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Letters

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## ADVERTISEMENT



## Enhancement of terahertz wave generation from laser induced plasma

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It is well known that air plasma induced by ultrashort laser pulses emits broadband terahertz waves. The authors report the study of terahertz wave generation from the laser induced plasma where there is a preexisting plasma background. When a laser beam from a Ti:sapphire amplifier is used to generate a terahertz wave, enhancement of the generation is observed if there is another laser beam creating a plasma background. The enhancement of the terahertz wave amplitude lasts hundreds of picoseconds after the preionized background is created, with a maximum enhancement up to 250% observed. © 2007 American Institute of Physics. [DOI: 10.1063/1.2719165]

Recent advances in nonlinear optics and ultrafast photonics have boosted the use of terahertz science and technology in various fields, including material characterization, nondestructive evaluation, biomedical and pharmaceutical testing, and national security applications.<sup>1</sup> However, the development of terahertz field applications has grown at a relative slow pace. One of the major issues encumbering attempts to use terahertz science and technology outside the laboratory environment is the intense water vapor absorption that exists in the ambient atmosphere. Compared to visible light, which has about 0.01 dB/km aerial attenuation, attenuation of a terahertz wave in ambient air is greater than 100 dB/km.

On the other hand, the state-of-the-art energy conversion efficiency of terahertz wave generation with the optical method is still extremely low. Martini *et al.* utilized the coherent nature of terahertz waves generated from a photoconductive antenna and succeeded in building a terahertz cavity.<sup>2</sup> In this design, superposition of a coherent terahertz wave and a coherent background can make fields add up before dephasing between these two waves sets in. By adding the background field, an enhancement (over 100%) based on coherent construction of the terahertz wave is realized.

Using air as an emitting medium to generate terahertz waves has attracted more attention in recent years<sup>3–7</sup> because of its potential applications for standoff distance terahertz wave sensing and imaging. Rather than sending the terahertz wave from a remote source, this method enables the terahertz wave to be generated close to the target. As a result, the strong water vapor absorption of terahertz waves in the ambient atmosphere is avoided and replaced by that of the optical laser beam. terahertz wave generation in the atmosphere was reported in the early 1990s by focusing an intense (peak power of 10<sup>12</sup> W) laser beam into air.<sup>3,4</sup> Through the mixing of an optical fundamental wave with its second-harmonic (SH) wave, the generation of intense terahertz wave pulses in air was later demonstrated.<sup>5-7</sup> Optical power dependence measurements across the air breakdown threshold suggested that the ionized air (plasma) plays an important role in the generation of terahertz radiation.<sup>6,7</sup> Recently, a terahertz field amplitude greater than 400 kV/cm has been reported by using a similar experimental arrangement with shorter laser pulses.

Ponderomotive force and the nonlinear process in laser induced air plasma are main mechanisms of terahertz wave generation with air medium.<sup>3,5–7</sup> Given this, one would expect to achieve stronger terahertz wave generation if a laser pulse excites on a precreated plasma background due to the conversion of more laser energy to the terahertz radiation. In this letter, we report the experimentally demonstrated enhancement of terahertz wave generation with precreated laser plasma.

Figure 1 is the schematic illustration of the experimental setup. Laser pulses from a Ti:sapphire amplifier (Spectra-Physics Hurricane i with 120 fs pulse duration, 850  $\mu$ J pulse energy, and 1 kHz repetition rate) are split into the probe beam and the pump beam. The pump beam is also split by a 50/50 beam splitter to form a Michelson interferometer. One beam is the control beam ( $I_c$ ) and the other beam is the signal beam ( $I_s$ ). The time delay between the control and signal beams is scanned by a temporal delay stage (delay-1). A mechanical chopper connected with a lock-in amplifier is placed in the signal beam path. The recombined control and signal beams are focused at the same point by a 2.5 in. focal length convex lens. The average powers of  $I_c$  and  $I_s$  at the



FIG. 1. Schematic illustration of experimental setup. An optical beam from an amplifier Ti:sapphire laser is split into three beams as probe, control, and signal beams. Both control and signal beams create plasma, where terahertz waves are generated through ponderomotive force. The signal beam is modulated by an optical chopper. Delay-1 varies the time delay between the control and signal pulses. Delay-2 is used for the probe pulse to sample the terahertz wave form.

