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Second- and third-harmonic parametric scattering in disordered quadratic media

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Abstract

We study frequency conversion via interaction of two fundamental beams in quadratic nonlinear media with random distribution of the ferroelectric domains. We demonstrate broadband second- and third-harmonic generation, analyse both spatial and polarization properties of the emitted harmonics and show that they closely follow the theoretical predictions.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

There is a natural common perception that coherent optical phenomena are always intimately associated with perfectly ordered media as is, for instance, the case for normal light propagation in crystals where particular ordering of atoms or ions and spatial symmetry associated with this ordering defines particular optical properties including the nonlinear optical response. In fact, the lack of this ordering immediately precludes certain optical properties from being observed. However it turns out that this is not always true and actually introducing disorder into an otherwise perfect optical system (structure) is in fact beneficial. As Baudrier-Raybaut *et al* [1] demonstrated recently the nonlinear optical process of non-phase-matched frequency conversion is much more efficient when realized in a polycrystalline medium consisting of micron-size randomly oriented crystallites than the same process conducted in a regular crystal of the same material. This beneficial role of disorder on the parametric process has been confirmed in a number of other works in recent years. Particularly interesting in this context is the use of ferroelectric crystals with disordered domain structure. Already Horowitz *et al* [2] and Kawai *et al* [3]

have shown that as-grown strontium barium niobate (SBN) crystals could be used to realize broadband second and sum-frequency generation. This was possible due to the presence of elongated antiparallel ferroelectric domains which leads to spatial random modulation of the sign of the quadratic nonlinearity. Such nonlinearity modulation provides a continuous domain of reciprocal wave vectors $\vec{G}$ which enable us to fulfil the phase-matching condition [4] which for second-harmonic generation (SHG) reads $2\vec{k}_{(\omega)} + \vec{G} = \vec{k}_{(2\omega)}$, where $\vec{k}_{(\omega)}$ and $\vec{k}_{(2\omega)}$ are the wave vectors of the fundamental (FW) and the second-harmonic (SH) beams, respectively. It appears that the multi-domain SBN crystal is particularly well suited for the broadband operation because its spatial random distribution of nonlinearity enables phase matching for the second-order nonlinear processes at practically any incident wavelength [5]. A number of useful applications have been demonstrated with this crystal including e.g. multi-frequency conversion [6–8], conical and planar emission [9–11] as well as the frequency mapping from infrared to visible [12]. It was also shown that the SBN crystal with disordered domains can be used to construct simple optical auto-correlator [11, 13]. It has been also recently demonstrated that such nonlinear

Figure 1. (a) Schematic of the experimental setup. The multi-domain structure of the SBN sample is schematically shown. WP—half-wave plate, L—cylindrical lens, BP—bi-prism. (b and c) Multi-domain structure of the SBN sample as visualized by the second-harmonic nonlinear microscopy; viewing area perpendicular (b) and parallel (c) to the optical (Z) axis.

medium with random domains can support cascaded third-harmonic generation [5, 14, 15].

As far as the application for pulse monitoring and characterization is concerned, the nonlinear interaction in the disordered crystal involves two beams (pulses) intersecting at a small angle inside the disordered medium. We have recently investigated this interaction scenario in the context of SHG [10] using fundamental beams from the very near-infrared part of the spectrum (1064 nm and less). In this work we extend our studies by considering simultaneous second- and third-harmonic generation via cascading. This is particularly relevant for interactions involving fundamental waves at the telecommunication wavelengths as the cascading leads to the emission of the third harmonic in the visible part of the spectrum, far from the absorption edge of the commonly used quadratic crystals. We investigate the polarizational properties of the emitted harmonics and model them using the concept of effective nonlinearity and taking into account the role of spatial randomness of the antiparallel ferroelectric domains. We support our experimental results with direct simulations of the Maxwell equation in a random quadratic medium.

