focused electron beam on a scanning electron microscope, it is possible to produce a spatial charge localised in the near-surface region that will induce electric fields sufficient for local switching of the sign of spontaneous polarisation [13, 14].

In this regard, of particular importance is the problem of domain structure diagnostics [15]. Visualisation of domains during chemical etching of crystals with acids is quite informative, based on the fact that surface areas perpendicular to the spontaneous polarisation vector have an electric charge, whose sign depends on the polarisation direction, which provides etching rate selectivity of domains of different sign [16, 17]. The presence of a surface charge also makes it possible to non-destructively diagnose domains by decorating them with colloidal particles [16] and nematic liquid crystals [18, 19]. The precise reconstruction of domain structures by atomic force microscopy [12] is actively developing, which allows domains with spontaneous polarisation along the surface to be distinguished [20]. Nevertheless, the above methods make it possible to identify the regions of emergence of domains to the surface, but do not provide information about the spatial arrangement of domain walls in the bulk of the crystal. In most cases, there is simply no reliable information about the depth and cross-sectional shape of microdomains formed in the near-surface regions of crystals.

The possibility of optical visualisation of bulk domain structures is due to the fact that the spontaneous polarisation direction does not directly affect the linear optical properties of crystals, but determines the sign of the key constants of quadratic nonlinearity. Thus, spatial changes in the direction of spontaneous polarisation are accompanied by the modulation of the sign of the components of the linear electro-optical tensor, making it possible to observe with an optical microscope the domains in a crystal to which a sufficiently strong electric field is applied [21]. On the one hand, this method provides the possibility of continuous monitoring of the movement and the switching of the sign of domain structures in an electric field, and on the other hand, the need to apply a field causes the measurement process to influence the result.

A.V. Kondratov A.V. Shubnikov Institute of Crystallography, Federal Scientific Research Centre 'Crystallography and Photonics', Russian Academy of Sciences, Leninsky prosp. 59, 119333 Moscow, Russia; e-mail: kondratov.aleksey@gmail.com;
M.V. Gorkunov A.V. Shubnikov Institute of Crystallography, Federal Scientific Research Centre 'Crystallography and Photonics', Russian Academy of Sciences, Leninsky prosp. 59, 119333 Moscow, Russia; National Research Nuclear University 'MEPhI', Kashirskoe sh. 31, 115409 Moscow, Russia

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In fact, the only possibility of non-destructive diagnosis of domains in the bulk of crystals is provided by nonlinear optics. In the simplest version, the crystal is subjected to a strong pump wave, whose front is not distorted and the nonlinear polarisation, which acts as a radiation source at the second harmonic (SH) wavelength, is phase modulated in space synchronously with the direction of the spontaneous polarisation vector. This allows one, in particular, to observe relatively large domains directly in the SH confocal microscopy regime [22], in which domains with different polarisation directions acquire different apparent contrast at the SH wavelength due to interference of the wave emitted by the polarised domain and the wave outgoing from the homogeneous nonlinear substrate or from an unpolarised crystal volume [23]. Diagnostics of smaller domain structures is possible according to the patterns of the distribution of SH intensity in the nonlinear scattering regime. Randomly alternating microdomains give a characteristic conical pattern of diffuse scattering [24], while periodic structures emit SH narrowly, in the form of peaks of nonlinear diffraction [25].

From general considerations it is clear that the intensity of the peaks of nonlinear diffraction contains valuable information not only about the period, but also about the depth and shape of the microdomain structures in the bulk of the crystal. In this paper, we solve the problem of nonlinear diffraction for two practically important cases of orientation of the polar axis of the crystal, i.e. parallel and perpendicular to its surface. The analysis of diffraction patterns is carried out and the possibility of reliable determination of the depth of microdomains, as well as of the restoration of the features of the shape of their cross section, is found.

