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Midwave Infrared Ultrashort Pulse Laser Frequency Conversion in Single Crystal, Polycrystalline, and Amorphous Optical Materials

Michael Tripepi[a], Michael Hastings[b], Aaron Schweinsberg[c], Laura Vanderhoef[c], Christopher Wolfe[c], Trenton Ensley[d], Miroslav Kolesik[b], Jerome Moloney[b], Enam Chowdhury[e], and Anthony Valenzuela[c]

[a]The Ohio State University, Department of Physics, 191 W. Woodruff Ave., Columbus, OH, 43224, USA
[b]University of Arizona, College of Optical Sciences, 1630 E. University Blvd., Tucson, AZ, 85721, USA
[c]CCDC-Army Research Laboratory, Weapons and Materials Research Directorate, Aberdeen Proving Ground, MD, 21005, USA
[d]CCDC-Army Research Laboratory, Sensors and Electron Devices Directorate, Adelphi, MD, 20783, USA
[e]The Ohio State University, Department of Material Science and Engineering, 201 W. 19th Ave., Columbus, OH, 43210, USA, and Department of Electrical and Computer Engineering, 2015 Neil Ave., Columbus, OH, 43210, USA

ABSTRACT

We investigate the nonlinear optical properties of transparent optical materials using ultrashort midwave infrared laser pulses between 3 and 4 microns. Random quasi-phase matching in polycrystalline materials generates multiple frequency harmonics of both odd and even orders throughout the transmission window of the target. We also investigate single crystal and amorphous materials and demonstrate a range of frequency conversion and pulse broadening. Simulations using a nonlinear polarization model enhanced with ionization and experimentally measured n_2 values provide good qualitative agreement with experimental data.

Keywords: Ultrashort pulse lasers, high harmonic generation, filamentation, UPPE

1. INTRODUCTION

Advancements in mid-infrared (MIR) wavelength ultrashort pulse lasers have allowed for new regimes of nonlinear propagation and frequency conversion in a wide range of materials. Our previous work with chalcogenide optical materials demonstrated the ability to generate all-order harmonics.[1,2] This work complemented the work of other groups that looked at a wide range of materials including fluorides,[3] chalcogenides[4] and silica glasses.[5] A pattern has emerged where at lower intensities, discrete harmonics are generated in these materials and, as the intensity is increased, an inter-harmonic continuum grows to the point where the spectral response is closer to that of a supercontinuum. This pattern was not readily observed using near-infrared (NIR) sources, particularly with a wavelength of 800 nm, since the higher harmonics are typically linearly absorbed by these materials. Further study of these materials at longer wavelengths has shown a wide and surprising set of resultant frequency conversion that demonstrates nonlinear mechanisms can play a significant role even beyond the linear regime.

