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MODEL STUDY OF THE CONDITIONS OF PHASE MATCHING FOR THz GENERATORS OF FORWARD AND BACKWARD WAVES IN GaSe CRYSTALS

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A comparative model analysis has been carried out for the first time of the phase matching conditions for generators of forward and backward terahertz waves in pure and highly sulfur-doped ($GaSe_{1-x}S_x$) *crystals of* GaSe*. Conservation of the phase matching conditions up to the maximum possible sulfur content x = 0.5 has been demonstrated. It has been established that doping makes it possible to partially overlap the range accessible to generators based on* GaSe *crystals. The figure of merit of sulfur-doped crystals has been determined.*

Keywords: nonlinear optics, phase matching, THz, $GaSe_{1-x}S_x$, backward-wave generator.

INTRODUCTION

A small number of works have been dedicated to the study of generation of backward waves by methods of parametric frequency conversion (PFC) in nonlinear crystals (NCs) in general and in GaSe NC in particular [1–3]. According to [4], parametric generation of backward waves (PGBW) requires a huge birefringence and pump intensity, and absence of fundamental absorption bands between the generated wavelengths of the signal and idler emission bands. On the other hand, PGBW can, in principle, be realized without the use of feedback (resonators) and anti-reflection coatings, the advantages of the use of which are ruled out by the fact that the idler wave propagates in the opposite forwardion to the pump beam. This being the case, because of the large birefringence and the corresponding large walk-off angle, achieving acceptable gain requires the use of long pump pulses. The enumerated rigid conditions of generation lead to a narrow line width of radiation from a PGBW in comparison with parametric generation of forward waves (PGFW). The very real possibility of creating cavity-free parametric generators with narrow line width has given rise to increased interest in PGBW.

The most favorable situation for the realization of PGBW should be expected from generation of a terahertz (THz) idler wave. In this case, the insignificant length of the wave vector (in the limit, the frequency of the idler wave is near zero) makes it possible to radically lower the requirements on the magnitude of the birefringence and make them achievable. This follows from the law of conservation of energy (one of the phase matching (PM) conditions, which ensures high efficiency of PFC) $h\nu_p = h\nu_s + h\nu_i$, where h is Planck's constant and ν_p, ν_s, and ν_i are the respective values of the frequency of pump radiation and the signal and idler waves. If the wavelengths of pump radiation and signal wave are close, then the required values of the refractive indices for PM will also be close in values, i.e., the value of the birefringence required to satisfy the PM conditions will be low. To pump PGBW, it is preferable to use long-wavelength (≥5 μm) sources, which, by virtue of the small difference in the energy of the pump and idler-wave photons,

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will ensure high potential efficiency of PFC with respect to its energy characteristics. Amongst known NCs, the requirements on the transmission spectrum and the magnitude of the birefringence for PGBW are satisfied to the greatest degree by nonlinear GaSe crystals, which are characterized by the transparency range 0.62–20 and ≥50 μm and $B = 0.35$ in the mid-IR and 0.79 in the THz range [5]. As a pump source for PGBW, greatest interest has been forwarded at using CO_2 lasers (λ = 9.2–10.8 μm), which are the most advanced in technological terms. There is also an interest in analyzing the characteristics of generation of THz radiation by the method of difference frequency generation (DFG) on widely used solid-state laser systems based on Nd:YAG lasers (λ = 1.064 μm).

The main drawback of pure layered GaSe nonlinear crystals – their extraordinarily low mechanical properties (almost zero hardness on the Mohs scale and ease of cleavage) – has held up their application in devices of applied spectroscopy, but it is eliminated to a significant degree by doping with isovalent impurities. The best result is obtained by doping with sulfur, which makes it possible to effectively control the transmission spectrum and PM conditions, and also significantly increases the laser damage threshold (by as much as fivefold for an optimum doping level) [6–8]. The possibilities of building forward- and backward-wave generators based on doped GaSe crystals, along with their potential characteristics, have not been investigated.

In this work we have investigated the comparative model possibilities of satisfying the PM conditions in order to create PGBW and PGFW based on GaSe and $GaSe_{1-x}S_x$ NCs pumped by a CO_2 laser, and also DFG on Nd:YAG laser radiation and its Raman-shifted radiation at 1.180 μm [9], with the figure of merit of the crystals serving as the criterion of potential PFC efficiency.