2. Amplitudes of nonlinear diffraction

Let us consider a crystal uniformly polarised in volume, in the near-surface layer of which domain structures with a period of $2d$ are formed (Fig. 1). We assume that the domains propagate under the crystal surface to a certain maximum depth h, with the neighbouring domains having oppositely directed spontaneous polarisation. These assumptions correspond to two experimental realisations of the recording of planar (near-surface) domains: on a nonpolar surface under electron beam irradiation [14] and on a polar surface in an AFM probe field [11, 26], in which the depth of the formed domains does not exceed a few micrometres. In the two most practical cases, the vector of spontaneous polarisation is oriented perpendicular or parallel to the surface of the crystal and can be expressed as $\boldsymbol{p}(\boldsymbol{r}) = p\boldsymbol{e}_z f(x,z)$ (Fig. 1a) or as $\boldsymbol{p}(\boldsymbol{r}) = p\boldsymbol{e}_x f(x,z)$ (Fig. 1b). In both cases, the function f is an integer, taking values of ±1 within the domain structure having a period $2d$ along the x axis and a constant value $f = 1$ away from the surface by more than some limit depth $z > h$.

Consider the problem of nonlinear diffraction, schematically presented in Fig. 1. Due to the fact that the SH generation (at a frequency of 2ω) is weak and occurs in a thin surface structure, we neglect its influence on the propagation of a plane pump wave at a frequency ω, incident on the surface of the crystal at an angle θ_0. We also neglect the distortions of the linear optical properties of the crystal caused by its deformation in the vicinity of the domain walls. From the point of view of symmetry, the SH generation is determined by the absence of a local centre of inversion in the crystal. In a ferroelectric crystal, the corresponding decrease in symmetry is directly related to the presence of a polar axis along the spontaneous polarisation vector $\boldsymbol{p}$. The SH generation is determined by the components of the third-rank tensor of the quadratic nonlinear optical susceptibility at the SH and pump frequencies, $\chi^{(2)}_{ijk}(2\omega;\omega,\omega)$, which is symmetric with respect the last two subscripts. The presence of three tensor combinations, $p_i p_j p_k$, $p_i \delta_{ik}$ and $(p_i \delta_{ik} + p_k \delta_{ij})$, defined by spontaneous polarisation and satisfying the symmetry requirements, indicates the existence of several independent nonlinear-optical coefficients. In practice, however, in the overwhelming majority of uniaxial ferroelectric crystals, one of these coefficients – the coefficient d_{33}, which is responsible for the interaction of light waves polarised along the vector $\boldsymbol{p}$ – far exceeds the others. For the sake of simplicity, we will further assume that the spatially inhomogeneous nonlinear susceptibility in the microdomain structure follows spontaneous polarisation

$$\chi^{(2)}_{ijk}(\boldsymbol{r};2\omega;\omega,\omega) = 2d_{33} f(x,z)\delta_{i\alpha}\delta_{j\alpha}\delta_{k\alpha}, \qquad (1)$$

where the index $\alpha = z, x$ denotes the orientations shown in Figs 1a and 1b, respectively.

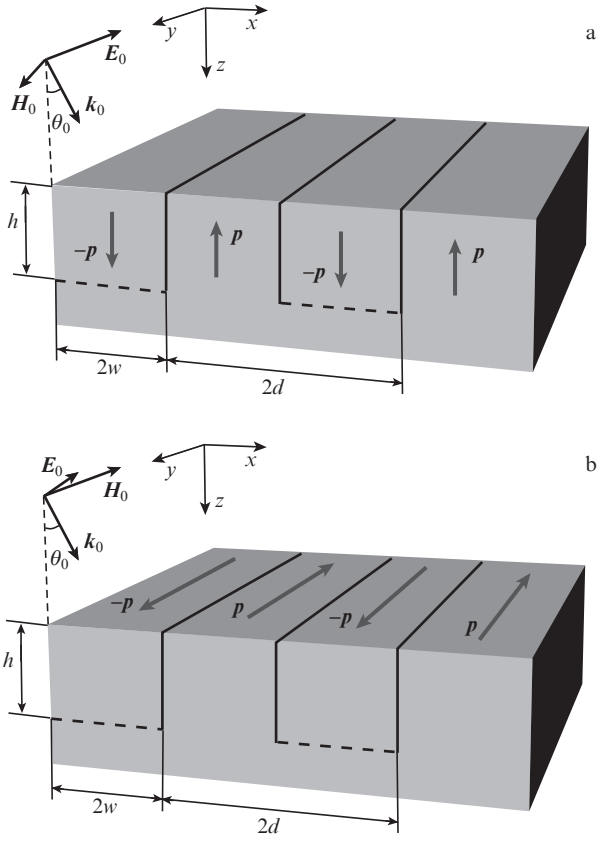

Figure 1. Domains with a vector of spontaneous polarisation (a) perpendicular and (b) parallel to the crystal surface.