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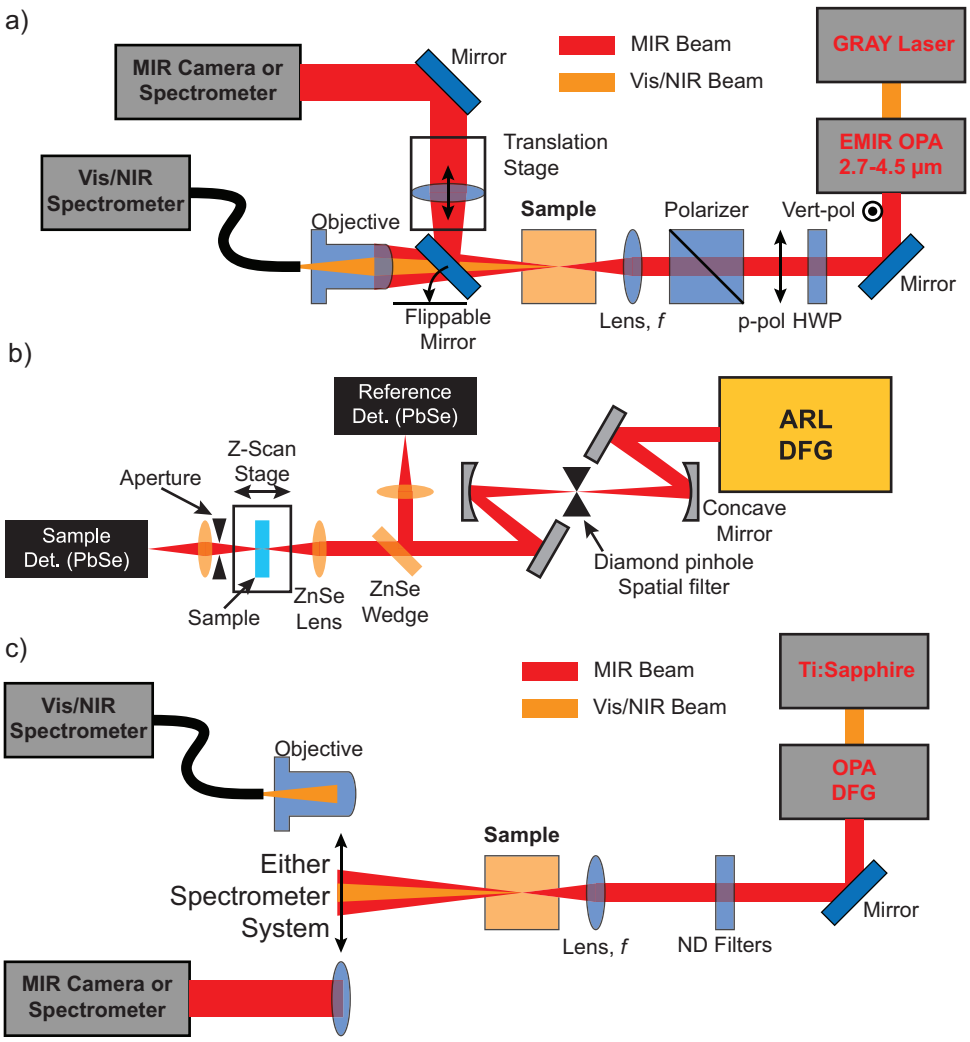

Figure 1. Experimental setups for a) OSU, b) ARL-ALC, and c) the AFRL laser systems.

2. EXPERIMENTAL SETUP

We utilized three USPLs to perform the experimental component, one at The Ohio State University (OSU), one at the US Air Force Research Laboratory (AFRL) and one at the US Army Research Laboratory (ARL-ALC). The OSU laser is the Extreme MIR (EMIR) optical parametric amplifier (OPA) pumped by the Ti:Sapphire OSU Gray Laser as shown in Fig. 1a. The EMIR output is continuously tunable between 2.5 and 4.5 µm with a pulse duration of 200 fs at a 500 Hz repetition rate. For the scope of our experiments, we utilized a central fundamental wavelengths at approximately 3.6 µm. Energy tuning of our pulse between 1 and 10 µJ is accomplished using a waveplate/polarizer system giving us peak powers ranging from 5 to 50 MW. The pulses were focused onto the sample using CaF_2 lenses with focal lengths varying from 20 to 105 mm, achieving spot sizes ranging from 24 to 98.5 µm FWHM for 20 and 105 mm lenses, respectively. Our focal intensities ranged between 0.06 and 11 TW/cm^2. A list of the samples used in this experiment is given in Table 1. The emission from the sample is sent into either a Visible (Vis) and NIR channel or a MIR channel by using a flippable mirror. The Vis/NIR emission was collected with a microscope objective that filtered out the MIR signal and fed into a fiber connected to to a suite of spectrometers covering 0.4 to 2.5 µm. The MIR channel focused the emission

Further author information: (Send correspondence to A.V.)
A.V.: E-mail: anthony.r.valenzuela6.civ@mail.mil, Telephone: 1 410 278 9876

Table 1. List of samples used for experiments including relevant parameters.

Name	Material	Type	Structure (AGS in µm)	Thickness (mm)	Bandgap (eV)
ZnS (100)	ZnS	Single Crystal	Cubic (100)	1	2.82
ZnS (110)	ZnS	Single Crystal	Cubic (110)	1	2.82
Cleartran	ZnS	Polycrystalline	Cubic (20-35)	3, 10, 20, 40	3.54
CaF_2	CaF_2	Single Crystal	Cubic	5, 10, 20, 40	12.1
BK-7	Borosilicate glass	Amorphous	n/a	5, 10, 20, 40	4.28

onto a MIR spectrometer from Acton Research with a FLIR cooled focal plane array capable of detecting 3–5 µm light.

