Broadband and high-sensitivity time-resolved THz system with grating-assisted noncollinear phase-matching

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***Abstract*— We demonstrate a time-resolved terahertz (THz) system taking advantage of noncollinear frequency mixing in thick generation and detection crystals to perform broadband and high-sensitivity spectroscopy. A phase grating etched at the surface of a 2 mm-thick GaP crystal is used to control phase-matching conditions by diffracting near-infrared pulses. Our dual grating configuration allows long nonlinear interaction lengths that maximize THz generation and detection efficiencies while maintaining an accessible spectral window up to 7 THz.**

I. INTRODUCTION

Broadband terahertz time-domain spectroscopy (THz-TDS) relying on optical rectification in second-order nonlinear crystals has been extensively exploited in studying low energy excitations in polymers [1], metamaterials [2], topological insulators [3] and superconductors [4]. The performance of a THz-TDS system is directly related to the signal-to-noise level and dynamic range, while the spectral bandwidth often decides the amount of information that can be acquired from a given material. Maximizing the nonlinear interaction length during the THz generation and detection processes can increase both the signal and detection sensitivity. However, there is a trade-off as the intrinsic material dispersion and related phase matching conditions will then reduce the effective spectral bandwidth. Here, we propose the use of phase gratings fabricated at the front surface of a 2 mm-thick gallium phosphide (GaP) crystal to tweak phase-matching conditions for THz generation and detection by changing the direction of propagation of an incident near-infrared (NIR) pulse inside the crystal (Fig. 1). Configurations relying on grating-assisted THz generation were previously studied by different groups [5–7], and last year we transposed this idea to the electro-optic sampling scheme and demonstrated an enhanced detection sensitivity over a large spectral bandwidth [8]. The combination of a grating-assisted noncollinear scheme for generation and detection processes allows us to overcome the spectral bandwidth limitations imposed by phase-matching conditions in thick crystals while still taking advantage of long nonlinear interaction lengths.

II. EXPERIMENT

In our experiment, a near-infrared optical source delivers 180 fs pulses centered at 1.035 µm. These pulses are launched into an argon-filled hollow-core photonic crystal fiber, where self-phase modulation broadens the NIR spectrum to 8.7 THz. A pair of chirped mirrors is then used to compensate for the dispersion to compress the pulse to a ~60 fs duration. The resulting pulse is then sent into a standard THz-TDS setup where THz is generated in a GaP crystal by optical rectification and detected inside an identical crystal by electro-optic sampling [9]. To obtain noncollinear phase matching, we fabricate a rectangular phase grating on the surface of a 2 mm-thick GaP crystal with a pitch of 1.635 µm. This value is selected to optimize generation and detection around 4 THz [9]. A precise modulation depth corresponding to an optical path difference of λ/2 allows optimal diffraction of the incident light into the ±1st orders where we measure more than 60% of the transmitted power, while less than 5% remains in the 0th order. A good diffraction efficiency in the 1st order is especially important for the generation process since the different diffraction orders lead to different phase-matching conditions and generated THz spectra.

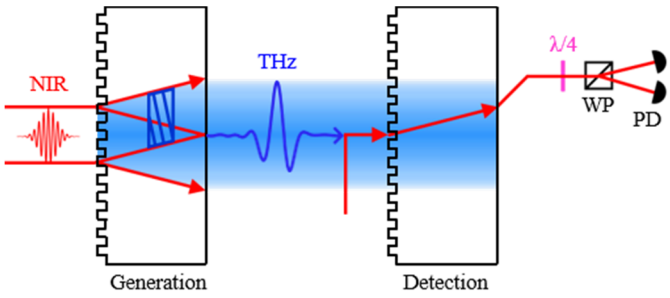

Fig. 1. Schematic representation of the experiment. We use THz generation and detection crystals with a periodically modulated surface to diffract the NIR pulse (while the direction of propagation of the longer THz wavelength remains unaffected) to optimize nonlinear phase matching conditions around 4 THz. Both diffracted first orders (m = -1, +1) contribute to the THz generation process while only one diffracted order on the detection crystal is monitored for electro-optical sampling. NIR: Near-Infrared; THz: Terahertz; λ/4: quarter-wave plate; WP: Wollaston prism; PD: photodetector.

III. RESULT

The system's sensitivity and spectral bandwidth is adjusted by changing the pair of nonlinear GaP crystals used for the THz generation and detection. We explore three different cases and present their results in Fig. 2: (i) A pair of thin 0.2 mm-thick crystals, which yields a relatively weak THz signal covering a large spectral bandwidth. (ii) A pair of 2 mm-thick crystals results in a larger THz amplitude signal, by a factor of 25, but the signal drops rapidly after 1.5 THz due to phase mismatch. (iii) Finally, a phase grating (PG) etched on the surface of a 2 mm-thick crystal enables noncollinear phase-matching conditions leading to a large THz spectral bandwidth and high signal amplitude due to the long nonlinear interaction length. More specifically, the spectral amplitude obtained with the PG crystals (blue line in Fig. 2) is >20 times higher at 3 THz than the one measured with the pair of thin 0.2 mm-thick crystals (black line). If we also compare the spectra obtained with PG crystals and the unpatterned 2 mm-thick crystal (red curve), we find that they have a similar peak amplitude but our new approach relying on a noncollinear configuration yields a much broader bandwidth: we can achieve a signal amplitude beyond 3 THz that is higher by more than one order of magnitude.