2. Experimental setup

The experimental setup is shown in figure 1(a). As a light source we used an optical parametric amplifier (TOPAS) generating 150 fs pulses ($\lambda = 1450$ nm, 250 Hz rep rate) with total energy of 0.788 mJ and beam diameter 5 mm (full width at half maximum). The beam was propagating through the half-wave plate (WP) in order to control its polarization, and then after focusing with a 100 mm cylindrical lens (L) was incident at the bi-prism (BP). Bi-prism converted the incoming beam into two symmetric beams which then intersected at the angle of $2 \times \alpha = 2 \times 2.344^\circ$ inside the SBN crystal with a random distribution of the antiparallel ferroelectric domains. This random domain pattern is illustrated in figures 1(b) and (c). These graphs were obtained by using the SH nonlinear microscopy [16] and depict the domain structure as seen along (figure 1(b)) and perpendicular (figure 1(c)) to the optical axis which coincides with the longer dimension of the domains. In the experiment, the orientation of the crystal was

such that both beams propagated in the X - Z plane, close to its crystallographic X -axis and nearly perpendicularly to the longer dimension of the domains (coinciding with the optical axis), as illustrated in figure 1(a). Interaction of these two beams inside the crystal resulted in the emission of second- and third-harmonic waves whose far field in the forward direction was recorded using a CCD camera located behind the SBN crystal.

In the experiment the input polarization of the fundamental beam was varied from ordinary to extraordinary by adjusting the WP. Band-pass filters (not shown) placed in front of the CCD camera were used to separate the fundamental beam and its harmonics so that their corresponding spatial intensity distribution could be separately recorded. In addition, a polarizer (not shown) located behind the SBN sample enabled measurements of the ordinary and the extraordinary components of the generated harmonics.

3. Experimental results

When the two intersecting linearly polarized fundamental beams propagate in the sample of the SBN crystal, the nonlinear interaction in the crystal leads to the emission of both second and third harmonics. Unlike our previous works with the fundamental wavelength of $1.064 \mu\text{m}$ or $0.8 \mu\text{m}$ when the third harmonic could not be observed since it was above the absorption edge of the medium, here it was in visible part of the spectrum and its intensity was high enough to be measured and characterized. Because of the randomness the overall generated signals were rather weak. The total emitted power of the second and third harmonics reached $35 \mu\text{W}$ and 16nW , respectively. A typical harmonic intensity pattern observed on the screen is depicted in figure 2. It consists of seven lines oriented perpendicularly to the optical (z) axis of the crystal. The three lines denoted as SH1, SH2 represent the second harmonic while the four lines marked as TH1 and TH2 represent the third-harmonic waves. The two second-harmonic signals (denoted as SH2) represent waves emitted due to the interaction of fundamental photons from each constituent fundamental beam (FW). On the other hand, the central line (SH1) is a result of interaction of fundamental photons from

Figure 2. Experimentally recorded planar emission of the second (SH) and third (TH) harmonics.

both FWs; hence, it represents the autocorrelation signal of incoming pulses [11]. Figure 3(a) illustrates the phase-matching condition for this process in the X - Z plane. In this case $\vec{G}_1$ denotes one of the reciprocal vectors provided by the randomness. Figure 3(b) depicts the phase matching in the three-dimensional (3D) setting. The number of differently oriented reciprocal vectors $\vec{G}$ located on an arc defined by the magnitude of the wave vector ($\vec{K}_{2\omega}$) of the SH reflects the planar emission of SH forming a vertical line on the screen. When such generated SH photons interact with photons from the fundamental beams via the sum-frequency mixing, the outcome is cascaded third-harmonic generation. Due to the presence of two non-collinear fundamental beams

