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Study of the second harmonic generation and optical rectification in a cBN crystal

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***Abstract*. Cubic boron nitride (cBN) – a kind of an artificial (synthetic) crystal with the band gap of ~6.3 eV, which has the zinc blende structure and the $\bar{4}3m$ symmetry, is studied. The optical rectification is obtained and the second harmonic generation (SHG) is observed in the cBN crystal for the first time by using a 1064-nm *Q*-switched Nd : YAG laser. The green light at 532 nm from the cBN sample can be seen with a naked eye.**

Keywords: *cBN, optical rectification, second harmonic generation.*

1. Introduction

Cubic boron nitride (cBN) is a kind of an artificial (synthetic) crystal with the band gap of about 6.3 eV. The heat conductivity of the cBN crystal is 13 W cm^{-1} K^{-1}, its heat resistance is very high, and it can endure 2000 °C [1]. This crystal is transparent in the near-UV, visible and near-IR spectral regions. Therefore, the wide band-gap semiconductor cBN crystal can find wide applications in high-frequency, heat-resistant and high-power electronic and UV optoelectronic devices.

Because the cBN crystal has the zinc blende structure and $\bar{4}3m$ symmetry, it should possess a linear electrooptic (EO) effect. We have studied the EO effect in this crystal by using a modifying transverse EO modulator, and have calculated its EO coefficient [2]. However, we have failed so far to find any reports devoted to the nonlinear optical properties of the cBN crystal in the literature.

In 1961, Franken and co-workers [3] observed for the first time the optical frequency doubling. This phenomenon was called the second harmonic generation (SHG). In 1962, Armstrong and co-workers [4] predicted the effect of optical rectification, which was experimentally observed by Bass and co-workers [5] in the same year.

In this paper, we report for the first time the results of the SHG and optical rectification in the cBN crystal.

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2. Theoretical derivation of equations for the SHG and the optical rectification in the ⟨111⟩ plane of the cBN crystal

Due to the zinc blende structure and $\bar{4}3m$ symmetry of the cBN crystal, its second-order nonlinear susceptibility tensor has the form

$$\begin{pmatrix} 0 & 0 & 0 & xyz & xyz & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & xyz & xyz & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & xyz & xyz \end{pmatrix} \quad (1)$$

in the principal coordinate system of the cubic crystal [6].

Only one element in the tensor is independent. Let us assume that $\chi = \chi_{xyz}$ and

$$\boldsymbol{E}(t) = \boldsymbol{E}_0 \cos(\omega t - \varphi) + \frac{1}{2}\boldsymbol{E}\exp[-\mathrm{i}(\omega t - \varphi)] + \frac{1}{2}\boldsymbol{E}\exp[\mathrm{i}(\omega t - \varphi)] \quad (2)$$

for the field of the fundamental frequency ω, where $\boldsymbol{E}(\omega) = \frac{1}{2}\boldsymbol{E}_0 \exp(\mathrm{i}\varphi)$.

The relation between the second-order nonlinear polarisation $\boldsymbol{P}^{(2)}(t)$ and $\boldsymbol{E}$ has the form

$$\boldsymbol{P}^{(2)}(t) = \sum_{m,n} \varepsilon_0 \chi^{(2)}(\omega_m, \omega_n) : \boldsymbol{E}(\omega_m)\boldsymbol{E}(\omega_n) \times \exp[-\mathrm{i}(\omega_m + \omega_n)t]. \quad (3)$$

Then, for the optical rectification, we have

$$\boldsymbol{P}^{(2)}(0) = 2\varepsilon_0 \chi^{(2)}(\omega, -\omega) : \boldsymbol{E}(\omega)\boldsymbol{E}^*(\omega), \quad (4)$$

and for the second harmonic generation, we have

$$\boldsymbol{P}^{(2)}(2\omega) = \varepsilon_0 \chi^{(2)}(\omega, \omega) : \boldsymbol{E}(\omega)\boldsymbol{E}(\omega). \quad (5)$$

According to these relations, the SHG and optical rectification occur simultaneously. In this case the intensity of optical rectification is twice as large as that of the SHG.

