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Fine Control of THz radiation from filamentation by molecular lensing in air

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We demonstrate a method to control remotely the Terahertz (THz) source in air based on the bifilamentation of femtosecond laser pulses. By fine tuning the time delay between the two pulses, a significant modulation of the THz intensity from bifilamentation is observed. The phenomenon is attributed to the molecule quantum lensing effect around the air molecule revival time which changes the separation between the two neighboring plasma producing filaments.

In the past few years, considerable progress has been achieved in the development of THz radiation sources and their applications. Many applications in security [1], health [2] or art preservation [3] require THz illumination of distant targets or samples. This raises a serious problem because of the strong absorption of THz radiation by water vapor. For instance, a THz pulse around 1.5 THz is reduced by 12.05 dB within 50 m of propagation in air with humidity content of only 39.4% [4]. Recently, several approaches have addressed this issue.
One approach exploits the femtosecond filamentation process in air. The plasma column formed by a filament emits bursts of THz radiation along a forward oriented hollow cone [5]. Since it is easy to displace the filament plasma longitudinally, the THz radiation source can be placed in the immediate proximity of a remote sample. However, this type of THz source has a low conversion efficiency, on the order of 10^{-9}, and suffers from its radial polarization pattern along the emission cone which makes it unsuitable in some applications.
The drawbacks of single filamentation THz source can be corrected in several ways. By applying a transverse DC bias on the plasma column, it is possible to obtain a linearly polarized THz emission with a three orders of magnitude enhancement and a maximum intensity emitted along the laser propagation axis [6]. However this requires an external electric field at the position of the filament plasma and therefore cannot be considered a remote method *stricto sensu*. In a different approach, one can use a two-color femtosecond laser pulse, at ω and 2ω with sufficient intensity to ionize air [7, 8]. Strong enhancement of the THz radiation with peak field amplitude as high as 400 kV/cm has been reported with this method using tightly focused laser beams. However, for remote illumination filamentation is unavoidable at high laser powers $P > 5$ GW. A saturation of the THz intensity occurs because of the dispersion between the ω and 2ω components of the bicolor laser field [9].

A last technique used is called bifilamentation; it involves the creation of two closely lying filaments, using a sequence of two single color laser pulses [10]. Bifilamentation radiates up to 60 times more than the transition-Cherenkov THz radiation from a single filament. Moreover, this enhanced THz emission is radiated in the forward direction and its polarization can be easily controlled by the orientation of the two filaments. The principle of this enhanced THz radiation can be qualitatively explained with a transmission line model. After the formation of a plasma column by the first filament, which remains for a few nanoseconds corresponding to the plasma recombination time, the second filament forms its own plasma. Due to the mutual conductance and inductance, these two parallel plasma columns can be viewed as a transmission line sustaining a perpendicular current with respect to the propagation axis. The transverse current oscillating at the characteristic plasma frequency of

the columns ~ 0.5-1 THz is at the origin of efficient THz emission. As the plasma lifetime is of a few nanoseconds, the THz enhancement appears for corresponding delays between the two pulses. On the other hand, alignment between the filaments is problematic when remote illumination is the objective: THz enhancement is suppressed if the two filaments are disjoint, or if they overlap completely.

To evaluate the required alignment accuracy, we first studied the influence of the distance *d* between the two filaments on the enhanced THz radiation. The laser used was a commercial femtosecond laser system (Alpha100, Thales), which delivers horizontally polarized pulses of 50 fs with up to 15 mJ energy at a repetition rate of 100 Hz. We used a Mach-Zender interferometer with an incorporated delay line to produce a sequence of two laser pulses. The two pulses were then focused by two f = 100 cm lenses in order to reduce the distance required for onset of filamentation. A set of mirrors and a three dimensions translation stage supporting one of the lenses, allowed us to precisely align the two filaments and to adjust the horizontal separation *d* between them. The forward THz radiation was collected with a Teflon lens and sent to a heterodyne THz detector, which is sensitive to the horizontally polarized 0.1 THz component of the THz radiation with a bandwidth of 4 GHz. The detector was 20 cm away from the end of the filament. All experimental data were averaged over three hundred laser shots.

In Fig. 1, the measured THz intensity is presented as a function of the filament separation. The delay between the two pulses was set to 1 ps. The acceptance angle of the detector was set to ~ 0.8° along the filament axis. These results show that there is an optimum distance *d* between the two filaments for maximum THz emission, as expected from the bifilamentation transmission line model. This maximum appears when the two plasmas filaments are just about to be separated. This alignment is critical since the separation between the maximum and minimum of THz output is on the order of 100 μm.