From relation (1), in particular, it follows that effective diagnostics of differently oriented domain structures requires different polarisation of the incident pump wave: Structures with spontaneous polarisation oriented perpendicular to the surface interact with the TM pump wave, and structures with planar oriented polarisation interact with the TE wave. Since the SH generation due to nonlinearity (1) does not change the polarisation state of light, for convenience we will further call the considered cases nonlinear TM or TE diffraction.

Let us examine in more detail the case of TM diffraction, presented in Fig. 1a. The components of the electric field of the pump wave inside the crystal have the form

$$E_{1x}(x,z,t) = E_{1x}\cos(k_{1z}z + k_{1x}x - \omega t),$$
$$E_{1z}(x,z,t) = E_{1z}\cos(k_{1z}z + k_{1x}x - \omega t), \quad (2)$$

where $k_{1x} = (\omega/c)\sin\theta_0$; $k_{1z} = \sqrt{\varepsilon_1\omega^2/c^2 - k_{1x}^2}$; ε_1 is the permittivity of the crystal at the pump frequency, the anisotropy of which we neglect; and E_{1x} and E_{1z} are the real amplitudes of the pump wave in the crystal, which are expressed through the amplitude E_0 of the wave incident on the crystal from the outside,

$$E_{1x} = \frac{c}{\omega}\frac{2k_{1z}\cos\theta_0}{\varepsilon_1\cos\theta_0 + \sqrt{\varepsilon_1 - \sin^2\theta_0}}E_0,$$
$$E_{1z} = -\frac{c}{\omega}\frac{2k_{1x}\cos\theta_0}{\varepsilon_1\cos\theta_0 + \sqrt{\varepsilon_1 - \sin^2\theta_0}}E_0. \quad (3)$$

Propagating into the crystal, this plane wave produces nonlinear polarisation along the z axis

$$P_z^{\mathrm{nl}}(x,z,t) = 2d_{33}f(x,z)E_{1z}^2(x,z,t), \quad (4)$$

which emits waves at the SH frequency. The system of Maxwell's equations for the SH fields contains a source in the form of a current distributed in a crystal, oscillating along the z axis at a frequency of 2ω with amplitude

$$j_{2z}^{\mathrm{nl}}(x,z) = -\mathrm{i}\omega d_{33}E_{1z}^2 f(x,z)\exp[2\mathrm{i}(k_{1z}z + k_{1x}x)]. \quad (5)$$

The specific form of the SH fields can be found by solving the wave equations for the amplitudes of the magnetic field H_{2y} in the crystal (i.e., at $z > 0$),

$$\left(\Delta + \frac{4\omega^2}{c^2}\varepsilon_2\right)H_{2y}(x,z) = \frac{4\pi}{c}\frac{\partial}{\partial x}j_{2z}^{\mathrm{nl}}(x,z), \quad (6)$$

and above the crystal (at $z < 0$),

$$\left(\Delta + \frac{4\omega^2}{c^2}\right)H_{2y}(x,z) = 0, \quad (7)$$

where ε_2 is the permittivity of the crystal at the SH frequency. The solutions of the equations satisfy the boundary conditions of continuity of H_{2y} and $(1/\varepsilon)\partial H_{2y}/\partial z$ at the surface of the crystal at $z = 0$.

In the framework of the formalism of Green's function, we find the magnetic field of the SH in the form

$$H_{2y}(x,z) = \frac{4\pi}{c}\int_{-\infty}^{\infty}\mathrm{d}x'\int_0^{\infty}\mathrm{d}z'\,G(x-x',z,z')\frac{\partial}{\partial x'}j_{2z}^{\mathrm{nl}}(x',z'), \quad (8)$$

where Green's function G in the crystal satisfies the wave equation with a source in the form of an infinitely thin filament

$$\left(\Delta + \frac{4\omega^2}{c^2}\varepsilon_2\right)G(x-x',z,z') = \delta(x-x')\delta(z-z'), \quad (9)$$

and outside the crystal, the homogeneous wave equation (7) and the same boundary conditions as the magnetic field. By representing Green's function as a Fourier integral along the longitudinal coordinate,

$$G(x-x',z,z') = \frac{1}{2\pi}\int \mathrm{d}q\exp[\mathrm{i}q(x-x')]\,G(q,z,z'), \quad (10)$$

and using the well-known general form of the dyadic Green's function for the interface between two media [27], we find that the field produced above the crystal ($z < 0$) by a thin filament emitting from the depth of the crystal ($z' > 0$) has the Fourier transform

$$G(q,z,z') = -\frac{\mathrm{i}}{\kappa_0(q)\varepsilon_2 + \kappa(q)}\exp[\mathrm{i}\kappa(q)z' - \mathrm{i}\kappa_0(q)z], \quad (11)$$

where $\kappa_0(q) = \sqrt{4\omega^2/c^2 - q^2}$ and $\kappa(q) = \sqrt{(4\omega^2/c^2)\varepsilon_2 - q^2}$.

