Third-Harmonic Generation In Organic Thin Films As An Alternative To Degenerate Four-Wave Mixing Ultrafast Optical Image Processing

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Abstract: We report on the use of noncollinear third-harmonic generation in thin organic films for ultrafast optical image processing using 80 fs pulses at 1550 nm and compare it with the traditional degenerate four-wave mixing approach.

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1. Introduction

The massive intrinsic parallelism of optical systems is attractive for applications that require computationally intensive operations. The problem of two-dimensional (2-D) image recognition is a good example where optical systems offer such an advantage. When these systems are implemented using ultrafast nonlinear optical (NLO) processes [1,2], computational rates faster than $10^{16}$ operations/second can be achieved. In comparison, BlueGene/L, the fastest supercomputer today, can perform $2.8 \times 10^{14}$ floating point operations per second [3]. Recently, we demonstrated a compact and self-aligned all-optical image correlator [4] that uses a diffractive optical element (DOE) to split the beams to be encoded with the input response ($h$, object ($g$), and reconstruction ($r$) functions. The modulated beams are introduced into a joint transform correlator (JTC) implemented as a 4-f system in a forward-folded BOXCARS geometry. The beams are Fourier transformed into the nonlinear media where their angular spectrum is coherently mixed through a third-order nonlinear optical (NLO) process, which, in turn, generates a signal field that after being Fourier transformed by the second lens is proportional to:

$$e_{\text{out}}(r') \propto \chi^{(3)}(r') \otimes h(r') \otimes g(r').$$

(1)

The use of a DOE and the single lens JTC geometry allows for a very compact setup with no moving parts and assures the spatial and temporal overlap of the interacting pulses, thus removing the need for delay lines and significantly easing alignment. In this paper, we report on the use of this compact correlator for the realization of ultrafast optical processing in the eye-safe and telecommunication compatible near-infrared region at 1550 nm with 80 fs pulses at 1 kHz. In this setup, we demonstrate imaging and image recognition (as seen in Fig 1) using the noncollinear third-harmonic generation (THG) in a thin amorphous polymer composite and compare its performance with degenerate four-wave mixing (DFWM) implemented through the ultrafast Kerr effect in a polyacetylene sample.

2. Results and Discussion

The THG experiments were carried out using a polymer composite with a $\chi^{(3)} \sim 6 \times 10^{-12}$ esu at 1550 nm [5]. A 10 µm polymer film was used for our experiments since the refractive index mismatch limits the coherence length of the collinear THG process to around 9 µm. When this sample is inserted into the correlator, the interacting pulses with wave vectors $k_i(\omega)$, $k_o(\omega)$, and $k_s(\omega)$ generate ten beams in directions determined by the combinations $k_i(3\omega)=k_i(\omega)+k_o(\omega)+k_s(\omega)$ with $i, j, k = h, g, r$. Using 1 mm apertures printed on overhead transparencies to encode $r, g$ and $h$, we measured a conversion efficiency $\eta_{\text{THG}} \sim 10^{-7}$ in the direction of $k_{\text{out}}(\omega)$ with a pump energy of 33 nJ per pulse/beam. The THG signals are strong enough to be seen by the naked eye and easily detected with low-cost Si-based electronic components. The images produced by the THG-based correlator were captured using a Genwac, GW-902H Si-based CCD, having a minimum detectable signal ~ 44 pW/cm². For the DFWM experiments, we used a ring opening metathesis polymerization method [6] to fabricate a 200 µm thick polyacetylene sample with an estimated $\chi^{(3)} \sim 5 \times 10^{-11}$ esu at 1550 nm. Under the same conditions used to evaluate $\eta_{\text{THG}}$, a diffraction efficiency...
$\eta \sim 1\%$ was measured. A MicronViewer 7290A vidicon camera with a minimum detectable signal of $\sim 200\text{ nW/cm}^2$ was used to capture the images produced by the DFWM-correlator. In both sets of experiments, only spatial filtering with an iris was used to isolate the signal beams.

Even though the THG and DFWM signals are both proportional to $|\chi^{(3)}|^2$, the nature of each process makes a direct comparison of the material performance a difficult one since each imposes different requirements on the material parameters and geometry of the setup, both of which will be discussed in detail. However, despite the fact that under current experimental conditions the DFWM process produces $\sim 10^5$ more photons than the noncollinear THG, it suffers from scattering from the fundamental beams which limits the signal-to-noise ratio (SNR) to ~2 with a 1mm reconstruction aperture and to < 1 with a 0.5mm aperture. The non-degenerate nature of the THG process allows for a SNR > 10, limited by the detector noise, even with a 0.5 mm aperture. Since, in our current setup the overall resolution is limited by the size of the aperture in $r$, better resolution was achieved using the THG-approach. As shown in Fig 1b), the enormous difference in photon detectivity of both image-capturing technologies (Si-based CCD ~ $10^4$ > vidicon) compensates for the differences in the signal strength making the images, or overall correlator signal, comparable in both cases. While detailed studies may reveal differences in the ability to reconstruct the angular spectra due to the different nature of the THG and DFWM processes, in our current setup scattering seems to be the ultimate limitation for the use of DFWM. However, scattering is a material dependent problem, so we expect that with the development of better materials and processing techniques, the cost/detectivity difference in imaging technologies will become the primary concern.

![DFWM and THG](image.png)

**Figure 1.** (a) Schematic of DOE-based correlator; Experimental demonstration of (b) image recognition through DFWM and noncollinear THG.

### 3. Conclusion

Despite having an order of magnitude difference in their $\chi^{(3)}$, the fact that a slightly better performance is obtained using the THG polymer composite with respect to the polyacetylene sample, makes this new approach an attractive alternative to the development of compact ultrashort optical image processors using DFWM. Even though the lack of ultrashort spatial light modulators continues to be the true bottleneck for the realization of this technology, the constant advances in portable ultrashort laser technologies and NLO material research continues to bring ultrashort optical processing closer to its realization.

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### 4. References


