PHOTONICS INSIGHTS

Making and breaking terahertz waves with fluid plasmas

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Vigorous research efforts during the past several decades have successfully closed the "terahertz gap" between microwaves and infrared light, offering new and increasingly efficient ways to produce, detect, and manipulate radiation fields in the terahertz (THz) frequency range. In our laboratory, THz time-domain spectroscopy (THz-TDS) and optical-pump THz-probe (OPTP) experiments have been routinely utilized as ultrafast spectroscopy tools to investigate a variety of emerging quantum materials[1] and metamaterials[2]. The ultrafast THz pulses are produced by femtosecond laser excitation of photoconductive antennas, semiconductor surfaces (InAs), and nonlinear crystals (ZnTe, GaSe, and $LiNbO_3$). Photoconductive antennas and nonlinear crystals also allow for the coherent detection of these pulses in the time domain, with amplitude and phase spectra obtained in the frequency domain via fast Fourier transform. Although such solid-state schemes are highly desirable in many aspects and thus also commonly utilized in many research laboratories and industrial applications, they typically suffer from limited bandwidths due to the absorption and frequency dispersion induced primarily by phonon resonances, limiting THz applications such as spectroscopy of emerging materials[3], imaging and detection[4,5], and biomedical characterization[6].

A prominent solution to this problem has been to use laser-induced plasma in gases (such as air, N_2, and noble gases) wherein phonon-related absorption and dispersion can be avoided. In their new review paper[7], Chen *et al.* provide a detailed look at developments in ultrafast THz wave generation, detection, and manipulation over the past three decades in laser-induced gas plasma. Although discovered within a similar period to other solid-state THz generation and detection methods[8], this approach suffered a much slower pace of development until the finding of substantially increased efficiencies in air plasma via two-color laser excitation[9]. After discussing a suite of experimental and theoretical works and the state of the art in THz gas plasma research, the authors proceed to review plasma-based THz generation and detection in liquids[7] (e.g., water), which has only recently emerged in the past several years[10] but with rapid progress that has stimulated exciting new research directions.

In general, THz emission requires that some directionality be defined in a system to produce a net current or nonlinear polarization, which thus requires inversion (or time reversal) symmetry breaking[11]. Because fluids consist of randomly oriented molecules that preserve inversion symmetry, the directionality must be defined by the optical field(s). In a simplified picture, transverse directional charge currents can be induced within a gas or liquid plasma by overlapping two laser pulses with different frequencies, e.g., at ω and 2ω, leading to a preferred field directionality that depends on the phase delay between the two pulses. In nonlinear weak- or strong-field ionization processes, this produces a net directional charge current that radiates THz pulses in the laser propagation direction[12]. Conversely, in plasma-based THz detection, the directionality is defined by the THz field itself, which transiently biases the fluid and breaks inversion symmetry to produce a measurable second harmonic (2ω) of the coincident optical pulse (ω)[13]. The authors describe these and other strategies for generation and detection.

Gas-plasma-based THz generation and detection has matured for practical applications. It exhibits a smooth spectrum covering the entire THz frequency range, which is extremely attractive for spectroscopy and imaging applications. This broad spectral support also introduces faster THz pulses in the temporal domain, leading to higher peak THz fields (hundreds of kV/cm and more) as well as the ability to probe faster dynamics in THz spectroscopies[14]. Beyond these features, coherent control over the THz radiation can be achieved by varying the relative phase between the ω and 2ω pulses[15], including the polarization state of the THz beam by varying the incident beam polarizations to generate complex electron trajectories in the fluid plasma[16,17].

Altogether, Chen *et al.* provide a comprehensive review of gas- and liquid-plasma-based THz wave photonics, illustrating a powerful set of capabilities of ultrabroadband, intense THz radiation for spectroscopy and nonlinear optics. In doing so, they offer a perspective on a field that clearly still has much room for growth. This perspective can be expected to stimulate further advances, as long as researchers are willing to go wherever the waves may take them.

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