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## THz Emissions from Air-Plasmas Created by Midand Far-Infrared Two-Color Femtosecond Pulses

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**Abstract:** We study THz emission by two-color femtosecond filaments in air using mid- to far-infrared pump wavelengths. 3D numerical simulations show that  $10.6-\mu$ m laser pulses can produce THz fields with unequaled mJ energies and GV/m amplitudes.

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#### 1. Introduction

Terahertz (THz) science is attracting a broad interest because many molecular fingerprints belong to the THz range [1]. A well-known technique to create efficient THz emitters is to focus a two-color femtosecond light pulse into air, creating a plasma that acts as frequency converter [2]. Recent studies [3,4] showed that increasing the pump wavelength can enhance the laser-to-THz conversion efficiency which is usually limited to about  $10^{-4}$  for a 800 nm pump pulse. Moreover, it is known that the pump power contained in a filament is limited to a few times the critical power for self-focusing  $P_{cr}$  before undesired spatial breakup and multiple filamentation kicks in [5]. Because  $P_{cr}$  scales quadratically with the pump wavelength, increasing the latter makes filaments with much higher energies possible. Longer laser wavelengths, e.g., 3.9  $\mu$ m, are nowadays accessible in the femtosecond regime and energetic, 100-fs CO<sub>2</sub> laser pulses operating at 10.6  $\mu$ m should be available in the coming years [6]. We here focus on intense, broadband THz fields produced by such long-wavelength pulses. By means of a three-dimensional (3D) unidirectional solver [4], we study THz generation by two-color femtosecond pulses ionizing air. Our numerical results evidence unprecedented THz energy yields and conversion efficiencies achieved by 10.6- $\mu$ m pump pulses along extended propagation distances.

#### 2. THz spectra and supercontinuum generation of mid- and far-infrared pulses

In our simulations, the input light field has a Gaussian profile in space and time with 2.5-mm  $1/e^2$  transverse radius and 100-fs FWHM duration. It is composed of the fundamental (FH) pulse operating at the carrier wavelength  $\lambda_0$  and 10% of second harmonic (SH) in energy. The overall beam is linearly focused by a converging lens of 30-cm focal length. All pulses have an input power equal to  $1.73P_{cr}$  of the FH pulse and the dispersion map includes HITRAN data for CO<sub>2</sub> and water absorption [8]. Peak intensities of ~ 200 TW/cm<sup>2</sup> produce electron densities above  $10^{17}$  cm<sup>-3</sup>. When increasing the pump wavelength and pump energy ( $P_{cr} \propto \lambda_0^2$ ), longer filaments are promoted and they start to self-focus at shorter propagation distances. Figure 1(a) illustrates the resulting THz energy yields below 10 THz that reach the 0.1 mJ level for 3.9- $\mu$ m pumps and several mJ for 10.6- $\mu$ m lasers. Figures 1(b,c,d) detail the THz spectra computed at the distance of maximum THz field production, i.e., where the on-axis THz fields attain their maxima. Because an important question has been the role of Kerr-induced four-wave mixing compared to that of photocurrents [7], Figs. 1(b,c,d) also compare the spectra computed with or without the plasma terms before the linear focus. The presence of plasma clearly increases the THz spectral intensity by at least three orders of magnitude and shifts the frequency centroid down to 0.2 THz for the three pump wavelengths. Hence, plasma generation – even when it involves low electron densities ~ 10<sup>15-17</sup> cm<sup>-3</sup> – is here the key player in THz pulse generation.

Figures 1(e,g,i) display the entire optical spectrum developed for the three FH wavelengths along the propagation axis. When increasing the pump wavelength, high-order harmonics are produced more efficiently and form a broader supercontinuum through the plasma nonlinearity and by self- and cross-phase modulation. They are able to contribute to photoionization. For a near-IR pump (0.8  $\mu$ m), the distance of maximum THz generation coincides with that of maximum plasma generation close to focus. For larger wavelengths, maximum THz generation shifts towards shorter propagation distances. For a pump wavelength of 10.6  $\mu$ m, THz production is maximum at  $z \approx 20$  cm only, well before



Fig. 1. (a) THz energies numerically computed for two-color Gaussian pulses with FH wavelengths  $\lambda_0 = 0.8, 3.9$  and 10.6  $\mu$ m. Vertical dotted lines indicate the distances of maximum THz production. (b,c,d) THz spectra at these distances, namely, (b) z = 30 cm, (c) z = 27 cm and (d) z = 19.5 cm. Gray curves correspond to the same plotted quantities without plasma. Note the absorption line  $\sim 6$  THz due to water vapor. (e,g,i) Intensity spectra along the propagation axis. (f,h,j) On-axis THz fields obtained by filtering (20th-order Butterworth) the total field in the frequency range v < 10 THz.

maximum plasma generation sets in, which is mainly due to high-order harmonics that do no longer contribute to THz generation. Figures 1(f,h,j) reveal that on-axis THz fields induced by a 10.6- $\mu$ m pump are two orders of magnitude higher than with a near-IR pump pulse, and twice as high as with a 3.9- $\mu$ m pump pulse. Moreover, the transverse THz field profile broadens with the pump wavelength (not shown). The THz conversion efficiency obtained at 10.6  $\mu$ m is thus three times the one at 3.9  $\mu$ m, that is 3% versus 1%, respectively. In addition, we report much smaller angles of the conical THz emission and a better coupling between the two colors along extended propagation ranges, which renders filamentation setups using CO<sub>2</sub> lasers particularly promising for further applications in THz spectroscopy.

#### 3. Conclusion

In summary, we numerically showed that two-color ultrashort  $CO_2$  laser filaments can provide THz conversion efficiencies up to 3%. Because far-infrared femtosecond filaments contain hundreds of mJ and are able to self-channel over long distances along which THz production is stable, they can supply THz energy yields of several mJ. Such energy levels are record values for 100-mJ pump pulses operating at moderately high (non-relativistic) laser intensities.

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