The ARL-ALC experimental setup, shown in Fig. 1b, was used for spectral measurements and characterization of n_2 for our samples using the Z-scan technique. The same equipment as described above was used for obtaining spectral information. For the Z-scan measurements,[6] MIR pulses at 3.9 µm were generated by using a Ti:sapphire laser system (Coherent, Hidra-F-100) to pump an OPA (Light Conversion, TOPAS-Prime-HE), which was then used for difference frequency generation, DFG (2 mm thick Type II KTA). The DFG pulses were spatially filtered by focusing into a 180 µm diameter single-diamond pinhole using an all-reflective geometry. A PbSe reference detector used the pickoff from a ZnSe wedge. The remaining light was focused into the sample using a 150 mm ZnSe focusing lens with the transmitted light from the sample focused onto a second PbSe detector. Energy readings in the Z-scan system were accomplished using a spectrally flat broadband THz radiometer with linearity ranging from 0.1 µW to 20 mW calibrated with a pyroelectric detector near the DFG output. The radiometer was then placed just before the sample without any filtering to record the actual energy. Germanium neutral density filters were used to attenuate the energy used in the Z-scan runs. The beam size was measured to be 48 µm gaussian waist radius using a knife-edge scan and the pulse duration was measured to be 260 fs (FWHM) using second-order autocorrelation.

The AFRL experimental setup is similar to the OSU setup and shown in Fig. 1c but with key difference in the laser parameters. The system uses a KM Labs Wyvern-1000 Ti:Sapphire laser operating at 1 kHz to pump a Light Conversion TOPAS-C OPA and a Light Coversion NDFG DFG. This system gives us up to about 20 µJ of energy at 3.6 µm with a pulse duration measured at about 80 fs while maintaining the 1 kHz repetition rate. For scaling laser energy and power, neutral density (ND) filters were used with the fundamnetal spectra recorded for each filter.

3. RESULTS AND DISCUSSION

We interrogated a wide range of materials with primarily 3.6 µm radiation and recorded the resultant spectra of around the fundamental and also at lower wavelengths. By varying the thickness of the samples and the laser energy, we see the interplay of both features on filament generation and frequency conversion. Expanding on previous work with polycrystalline chalcogenides,[1,2] we examined the spectra generated through single crystal ZnS with either (100) or (110) orientation. As shown in Fig. 2, we see continuation of odd harmonics of the 3.6 µm fundamental. In addition, the (110) crystal exhibited a continua in the 0.4-0.6 µm region. This contrasts with the polycrystalline Cleartran samples in our previous work that demonstrated all order harmonics with a broadening continua between the harmonics with increased laser power.

A more commonly used single crystal optical material is calcium fluoride (CaF_2) which has a much broader linear transmission window than the chalcogenides we examined. By propagating through a variety of thicknesses and laser powers in Fig. 4 we see how rapidly the fundamental spectrum is broadened on both sides of the original 'No Sample' trace. As well, Figs. 5 and 6 show how varying laser power and material thickness affect the generation of harmonics and continua. Of note is strong generation of third harmonic radiation that requires both higher powers and thicknesses. There is also a lack of appreciable second harmonic radiation. We also

