Theoretical study of second harmonic generation properties in Strontium and Calcium Tartrato-Antimonates

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In 1999, Bohatý et al. [1] reported the growth and linear optical properties of the nonlinear strontium and calcium tartrato-antimonates, which they referred to as SrSbT and CaSbT, respectively. Larger than 1-cm^3-sized crystals were synthesized, exhibiting transparency from ~300nm to ~2μm. It was recognized that second harmonic generation (SHG) under type-I geometry is allowed throughout the entire transparency range of the compounds. However, SrSbT and CaSbT exhibit modest nonlinear coefficients [2]. For example, the relevant tensor element for SrSbT (d_{311} = 0.09pm/V) amounts three times less than the respective value for α-quartz. This efficiency constraint has discouraged further employment of SrSbT and CaSbT in wave-mixing applications.

In the present article, we report theoretical calculation of type-I SHG in SrSbT and CaSbT. These point-group-6 negative-uniaxial crystals share nearly identical dispersion characteristics. Fig. 1a shows phase-matching curves indicating type-I SHG possibilities within the entire transparency range. The corresponding angle adjustment varies from ~20 to 90degrees. Phase-matching angle exhibits retracing behaviour near 1.8μm.

Fig. 1b presents temporal walk-off calculations versus fundamental wavelength. It is observed that temporal walk-off crosses the zero-point at a wavelength of 1828nm (1795nm) for SrSbT (CaSbT), respectively. At this wavelength, fundamental and SHG pulses travel with equal group-velocities, leading to large interaction lengths. This effect is also depicted in the spectral domain via acceptance bandwidth considerations. Spectral acceptance reaches infinity at 1828nm (1795nm) for SrSbT (CaSbT), respectively (Fig. 1b). Moreover, spectral acceptance exceeds 500nm·mm for wavelengths longer than 1.5μm. This value suggests that mid-infrared single-cycle pulses may be frequency doubled in crystals longer than 1mm.

Apart from temporal walk-off, useful interaction length may be limited also by spatial walk-off. Spatial walk-off (that is, a birefringence-induced angle between Poynting- and wave-vectors) manifests itself in the phase–matching condition as tolerance to angular variations (angular acceptance). Fig. 1c illustrates scaling of spatial walk-off and angular acceptance versus wavelength. Coincidentally, spatial walk-off reduces (angular acceptance increases) at the long wavelength end of the spectrum, acquiring values of < 50mrad (>70 mrad·mm) at the retracing region, respectively. These values compare well with other common nonlinear crystals such as β-BBO.

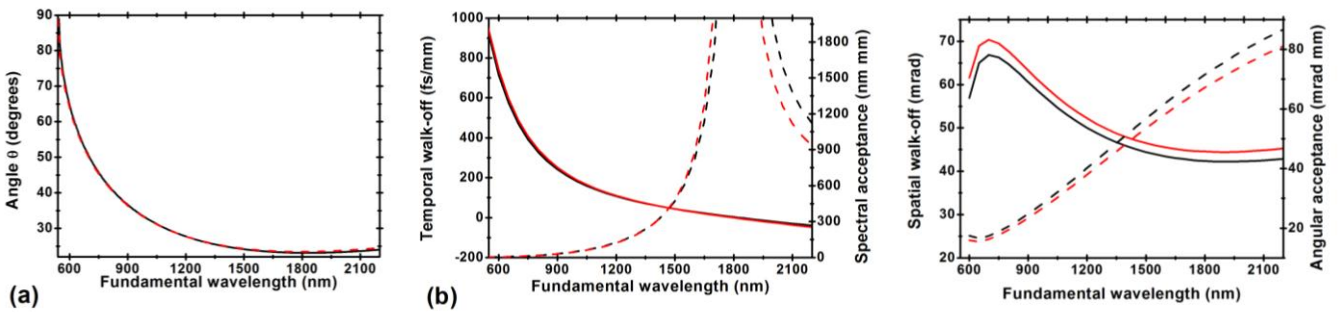

Figure 1 : Theoretical calculation for type-I SHG in SrSbT (black lines) and CaSbT (red lines). (a) Phase matching curves. (b) Temporal walk-off (solid lines) and fundamental spectral acceptance (dashed lines). (c) Spatial walk-off (solid lines) and angular acceptance (dashed lines).

In summary, it was found that type-I SHG in SrSbT and CaSbT crystals exhibits retracing behavior near 1800nm. Ultra-broadband wavelength acceptance and modest spatial walk-off were calculated in the spectral range between 1.5μm and 2μm. This observation reveals that extremely short pulses in this mid-infrared band may be frequency doubled in SrSbT and CaSbT crystals with practical lengths. The 1.5μm – 2μm band includes the telecommunications window and corresponds to the emission of numerous optical parametric oscillators, fiber devices, and other laser systems. As a result, despite the modest nonlinear coefficients, the compounds under examination may be of particular interest to various applications with stringent bandwidth - but flexible output power - requirements. Such applications include, but are not limited to, temporal SHG autocorrelation measurements as well as seeding of parametric amplifiers.

[1] L. Bohatý, H. Hellwig, S. Ahrweiler, A. Möller, "*Linear optical properties of the nonlinear optical calcium and strontium tartrato-antimonates(III),* $Ca[Sb_2\{(+)\text{-}C_4H_2O_6\}_2]\times 2H_2O$ and $Sr[Sb_2\{(+)\text{-}C_4H_2O_6\}_2]\times 2H_2O$", Cryst. Res. Technol., **Volume 34**, page 533, 1999

[2] L. Bayarjargal, P. Becker, L. Bohatý, "*Nonlinear optical properties of strontium tartrato-antimonite(III) dihydrate,* $Sr[Sb_2\{(+)\text{-}C_4H_2O_6\}_2]\times 2H_2O$ ", Cryst. Res. Technol., **Volume 43**, page 508, 2008