Pulse Measurement of Second-Harmonic Generation from Random Quasi-Phase-Matching in ZnS

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Abstract: We use 2.4-micron laser pulses to produce second-harmonic generation via random quasi-phase-matching in ZnS. Using a frequency-resolved optical gating system, we reconstruct the complex temporal profile of the second-harmonic pulses. © 2024 The Author(s)

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Quasi-phase-matching (QPM) enables efficient nonlinear optical processes in a wide variety of materials. QPM involves periodically modulating a material property to overcome the phase mismatch between interacting waves. Typically, QPM schemes use engineered, periodically-poled materials. Interestingly, an effect similar to QPM can be observed (without periodic poling) in polycrystalline materials. This effect, called random quasi-phase-matching (RQPM), occurs when the randomly oriented grains in a polycrystalline material disrupt the phase mismatch accumulation between nonlinearly interacting waves [1]. RQPM has garnered significant attention in recent years. For example, researchers have recently demonstrated broadband, intra-pulse difference frequency generation via RQPM [2], as well as octave-spanning supercontinua supported by RQPM [3]. Although interesting, these results have primarily relied on measurements of the spectrum produced by the RQPM process, and questions remain about the temporal structure of the nonlinear light generated via this random phase-matching process [4]. Here, we use frequency-resolved optical gating (FROG) to reconstruct the temporal pulse profile of second-harmonic generation (SHG) produced via RQPM driven by femtosecond laser pulses in ZnS.

Our experimental setup consists of a Cr:ZnS laser system, in which SHG is produced via RQPM, and a pulse measurement apparatus. The laser system is similar to the one described in Ref. [5]. The system consists of a mode-locked, polycrystalline Cr:ZnS master oscillator (MO) with a central wavelength of 2.4 µm and a pulse repetition rate of $f_R = 80$ MHz. Laser pulses from the MO are sent into a single-pass, polycrystalline Cr:ZnS power amplifier (PA). Both the MO and PA are pumped by off-the-shelf Er-doped fiber lasers (EDFL). The complete system outputs a pulse train with 4-W average power (50-nJ pulse energy), and the output spectrum supports 16-fs pulses. The measured pulse width is about 24 fs due to uncompensated third-order dispersion of the amplifier's gain element. In the polycrystalline gain element of the PA, SHG (1070 nm – 1230 nm) is produced via RQPM. The average power of the SHG is ≈154 mW.

The pulse measurement system is based on a collinear FROG arrangement. The recorded FROG trace was preprocessed into an SHG FROG trace by selecting the baseband signal via filtering, and the result was interpolated into a 1024 x 1024 matrix whose increment and extent satisfied the FROG sampling rate. Additional noise filtering included background subtraction and corner suppression in both image and frequency space. Examination of the prepared trace indicated fine structure and complexity in the pulse, a problem which necessitated a good guess to seed the PCGPA algorithm. In order to produce a high quality guess, we implemented the RANA approach [6], wherein we computed the spectral intensity directly from the FROG trace. We projected the computed intensity onto a number of random guesses that were tested in parallel on smaller traces in a subroutine called multigrid. The best guesses were culled, with preference given to those which produced the lowest G error. Specifically, we executed multigrid using 250 initial guesses at 50 iterations each on a 256 x 256 trace, and the 25 best guesses were selected to run for 25 iterations each on a 512 x 512 trace. The algorithm was seeded with the optimal guess, and ran on the full size trace for a total of 100 iterations, whereupon we achieved a minimum G error of 0.008 on the first attempt.

The measured and recovered FROG traces are shown in Fig. 1a and Fig. 1b, respectively. The recovered pulse temporal profile exhibits detailed pulse structure and indicates a FWHM of the central double-peak of around 120 fs (Fig. 1c). Since an SHG FROG trace is symmetric in the delay variable, our recovery includes a built-in direction-of-time ambiguity, which in the frequency domain amounts to an ambiguous sign in the spectral phase of the pulse. We resolved this ambiguity by measuring a pulse after propagation through 5 mm of ZnSe. Using this pulse, the pulse measurement without the ZnSe, and the known refractive index of the ZnSe, we were able to resolve the phase ambiguity. Additionally, this measurement enabled us to recover the expected spectral phase shift after propagation through ZnSe. This result agrees well with that predicted from the refractive index of ZnSe (see the inset of Fig. 1c) and thereby provides confidence in the performance of our FROG system. This confidence in our FROG system and our pulse reconstruction was strengthened by comparison of an interferometric autocorrelation (IAC) that was computed from the recovered pulse against an IAC that was measured separately on a silicon photodiode (Fig. 1d). Lastly, we should note that preliminary analysis of our reconstructed pulse indicates the possibility that the pulse may be compressible with a simple prism compressor. Initial simulations suggest that a simple, fused-silica prism compressor could enable ~35-fs pulses, albeit with significant satellite pulses and additional structure.