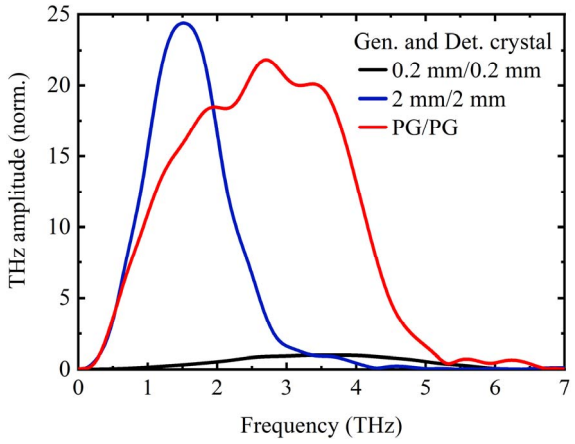

Fig. 2. THz spectral amplitude measured with 3 pairs of identical generation and detection GaP crystals having a thickness of 0.2 and 2 mm. A phase grating (PG) on the incident surface of a 2 mm-thick crystal modulates phase matching conditions to enhance THz generation and detection above 2 THz. The THz amplitude is normalized to the total power detected on the balanced photodiodes and then to peak amplitude measured with the pair of thin GaP crystals.

To quantify the ability of a system to perform spectroscopy of a highly absorptive material, we also investigate the noise floor measured in the absence of THz signal. Using the two configurations involving a pair of thin crystals (Fig. 3a) and a pair of patterned (PG) crystals (Fig. 3b), we collect measurements with and without the THz signal. Each line in Fig. 3 corresponds to the Fourier transform of a single time-domain scan using a 30 ms time constant for each data point and a 2 min acquisition time to measure the full transient. Figure 3 shows that both configurations yield similar spectral bandwidth, with a signal extending up to 7 THz, after which the spectral amplitude approaches the noise caused by signal fluctuations. We then block the THz beam in the setup and perform another scan to measure the noise floor corresponding, which is calculated by averaging the recorded amplitude across the spectral range. Using this value and the spectral peak amplitude, we extract the intensity dynamic range (DR) as defined in [10], which has a maximum of 55 dB for the thin crystal scheme and 70 dB when we use a pair of PG crystals. This 15 dB improvement is mainly due to the higher amplitude signal generated and detected in a thicker nonlinear crystal, but it is also partially due to an intrinsic polarization filtering effect occurring at the back surface of the detection crystal and described in [8]. Briefly, partial reflection of the s-polarized component of the diffracted NIR pulse at the back surface of the detection crystal reduces the total power on the photodiodes, and therefore the experimental noise, while the THz information, which is contained in the preferentially transmitted p-polarization, is not significantly reduced. We believe that our setup relying on a pair of PG crystals can show an even larger improvement over conventional configurations once we have addressed other experimental sources of noise, such as electronic noise and beam pointing instability.

IV. Summary

Our configuration based on a pair of nonlinear crystals with a surface phase grating overcomes some limits imposed by phase-matching conditions at normal incidence, while allowing a long nonlinear interaction length to achieve high signal amplitudes over a large spectral bandwidth. We notably achieve a dynamic range above 70 dB between 1.5 and 3 THz. Our concept can be easily implemented in a standard THz-TDS system, since the PG crystals simply replace the conventional generation and detection crystals. Only minimum adjustments are required in the detection setup to pick up one of the diffracted orders. Although we focused only on improving spectroscopy applications in the region between 3 and 7 THz with GaP as a nonlinear crystal, the idea could be extended to other materials to improve THz generation and detection in other spectral regions. Finally, our results pave the way towards efficient THz spectroscopy across a broad bandwidth and could enable high-field THz generation at frequencies above 3 THz.

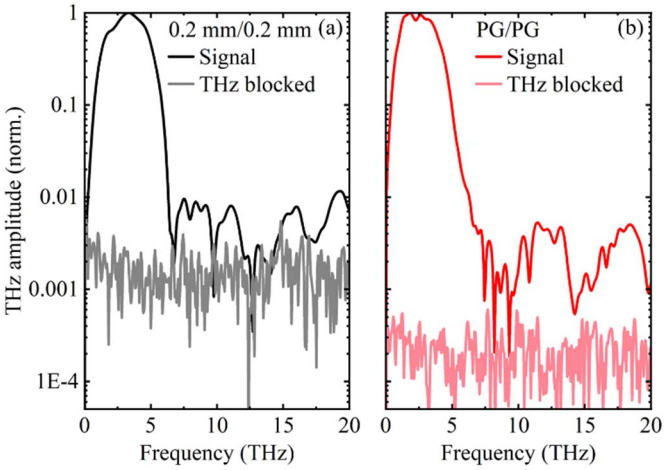

Fig. 3. (a) THz spectral amplitude measured with a pair of thin 0.2 mm thick generation and detection GaP crystals. (b) Same measurement performed with a pair of 2-mm thick GaP crystals with a phase (PG) etched on the incident surface to enable non-collinear phase matching conditions. The dashed lines correspond to the noise floor measured by blocking the THz beam.

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