Substitution of expression (11) into the Fourier integral (10) makes it possible to represent in general form the SH magnetic field (8) for current (5) corresponding to nonlinear scattering on an arbitrary domain structure defined by some function $f(x, z)$. To describe narrowly directed nonlinear diffraction on a periodic domain structure, we consider a periodic function f with a period $2d$, which can be represented as a discrete Fourier series

$$f(x,z) = \sum_{n=-\infty}^{\infty} f_n(z)\exp\left(\mathrm{i}\pi n\frac{x}{d}\right), \quad (12)$$

where

$$f_n(z) = \frac{1}{2d}\int_{-d}^{d} f(x,z)\exp\left(-\mathrm{i}\pi n\frac{x}{d}\right)\mathrm{d}x.$$

In this case, the SH magnetic field (8) also takes the form of a discrete sum of plane waves:

$$H_{2y}(x,z) = \sum_{n=-\infty}^{\infty} H_n\exp[\mathrm{i}q_n x - \mathrm{i}\kappa_0(q_n)z],$$

emerging from the crystal and having a wave vector component along the surface, $q_n = 2k_{1x} + \pi n/d$, i.e. emerging at a reflection angle of θ_n, satisfying the condition for nonlinear Bragg diffraction

$$\sin\theta_n = \sin\theta_0 + \frac{\lambda n}{d}, \quad (13)$$

where $\lambda = 2\pi c/\omega$ is the wavelength of light in vacuum at the pump frequency.

The magnetic field amplitude of a plane wave of the nth order of nonlinear diffraction is given by

$$H_n = -4\pi\frac{\mathrm{i}\omega}{c}d_{33}E_{1z}^2\frac{q_n}{\varepsilon_2\kappa_0(q_n) + \kappa(q_n)} \times\int_0^{\infty}\mathrm{d}z'\exp\{\mathrm{i}[\kappa(q_n) + 2k_{1z}]z'\}f_n(z'), \quad (14)$$

and the intensity of this wave, $I_n = (c/4\pi)|H_n|^2$ is proportional to the square of the pump wave intensity $I_0 = (c/4\pi)|E_0|^2$.

Nonlinear TE diffraction on the domain structure shown in Fig. 1b is considered in a similar way. The nonlinear current directed along the y axis acts as a source of the SH radiation and reads as:

$$j_{2y}^{\mathrm{nl}}(x,z) = -\mathrm{i}\omega d_{33}E_{1y}^2 f(x,z)\exp[2\mathrm{i}(k_{1z}z + k_{1x}x)], \quad (15)$$

where the amplitude of the electric field of the pump wave in the crystal has the form

$$E_{1y} = \frac{2\cos\theta_0}{\cos\theta_0 + \sqrt{\varepsilon_1 - \sin^2\theta_0}} E_0.$$

The fields of the generated SH can be calculated by solving the wave equations for the amplitudes of the electric field E_{2y} in the crystal (at $z > 0$),

$$\left(\Delta + \frac{4\omega^2}{c^2}\varepsilon_2\right) E_{2y}(x,z) = -\frac{8\pi i\omega}{c^2} j_{2y}^{\mathrm{nl}}(x,z), \tag{16}$$

and above the crystal (at $z < 0$),

$$\left(\Delta + \frac{4\omega^2}{c^2}\right) E_{2y}(x,z) = 0, \tag{17}$$

with the boundary conditions of continuity of E_{2y} and $\partial E_{2y}/\partial z$ at the crystal surface at $z = 0$. The Fourier transform of Green's function for this problem also follows from the general form of the dyadic Green's function for the interface between two media [27]

$$G(q,z,z') = -\frac{i}{\kappa_0(q)\varepsilon_2 + \kappa(q)} \exp[i\kappa(q)z' - i\kappa_0(q)z], \tag{18}$$

and calculations completely analogous to the previous ones allow us to find the electric field amplitudes of a plane wave of the nth order of nonlinear diffraction in the form

$$E_n = -8\pi\frac{\omega^2}{c^2} d_{33} E_{1y}^2 \frac{1}{\kappa_0(q_n) + \kappa(q_n)} \times \int_0^\infty dz' \exp\{i[\kappa(q_n) + 2k_{1z}]z'\} f_n(z'), \tag{19}$$

which determine the intensities of the TE diffraction waves, $I_n = (c/4\pi)|E_n|^2$.

