Surface second-harmonic generation in $Sr_{0.6}Ba_{0.4}NbO_3$ with a nonlinear diffusion mechanism

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Surface second-harmonic generation excited by photorefractive surface electromagnetic wave with a diffusion mechanism of nonlinearity has been observed at the surface of the negative c axis of a $Sr_{0.6}Ba_{0.4}NbO_3$(SBN:60) experimentally. The second-harmonic 532 nm wavelength light is generated by 1064 nm laser in a passive guiding manner in the experiment, for the wavelength of the fundamental beam is insensitive to the SBN crystal. The transfer efficiency of surface second-harmonic generation is 1%/W.

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I. INTRODUCTION

Surface second-harmonic generation (SHG) is a useful tool for the investigation of material surface properties and surface processes because of its intrinsic sensitivity to the structure and symmetry of material and interfaces. It is extremely sensitive to the surface structure down to atomic scale, such as surface defects, structural inhomogeneity, and adsorbates, etc.[1–3]

Phase matching is a necessary condition to generate the optical SHG in the noncentrosymmetric crystal. However, as for $Sr_{0.6}Ba_{0.4}NbO_3$ (SBN) crystal it is because it has a relatively small birefringence of the crystal $\Delta n \approx 0.02$, which does not allow for birefringence phase matching for wavelengths lower than 2 μm.[4] Quasiphase-matched SHG has been realized in periodically domain-inversed SBN crystal plate induced by an external electric field in 1997.[5] Lee *et al.* have obtained the SHG in the bulk of SBN by illuminating needlelike ferroelectric domains structure, but it is unsuitable for power application because of its diffuse nature.[6] Later Romero and Jaque apply a resonator to increase the efficiency of the SHG and make the SHG radiate out of the crystal.[7]

The SHG excited by a photorefractive spatial soliton was put forward by Song *et al.* and was implemented in $KNbO_3$ crystal with a drift mechanism. The phase matching can be satisfied easily in a soliton induced waveguide because it occurs among propagation constants of the interacting guided modes, rather than wave vectors in a bulk.[8] A spatial soliton can form in SBN crystal with a drift mechanism. However the SHG excited by the spatial soliton in the bulk of SBN have not been reported. It may be because of the destructive interference formed by the strong diffusion in SBN.

Recently, a new kind of nonlinear surface electromagnetic wave (SEW) called photorefractive surface wave has been predicted and experimentally observed.[9–11] A photorefractive SEW occurs due to the interference between the two waves reflecting into each other from the boundary of the sample and form the Bragg-type grating formed in the sample volume. Then a surface waveguide formed at the interface between SBN and air. As a result, all nonlinear surface optical phenomena may be expected to be strongly enhanced. SHG could satisfy the conditions of phase matching in the surface waveguide induced by the surface waves.[12]

In this paper we report the strongly enhanced PR SSHG in $Sr_{0.6}Ba_{0.4}NbO_3$ crystal excited by PR SEW with diffusion nonlinearity.

II. THE PR SURFACE WAVE

A theoretical explanation of PR SEW solutions of Maxwell equations in a photorefractive medium has been presented.[9–11] Given the c axis of the crystal is along the x axis, when a surface wave with propagation constant β propagates along the z axis. The optical field $E(x,z,t) = E(x)\exp i(wt-\beta z)$ is inserted into the scalar wave equation

$$[(d^2/dx^2 + d^2/dz^2) + \omega^2\varepsilon_o u_0 \varepsilon(x,z)]E(x,z) = 0 \qquad (1)$$

in photorefractive medium ($x>0$). Here in photorefractive crystal (PRC) $\varepsilon = \varepsilon_0 + \delta\varepsilon$,

$$E_{sc}(x) = -\frac{k_B T}{e}\frac{1}{I(x)+I_d}\frac{dI(x)}{dx}, \qquad (2)$$

$$\delta\varepsilon(x) = 2n^4 r(k_B T/e)[dE_{sc}(x)/dx]/E_{sc}(x), \qquad (3)$$

$$\left[\frac{d^2}{dx^2} + 2\gamma k_2\frac{d}{dx} - (\beta^2 - k_2^2)\right]E(x) = 0, \qquad (4)$$

where $I(x)$ is the light intensity. I_d is equivalent dark irradiance. Possible thermal ionization is neglected. $k_1 = 2\pi n_1/\lambda$, $k_2 = \omega(\varepsilon\varepsilon_0\mu_0)^{1/2} = 2\pi n/\lambda$, and $\gamma = k_2 n^2 r k_B T/e$. $\delta n = n - n_1$ the refractive index is n_1 in dielectric and n in PRC, $n_1 < n$. r is the linear electro-optic coefficient.

