Generation of terahertz pulses by photoionization of electrically biased air

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We present an experimental demonstration of the generation of far-infrared (terahertz) pulses by photoionization of electrically biased air with amplified laser pulses. The current surge following photoionization of the air with an applied bias field of 10.6 kV/cm leads to the emission of THz pulses with an intensity which can be almost as high as that of THz pulses radiated from a large-area intrinsic-field GaAs emitter. The spectra peak at higher frequency than those of biased large-area GaAs emitters. © *2000 American Institute of Physics.* [S0003-6951(00)01229-8]

The generation of intense pulses of far-infrared or terahertz (THz) electromagnetic radiation with high-energy femtosecond pulses from amplifier laser systems is usually based on electro-optic conversion in nonlinear crystals[1–5] or on photomixing in semiconductors with[6–12] and without[12] large-aperture antenna. Besides these solid-state-based conversion methods, ionization processes in laser-excited gases are known to be associated with the emission of intense THz pulses since Hamster *et al.* first demonstrated emission of THz radiation from noble gases subsequent to excitation by intense optical pulses.[13,14] In this experiment the emission of the electromagnetic transients results from the ponderomotive forces within the laser focus with peak intensities of $10^{19}\,\mathrm{W/cm^2}$ inducing an ultrafast charge separation in the plasma which leads to emission of THz pulses in a cone-shaped radiation pattern. Besides this down-conversion scheme the literature also reports an up-conversion approach for the generation of high-frequency pulses in gases[15] and semiconductors.[16] It is based on the passage of a photoionization front through a capacitor array converting energy of the dc bias field of the capacitors into an ac transient propagating with the front. In this letter, we study the up-conversion technique in air for the case of a single capacitance, which conceptually allows for the generation of the shortest THz pulses.

The experimental set-up is sketched in Fig. 1. A CLARK CPA 2001 laser system provides pump and probe (10%) laser pulses of 150 fs duration at a wavelength of 775 nm at a repetition rate of 1 kHz. The pump beam is chopped at half the laser-pulse repetition frequency. The pump pulses with a maximum energy of 500 μJ are focused into the air gap between two copper electrodes of 6 mm diameter separated by 0.8 mm from each other, thus forming a capacitor. The electrodes are not short circuited by the laser-generated plasma. A dc bias voltage of up to 850 V is applied between the electrodes, the maximum voltage being limited by dielectric breakdown of the air. The THz radiation generated in the plasma is focused on the detector by two off-axis paraboloidal mirrors. For detection, a 1 mm thick ⟨110⟩ ZnTe crystal is employed in an electro-optic detection scheme.[3] 15 time scans are averaged with a lock-in time constant of 30 ms each.

Figure 2(a) shows the measured temporal waveform of the electromagnetic transient emitted by an air plasma at a bias field of 10.6 kV/cm and at maximum pump intensity of 500 μJ. Time delay zero is chosen arbitrarily. The leading signal is followed by various oscillations over a time period of more than 10 ps which are a characteristic signature of water vapor absorption and of reflections at the electrodes. In order to compare the air plasma emitter with a standard emitter, we perform additional measurements with a semi-insulating large-area GaAs surface emitter. The energy of the pump beam remains the same as in the preceding measurement, but it is now distributed with the help of a telescope in order to illuminate the GaAs wafer of 3 cm×3 cm size. The THz radiation is rather well collimated allowing us to employ only a single paraboloidal mirror to focus the radiation onto the detector. When the GaAs wafer is not biased externally, the photomixing effect is a result of the built-in surface field alone. In order to achieve a nonvanishing transverse dipole moment the GaAs wafer is placed at an angle of 45° with respect to the beam path. The result of a measurement without external bias is depicted in Fig. 2(b). The trace con-

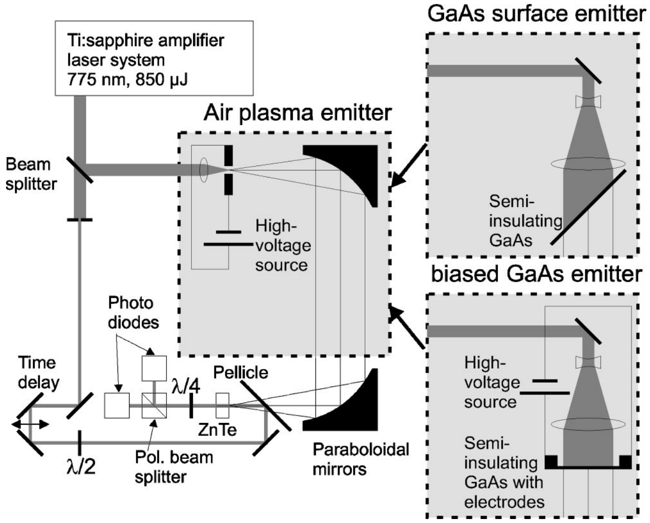

FIG. 1. Experimental set up for the detection of THz radiation from photoionized air. The boxes indicate the alternative set-ups for the detection of the emission from a GaAs surface and from the biased GaAs wafer.