3. Role of depth and width of domains

As follows from the obtained amplitudes of nonlinear diffraction (14) and (19), for both considered geometries, the nth order diffraction intensity is proportional to the square of the modulus of the dimensionless complex structural factor

$$F_n = \frac{1}{2d}\frac{\omega}{c} \times \int_0^\infty dz \int_{-d}^{d} dx\, f(x,z) \exp\{i[\kappa(q_n) + 2k_{1z}]z - i\pi n x/d\}, \tag{20}$$

which contains information about the shape of the domain structure defined by the integer function $f(x,z)$.

Let us consider the simplest case of a lattice of domains of rectangular cross section with a depth h and width $2w$, less than the period of structure $2d$. In fact, in the bulk of the crystal, domains can be of completely different shapes: both semi-ellipsoidal and conical, as follows from theoretical calculations based on minimising the corresponding free energy [28]. However, as will be shown below, in the case of a rectangular cross section, the nonlinear diffraction problem has an exact analytical solution that allows us to trace the main regularities, whereas consideration of domains of arbitrary shape requires the introduction of the function of profile deviation from the rectangular one. Thus, within the same period for $|x| < d$ we obtain

$$f(x,z) = \begin{cases} 1, & z > h, \\ -1, & |x| \leqslant w,\ z \leqslant h, \\ 1, & |x| > w,\ z \leqslant h. \end{cases} \tag{21}$$

This corresponds to the expression

$$|F_n|^2 = \frac{16}{\pi^2 n^2}\frac{\omega^2}{c^2}\frac{1}{[\kappa_0(q_n) + 2\kappa_{1z}]^2} \times \sin^2\left(\pi n\frac{w}{d}\right)\sin^2\left\{\frac{h}{2}[\kappa(q_n) + 2k_{1z}]\right\}, \tag{22}$$

which determines the simple dependence of the nonlinear diffraction intensity on the domain structure depth h and duty cycle w/d.

In particular, $|F_n|^2 = 0$ when the condition

$$n\frac{w}{d} = l \tag{23}$$

with an arbitrary integer l is fulfilled, which means the complete disappearance of the nth order of diffraction, regardless of the angle of incidence of the pump wave, if the domain structure duty cycle w/d is equal to a certain rational number with denominator n. Since the ratio w/d is always less than unity, such a complete disappearance of a certain diffraction order is possible only for $|n| > 1$.

The last factor in expression (22) indicates the possibility of non-destructive diagnostics of the depth of the domains due to the fact that the intensity of the nth order of diffraction identically vanishes when the condition

$$[\kappa(q_n) + 2k_{1z}]h = 2\pi m \tag{24}$$

with an arbitrary integer m is fulfilled. Taking into account the notations adopted above, this takes place at such an angle of pump wave incidence, θ_0, that

$$h = \frac{\lambda m}{2}\left[\sqrt{\varepsilon_2 - \left(\sin\theta_0 + n\frac{\lambda}{4d}\right)^2} + \sqrt{\varepsilon_1 - \sin^2\theta_0}\right]^{-1}. \tag{25}$$

Consequently, the depth of the domains of rectangular cross section can be found easily if we know the set of values of angles θ_0 at which a given diffraction order disappears.

