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To cite this article: E Dimova et al 2018 J. Phys.: Conf. Ser. 992 012007

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# **Experimental demonstration of efficient and robust second** harmonic generation using the adiabatic temperature gradient method

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Abstract. We propose a way of achieving efficient and robust second-harmonic generation. The technique proposed is similar to the adiabatic population transfer in a two-state quantum system with crossing energies. If the phase mismatching changes slowly, e.g., due to a temperature gradient along the crystal, and makes the phase match for second-harmonic generation to occur, then the energy would be converted adiabatically to the second harmonic. As an adiabatic technique, the second-harmonic generation scheme presented is stable to variations in the crystal parameters, as well as in the input light, crystal length, input intensity, wavelength and angle of incidence.

#### 1. Introduction

Second-harmonic generation (SHG) was first reported for the most common nonlinear optical process for crystal materials with a  $\chi^{(2)}$  nonlinearity [1]. It is a special case of sum-frequency generation [2-4]. SHG usually requires the phase matching to be efficient; the SHG efficiencies are typically extremely small when a phase matching does not occur. Traditionally, there are three ways to compensate the phase mismatching: by using different orientation of birefringent crystals, by temperature compensation, or by quasi-phase matching (QPM) [5, 6]. These ways of compensations are tunable for different wavelengths, either due to different orientation of the crystal or due to the temperaturecontrollable phase-matching of the crystal; unfortunately, these methods do not lead to a broadband SHG. A theoretical prediction of an efficient and stable SHG in the adiabatic limit was suggested first by Baranova et al. [7]. Later, chirped QPM gratings were used to achieve broadband SHG [8-10], but due to the lack of adiabaticity, the efficiency remained low.

In this paper, we propose a way of achieving an efficient, robust and stable SHG by adiabatically chirped phase-mismatching using a temperature gradient method. The technique proposed uses the analogue of adiabatic evolution in quantum physics [11-14]. A similar technique, one using chirped quasi-phase match gradients in the case of sum-frequency generation was recently demonstrated experimentally [15-18]. In contrast to the previous work, we show that an efficient and robust SHG is possible with a proper temperature gradient.

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#### 2. Experiment

The light source used was a Quanta-Ray GCR3 high-power Q-switched Nd:YAG laser system oscillating at the fundamental wavelength of 1064 nm, with pulse duration from 7 to 9 ns and maximal energy per pulse of 850 mJ. This laser system can also emit the second harmonic wavelength of 532 nm, pulse duration 5-7 ns, maximal energy per pulse 400 mJ; and the third harmonic at 355 nm, pulse duration 4-6 ns, maximal energy 200 mJ. The repetition rate is in the range 1-20 Hz. The crystal (MgO:LiNbO<sub>3</sub>) chosen for SHG had a square cross-section (5×5 mm) and a length of 40 mm. The laser beam had a diameter of 6 mm; it was restricted by a diaphragm to fit the cross-section of the crystal (beam diameter of about 4 mm). The laser beam was well collimated to a distance of 4 m. The alignment into the active media was performed by means of two mirrors to ensure that the light beam passes through the middle of the sample. To separate the light of different wavelengths, a prism was placed in the output light beam path. We found that, together with the main frequency of 1064 nm polarized along the ordinary polarization axis, there was another light beam with a wavelength of 532 nm and polarized along the extraordinary polarization axis, as it is predicted theoretically [1]. The wavelengths were measured by an AvaSpec-2048 monochromator with AvaSoft 7.5 controlling software. The pulse energy of each beam was measured by laser power meter (Edmund),

$$1064 \text{ nm}(e) + 1064 \text{ nm}(e) \rightarrow 532 \text{ nm}(o).$$
 (1)

The temperature control was achieved by heating the crystal in an oven. Two ovens were built for this purpose, each made from a quartz cylinder with a diameter of 20 mm and a length of 60 mm. A tantalum wire was used to form a suitable heating coil on the outer surface of the cylinder as necessary to conduct the experiment. The coil turns on the first oven were placed equidistantly, while on the second oven the distance between the wire turns was decreased gradually from one end to the other. We could thus follow the effect of the temperature on the nonlinear response of the crystal medium at both a constant temperature and a temperature varying along the crystal sample. The temperature was measured by a thermocouple on the sample surface. To keep the temperature independent of the surrounding conditions, both sides of the oven were closed except for a hole in the middle for the input laser beam and the output light.

The experiment was carried out follows: the laser was oscillating at the IR wavelength of 1064 nm. The laser pulse energy was varied via the pumping lamp power. The losses due to the transfer of the laser beam through the whole optical set-up were accounted for by measuring the output power at the optical path end (figure 1) by using a prism separating the light with different wavelengths, and a power meter placed on the path of each of them.

As a first step of the experiment, we studied the effect of the temperature on the nonlinear response at a constant heating along the crystal medium. To ascertain that the effect measured was indeed due to the temperature difference, we examined the output light from the nonlinear crystal at room temperature (22 °C); no output light at 532 nm was detected. We followed the second harmonic generation at 532 nm in the temperature range 100 - 130 °C) for different pulse laser energies and found that the maximal nonlinear effect occurred at



Figure 1. Experimental setup.

 $110 \,^{\circ}$ C (figure 2). Note that there is a non-critical phase matching along the propagation direction at this temperature.

Bearing the theoretical model in mind, we applied a temperature gradient along the crystal by the second oven. In figure 3 one can see the effect of the temperature on the chirp along the nonlinear

crystal at different temperature gradients. The optimal effect was measured was at the temperature gradient 121  $^{\circ}$ C – 110  $^{\circ}$ C. We were not able to achieve the expected full transfer of the laser light at 1064 nm to the SHG light at 532 nm. The reasons could be the high dispersion of the non-linear medium, the less-than-perfect temperature control along the crystal sample, or the limited laser pulse energy at 1064 nm due to technical problems.



**Figure 2.** Second harmonic generation at 532 nm light in the temperature range 100 - 130 °C for different laser pulse energies. Non-critical phase matching along the propagation direction at 110 °C.



**Figure 3.** Second harmonic generation at 532 nm with a temperature gradient along the crystal at different laser pulse energies (achieved by varying the laser pump lamp power).

### 3. Conclusions

We used the analogy between the time-dependent Schrödinger equation and the SHG equations beyond the laser wave's regime. A temperature gradient at the two ends of the crystal produces a phase mismatch chirp along the light propagation path and creates a crossing in the phase matching, which in combination with adiabatic evolution conditions, allow for an efficient and robust SHG. The adiabatic temperature chirp offers stbility against variations of the parameters of both the crystal and the electromagnetic field, which include variation in the temperature, the wavelengths of the input electromagnetic field, the crystal length and the angle of incidence. The adiabatic SHG suggested is stable due to the absence of an inverse process (the fundamental signal becomes low when the phase mismatch is large and thus the inverse process that may start is very weak). The price that has to be paid for this stability, efficiency and robustness is a large crystal or/and high input-intensity at the fundamental wavelength.

#### Acknowledgments

The authors would like to thank Dr. A. Rangelov for the fruitful discussions and theoretical interpretations of the experimental results. Dr. E. Kyoseva acknowledges the funding from the European Union's Horizon 2020 Research and Innovation Program under the Marie Skłodowska-Curie grant agreement No 705256-COPQE. Dr. E. Dimova acknowledges the Polish-Bulgarian Joint Research Project (2015-2017) under the Scientific Agreement between the Bulgarian Academy of Sciences and the Polish Academy of Sciences.

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IOP Conf. Series: Journal of Physics: Conf. Series **992** (2018) 012007 doi:10.1088/1742-6596/992/1/012007

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