Broadband frequency doubling in unpoled SBN crystals in the thermal focusing regime

Robert Fischer[1], Solomon M. Saltiel[1,2], Dragomir N. Neshev[1], Wieslaw Krolikowski[1], and Yuri S. Kivshar[1]

[1]) *Nonlinear Physics Centre and Laser Physics Centre,*
Centre for Ultrahigh-bandwidth Devices for Optical Systems (CUDOS),
Research School of Physical Sciences and Engineering,
Australian National University, Canberra, Australia
[2]) *Dept. Quantum Electronics, Faculty of Physics, Sofia University, 1164 - Sofia, Bulgaria*

We study experimentally broadband noncollinear second-harmonic generation in unpoled Strontium Barium Niobate crystals. We show the effects of thermal self-focusing on the parametric conversion, including spatial localization of the second-harmonic, increased efficiency, and spectral broadening.

OCIS codes: 190.2620;190.4870;

Naturally disordered ferroelectric domain structure of Strontium Barium Niobate (SBN) crystal allows to phase-match any second-order parametric processes such as second-harmonic generation (SHG) over practically the whole range of the crystals transparency window[1,2]. This property is particularly relevant in the context of ultra-short pulses where a constant efficiency of the nonlinear process over the full pulse spectra is required[3]. In naturally grown SBN this can be achieved without the need of poling or temperature tuning. Moreover, beam alignment in the crystal is very uncritical, an aspect of special importance for many technical applications.

Broadband phase matching (PM) in SBN originates from a random distribution (in size and position) of needle-like anti-parallel ferroelectric domains, all of them orientated along the optical z-axis of the crystal. These domains form a two-dimensional nonlinear structure with a constant linear refractive index but randomly modulated quadratic response, thus providing an infinite pool of grating vectors to fulfill the quasi-phase-matching condition[4]. The second harmonic (SH) radiation is emitted either in the plane or as a cone[2] depending on whether the fundamental beam propagates perpendicularly or along the z-axis of the crystal. While the PM with the random structured SBN overcomes the bandwidth limitation of standard methods like birefringence, periodical poling or mode-matching (in fibers), it goes along with a comparatively low efficiency. Therefore methods to increase the conversion efficiency without compromising on bandwidth will open up possibilities for a wide range of new applications.

In this paper we demonstrate experimentally that thermally-induced self-focusing of the fundamental wave leads to a four-fold increase of the SHG efficiency. We also discuss the impact of this effect on other properties of the SH radiation which include its spatial localization and spectral broadening.

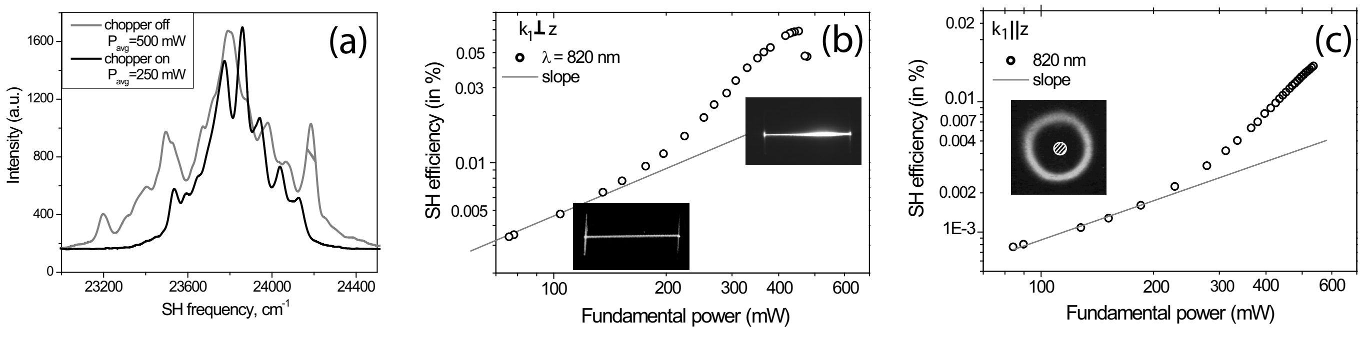

Fig. 1. (a) Spectra of the SH signal for different average power but identical peak intensity. (b,c) Efficiency of the SHG process for (b) plane emission and (c) cone emission as a function of average power of the fundamental beam. Insets in top graph depict the SH intensity distribution emitted from a facet parallel to the (k,z) plane of the crystal at low (b, LP) and high (b, HP) input fundamental power, respectively. The inset in (c) shows a cut through the SH cone, with the dot in the middle blocking the fundamental.

