

Second Harmonic Generation and Optical Rectification Using Intense Terahertz Pulses

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Abstract: Nonlinear optical processes with THz frequency light have rarely been measured and characterized. Using 2D THz spectroscopy, we clearly observe THz-optical rectification and THz-second harmonic generation in a variety of materials. © 2022 The Author(s)

1. Introduction

Terahertz (THz) spectroscopy is an emerging field that has many important and powerful applications in bio-imaging, security imaging, non-destructive analysis, and high-speed computing and communications. Many of these applications rely on the generation of high intensity and broadband THz pulses. Efficient generation of THz frequencies has been a topic of interest in recent years, and intense pulses of THz light enable nonlinear processes that may be the basis for high-speed technologies to occur. The ability of this THz light to generate new frequencies through optical rectification (OR) and second harmonic generation (SHG) has only been studied in limited situations. The ability of these THz frequencies to cause nonlinear phenomena is of interest and has potential as a useful method of frequency conversion—leading to long term applications in electronic devices. Using 2D THz transmission spectroscopy [1-5] on THz generation crystals, we can generate and detect new frequencies of THz light. 2D spectroscopy enables us to identify clear signatures of optical rectification and second harmonic generation of broadband THz frequency light [1-5].

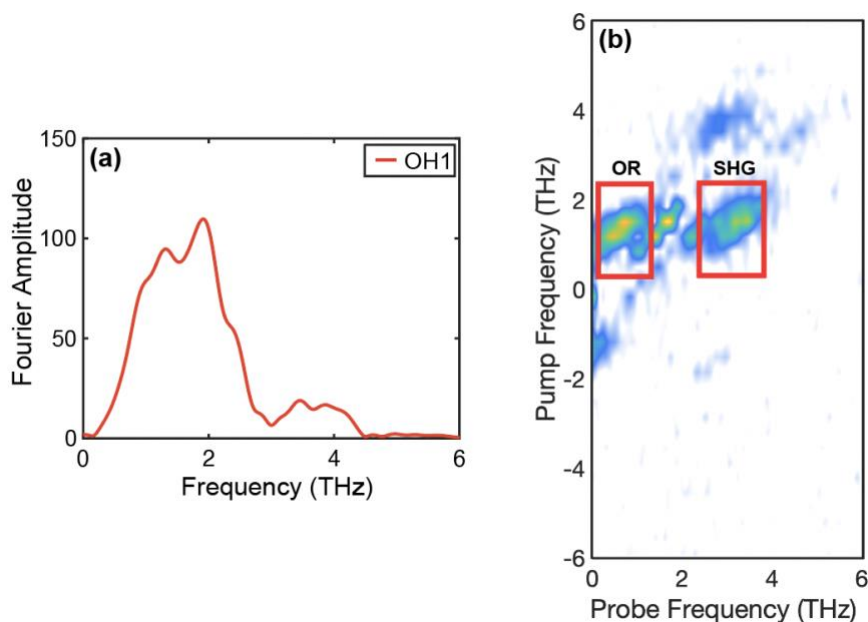


Figure 1. (a) The THz generation spectrum of the NLO crystal OH1. **(b)** A 2D transmission spectrum of the nonlinear signal in the THz generation crystal PNPA when pumped with THz generated from OH1 crystals.

2. Results

Using a 2D THz transmission measurement, we can isolate new frequencies of THz emitted from the sample as a function of the incident THz pump frequencies. This makes it straightforward to observe optical rectification and second harmonic generation with THz frequency light as our pump source. Using a two-dimensional setup with

OH1 THz crystals [6] as the THz generators (see OH1 generation spectrum in **Figure 1a**), and PNPA [7,8]—a new, highly efficient organic THz generation crystal—as the sample NLO crystal, we obtain the 2D spectrum shown in **Figure 1b**. The two THz pulses from the OH1 interact with the PNPA, and we detect the nonlinear response that results in new THz frequencies emitted from our sample crystal.

The probe axis shows new frequencies of THz light that are generated when the pump THz pulses interact with the PNPA crystal. The pump axis shows which THz frequencies from the OH1 crystals are mixing to give us these specific emitted (probe axis) frequencies. As shown in **Figure 1a**, the THz spectrum of OH1 peaks at ~ 1.5 THz. We observe nonlinear signal at a pump frequency from 1-2 THz (corresponding to the peak in the OH1 spectrum) that corresponds to a probe frequency of ~ 0 THz (the red box on the left). This shift to lower frequencies corresponds to THz-optical rectification that is occurring in the PNPA crystal. We also observe second harmonic generation. The red box on the right highlights the nonlinear signal present at ~ 1.5 THz (pump axis) and ~ 3 THz (probe axis). This doubling of the input frequency from the OH1 corresponds to SHG.

We will present additional measurements of THz-nonlinear optical effects in a variety of organic and inorganic materials, as well as modeling that helps describe phase-matching considerations that, in part, govern the efficiency of THz nonlinear optical effects in these materials.

3. Summary

We will present on the nonlinear optical processes achievable with THz frequency light. 2D THz transmission spectroscopy enables us to easily isolate THz-optical rectification and THz-second harmonic generation in a variety of materials. These nonlinear phenomena have not yet been characterized when THz frequency light is used as the pump source. A better understanding of these nonlinear THz processes will be helpful in developing new technologies based on nonlinear optical effects with THz light.

4. References

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