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FIG. 2. Terahertz wave form from the plasma generated by the signal pulse and enhanced by the control pulse: (a) the control beam is blocked and the signal beam has 160  $\mu$ J pulse energy. (b) The control pulse is 11 ps after the signal pulse. [(c) and (d)] Control beam is 22 and 175 ps before the signal beam, respectively. For the curves (b), (c), and (d), both the signal beam and the control beam have 160  $\mu$ J pulse energy.

focal spot are both about 160 mW (160  $\mu$ J pulse energy).

In the experiment, the temporal delay between the control and signal beams (delay-1) is adjusted. At each specific delay of the delay-1, the probe beam is scanned (delay-2) to obtain the terahertz temporal wave form.<sup>9</sup> The terahertz radiation is measured by using a 3 mm thick  $\langle 110 \rangle$  oriented ZnTe crystal through electric optical sampling.<sup>10</sup> A lock-in amplifier analyzes the signal from the balanced detector pair. We decided not to use the four-wave-mixing method with the signal beam  $I_s$  to generate terahertz wave<sup>7</sup> in order to avoid the optical interference on the barium borate crystal. Therefore, only an 800 nm laser beam is used in the experiment.

Figure 2 plots terahertz temporal wave forms in the absence of the control beam  $I_c$  [curve (a)] and with different time delays (delay-1) between the control beam  $I_c$  and the signal beam  $I_s$  [curves (b)–(d)]. Both the control beam and the signal beam are p polarized. Here the negative timing of delay-1, illustrated by curves (c) and (d), means that the control beam leads the signal beam. Therefore, the control beam  $I_c$  creates the plasma (preionization) and the signal beam  $I_s$ generates the terahertz wave on the preionization background. Comparing curves (a) and (c) an enhancement factor of 2.5 of terahertz wave amplitude is shown when the control beam  $I_c$  leads the signal beam  $I_s$  by 22 ps. This enhancement is not due to the constructive interference of the terahertz waves generated by the control beam and the signal beam. As shown by curve (d), when delay-1 is at -175 ps, the interference between the two optical pulses and the interference between the two generated terahertz pulses are both negligible. The observed terahertz waveform is still enhanced compared to the case of curve (a) which is obtained in the absence of the control beam.

It should be pointed out that, as shown by curve (b), when the control beam arrives after the signal beam, two terahertz temporal wave forms are observed although only the signal beam is modulated and their separation is just delay-1. The phases of the two wave forms are different. In this case, when the signal beam arrives earlier, the plasma created by the signal beam acts as the ionized background and enhances the terahertz wave generated by the control beam. So the second observed wave form can be understood as the pure enhancement. Considering the lock-in amplifier is set at the phase of the mechanical chopper modulating the signal beam, while the enhancement of terahertz wave generated by the control beam is modulated by the signal beam



FIG. 3. Enhancement as a function of the relative timing between the control pulse and the signal pulse (delay-1). The dash-dot line is the zero offset and the dash line is the exponential decay fit. A 185 ps decay time is extrapolated.

induced plasma, these two wave forms have different phases.

Hamster et al. explained the mechanism of terahertz wave generation from laser induced air plasma as the effect of ponderomotive force.<sup>3,4</sup> When air is ionized by an intense laser beam, charged particles, both electrons and ions, experience ponderomotive forces near the laser focus due to the large inhomogeneity of the laser field. These forces generate a density difference between electric and ionic charges, and this separation results in a powerful electromagnetic transient, i.e., terahertz wave radiation.<sup>3,4</sup> So if one laser beam ionizes air, other laser beams will benefit from the precreated air plasma. Here we also attribute the enhancement phenomena to the ponderomotive force instead of a third order nonlinear process due to the following reasons. Firstly, other observed phenomena reveal enhanced  $\chi^{(3)}$  in air plasma<sup>11</sup> but this third order enhancement only happens in picosecond time scale and cannot explain the long time  $(10^2 \text{ ps})$  enhancement (as shown in Fig. 3, details follow). Secondly, we also observed that the enhancement is not sensitive to the control beam's polarization. When we rotated the polarization of the control beam, similar terahertz wave enhancement was also observed.