Recently it has been shown that one can deflect a filament at distance by exploiting field-free refractive index revivals [11, 12]. A short intense optical pulse induces a transient alignment of dipolar molecules along the laser field direction. Quantum mechanically, most of the rotational states which have a component along the laser field will be excited with a fixed phase relationship. Similarly to the mode locking process in laser oscillator, the sum of these rotational modes produces periodic, sharp molecular alignments with a period determined by the constant of rotation of each considered molecule. Due to the different polarizabilities of the molecules with respect to their orientation, these alignments are accompanied by retarded changes of the index of refraction.

Fig. 2 shows the calculated response *R(t)* of oxygen and nitrogen as a function of time given by the expression, $R(t) = n_{2,rot} \sum_{J=0}^{\infty} F_J \sin(-\omega_J t)$, where n_{2rot} is the overall magnitude of the effect given by: $n_{2,rot} = 32\beta^2(\omega)\pi^2 N/hcn_0$, with $\omega_J = 4\pi Bc(2J+3)$ the angular frequency difference between the coupled rotational levels, *B* the rotational constant of the molecule, *N* the density of the considered gas, *h* the Planck's constant, *c* the speed of light, n_0 the linear refractive index at the laser wavelength and $\beta(\omega)$ the anisotropy of the molecular polarizability [13]. The response of the molecular gas is the coherent sum of the response of each level of rotation. F_J corresponds to the contribution of these rotation levels which have been excited by the laser pulse: $F_J = (\rho_{J+2} - \rho_J) Z_J (J+1)(J+2)/(2J+3)$, with ρ_J the population of the *J* level and Z_J is a coefficient which depends on the molecule considered and on the parity of *J*. In air, this phenomenon is mainly arising from nitrogen and oxygen molecules which have revival times of 8.4 ps and 11.6 ps respectively.

A second probe pulse will be influenced by the field free change of refractive index and will be attracted or repelled depending on the sign of the change of index [11, 12]. This suggests that one should be able to control remotely the distance between the two filaments in the bifilamentation process just by finely tuning the delay between the two pulses. This would provide a technique to correct at distance a default in the alignment of the bifilament, or to control precisely the gain of the THz radiation by bifilamentation.

In Fig. 3 (a), we present the intensity of the THz signal in air as a function of the delay between the two pulses around the half revival time of N_2. As can be seen there is a strong modulation of the THz around the half revival time of N_2 in air. Similar measurements performed in argon are shown in Fig. 3 (b) and no modulation is observed. This confirms that THz emission can indeed be controlled by the quantum wake effect since the phenomenon appears at the precise time of revival and only in molecular gases. In Fig. 3 (a), we can see that the THz radiation was not at its maximum before the time of the revival but this maximum could be reached by increasing the delay between the two pulses.

We have performed systematic measurements around half and full revival in pure N_2 and O_2 (see Fig. 4). For these experiments, the filaments separation was first optimized for maximum THz emission at a time delay without molecular lensing. As a result, a fine tuning around the revival time can only lead to a decrease of the THz radiation, since it will either way deviate from the optimum distance. There is an exception at a singular delay time when no net refractive index change occurs and for which the optimal THz signal is recovered. This is indeed the behavior observed experimentally.

To confirm that the THz intensity modulation is due to the molecule lensing effect, we have performed damage pattern experiments with glass plates. The damages allow monitoring the distance between the two filaments. Fig. 5 presents damage patterns obtained in nitrogen at three characteristic time delays specified in Fig. 4 (c). The damage in Fig. 5 (a) was taken at time delay away from the molecule alignment revival (τ_d = 8.15 ps), displaying two neighboring filaments with an optimal separation. At time delay τ_d = 8.32 ps, when the THz intensity shows a minimum (position 2), the damage pattern in Fig. 5 (b) clearly indicates a fusion of the two filaments due to positive molecule lensing effect. In the middle of the N_2 molecule revival (τ_d = 8.42 ps), the local refractive index comes back to its initial value and a maximum THz intensity is recovered (see Fig. 4 (c)).

To conclude we have demonstrated the influence of quantized molecule rotation states on the THz radiation created by two copropagating filaments. A fine tuning of the delay between two laser pulses provides an effective remote technique for optimization of the distance between two plasma channels created by filamentation and thereby of the THz intensity. An extension to multifilament patterns should be possible.

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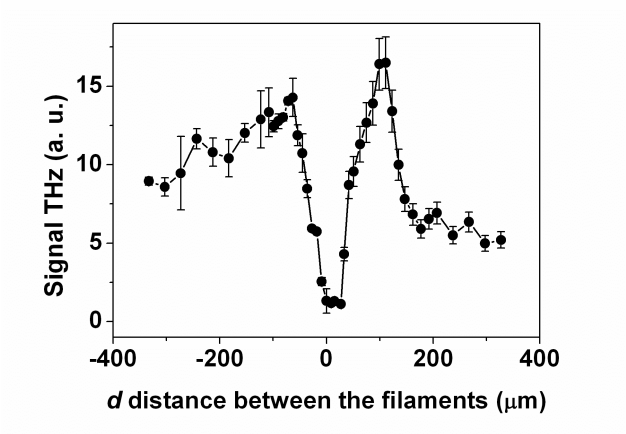


Fig. 1. THz intensity as a function of the distance between the two filaments. The two filaments were created by two laser pulses of 50 fs duration and 1.5 mJ energy. The maximum enhancement of the THz radiation is on the order of 30 as compared to a single filament. .