An ideal cBN crystal has an isotropic octahedron structure, however, most crystals synthesised by us have an anisotropic octahedron structure whose facets are all {111} planes. In these crystals the top and bottom planes are larger than the side planes, and they cannot intersect at right angles. The real shape of the cBN crystal in the experiment is shown in Fig.1. The upper cBN sample shown in Fig.1, whose opposite planes were parallel and approximately equal, was used in the experiment. According to the real shape of the cBN crystal, the incident beam can be only perpendicular or parallel to the $\langle 111 \rangle$ plane.

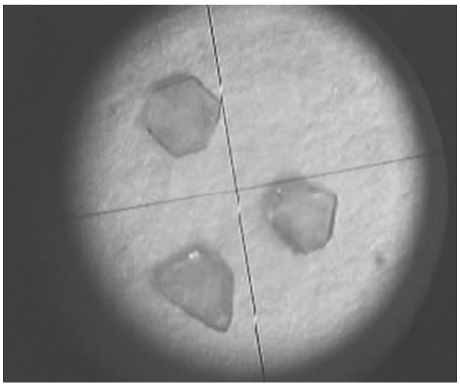

Figure 1. Microscope photograph of cBN samples. The top left corner cBN sample with the area of $S = 0.1429$ mm^2 and length of the facet of $L = 0.06$ mm was used in our experiment.

2.1 The incident beam perpendicular to the $\langle 111 \rangle$ plane

Let us assume that the angle between the polarisation orientation of the incident beam and the $[11\bar{2}]$ orientation in the $\langle 111 \rangle$ plane is θ. In the $\langle 111 \rangle$ plane, the orientations $[11\bar{2}]$ and $[1\bar{1}0]$ are perpendicular to each other, and, hence, the equations

$$E_{[11\bar{2}]} = E_0 \cos\theta,$$
$$E_{[1\bar{1}0]} = E_0 \sin\theta \qquad (6)$$

are valid.

The three components of the vector $\boldsymbol{P}^{(2)}(t)$ in the principal coordinate system have the form

$$P_x^{(2)}(0) = -\frac{\sqrt{3}}{6}\varepsilon_0\chi\left(\frac{\sqrt{3}}{3}\cos^2\theta - \sin\theta\cos\theta\right)E_0^2,$$

$$P_y^{(2)}(0) = -\frac{\sqrt{3}}{6}\varepsilon_0\chi\left(\frac{\sqrt{3}}{3}\cos^2\theta + \sin\theta\cos\theta\right)E_0^2, \qquad (7)$$

$$P_z^{(2)}(0) = \frac{1}{4}\varepsilon_0\chi\left(\frac{1}{3}\cos^2\theta - \sin^2\theta\right)E_0^2.$$

Let us expand $\boldsymbol{P}^{(2)}(0)$ into components $\boldsymbol{P}_\parallel$ and $\boldsymbol{P}_\perp$ in the $\langle 111 \rangle$ plane, $\boldsymbol{P}_\parallel$ being parallel to the $\langle 111 \rangle$ plane, and $\boldsymbol{P}_\perp$ – perpendicular to it. We will locate in the $\langle 111 \rangle$ plane the coordinate system with axes corresponding to $[11\bar{2}]$ and

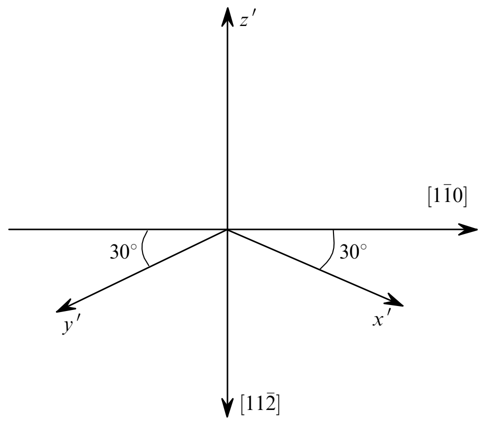

Figure 2. The scheme of the polarisation in the $\langle 111 \rangle$ plane.