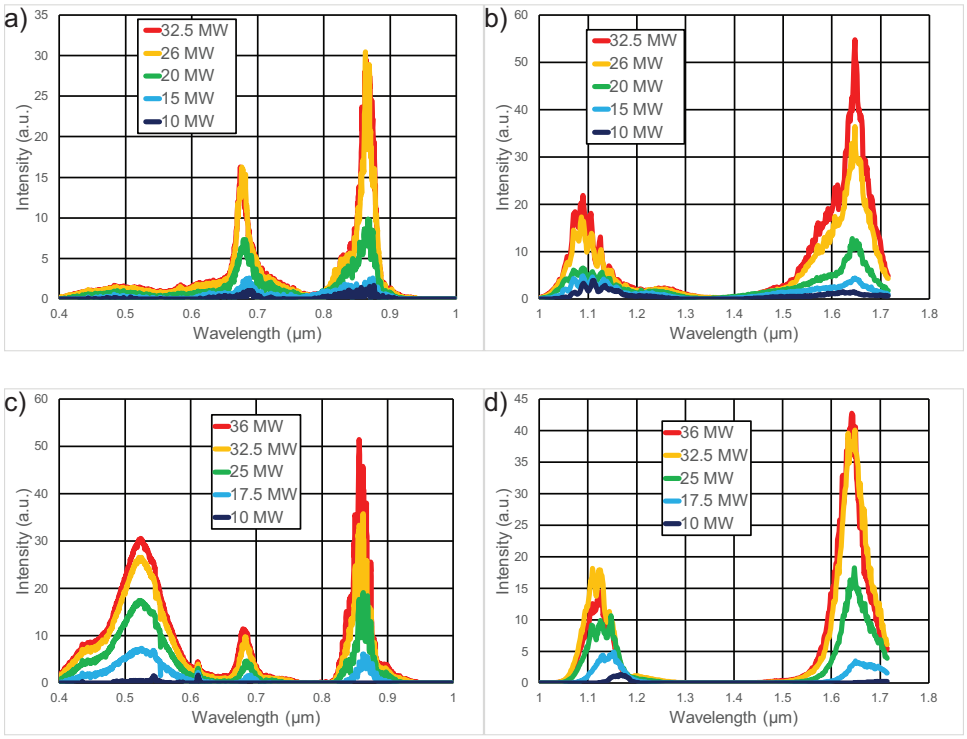

Figure 2. Spectra through 1 mm of single crystal ZnS with 3.6 µm laser light showing for (100) orientation a) visible to NIR and b) NIR to SWIR regions and for (110) orientation c) visible to NIR and d) NIR to SWIR regions as compared to laser power.

recorded a complex continua between 0.3 and 0.6 µm for a wide range of powers and thicknesses. This is similar to previously published results in Ref. 3 and also represents a striking visual image in Fig. 3.

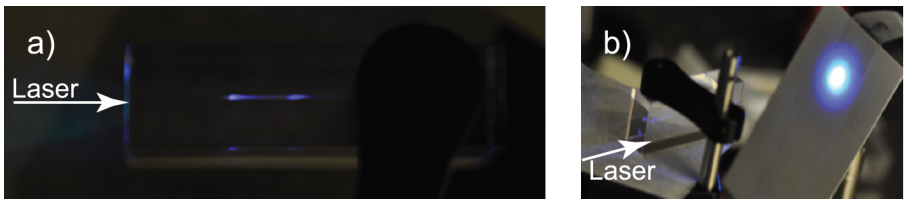

Figure 3. Photographic images of a) laser focused into 40 mm of CaF_2 showing fluorescence from filament formation and the resultant polychromatic emission is b) imaged on a white paper card.

The next class of materials we examined were amorphous glasses, in particular the commonly used borosilicate BK-7. In contrast to the other materials, the linear transmission of BK-7 at 3.6 µm is very low (typically ¡5%). However, as we see in Fig. 7 a strong nonlinear response is still present at this wavelength to generate a distinct third harmonic signal. It should be noted that no clear signal was detected below 1 µm.

To complement the spectral data acquired, we also measured the nonlinear index of refraction and used that data to inform models of these interactions. Further details of Z-scan measurements can be found in Ref. 6 and relevant n_2 data is given in Table 2. Modeling of these interactions involves using measured n_2 values as inputs into the Unidirectional Pulse Propagation Equation (UPPE).[7] Our previous work has demonstrated incorporating Random Quasi Phase Matching[8] into UPPE produces good qualitative agreement with experimental measured spectra of polycrystalline chalcogenides. Further improvements to the modeling component involves better understanding of the relationship between the focal spot size and the grain size of a polycrystalline material. An