and three SH signals, the third-harmonic emission acquires the four-line pattern. Similar to SH generation, the sum-frequency mixing also utilizes the reciprocal wave vectors $\vec{G}$ to fulfil its corresponding phase-matching conditions. In typical fabricated structures the simultaneous phase matching of two such processes would require careful design of the ferroelectric domain pattern and would work for only one set of frequencies. Here, the randomness ensures that there are always reciprocal vectors available which can phase match practically all quadratic processes for the broad spectrum of the fundamental waves. The emission of the four third-harmonic lines can be explained by considering again the respective phase-matching conditions of the underlying wave-mixing mechanism. In particular, the two lines denoted as TH2 are formed by the interaction of the fundamental beam with its corresponding second harmonic (SH2). On the other hand, the third-harmonic signal marked as TH1 is emitted via two different processes which are illustrated in figure 3(c). The first one involves the fundamental wave $\vec{K}_{1\omega}$, the SH signal $\vec{K}_{2\omega}$ originating from the second fundamental wave $\vec{K}'_{1\omega}$ and the reciprocal wave vector $\vec{G}_4$. The resulting third-harmonic signal is emitted in the direction indicated by the vector $\vec{K}_{3\omega}$. However, this TH signal will also receive a contribution originating from the interaction of the auto-correlation SH signal (with the wave vector $\vec{K}_{2\omega}$) with one of the fundamental wave $\vec{K}'_{1\omega}$ employing the reciprocal vector $\vec{G}_5$. The graph in figure 3(d) illustrates the full 3D character of the interaction leading to the emission of the third-harmonic signal in a form of the line TH1. As can be seen in this figure, the TH emission in a particular direction is due to the contributions of the SH signals in all directions. Also a single SH signal will contribute to the TH emission in different spatial directions. An experimentally recorded image of the emission lines (see figure 2) shows that the spatial (vertical) extent of the third harmonic is significantly larger than that of the SH. This is a consequence of the fact that in each of the quadratic processes such as SHG or

Figure 3. Diagrams illustrating the phase matching-scenarios for second- (a and b) and cascaded third-harmonic (c and d) generation in a random SBN crystal in the presence of two fundamental beams. $\vec{G}_1 \dots \vec{G}_8$ represent reciprocal vectors provided by the randomness of the nonlinearity. See the text for details.

Figure 4. Intensity of the second- (left) and third-harmonic (right) signals as a function of the polarization angle of the input fundamental beams. Filled and open circles denote the experimental points for the central (SH1,TH1) and peripheral (SH2,TH2) harmonic signals, respectively. The solid line represents the theoretical fit (equations (1)–(4)). The red and blue colours refer to the extraordinary and ordinary components of the corresponding harmonics.

sum-frequency mixing the randomness of the nonlinearity results in delocalized emission of the generated frequency [17–19]. As the third harmonics is formed via cascading of two quadratic processes its spatial distribution is broader than each of the constituent interactions.

The strength of the nonlinear interaction in quadratic media is determined by the effective nonlinearity which itself is a function of polarization of the interaction waves. Hence, by studying the polarization characteristics of the emitted harmonics one can extract information about details of the particular interaction process. In the experiment we varied the azimuthal angle θ (θ is the angle with respect to the y-axis of the sample) of the linearly polarized fundamental wave from 0° to 360° , while measuring the power of the extraordinary and ordinary components of the emitted harmonics. The results of these measurements are shown in figure 4, where the experimental data are represented by filled and open circles for the central (SH1,TH1) and peripheral (SH2,TH2) harmonic signals, respectively. Figures 4(a) and (b) show the dependence of the ordinary and extraordinary components of the SH and TH on polarization of the fundamental beam. The plots show that the polarization properties of the SH and TH beams do not depend on the particular emission directions. This is because the angle between two fundamental beams is relatively small.