BASIC ANALYTICAL EXPRESSIONS AND CALCULATIONAL ALGORITHMS

The PM conditions in GaSe NC were calculated using adequate dispersion equations from a number of known versions [10,11] for waves of ordinary (o) and extraordinary (e) polarization:

$$n_o(\lambda) = \sqrt{7.443 + \frac{0.050}{\lambda^2} + \frac{0.0186}{\lambda^4} + \frac{0.0061}{\lambda^6} + \frac{3.1485\times\lambda^2}{\lambda^2 - 2194}},$$
$$n_e(\lambda) = \sqrt{5.760 + \frac{0.3879}{\lambda^2} - \frac{0.2288}{\lambda^4} + \frac{0.1223}{\lambda^6} + \frac{1.8550\times\lambda^2}{\lambda^2 - 1780}}. \quad (1)$$

These equations are also suitable for use in the THz range [7]. Estimates of the PM angles in $GaSe_{1-x}S_x$ NC were calculated according to Eqs. (1) and data of forward measurements of the dispersion properties of $GaSe_{0.6}S_{0.4}$ NC [12], approximated in the form of the dispersion equations

$$n_o^2 = 9.550489 + 0.303723/(\lambda^2 - 0.054331) + 3738.138/(\lambda^2 - 1408.38),$$
$$n_e^2 = 97.335355 + 0.247335/(\lambda^2 - 0.037580) + 2580.856/(\lambda^2 - 1268.56). \quad (2)$$

The dispersion properties for NCs of intermediate composition were determined by an algorithm proposed in [13]. The PM conditions for generation of THz forward and backward waves and difference-frequency waves were calculated using standard expressions having respectively the following forms for conversions via type-I (e-oo) three-wave interactions:

$$\frac{n_e(\lambda_p,\theta)}{\lambda_p} = \frac{n_o(\lambda_s)}{\lambda_s} \pm \frac{n_o(\lambda_i)}{\lambda_i}, \text{ (PFC)}, \quad (3)$$

$$\frac{n_o(\lambda_p)}{\lambda_p} - \frac{n_o(\lambda_s)}{\lambda_s} = \pm\frac{n_e(\lambda_i,\theta)}{\lambda_i}, \text{ (DFG)}, \quad (4)$$

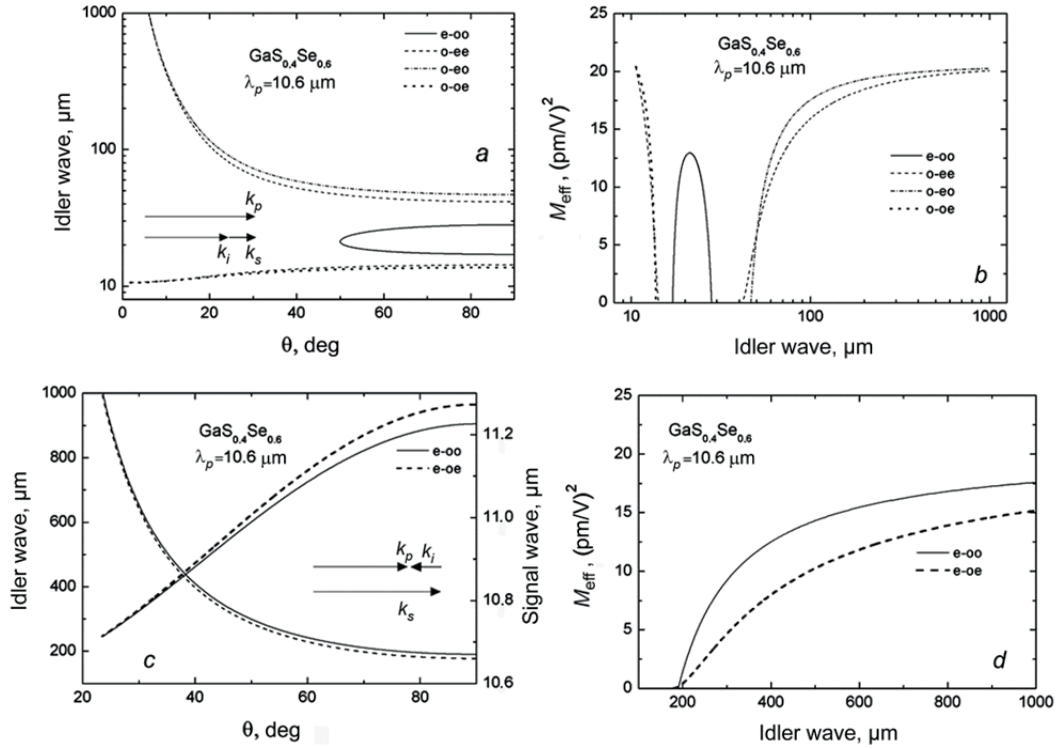

Fig. 1. PM conditions (*a*, *c*) and figure of merit (*b*, *d*) for PGFW (*a*, *b*) and PGBW (*c*, *d*). The legends identify the corresponding NCs, the types of three-wave interactions, and show the vector schemes of the corresponding generation processes; k_p, k_s, and k_i are the wave vectors of the pump, signal, and idler waves, respectively.