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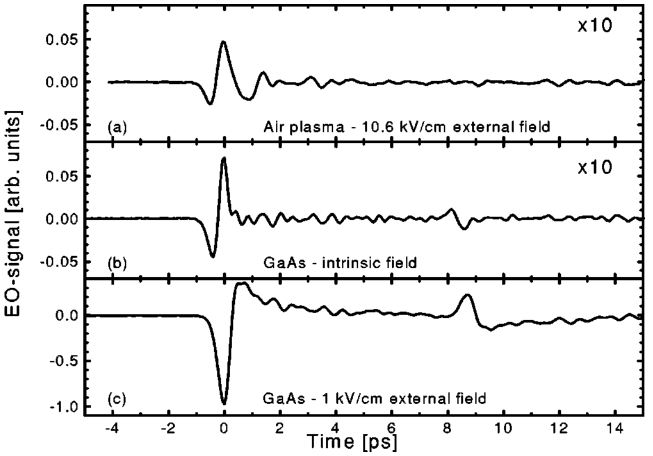

FIG. 2. (a) Detected THz waveform from the photoionized air biased at 10.6 kV/cm; (b) THz emission from a GaAs surface without external bias; (c) THz emission from a biased GaAs wafer (same scaling for all emitters).

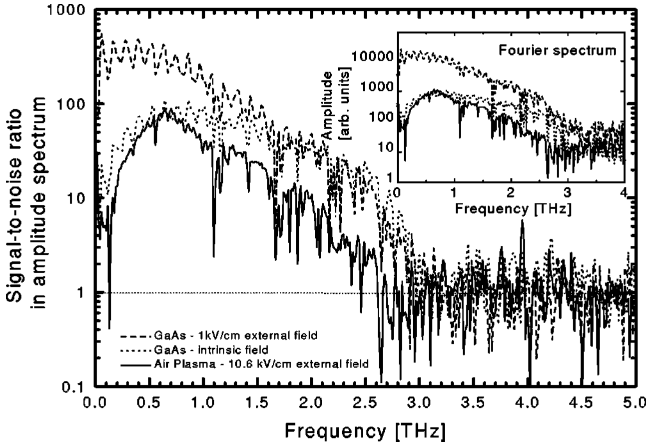

FIG. 3. Signal-to-noise ratio in Fourier spectrum of the three emitter types. The inset shows the Fourier spectrum with the same scaling for the three emitters. Both the GaAs spectra show a strong periodic modulation which is due to the reflection of the backside of the emitter in the time domain data.

sists of a bipolar transient and a signal contribution at around 8.5 ps resulting from a reflection of the THz radiation at the backside of the GaAs emitter. The peak amplitude of the THz signal is 60% larger than that from the air plasma. This is remarkable as significantly more power of the pump beam is absorbed in the GaAs wafer than in the air, where the measurement of the degree of absorption is at the limit of our detection capability. Additionally, a measurement is performed with an external field of 1 kV/cm applied horizontally to the GaAs wafer by two copper electrodes in contact to the wafer. In this case, the wafer face is normal to the beam path. The result of this measurement is shown in Fig. 2(c). The signal amplitude is about one order of magnitude higher in comparison with the radiation from biased air and from unbiased GaAs.

The temporal waveforms of the three transients in Fig. 2 show dictinct differences resulting from the different temporal evolution of the respective photoinduced polarizations $P(t)$, which determine the amplitude $E(t)$ of the emitted electric fields according to $E(t) \sim \partial^2 P / \partial t^2$. The air plasma emission indicates that the dipole moment possess an ultrafast rise and decay. Such a time dependence is likely to result from acceleration of the photogenerated charge carries in the presence of the bias field terminated either by rapid deceleration or charge recombination. In fact, recombination times of an air plasma at ambient pressure by dissociative recombination are estimated to be in the order of 1 ps.[17] In contrast the emission from unbiased GaAs is typical for the rapid build up of a polarization in the surface field with a rise time on the time scale of the laser pulse length. The polarization afterwards remains constant on a long (nanosecond) time scale. The emission from biased GaAs, on the other hand, is characterized by an expanded positive slope which indicates a decay of the polarization on a time scale of several picoseconds. This behavior is known to result from screening of the bias field in the wafer by the photocurrent.

