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(G*) Broadband and high sensitivity THz system with grating-assisted noncollinear phase-matching

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Time-domain terahertz (THz) spectroscopy has been widely exploited in studying semiconductors, superconductors, topological insulators, and metal-organic frameworks. A high-sensitivity THz system can resolve weak spectroscopic features and a broadband system allows experimentalists to rely on additional spectral information to investigate novel phenomena in materials. In a standard configuration relying on difference frequency mixing to generate and detect THz radiation, the spectroscopy signal can be improved by increasing the nonlinear interaction length inside a nonlinear crystal. However, the accessible spectral bandwidth is then limited by phase-matching conditions. Here we demonstrate a time-resolved THz system relying on noncollinear THz generation and detection schemes in thick nonlinear crystals to perform high-sensitivity and broadband spectroscopy. This concept relies on a phase grating etched on the front surface of two 2-mm thick gallium phosphide (GaP) crystals (THz generation and detection crystals) to diffract the incident near-infrared pulses. Our scheme exploits the long interaction length in these crystals to improve the signal strength and dynamic range in the system. In addition, the noncollinear geometry yields optimizable phase-matching conditions to access a broad spectral bandwidth. We compare our results with those obtained with a traditional broadband collinear system using a pair of thin GaP crystals without gratings. The noncollinear geometry shows a significant increase of the maximum signal amplitude, by a factor of 20, while also achieving a large spectral bandwidth reaching up to 7 THz. We also achieve a dynamic range above 80 dB between 1.1 and 4.3 THz. Our concept could be extended to other nonlinear crystals besides GaP to improve THz generation and detection in different spectral regions. In conclusion, this work paves the way towards high-sensitivity THz spectroscopy over a broad bandwidth in low power experiments and could enable high-field THz generation above 3 THz.

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