The solid lines in figure 4 represent the theoretically predicted dependence. It has been obtained by considering all possible processes contributing towards the second- and third-harmonic signals. Let us denote by O_1 and E_1 (O'_1 and E'_1) the ordinary and extraordinary components of the two fundamental waves and by O_j and E_j the ordinary and extraordinary components of second ($j = 2$) and third ($j = 3$) harmonics, respectively. Then, for arbitrary polarized fundamental beams, the second and third harmonics will contain ordinary and extraordinary components. In the case of e.g. SH1 second-harmonic generation, its extraordinary component is formed via the following two processes:

$$E_1 E'_1 \rightarrow E_2 \quad \text{and} \quad O_1 O'_1 \rightarrow E_2.$$

On the other hand, the ordinary component of the same SH signal is obtained via the following interaction:

$$E'_1 O_1 \rightarrow O_2 \quad \text{and} \quad E_1 O'_1 \rightarrow O_2.$$

The third harmonic is formed due to mixing of photons from the fundamental and SH beams. In particular, for the light emitted into the TH1 line, its extraordinary component is created via the following processes:

$$\begin{aligned} E_1 E_{2\text{SH}2} &\rightarrow E_3 & \text{and} & & O_1 O_{2\text{SH}2} &\rightarrow E_3 \\ E'_1 E_{2\text{SH}1} &\rightarrow E_3 & \text{and} & & O'_1 O_{2\text{SH}1} &\rightarrow E_3 \end{aligned}$$

while the ordinary component appears due to the following interaction:

$$\begin{aligned} E_1 O_{2\text{SH}2} &\rightarrow O_3 & \text{and} & & O_1 E_{2\text{SH}2} &\rightarrow O_3 \\ E'_1 O_{2\text{SH}1} &\rightarrow O_3 & \text{and} & & O'_1 E_{2\text{SH}1} &\rightarrow O_3. \end{aligned}$$

In an analogous way one can identify all remaining processes contributing towards the SH2 and TH2 emissions. We have shown earlier [10, 15] that unlike homogeneous media where all these processes contribute coherently to the total amplitude of the generated harmonic, the randomness of nonlinearity induced by the multi-domain structure makes these constituent processes mutually incoherent. Therefore one can show that the intensities of the second and third harmonics depend on the polarization angle θ of the fundamental beam as follows:

$$I_{(2\omega)}^{(e)} = I_{(\omega)}^2 (\sin^4 \theta + R_{21} \cos^4 \theta) \quad (1)$$

$$I_{(2\omega)}^{(o)} = I_{(\omega)}^2 (R_{22} \cos^2 \theta \sin^2 \theta) \quad (2)$$

$$I_{(3\omega)}^{(e)} = I_{(\omega)}^3 (\sin^6 \theta + R_{31} \cos^4 \theta \sin^2 \theta) \quad (3)$$

$$I_{(3\omega)}^{(o)} = I_{(\omega)}^3 (\cos^6 \theta + R_{32} \cos^2 \theta \sin^4 \theta), \quad (4)$$

where R_{2j} ($j = 1, 2$) and R_{3k} ($k = 1, 2$) denote the strengths of the constituent nonlinear processes and are determined by the corresponding effective nonlinearity [11]. It is worth noting that the effective nonlinearity for all constituent processes depends not only on the material parameters, polarization and geometry of interaction but is also affected by the disorder as has been shown by Le Grand *et al* [20]. We have recently extended this theory to cascaded processes [21]. It appears that disorder generally weakens the strength of the quadratic interaction and its effect depends on the phase mismatch of the process. In fitting the theoretical formulae to the

experimental data we used the following values for the R_{2j} and R_{3k} parameters: $R_{21} = 0.75$, $R_{22} = 4.10$ and $R_{31} = 0.25$, $R_{32} = 1.40$ for the harmonic lines SH1 and TH1; $R_{21} = 0.82$, $R_{22} = 4.20$ and $R_{31} = 0.04$, $R_{32} = 1.31$ for the harmonic lines SH2 and TH2;