The solution of Eq. (4) in the case of $k_2^2 - k_2^2\gamma^2 > \beta^2$ is

$$E(x) = \exp(-\gamma k_2 x)\cos(-x\sqrt{k_2^2 - k_2^2 - \beta^2}) \qquad (5)$$

$x>0$, $\beta \ll k_2$. According to this result, a surface waveguide is formed. The photorefractive SEW field penetration depth is

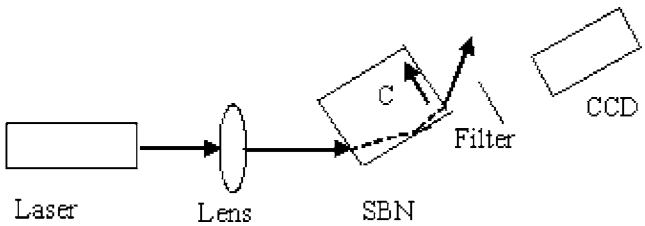

FIG. 1. Setup for observing the generation of surface SHG excited by a PR SEW at the interface between SBN and air with a diffusion mechanism.

$d_z = (\gamma k_0)^{-1} \approx 30\ \mu$m. The PR SEW would propagate in a relative narrow layer at the surface of the negative *c* axis. Then surface SHG can be excited in the waveguide induced by the PR SEW.

III. EXPERIMENT AND RESULTS

The crystal we used in experiment is SBN:60 which has a tungsten-bronze structure. It is ferroelectric with a 4 mm symmetry below the ferroelectric Curie temperature $T_c = 80\,^\circ$C, and Para electric with 4*mmm* above T_c. The dielectric constants are $\varepsilon_{11} = 450$, $\varepsilon_{33} = 900$. The electro-optical coefficients (Pm/V)r_{33} and r_{51} are 420 and 42, respectively.[13]

The experiment setup is shown in Fig. 1. The pure 90-deg-cut poled SBN:60 crystal is $4.62\ \text{mm} \times 6.6\ \text{mm} \times 7.62$ mm cube with two opposing faces perpendicular to the optical axis (*c* axis) and poled along the *c* axis. A 1064 nm *e* polarization laser beam is focused onto the surface of the negative *c* axis by a lens with a focal distance of 4.7 cm. The power of the incident beam is 60 mw. The incident angle to the input face is 44°, accordingly the incident angle to the negative *c* axis face is 71°, where the total reflection occurs. A uniform background illumination is provided by an incoherent beam $\lambda = 650$ nm. The image of the PR SEW and the excited surface SHG at the output face is recorded by a charge coupled camera (CCD).

As the transverse position along the *c* axis of the crystal was imaged, a green light can be seen at the edge of the exit face, which can radiate out. The brightness of SH emission was evident from Fig. 2. Figure 2(a) shows the image of PR SEW at the exit face of the SBN crystal, which is recorded by CCD with an infrared transmitting filter. The slit area on the left is the PR SEW at the crystal edge, which corresponds to the surface of the negative *c* axis in the crystal. The other two spots are images of the cast shadow of incident point at the input face on the output face and the reflected beam at the output face respectively. Figure 2(b) shows the surface SH at the exit face, which is recorded by CCD with a 532 nm bandpass filter. The site of surface SH corresponds to the site of PR SEW. The results indicated that the photorefractive SEW is formed at the surface of the negative *c* axis, by which SH could be excited at the same time and propagates along surface. Figure 3 shows the intensity of the PR SEW and the SH as a function of the transverse position of the crystal corresponding to Fig. 2. The full width at a half maximum of the SH′ peak is equal to 40 μm.

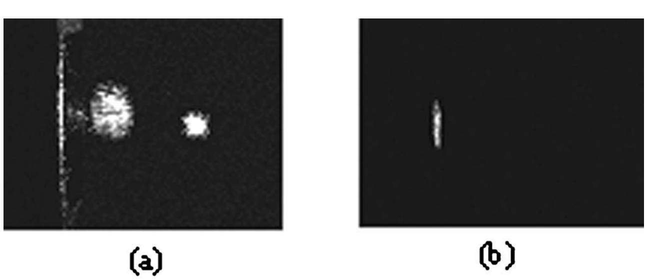

FIG. 2. (a) Shows the image of output face by CCD. The slit area on the left is the intensity of the PR SEW; (b) surface SHG in the SNB crystal. A bandpass filter is used to cut off the effect of the signal beam.

