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Generation of fs-pulses down to 121 nm by frequency doubling using random quasi-phase-matching in strontium tetraborate

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Abstract: Tunable femtosecond pulses are generated in the VUV down to 121 nm using random quasi-phase-matching in strontium tetraborate, the shortest wavelength ever produced with a second-order nonlinear optical crystal.

I. INTRODUCTION

Random variation of the second-order susceptibility in acentric crystals leads to modification of the quasi-phase-matching (QPM) conditions and broadband spectral acceptance which is useful for frequency conversion of tunable femtosecond laser pulses. This is especially true for the VUV spectral region below 150 nm where no transparent optical crystals exist that combine non-zero 2nd order nonlinearity with sufficient birefringence. Some attempts to fabricate QPM structures have been reported in the literature, e.g. by electric-filed poling of the ferroelectric $MgBaF_4$ which has mm2 symmetry,[1] or by mechanical twinning of crystalline quartz (SiO_2) which possesses trigonal 32 symmetry,[2] however, both materials exhibit rather low nonlinear coefficients and the shortest wavelength demonstrated so far by second-harmonic generation (SHG) with a 5th order grating is 194 nm.[2]

Strontium tetraborate, SrB_4O_7 (SBO), exhibits orthorhombic mm2 symmetry, the same as KTP or $MgBaF_4$ but its birefringence is too low (<0.005) for phase-matching and it is not ferroelectric. Non-phase-matched SHG has been used in SBO for diagnostics (autocorrelation measurements) but the efficiency of this process is extremely low when only one coherence length is utilized and a practical detection limit was estimated to be of the order of 2 μJ for 120-fs pulses at 267 nm.[3] However, the band-gap wavelength (<120 nm from absorption measurements[4]), the good damage resistivity and chemical stability, as well as extraordinary high (1.5-3.5 pm/V) value of the diagonal d_{33} element[3,5] (compared to the band-gap value) are features that make SBO very attractive for random QPM. Recently, this technique has been demonstrated to be very useful for frequency doubling of femtosecond pulses in the UV, such as generation of the 4th harmonic of a mode-locked 82 MHz Ti:sapphire oscillator in the 187.5-215 nm range.[6] In this case an average power of 1 μW has been generated at 200 nm by frequency doubling 280-fs pulses which corresponds to a conversion efficiency of 10^{-5} and enhancement factor of 320 compared to SHG within one coherence length. Here we study the potential of random QPM SBO for frequency doubling of amplified femtosecond pulses at shorter wavelengths, covering the 121 to 172 nm spectral range with the second-harmonic.

II. EXPERIMENTAL RESULTS

It was found experimentally that as-grown boules of SBO obtained via Czochralski method contain domain structures comprising oppositely poled domains.[5] Shape and orientation of domains are identical to those occurring in KTP: they have the form of layers perpendicular to *a* crystallographic axis and the static polarization is parallel to *c* axis. Thus, the largest nonzero nonlinear coefficient that can be employed is d_{33}. Figure 1a shows the SBO sample available for the present experiment. After removal of the initial and final part along *a*-axis where no domains are seen, the thickness of the sample became 0.9 mm. The transversal dimensions were 6.4×5.6 mm^2 (*b*×*c*).

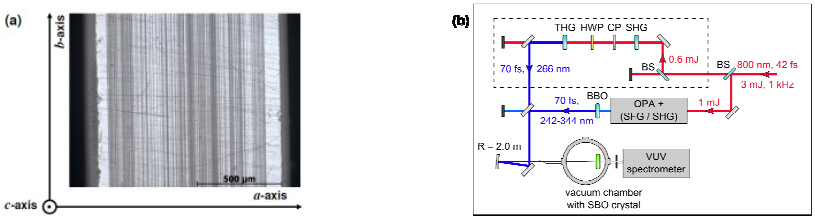

Fig. 1. Photograph of a part of the (001) surface of the SBO sample after etching (a) and experimental set-up (b).

While for longer wavelengths the individual domains could be identified and the SHG process directly simulated, the domain structure responsible for conversion to the VUV is unknown. The transmission of the sample was measured prior to the experiment at the 5th harmonic of the Ti:sapphire regenerative amplifier at ~160 nm (available from FWDFM in Ar) and amounted to ~45%. The pump source was either the 3rd harmonic (at 266 nm) of the same regenerative amplifier at 1 kHz or the frequency-doubled output of a commercial optical parametric amplifier (OPA), Fig. 1b. The 3rd harmonic was generated in a tripling stage (SHG+THG) pumped by 0.6 mJ of the output of the laser system and consisting of two 0.2-mm thick type-I BBO crystals (θ=29° and θ=44° cuts), a calcite plate (CP) to compensate the group-velocity mismatch and a half-wave plate (HWP) which rotates the polarization of the ~800 nm wave. The visible output of the OPA (equipped with internal frequency-doubling of the signal wave or sum-frequency mixing of signal and pump waves) was additionally frequency-doubled in a 0.2-mm thick type-I BBO crystal (θ=45° or 65° cut). In both cases the resulting fundamental UV (242-344 nm) pulse duration was of the order of 70 fs. Depending on the wavelength, the fundamental UV energy varied from 0.2 to few µJ. These UV pulses were focused onto the SBO crystal by a curved Al-mirror. The spot size (diameter) in the crystal was ~220 µm. The generated VUV pulses were detected by a McPherson monochromator Model 234/302 equipped with a VUV-optimized CCD Camera Andor D0420-BN-995.

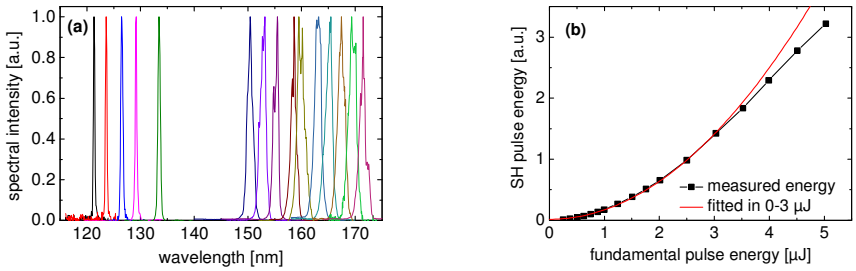

Fig. 2. Spectra of the second-harmonic generated in SBO (a) and conversion efficiency at 133 nm (b).

Figure 2a shows the obtained VUV spectra extending down to 121 nm. The conversion efficiency in terms of energy is shown in Fig. 2b for SHG at 266 nm. The dependence on the input energy is quadratic, as should be expected for random QPM, but at fundamental energies exceeding 3 µJ it tends to be more linear which can be attributed to nonlinear losses, most probably with both 266 and 133 nm waves involved. Although comparison with a non-phase-matched single crystal of SBO with 0.1 mm thickness indicated that much stronger second-harmonic is generated by random QPM, quantitative estimate of the VUV energy, the absolute conversion efficiency, and the actual interaction length remain the main tasks to be solved in the next experiments. This will reveal the actual potential of random QPM in SBO for coverage of the difficult part of the VUV spectral range down to ~120 nm.

III. CONCLUSION

In conclusion, random QPM in SBO is feasible for SHG down to 121 nm and could find applications in generation of femtosecond pulses deep in the VUV or their temporal diagnostics through a reverse nonlinear process.

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