$[1\bar{1}0]$ orientations (Fig. 2). The projections of x, y, z axes in the $\langle 111 \rangle$ plane are x', y', z' axes, respectively. Then, let us denote the projections of components $P_x^{(2)}(0)$, $P_y^{(2)}(0)$, and $P_z^{(2)}(0)$ on the $\langle 111 \rangle$ plane as $P_{x'}^{(2)}(0)$, $P_{y'}^{(2)}(0)$, $P_{z'}^{(2)}(0)$.

Expressions for the latter have the form

$$P_{x'}^{(2)}(0) = -\frac{\sqrt{2}}{6}\varepsilon_0\chi\left(\frac{\sqrt{3}}{3}\cos^2\theta - \sin\theta\cos\theta\right)E_0^2,$$

$$P_{y'}^{(2)}(0) = -\frac{\sqrt{2}}{6}\varepsilon_0\chi\left(\frac{\sqrt{3}}{3}\cos^2\theta + \sin\theta\cos\theta\right)E_0^2, \qquad (8)$$

$$P_{z'}^{(2)}(0) = \frac{\sqrt{6}}{12}\varepsilon_0\chi\left(\frac{1}{3}\cos^2\theta - \sin^2\theta\right)E_0^2.$$

Therefore,

$$P_{[1\bar{1}0]}^{(2)}(0) = P_{x'}^{(2)}(0)\cos 30^\circ - P_{y'}^{(2)}(0)\cos 30^\circ$$
$$= \frac{\sqrt{6}}{6}\varepsilon_0\chi E_0^2 \sin\theta\cos\theta, \qquad (9)$$

$$P_{[11\bar{2}]}^{(2)}(0) = P_{x'}^{(2)}(0)\sin 30^\circ + P_{y'}^{(2)}(0)\sin 30^\circ - P_{z'}^{(2)}(0)$$
$$= -\frac{\sqrt{6}}{12}\varepsilon_0\chi E_0^2\left(\frac{2}{3}\cos^2\theta - \sin^2\theta\right). \qquad (10)$$

The modules of the components of the polarisation vector parallel and perpendicular to the $\langle 111 \rangle$ plane have the from:

$$P_\parallel^{(2)}(0) = \left[\left(P_{[1\bar{1}0]}^{(2)}(0)\right)^2 + \left(P_{[11\bar{2}]}^{(2)}(0)\right)^2\right]^{1/2}$$
$$= \frac{\sqrt{6}}{72}\varepsilon_0\chi E_0^2\left[\frac{432}{11} - 11\left(\cos 2\theta + \frac{5}{11}\right)^2\right]^{1/2}, \qquad (11)$$

$$P_\perp^{(2)}(0) = \frac{\sqrt{3}}{3}P_x^{(2)}(0) + \frac{\sqrt{3}}{3}P_y^{(2)}(0) + \frac{\sqrt{3}}{3}P_z^{(2)}(0)$$
$$= -\frac{\sqrt{3}}{12}\varepsilon_0\chi E_0^2. \qquad (12)$$

When the incident beam is perpendicular to the ⟨111⟩ plane, the components of the polarisation exciting the SHG wave can be obtained by using expression (5),

$$P_{\parallel}^{(2)}(2\omega) = \frac{\sqrt{6}}{144}\varepsilon_0\chi E_0^2\left[\frac{432}{11} - 11\left(\cos 2\theta + \frac{5}{11}\right)^2\right]^{1/2}, \quad (13)$$

$$P_{\perp}^{(2)}(2\omega) = -\frac{\sqrt{3}}{24}\varepsilon_0\chi E_0^2. \quad (14)$$