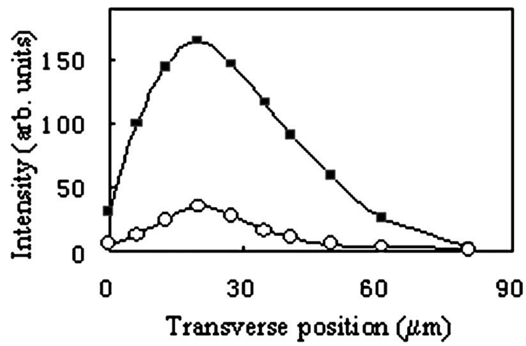

FIG. 3. Shows the intensity of the PR SEW and the SH as a function of the transverse position of the crystal. The curve with black squares represents the intensity of the PR SEW, the curve with blank circles represents the intensity of the SH excited by the PR SEW.

The surface SHG is an extraordinary polarization detected by a polaroid, which is the same with the signal beam. The power of surface SHG was measured by a laser power meter, which is about 37 μw when the intensity of the incident beam is about 60 mw, and the transfer efficiency of surface SHG is about 1%/W of the incident fundamental beam. The power of the PR SEW is about 300 μW in our experiment. Then the transfer efficiency of the surface SHG may reach 13% of the PR SEW.

IV. DISCUSSION

The phase matching conditions for SHG is necessary. In the waveguide induced by nonlinear soliton, the phase matching is further modified as it occurs among propagation constants of the interacting guided modes, rather than wave vectors in a bulk.[8] For the case of surface waveguide induced by PR SEW similar to SHG excited in waveguide by soliton theory there is always a SH waveguide mode phased matched with the fundamental light, which is at a peculiar incident angle.

In the experiment we find that exit angle of SHG is about 44° to the exit face, which is parallel with the incident beam. The SHG can be excited by the fundamental beam in a certain angular region corresponding to the incident beam which is a focused beam with a convergence angle about 3°. It is because upon resonant excitation, the SH waveguide mode will be coupled to many other modes existing in the waveguide at the same frequency. Thus, the SHG can be excited in a certain range of the incident angle, while the phase-matching can be satisfied.

The background illumination to the surface SHG will accelerate the formation of PR SEW as well as the surface SHG guided by the PR SEW. Generally, the formation time is about 50 min without the background illumination. It will reduce to several minutes with a background illumination. However, the time for generation of the soliton is often less than a minute in SBN crystal. That difference may be caused by the difference of the intensity between PRC SEW and soliton in the bulk. The PR SEW originates from partial incident beam, most of which is reflected; but the solitons in bulk media often include whole energy of the incidence light beam. The generation speed of the photoinduced carrier and establishment time of the space charge field are proportional to the light intensity. Thus, a background illumination will decrease the time for the formation of PR SEW as well as the surface SHG.

The PR nonlinearity of SBN crystal is insensitive to a 1064 nm wavelength beam. On the other hand, it is sensitive to a 532 nm wavelength beam. In the experiment the SHG should be followed in a passive guiding manner. The SH beam forms a soliton and guides the fundamental beam in its induced waveguide, which leads to a lower transfer efficiency of the surface SHG. An active method maybe improves the transfer efficiency SHG, in which the interacting fundamental and SH beams is guided by a waveguide which has been written.

V. CONCLUSION

In conclusion, SHG have been observed in a SBN: 60 with diffusion nonlinearity when the wavelength of the incident beam is 1064 nm. The intensity of the surface SHG reaches 37 μw with an incident power of 60 mw. The transfer efficiency of the SHG is about 1%/W to the fundamental beam. The surface SHG can be excited at the interface between SBN and air induced by the PR SEW with diffusion mechanism in a passive method. The phase matching can be satisfied in a certain region corresponding to the angular distribution of the focused fundamental beam. Application of a pulse light source will improve the transfer efficiency. Surface SHG may be applied in nonlinear surface spectroscopy and the surface photonics apparatus.

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