In our experimental setup we use a Ti:Sapphire oscillator as a light source, emitting 150 fs pulses of 6 - 7 nJ energies and an average power of up to 550 mW. The laser beam is focused to a spot size of 74 μm inside the 10 mm long SBN crystal. The conversion efficiency is measured for two different geometries, namely with the fundamental wave vector perpendicular to the z-axis (k⊥z, emission in a plane) and with the fundamental wave vector orientated along the z-axis (k||z, emission in a cone). In the former case, the input polarization is chosen to be parallel to the z-axis. In the latter case, the SHG process is independent of the polarization of the fundamental beam. Dependence of the efficiency on input power [Fig. 1(b,c)], for both geometries exhibits a dramatic change from linear to quadratic at an average power of 200-250mW. In the same time the conversion efficiency increases four times compared to a pure quadratic process. Analogous threshold behavior can be found in the fundamental beam divergence that increases from 6.5 mrad at 100 mW average power up to 34 mrad at 450 mW. This behavior is accompanied by a significant spectral broadening of the fundamental beam from $70\,cm^{-1}$ (FWHM, at 100 mW) to $150\,cm^{-1}$ (at 450 mW). Due to the broadband phase-matching, the SH spectra at any power are almost exact copies of the fundamental spectra[3].

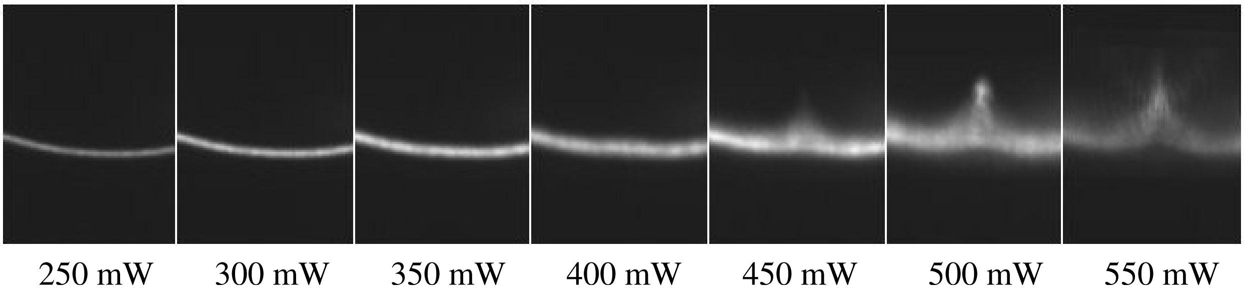

Fig. 2. Effect of a thermal lens on the SH plane emission in the forward direction: As the power increases and the thermal lens gets stronger, the part of the plane formed by the SH in the vicinity of this lens becomes increasingly distorted until at around 500 mW substantial part of the SH wave gets localized in the vicinity of the fundamental beam.

In order to identify the origin of this dramatic changes in the frequency conversion process we place a chopper in front of the SBN crystal to control the average input power while keeping the peak intensity unchanged. The observed decrease in both beam divergence and its spectral width [Fig. 1(a)] with decreasing the average power point directly towards a thermal self-focusing of the fundamental wave inside the SBN crystal. It appears that this process is initiated by the two-photon absorption of the fundamental beam which then leads to a local heating of the crystal and formation of a thermal lens. Subsequent focusing of the fundamental beam increases its intensity and leads to self-phase modulation via the Kerr nonlinearity[5].

The induced thermal lens has a beneficial effect on the conical emission, namely it decreases the divergence angle from 131 degree at 100 mW average power to 125 degree at 382 mW for a fundamental wavelength of 860 nm. Since the SH light emitted in this geometry is radially polarized, this conversion process is a suitable source for focusing beyond the theoretical limit of linear polarized laser beams[6]. A pre-focusing within the crystal thus reduces the necessary aperture requirements for the external focusing optics.

An interesting effect can be observed when the direction of the fundamental beam departs slightly from being perpendicular to the z-axis. For high input powers, i.e. in the thermal focusing regime, the emitted SH plane in forward direction becomes distorted by the thermal lens. The effect is depicted in Fig. 2, where the SH signal with increasing power and thus stronger thermal lens bends toward the fundamental beam until at around 500 mW the SH is localized on top of the fundamental beam.

In conclusion, we have studied SHG in media with random spatial distribution of the ferroelectric domains. We have found that two-photon absorption-mediated thermal self-focusing of the fundamental beam leads to a significant increase of the efficiency of the frequency conversion process as well as to a geometrical and spectral reshaping of the emitted SH signal. We have also demonstrated that the thermal lens can be employed to reduce the angle of conical emission or to spatially localize the SH signal depending on particular interaction geometry. We believe that our findings will broaden the possible applications of broadband SHG in media with random spatial modulation of the nonlinear properties.

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