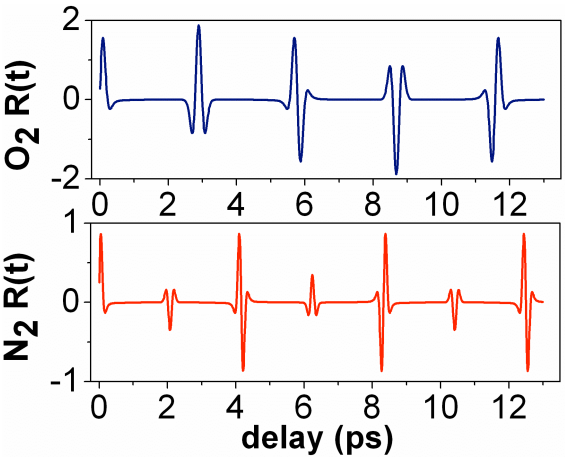


Fig. 2. Calculated response of the O_2 (top) and N_2 (bottom) molecules.

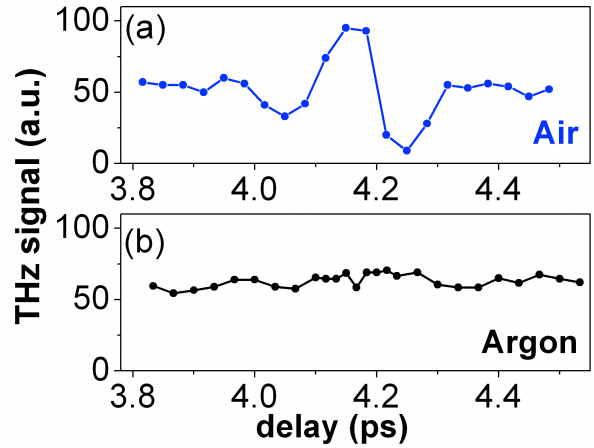


Fig. 3. THz intensity produced by bifilamentation as a function of the delay between the two laser pulses (pulse energy: 2 mJ; pulse duration: 50 fs) around the half-revival period of N_2 molecule in air (a) and in Argon (b). The enhancement factor of the THz radiation in case (a) is about 10.

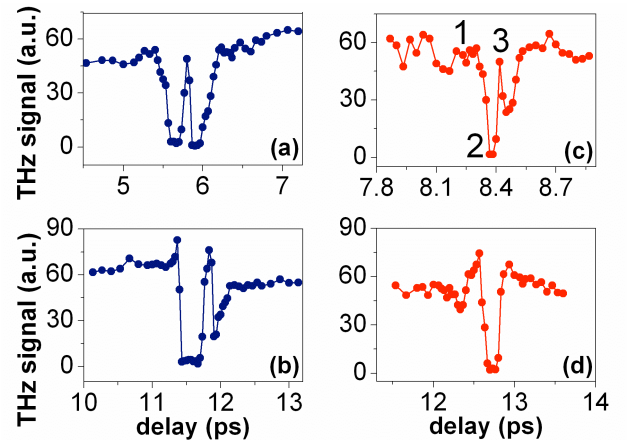


Fig. 4. (a) and (b), measurement of the variation of the THz intensity as a function of delay between the two laser pulses around the half and full revival period of O_2. (c) and (d) show the same measurements in N_2 for time around 1 and 3/2 of the revival period. Enhancement of the THz radiation is about 20 for each figure.

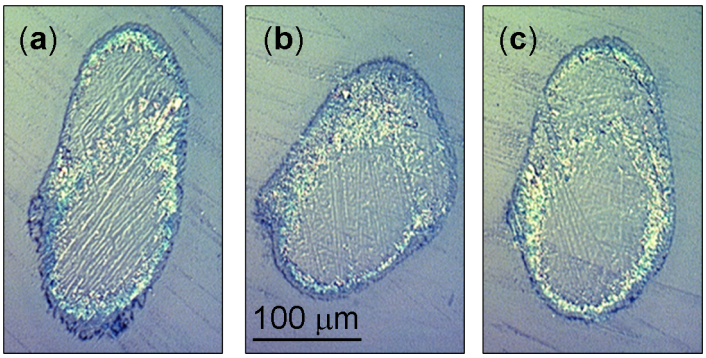


Fig. 5. (a), (b) and (c), damage patterns produced by the bifilament on a glass plate sample in N_2 at the three sequential time delays indicated in Fig. 4 (c).