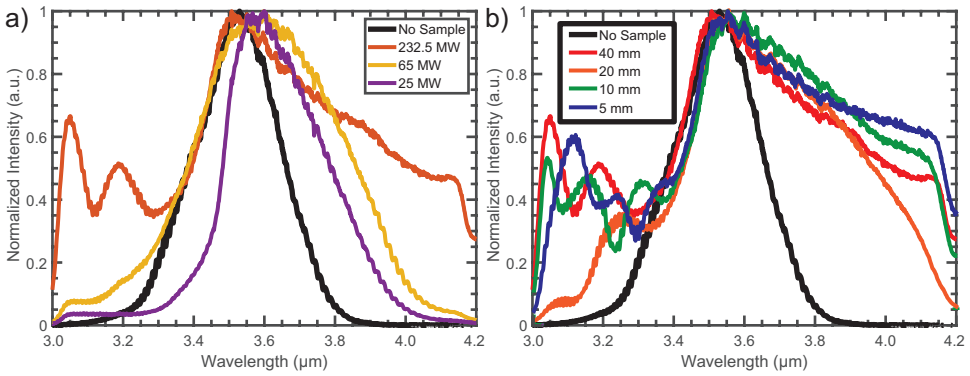

Figure 4. Mid-IR spectra of CaF_2 with 3.6 μm laser light showing a) broadening with increasing laser power and b) broadening with respect to target thickness.

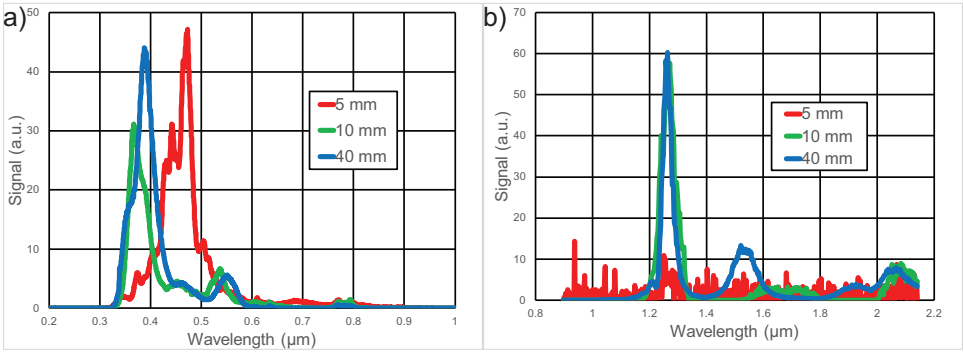

Figure 5. Spectra of CaF_2 with 232.5 MW of 3.6 μm laser light showing a) visible to NIR and b) NIR to SWIR regions as compared to depth of the sample.

Average Grain Size (AGS) is compared with the focal spot, w_0, in the material and simulated over a variety of thicknesses. As shown in Fig. 8a, an AGS of 60 μm versus w_0 of 20 μm leads to very distinct harmonics whereas in Fig. 8b an AGS of 20 μm and w_0 of 60 μm has harmonics on top of a broader continua with increasing material thickness. This is qualitatively compared with experimental data of 3 mm Cleartran samples in Fig. 8c where a wider focus shows a more intense continua though the harmonics are of approximately the same intensity.

4. CONCLUSIONS

Our data demonstrates that a broad range of material types all respond to intense MIR radiation through frequency conversion into harmonics or a continua. The propensity of these materials to react to MIR radiation, even when the linear transmission is very low demonstrates that these interactions require careful consideration in the nonlinear regime. These results could impact applications of a wide range of materials for filtering when using

Table 2. Measured values of n_0 and n_2 from Ref. 6, measurement errors and references therein.

Material	n_0		n_2 (10^{-15} cm^2/W)		
	0.8 μm	3.9 μm	1.0 μm (*0.8 μm)	2.0 μm	3.9 μm
ZnS	2.31	2.41	5.5	-	5
CaF_2	1.43	1.41	0.18*	0.17	-
BK-7	1.51	-	0.33	0.35	-