In summary, we have made measurements of the temporal structure of second-harmonic generation produced via RQPM by 2.4-μm laser pulses in ZnS. Using FROG, we found that the SHG pulse is finely structured with a duration of around 120 fs. Additionally, these pulses may be amenable to simple compression techniques.

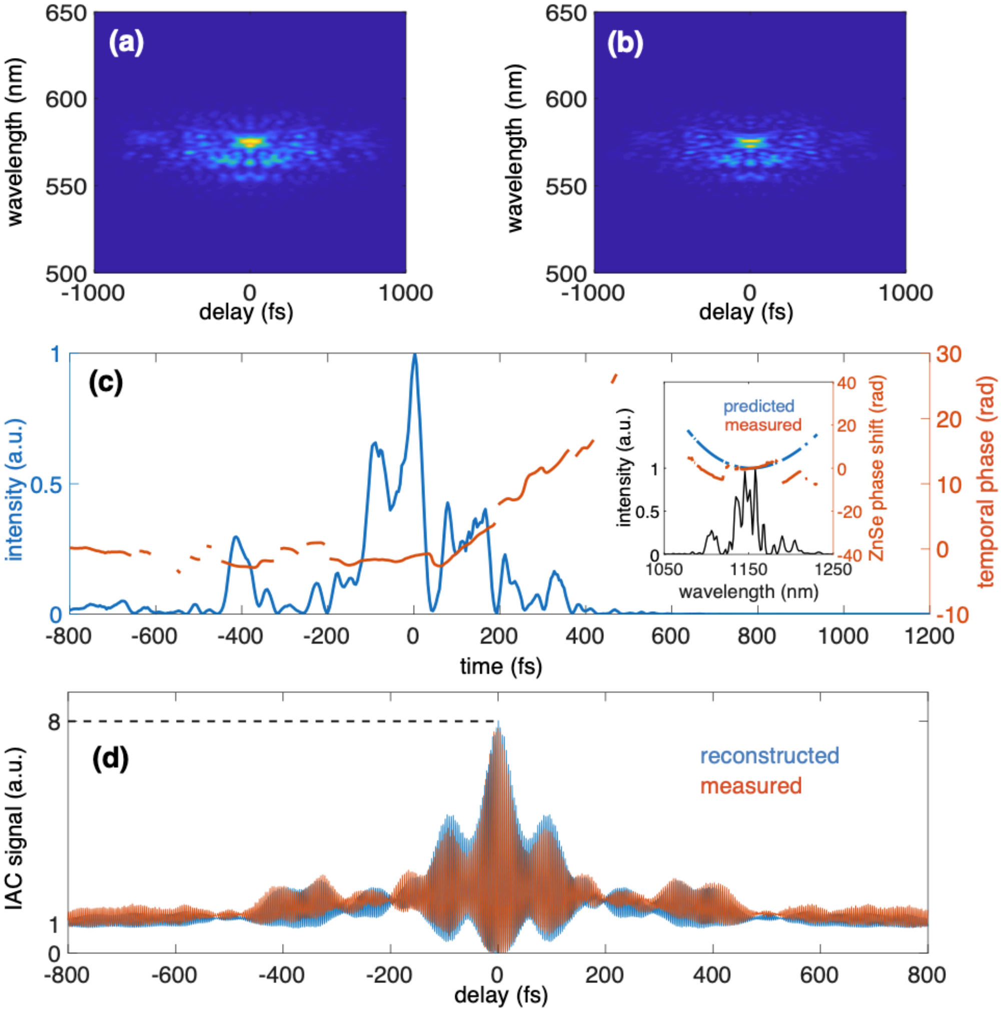

Figure 1: FROG measurements of SHG from RQPM in ZnS. **(a)** Measured SHG FROG trace. **(b)** Recovered SHG FROG trace. **(c)** FROG-recovered pulse temporal intensity and phase. The inset shows the recovered spectral intensity (black trace); also shown is the phase shift of a 5-mm thick sample of ZnSe, both predicted (blue trace) and recovered with a separate FROG retrieval (red trace). (Note: the linear phase has been removed.) **(d)** A measured IAC is overlaid with an IAC reconstructed from the recovered pulse.

References

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