As a simple application, we used the enhancement to estimate the plasma lifetime. The method is to let delay-1 stop where the amplitude of the terahertz waveform is at its maximum, and then scan delay-2. As shown in Fig. 3, zero timing of delay-1 means that the signal and control beams are temporally overlapped, and negative delay-1 gives an earlier control beam. The dash-dot line in the figure is the zero offset. Only in the initial several picoseconds after zero timing is interference between the two terahertz waves observed. After about 20 ps, the enhancement reaches its maximum. As of this writing, the mechanism of this time characteristic is not clear to us. We believe that a possible explanation of the phenomenon is the redistribution of plasma to attain the optimum condition for enhancement. In comparison to the peak terahertz amplitude at positive delay-1, one can clearly see enhancement lasting for over 175 ps. An exponential fit of 1/e gives 185 ps decay time as shown by the dash curve. Under our experimental conditions, carrier recombination should have a stronger effect on plasma lifetime than the plasma diffusion in a radical direction. By using parameters from Ref. 12 with an estimated initial plasma density of  $2 \times 10^{17}$  cm<sup>-3</sup> and plasma temperature of 1 eV, the simulation shows a plasma lifetime of about 200 ps. This number agrees with the measured enhancement decay time of 185 ps.



FIG. 4. Dependence of the enhancement on the control beam pulse energy. The terahertz pulse energy is obtained from integrating over the square of the whole wave form. The solid curve is the power fit with slopes of 1.2 (when  $I_s$ =168  $\mu$ J) and 1.4 (when  $I_s$ =84  $\mu$ J). An estimated threshold of  $I_c$ =20  $\mu$ J corresponding to laser intensity of 10<sup>14</sup> W/cm<sup>2</sup> is observed.

Figure 4 plots the power dependence of the terahertz energy enhancement on the control beam. The delay-1 is set at -22 ps and the signal pulse energy I<sub>s</sub> at 84 and 168  $\mu$ J, respectively. At each control pulse energy level, a whole terahertz wave form is recorded by scanning the probe beam (delay-2). Then the integral over the square of the whole terahertz wave form gives the energy of the terahertz pulse. In Fig. 4, the enhancement shows the threshold behavior with an estimated value of 20  $\mu$ J of laser pulse energy corresponding to 10<sup>14</sup> W/cm<sup>2</sup> intensity at the laser focus, which is consistent with the previously reported air breakdown threshold.<sup>6</sup> This observation provides further evidence to support plasma enhanced terahertz wave generation. Furthermore, when increasing control pulse energy, enhanced terahertz pulse energy does not increase linearly. The two solid curves in the figure are the power fits of 1.2 (with  $I_s = 168 \ \mu\text{J}$ ) and 1.4 (with  $I_s = 84 \ \mu\text{J}$ ) obtained from a leastsquares fit. We explain the enhancement as a product of the preionized air plasma. When the signal beam arrives at the ionized background created by the control beam and excites all dipoles in its beam path, radiation from each dipole will coherently add up. Quantitive analysis is possible if the number of ions in the plasma background can be measured and the relationship between it and the enhancement can be studied.

In conclusion, with ambient air as the medium we demonstrated enhancement of terahertz wave generation through preionized air plasma. The amplitude of enhancement increases following the power law of the control laser beam intensity. Enhancement lasting up to 175 ps is observed. It is possible to enhance a terahertz wave generation by using plasma created by other methods such as gas discharge and laser ablation. By measuring the dependence of terahertz wave enhancement on plasma density and temperature, conditions for the enhancement can be optimized. This method can also be a tool for plasma diagnosis.

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