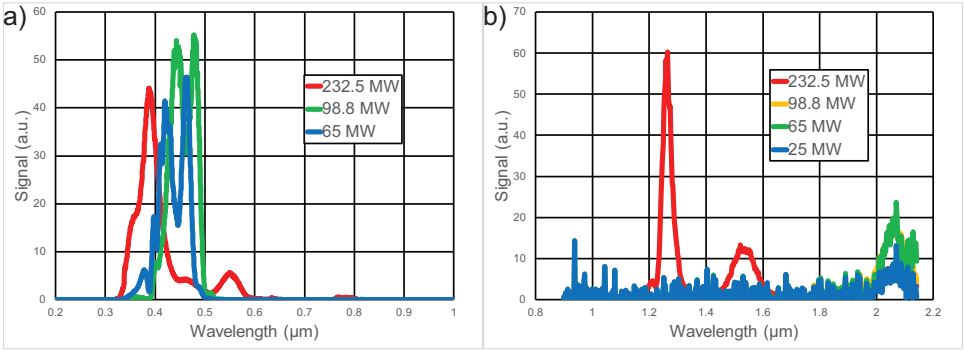

Figure 6. Spectra through 40 mm of CaF_2 with 3.6 μm laser light showing a) visible to NIR and b) NIR to SWIR regions as compared to laser power.

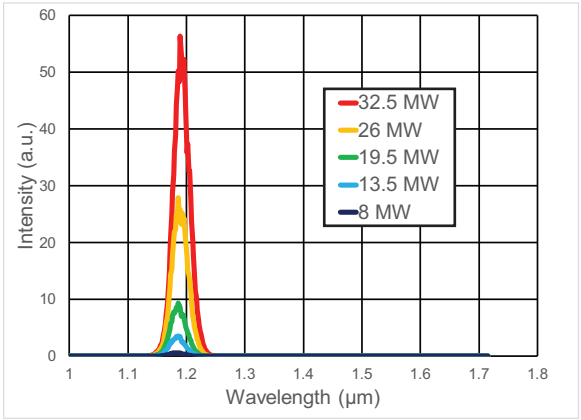

Figure 7. Spectra through 40 mm of BK-7 glass with 3.6 μm laser light showing NIR to SWIR regions as compared to laser power.

high laser powers in this wavelength range. Care must be taken to consider the nonlinear effects of these materials as well as the thickness of a substrate for applications like controlling laser energy. This work is supported with experimental measurements of the nonlinear index of refraction and improvements to UPPE. Further work will expand the range and capabilities of UPPE to simulate these experiments and to better understand the effects of angle of incidence on single crystal targets.

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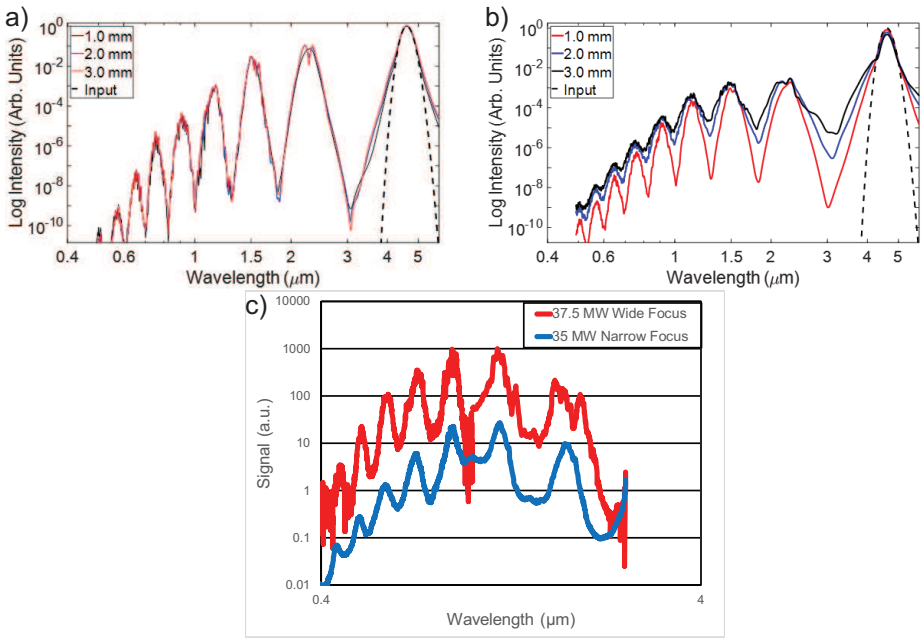

Figure 8. Comparison of simulated spectra between a) narrow focus and b) wide focus spot sizes compared with average grain size of the material. This is compared to c) experimentally measured spectra of 3 mm Cleartran (polycrystalline ZnS) using different focal parameters.

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