2.2 The incident beam parallel to the ⟨111⟩ plane

Let the incident beam be parallel to the ⟨111⟩ plane. We assume that the angle between the polarisation vector of the incident beam and the [111] orientation in the ⟨111⟩ plane is α, and the angle between the direction of the propagation of the incident beam and the $[11\bar{2}]$ orientation is β. In this case,

$$E_{[111]} = E_0 \cos\alpha,$$
$$E_{[1\bar{1}0]} = E_0 \sin\alpha\cos\beta, \quad (15)$$
$$E_{[11\bar{2}]} = E_0 \sin\alpha\sin\beta.$$

In this case the components of the vector polarisation of the optical rectification are obtained from expression (4):

$$P_x^{(2)}(0) = \frac{1}{2}\varepsilon_0\chi E_0^2\left[\frac{1}{3}\cos^2\alpha - \frac{\sqrt{2}}{6}\sin\alpha\cos\alpha(\sin\beta + \sqrt{3}\cos\beta) - \frac{\sqrt{3}}{3}\sin^2\alpha\sin\beta\left(\frac{\sqrt{3}}{3}\sin\beta - \cos\beta\right)\right], \quad (16)$$

$$P_y^{(2)}(0) = \frac{1}{2}\varepsilon_0\chi E_0^2\left[\frac{1}{3}\cos^2\alpha - \frac{\sqrt{2}}{6}\sin\alpha\cos\alpha(\sin\beta - \sqrt{3}\cos\beta) - \frac{\sqrt{3}}{3}\sin^2\alpha\sin\beta\left(\frac{\sqrt{3}}{3}\sin\beta + \cos\beta\right)\right], \quad (17)$$

$$P_z^{(2)}(0) = \frac{1}{2}\varepsilon_0\chi E_0^2\left[\frac{1}{3}\cos^2\alpha + \frac{\sqrt{2}}{3}\sin\alpha\cos\alpha\sin\beta + \frac{1}{3}\sin^2\alpha\left(\frac{1}{3}\sin^2\beta - \cos^2\beta\right)\right]. \quad (18)$$

Then, the polarisation of the optical rectification in the [111] orientation is

$$P_{[111]}^{(2)}(0) = \frac{\sqrt{3}}{3}\left[P_x^{(2)}(0) + P_y^{(2)}(0) + P_z^{(2)}(0)\right] = \frac{\sqrt{3}}{72}\varepsilon_0\chi E_0^{(2)}(5\cos 2\alpha - 1). \quad (19)$$

When the incident beam is parallel to the ⟨111⟩ plane, the polarisation of SHG can be also obtained by formula (5):

$$P_x^{(2)}(2\omega) = \frac{1}{4}\varepsilon_0\chi E_0^2\left[\frac{1}{3}\cos^2\alpha - \frac{\sqrt{2}}{6}\sin\alpha\cos\alpha(\sin\beta + \sqrt{3}\cos\beta) - \frac{\sqrt{3}}{3}\sin^2\alpha\sin\beta\left(\frac{\sqrt{3}}{3}\sin\beta - \cos\beta\right)\right], \quad (20)$$

$$P_y^{(2)}(2\omega) = \frac{1}{4}\varepsilon_0\chi E_0^2\left[\frac{1}{3}\cos^2\alpha - \frac{\sqrt{2}}{6}\sin\alpha\cos\alpha(\sin\beta - \sqrt{3}\cos\beta) - \frac{\sqrt{3}}{3}\sin^2\alpha\sin\beta\left(\frac{\sqrt{3}}{3}\sin\beta + \cos\beta\right)\right], \quad (21)$$

$$P_z^{(2)}(2\omega) = \frac{1}{4}\varepsilon_0\chi E_0^2\left[\frac{1}{3}\cos^2\alpha + \frac{\sqrt{2}}{3}\sin\alpha\cos\alpha\sin\beta + \frac{1}{2}\sin^2\alpha\left(\frac{1}{3}\sin^2\beta - \cos^2\beta\right)\right]. \quad (22)$$

Therefore, the SHG and optic rectification occur independently no matter if the incident beam is perpendicular or parallel to the ⟨111⟩ plane.