Finally, we investigated numerically the second- and third-harmonic generation in disordered quadratic media with two intersecting fundamental beams with Gaussian spatial and temporal profiles. Starting from Maxwell's equations and considering monochromatic stationary waves we can write the generic and well-known equation describing the electromagnetic interaction that for the transverse-electric case, for example, holds as

$$\nabla^2 \mathbf{E}_j - k_{0j}^2 n_j^2 \mathbf{E}_j = \mu_0 \omega^2 \mathbf{P}_{\text{NL},j},$$

where $k_{0j} = \omega_j/c$, ω_j is the frequency, n_j is the index of refraction and $j = \text{FB1, FB2, SH, TH}$ discriminates for the two noncollinear fundamental beams, second and third harmonic respectively. The Gaussian fundamental beams, assumed to be undepleted, are tuned at 1450 nm, are 24 μm in diameter and impinge at the nonlinear medium at an internal angle $\alpha = \pm 2.35^\circ$ with respect to the X axis. The second- and third-harmonic nonlinear polarizations can be simply expressed as

$$\begin{aligned} \mathbf{P}_{\text{NL,SH}} &= \epsilon_0 \chi^{(2)}(x, y) [\mathbf{E}_{\text{FB1}}^2 + \mathbf{E}_{\text{FB2}}^2 + 2\mathbf{E}_{\text{FB1}}\mathbf{E}_{\text{FB2}}] \\ \mathbf{P}_{\text{NL,TH}} &= 4\epsilon_0 \chi^{(2)}(x, y) [(\mathbf{E}_{\text{FB1}} + \mathbf{E}_{\text{FB2}})\mathbf{E}_{\text{SH}}], \end{aligned}$$

where we neglected the cubic nonlinearity and did not impose any constrain on the 2D spatial distribution of the $\chi^{(2)}$ nonlinearity. The equations were solved with a standard 2D finite-element method considering several realizations of Gaussian distributions of the nonlinear domain pattern. As example, in figure 5 we report the case of domain distribution with 1 μm mean value and 0.15 μm standard deviation. Plots in figure 5 depict the far-field intensity at the exit surface of the medium of both second- and third-harmonic signals. The different emission angles are due to the noncollinear phase-matching conditions and are in good agreement with the above discussion. For the peripheral emission lines, the SH and TH overlap as in the experiment while for the centrally emitted (TH1) third harmonic, the simulated emission angle is $\beta = 1.70^\circ$, corresponding very well to the measured angle of 1.75° . It is clear from these plots that the signals corresponding to the noncollinear interactions are always stronger than those originating from the collinear interaction. This effect, also clear from the experimental data, results from the fact that the corresponding effective second-order nonlinearity is at least four times stronger in the former case [11]. Varying the parameters of the domain distribution (i.e. mean value and standard deviation) does not affect the positions of the peaks but only changes the relative emitted intensity, since it will only change the number of $\vec{G}$ vectors available to satisfy that particular phase-matching relation.

4. Conclusions

In conclusion, we studied the second- and third-harmonic generation in a quadratic crystal with disordered ferroelectric domains via mixing of two fundamental beams. The

Figure 5. Numerically simulated far-field intensity of the second (red-dashed) and third (blue-solid) harmonics generated via interaction of two fundamental waves in the SBN crystal with random domain distribution.

nonlinear interaction leads to the second and third harmonics being emitted in a form of multiple lines whose positions are determined by the corresponding phase-matching condition associated with particular constituent interaction mechanism. We analysed the polarizational properties of the emitted harmonics and showed that they can be reproduced theoretically by considering effective second-order nonlinearities and taking into effect the randomness of the system which leads to incoherent character. Our experimental results agree well with the theoretical model incorporating the effect of nonlinearity disorder in the parametric wave interaction.

Since the second- and third-harmonic emission patterns depend on the randomness of the domain distribution they can be used to characterize the degree of randomness of the domain pattern in quadratic crystals. Moreover, simultaneous polarization measurements of SH and TH may be employed to determine the various nonlinear coefficients of the medium. In addition, as the TH1 third-harmonic signals represent actually the third-order correlation of the input pulses they can be used, together with the autocorrelation SH1 signal, in short pulse characterization and monitoring.

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