$$\frac{1}{\lambda_p}=\frac{1}{\lambda_s}+\frac{1}{\lambda_i},\quad n_e(\lambda,\theta)=\frac{n_o(\lambda)\,n_e(\lambda)}{\sqrt{n_e^2(\lambda)\cos^2(\theta)+n_o^2(\lambda)\sin^2(\theta)}}, \tag{5}$$

where $\lambda_{p,s,i}$ are respectively the wavelengths of the pump, signal, and idler waves. For a comparative estimate of the potential efficiency of PFC, we calculated the effective figure of merit of the NCs $M_{eff} = d_{eff}^2/(n_p \cdot n_s \cdot n_i)$, where n_p, n_s, and n_i are the refractive indices for the corresponding waves. The effective second-order nonlinear susceptibility coefficient d_{eff} for e → o + o, e → o + e, e → e + e, and o → o + o types of interactions was determined respectively using the expressions $d_{eff} = -d_{22}\cos\theta\sin 3\varphi$, $d_{eff} = d_{22}\cos^2\theta\cos 3\varphi$, $d_{eff} = d_{22}\cos^3\theta\sin 3\varphi$, and $d_{eff} = -d_{22}\cos 3\varphi$. The value of d_{22} for GaSe crystals in the THz range was taken to be equal to 24.3 pm/V [14]. The decrease in d_{22} with sulfur doping was determined using a technique suggested by Feng *et al.* [15]. In particular, $d_{22}(GaSe_{0.6}S_{0.4})$ was determined to be equal to $0.8d_{22}$(GaSe) or 19.44 for the THz range.

RESULTS OF CALCULATIONS

Birefringence in the THz range is very high (as high as 0.79 in GaSe NC), which gives wide possibilities for the realization of PM conditions both for PGBW and PGFW. Note that the large drop in the birefringence with increase in the doping level, established in the calculations, makes it possible to control the PM angle at the stage of processes of technological growth of the NCs and thereby to optimize the conditions of generation. Figures 1 and 2 display examples of calculated dependences of PM conditions and values of the figure of merit for the instances of PGBW, PGFW, and DFG under consideration.

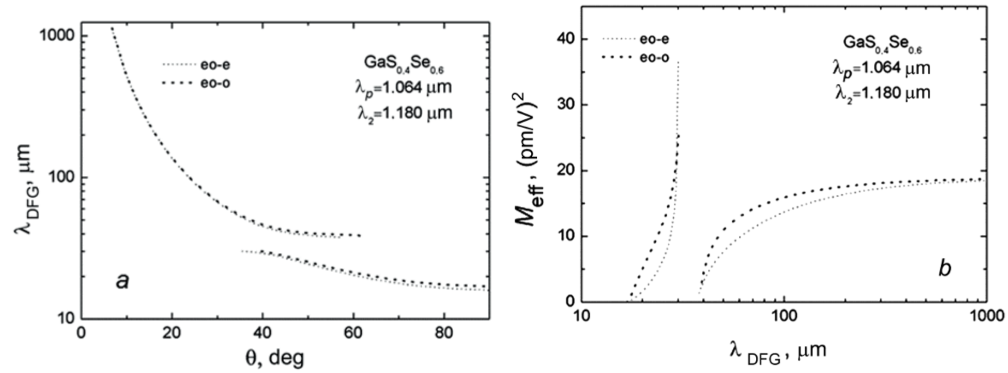

Fig. 2. PM conditions (*a*) and figure of merit (*b*) for difference frequency generation of the backward wave. The legends identify the corresponding NCs and the types of three-wave interactions.

It can be seen from Fig. 1*a* that in spite of the decrease in birefringence of GaSe crystals with doping by sulfur the realization of parametric generation is still possible up to doping levels close to the limiting value $x = 0.5$. An analogous result was also obtained for DFG. The range of parametric generation based on $GaSe_{1-x}S_x$ NC shifts with doping toward shorter wavelengths, enabling a partial overlap of the gap in the spectrum of parametric generators based on GaSe NC. The figure of merit of these crystals varies insignificantly: a decrease in the nonlinear coefficient is partially compensated by a decrease in the values of the refractive indices. This gives reason to assume that the efficiency of actual parametric generators of backward waves can be higher due to absence of the need to use dielectric mirrors and anti-reflection coatings, which introduce additional optical losses.

CONCLUSIONS

We have carried out the comparative model analysis here for the first time of the phase matching conditions for generation of terahertz forward and backward waves in nonlinear GaSe crystals doped with sulfur. We have demonstrated the possibility of realizing parametric generators of light and difference frequency generators of forward and backward waves up to the maximum possible value of the sulfur content $x = 0.5$ in $GaSe_{1-x}S_x$ crystals. We have shown that doping makes it possible to partially overlap the range of generation accessible to generators based on pure GaSe crystals. The spectral dependences of the figure of merit of crystals doped with sulfur have been determined.

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