3. The experimental measurement of SHG and the optical rectification in the ⟨111⟩ plane of the cBN crystal

According to the theoretical analysis, the polarisations of the SHG and optical rectification are nonzero no matter if the incident beam is perpendicular or parallel to the ⟨111⟩ plane. Therefore, the cBN crystal may produce the SHG and optical rectification effects under the action of high-power laser radiation. In our experiment we observed second harmonic generation and measured the optical rectification signal.

Due to the small size of the cBN crystal, we designed a special structure in which the cBN sample was sandwiched between two conducting glass plates. The conducting glass plates, used as two electrodes, were attached to each other with an insulating glue (Fig. 3).

We used in the experiment a 1064-nm, 700-W Q-swit-

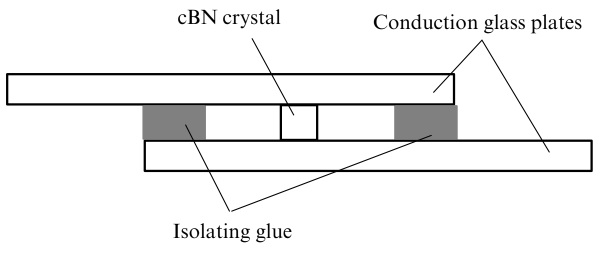

Figure 3. The structure of the sample under study.

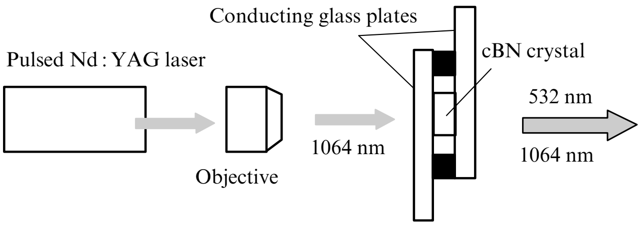

Figure 4. Scheme of the experimental setup for the SHG.

ched Nd : YAG laser with a pulse repetition rate of 2 kHz and a pulse duration of less than 20 ns.

3.1 The SHG in the cBN crystal

The experiment setup is shown in Fig. 4. The incident beam was perpendicular to the ⟨111⟩ plane and the output radiation contained components at 1064 nm and 532 nm. The photograph of the radiation spot of SHG is shown in Fig. 5.

Figure 5. Photograph of the radiation spot of the SHG.

In this experiment we failed to obtain the optical rectification in a pure form because of the possibility to damage the electrodes by the high-power laser radiation.

3.2 Optical rectification in the cBN crystal

The experiment setup is shown in Fig. 6. The incident beam is parallel to the ⟨111⟩ plane. A part of the beam splitted by the beamsplitter is used as a reference signal for the lock-in amplifier. The main part of radiation is focused by the objective, then passes through the cBN crystal and is frequency doubled (see Fig. 5). The voltage of the signal of the optical rectification recorded with the lock-in amplifier was about of $\sim 1\ \mu$V.

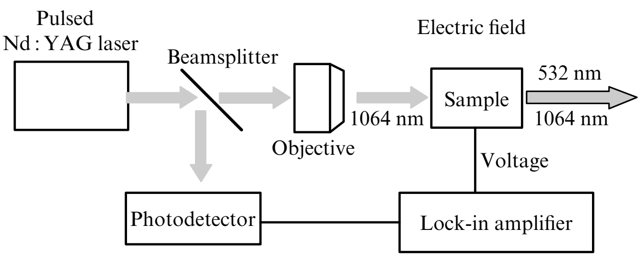

Figure 6. Scheme of the experimental setup to study optical rectification.

4. Conclusions

We have shown theoretically the possibility of SHG and optical rectification in the cBN crystal. We have observed for the first time both these effects in the experiment. Because the cBN crystal is transparent within the whole visible and most of the UV spectra, it is possible to develop an ultraviolet laser by using the SHG of the visible laser. When femtosecond pulses are incident on the crystal, terahertz waves can be obtained due to optical rectification. The research is being conducted at present in both directions.

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