As a practical illustration, Fig. 2 presents the intensities of the first orders of nonlinear diffraction, $I_{\pm1}$, on a periodic domain structure in a lithium niobate crystal. The pump and SH wavelengths are assumed to be 1064 and 532 nm, respectively, and the weak anisotropy of the linear optical properties of this crystal makes it possible to describe it for simplicity with isotropic permittivities equal to the values of ordinary components of the corresponding tensor at these wavelengths: $\varepsilon_1 = 4.97$ and $\varepsilon_2 = 5.23$ [29]. Among the known values of the three nonzero components of the nonlinear susceptibility tensor for lithium niobate [30], $d_{33} = -98 \times 10^{-9}$ CGSE units, $d_{31} = 14 \times 10^{-9}$ CGSE units, $d_{22} = 7.4 \times 10^{-9}$ CGSE units, the first component is dominant, which justifies the use of a simplified form of the nonlinear susceptibility tensor (1). The intensities $I_{\pm1}$ for the geometries corresponding to TE and TM diffractions are shown in Figs 2a and 2b, respectively. In the case of

TE diffraction, there are clear minima in the regions of 15°–20° and 45°–50°, corresponding to the angles of incidence obtained from expression (25). For TM diffraction, the minima in the region of 15°–20° are not visible because the intensity is close to zero at small angles due to the specificity of the geometry and the proportionality of the values of $I_{\pm1}$ and $|E_{1z}|^4$. In both cases, the intensity of the diffracted wave for $n = 1$ becomes zero when the angle of incidence is less than 90°, which corresponds to a negative root expression in the formula for $\kappa_0(q_n)$, i.e., under total internal reflection.

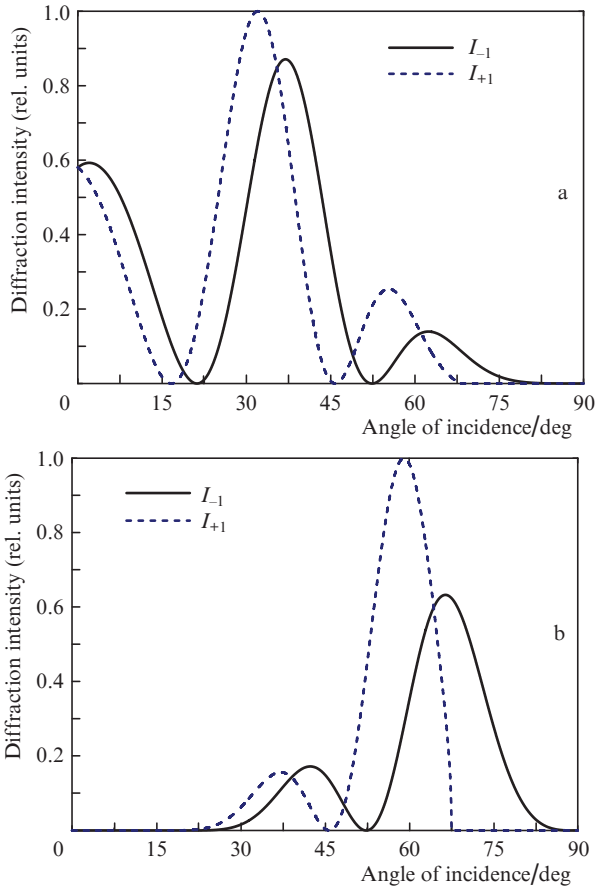

Figure 2. Dependences of the relative intensities of the first orders of nonlinear diffraction, $I_{\pm1}$, on the angle of incidence of the pump wave on a lithium niobate crystal with a rectangular domain structure having a depth of 2.5 μm, a period of 7 μm and a duty cycle of 1/7 for geometries corresponding to (a) TE and (b) TM diffraction.

4. Role of the cross-sectional shape of domains

The explicit dependence of the structural factor (20) on the cross section of the domains described by the function $f(x,z)$ allows one to consider nonlinear diffraction as a way to diagnose the shape of the domain walls inside the crystal. For visual illustration, we use a set of cross sections of domains that have the same duty cycle w/d at the output to the crystal surface and the same maximum depth h, but differing in the shape of the walls. This shape is described at $z > 0$ within one period of the structure ($|x| < d$) by the function

$$f(x,z) = \begin{cases} 1, & z > h, \\ -1, & |x| \leqslant w - \Delta w(z), \quad z \leqslant h, \\ 1, & |x| > w - \Delta w(z), \quad z \leqslant h, \end{cases} \tag{26}$$

where

$$\Delta w(z) = w\left[1 - \left(1 - \frac{z}{h}\right)^{1/N}\right] \tag{27}$$

is the deviation of the domain walls from the vertical form; and N is a positive integer specifying various wall shapes: from triangular with $N = 1$ and smooth parabolic with $N = 2$ to sharp rectangular in the limit $N \to \infty$. The corresponding structural factors (20) can then be represented as

$$F_n = -\frac{\omega}{c}\frac{2}{\pi n}\int_0^h \mathrm{d}z \sin\left[\frac{w - \Delta w(z)}{d}\right]\exp\{\mathrm{i}[\kappa(q_n) + 2k_{1z}]z\}, \tag{28}$$

and finding their numerical values allows us to calculate the intensities of various orders of nonlinear diffraction.