The Fourier spectra of the three emission signals are depicted in the inset of Fig. 3. All spectra show a roll off which cuts the noise level at about 3 THz. This is a result of the detector bandwidth being limited by the phase mismatch of the THz pulse and the optical signal within the 1 mm thick ZnTe crystal.[18] Also, water absorption lines (dominant transitions are at 0.6, 1.1, and 1.6 THz) can be identified.

The important quantity for the use of any emitter in spectroscopy or imaging systems is the signal-to-noise ratio (SNR) in the Fourier spectrum, shown in the main panel of Fig. 3 in the frequency domain. The normalization to the noise floor reveals that biasing the GaAs emitter leads to an enhancement of the SNR only in the frequency range below 1.5 THz. That the SNR is not improved further is a result of the noise increase by the charging and discharging of the GaAs emitter at high bias. The plasma emitter has similar SNR characteristics to the biased GaAs in the upper frequency range from 1 to 3 THz, however with a factor 3 lower SNR value. In the lower frequency range, the plasma emitter is comparable with the intrinsic GaAs emitter.

In Fig. 4(a) the variation of the peak-to-peak amplitude of the THz radiation from biased air is depicted as a function of the applied field. Starting with an offset at zero bias the amplitude increases linearly with the applied bias field. At high bias fields, approaching the threshold for dielectric breakdown, we observe a slight deviation from the linear dependence. This is explained by an increase in the efficiency of the generation process when the bias field is close to the field ionization threshold without optical excitation.

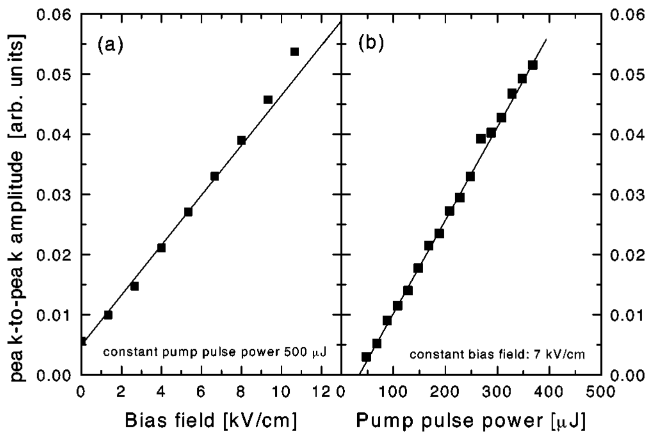

FIG. 4. Peak-to-peak amplitude of the THz signal: experimental data (points) and a linear fit (line) as a function of (a) the bias field and (b) the pump pulse energy.

The remaining emission at zero bias originates from the ponderomotive forces.[13,14] This interpretation is supported by the observation that the waveform of the THz transients is nearly independent of the bias field except close to zero bias where the pulse spectrum is shifted to higher frequencies (data not shown).

A variation of the pump-pulse energy reveals a threshold for the THz-emission process with an applied field of 10.6 kV/cm at a pump-pulse energy of about 40 μJ. Above the threshold, the relationship between the amplitude of the THz transients appears to be fairly linear with respect to the pump-pulse energy. Assuming a focal diameter on the order of 10 μm one estimates a threshold intensity of about $10^{15}\,\mathrm{W/cm^2}$ for the THz emission. Assuming a classical current-surge model where the THz emission results from the acceleration of the photogenerated charge carriers by the applied bias field, one expects a linear dependence of the emitted THz field on the number of photogenerated charge carriers provided reabsorption and reflection of the THz signal by the plasma itself are negligible. Hence, the number of free electrons would have to first exhibit a threshold behavior and then to depend linearly as a function of the pump-pulse energy in order to yield the measured relationship. In fact, various calculations and experimental data are available on the number and charge state of ions created in a pulsed-laser focus, at least for the case of noble gases if not for air.[19,20] The picture emerges that photoionization occurs predominantly by multiphoton absorption if the laser intensity is below $10^{14}\,\mathrm{W/cm^2}$, and by tunneling escape if the intensity is higher. Taking this into account, we can conclude that we observe THz radiation only when the laser intensity has risen well into the tunneling regime; at lower intensities, the number of photogenerated charge carriers apparently is so low that THz emission is below our noise floor. In the domain of tunneling escape, however, the literature data show that the number of ions created depends in a fairly linear way on the laser intensity. This is in accordance with our expectation as discussed above.

In summary, we have presented the observation of THz radiation from photoionized air under a strong bias field. The THz pulses reach amplitudes and pulse widths comparable to the THz signals from intrinsic-field semiconductor emitters. The emission is still much weaker than that from GaAs surface emitters with an external bias field applied by a large-aperture antenna. There is much room for improvement, by harder focusing, pulsed high-voltage biasing to achieve higher breakdown fields, controlling the gas pressure, and by the choice of potentially better-suited gases.

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