As an example, Fig. 3 presents dependences similar to those shown in Fig. 2b, with identical geometrical parameters of the domain structure h, w and d, but for different cross-sectional profiles. It can be seen that for small deviations of the shape of the domains ($N = 20$), the angular dependence of the diffraction intensity is almost equivalent to that in the case of ideal rectangular domains. For the shape of the domains corresponding to $N = 5$, the dips in the angular dependence of intensity are further smoothed, and its zeros become local minima; however, their position still corresponds to expression (25) and allows the domain structure depth to be determined. In the case of both parabolic ($N = 2$) and triangular ($N = 1$) domains, there is a complete blurring of dips in the angular dependence of the diffraction intensity, and the determination of their depth turns out to be impossible.

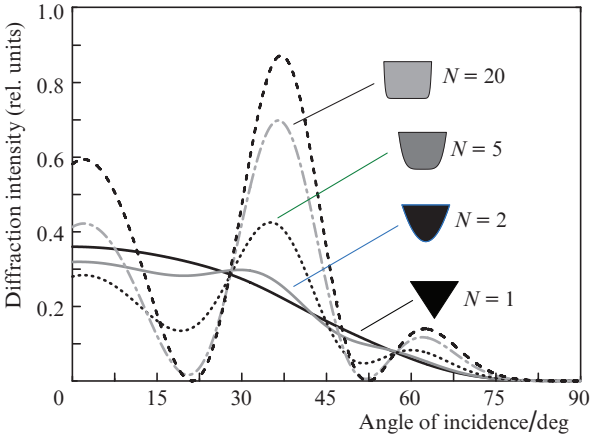

Figure 3. Influence of domain cross-sectional shape on nonlinear diffraction: dependences of the relative first-order intensity of TE diffraction, I_{-1}, on the angle of incidence of the pump wave on a lithium niobate crystal with a domain structure having a depth of 2.5 μm, a period of 7 μm, a duty cycle of 1/7 and cross-sectional profiles corresponding to the analytical representation (26) for $N = 1$, 2, 5 and 20. The dashed curve is the intensity in the case of domains with an ideal rectangular shape.

5. Conclusions

Thus, nonlinear diffraction allows one to reconstruct in detail the most important characteristics of periodic domain structures for different orientations of spontaneous polarisation

relatively to the surface. Along with the simple condition (13), which makes it possible to determine the period of structures in the directions of propagation of nonlinearly diffracted waves, the dependences of the intensity of these waves on the angle of incidence of the pump wave allow one to accurately estimate the depth of the domain structure and even reveal the shape of the domain walls inside the crystal.

The most accurate determination of the depth is possible for periodic domains of rectangular cross section, for which, at given angles of incidence of the pump wave, condition (25) is satisfied and some diffraction orders disappear. Being a consequence of the destructive interference of SH fields generated by nonlinear currents at different depths, the essence of the used SH interference suppression is the same as when contrasting individual large domains in the SH confocal microscopy regime [22, 23]. Nonlinear diffraction, however, makes it possible to measure the depth of periodically repeating domains, the width of each of which can be substantially less than the wavelength of light.

The deviation of the cross-sectional shape of the domains from the ideally rectangular one leads to the violation of the conditions necessary for the complete interference supression of the SH. As shown above, the depth of the dips in the dependence of the intensity of the diffracted waves on the angle of incidence of the pump wave turns out to be quite sensitive to changes in the shape of the domain wall, even with a constant two-dimensional pattern of the emergence of the domains on the surface.

We also note that the theoretical formalism developed on the basis of simple domain structures is applicable to structures with a much more complex unit cell. The fabrication of such structures with arbitrary regular, including superperiodic, domain ordering is possible using modern atomic force and scanning electron microscopy devices. The obtained relations allow us to calculate the efficiency of nonlinear diffraction on such structures and, for example, to find out the conditions for the suppression and amplification of its certain orders, which has an independent practical value. Thus, the formation of a narrowly directed SH, emitted in one direction, can actually be considered as nonlinear refraction of light.

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