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Few-Cycle Laser Pulses: The Carrier-Envelope Phase, Its Role in the THz Emission from Laser-Generated Plasmas and a New Way to Measure It

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Terahertz emission from laser-generated air plasmas has recently been identified as an interesting source for THz radiation. High intensities and a large bandwidth of the THz pulses can be achieved. We briefly review several mechanisms which were employed to generate the quasi-static dipole moment needed for the optical rectification process. This leads us to a discussion of a specific application of THz emission from an air plasma, namely the investigation of the carrier-envelope phase of few-cycle optical pulses. Such pulses of a duration of less than 10 fs induce a spatial charge asymmetry in the plasma directly via non-linear tunneling ionization. The asymmetry, and with it the emission of the THz radiation from the plasma, depend on the carrier-envelope phase, with the consequence that one can determine the phase by measurement of the amplitude and polarity of the THz pulse.

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1. THz emission from laser-generated air plasmas

Inspired by the seminal work of Hamster et al. [1, 2] which had been the first to study the terahertz (THz) emission properties of laser-generated plasmas in the first half of the 1990's, several research groups began to explore this novel THz radiation source in the new century with a view towards the development of a practical emitter for high-power THz radiation. While the work of Hamster et al. concentrated on ponderomotive forces in the plasma as the driving forces needed for the radiating current surge, our subsequent work was guided by the operation principle of standard photomixer devices. There, a bias voltage is applied across a highly resistive semiconductor in which an ultrashort current surge is induced by photoexcitation of charge carriers. The semiconductor material was replaced by air which was ionized by intense laser pulses from an amplifier laser system. A current was induced via high-voltage electrodes positioned next to the plasma generation zone [3]. It was found that ambient pressure is quite optimal with respect to the THz-emission efficiency [4], with the consequence that the air plasma emitter is a very convenient THz radiation source. However, the power conversion efficiency turned out to be rather low because of ultrafast screening of the bias field by the plasma itself. THz fields of 1 kV/cm were measured for pump laser pulses with an energy of 400 μJ and a pulse duration of 150 fs (pulse repetition rate: 1 kHz) [5].

A way to overcome the screening is to apply an ultrahigh-frequency "AC bias" instead of a DC voltage to the plasma. This frequency has to be larger than the plasma frequency. With a molecule density of air of 2.4×10^{19} cm^{-3}, the plasma frequency ν_p of a fully ionized air plasma (one electron per molecule ionized) at ambient conditions is 44 THz. A "bias modulation" at optical frequencies is achieved if the laser pulse is superimposed by its own second harmonic radiation. This approach was first implemented by Cook and Hochstrasser [6] and has been applied in a number of experiments to be addressed later. The laser pulse propagates through a nonlinear crystal before generating the plasma in its focus in air. The second-harmonic (SH) signal generated in the crystal is directly superimposed on the fundamental wave thus creating an asymmetry of the temporal waveform. For a suitable phase between the two waves, the total field is stronger for one polarity than for the other. This translates into an asymmetry of the ionization process in air in that way that the ionization rate is larger for one field polarity than for the other, resulting in a quasi-DC unidirectional net current in the plasma oriented perpendicular to the propagation direction of the optical pulse.

The emission of THz radiation by this current is substantial. While the first studies indicated an enhancement of the THz field strength by one order of magnitude over that of the DC-bias variant reaching values on the order of 10 kV/cm [5–7], subsequent work has shown that the conversion efficiency can be much larger. It appears that an important requirement is a shorter width of the optical pulses ($<$ 50 fs). The literature reports of THz field amplitudes of 400 kV/cm for plasma excitation with 25 fs pulses from a 1 kHz Ti:sapphire laser (maximal pulse energy: 0.5 mJ) [8], and of 150 kV/cm if the plasma is generated with 50 fs pulses from a 10-Hz Ti:sapphire laser (maximal pulse energy employed:

20 mJ) [9]. The data base is not very consistent yet, leaving many questions open with respect to the differences in the values obtained for the THz field strength, the exact temporal and spatial shape of the THz radiation, its bandwidth, and the exact physical mechanisms at work.

However, it is undisputed that the performance of this novel THz source is superior to that of semiconductor-based high-THz-field sources. For comparable laser parameters, the achieved field amplitude is at least as large [5], the bandwidth is higher, and the THz emitter (with the exception of the nonlinear crystal used for SH generation) is indestructible, quite in contrast to semiconductor sources. It is highly probable that it will replace the latter in experiments where intense broadband THz pulses are required [10, 11]. The versatility of air-plasma-based THz systems has been underscored by experiments of the group of Zhang, who has demonstrated that not only THz emission, but also the detection of THz pulses is possible via SH generation in an air plasma [12]. Also, enhancement of THz waves in plasmas has been observed [13]. For an overview of the state of the art of THz-pulse generation by laser-induced ionization of gasses, see Ref. [14].

Plasma-based THz emission remains a vibrant field of THz research. Aiming at an optimization of the plasma emitter, researchers explore different atomic and molecular gaseous substance (such as alcohols) and investigate the influence of pressure, temperature, etc. [15]. Solid targets in vacuum are being discussed because they should allow to generate denser, less extended plasmas with less possibility for destructive interference for the generated THz waves (an aspect to consider being the concomitant emission of intense X-rays which may be the cause of practical concern if this emission is not desirable for research). Novel THz-generation mechanisms are being presented for inhomogeneous plasmas generated in gas jets [16] and for laser-induced beam filaments in air [17]. Beyond these advances, the strong nonlinear-optical properties of plasmas hold much promise for the optimization of THz emission especially if it should become possible to utilize nonlinear effects for a self-organized optimization of the emission process. The self-compression of optical pulses in filaments is such an effect [18, 19] which has been proposed to deliver THz pulses remotely which is possible because of the fact that a suitably chirped pulse generates THz radiation strongly only after it has shortened sufficiently after some distance of travel [17].

2. THz emission and the carrier envelope phase

In the following, we address another mechanism for THz emission from laser-ionized air which is unique for extremely short optical pulses of a duration of not more than 10 fs. This mechanism does not require superposition of a SH pulse on the fundamental pulse. The asymmetry needed to generate a quasi-DC dipole moment for THz emission results directly from the ionization dynamics of few-cycle laser pulses themselves. The emission depends on the carrier-envelope (CE) phase of the pulses which opens the way to measure it via THz-emission techniques [20].

Figure 1 (upper parts) displays bandwidth-limited optical pulses of a duration of 6 fs and a center wavelength of 800 nm. The pulses differ by the CE phase, i.e., the relative phase between the carrier wave and its envelope function. If such pulses are focussed into air, this difference has strong implications for the photo-ionization processes of the atoms and molecules of air. This is illustrated in the lower parts of Fig. 1. Here, we plot the ionization rate predicted by static tunneling theory which, for the conditions of our experiments, is known to describe ionization well. In this theory, the ionization rate depends only on the laser beam intensity at any given moment in time, and not on the history of the pulse or of the electrons in the atoms and molecules. Substantial ionization during a cycle of the laser carrier wave occurs for laser intensities exceeding 10^{14} W/cm^2. The laser field strength for which the probability is 1% per fs is 3.3×10^{10} V/m. This value is indicated in the upper parts of Fig. 1 as the arbitrarily defined ionization threshold. The lower parts show the corresponding ionization rate as a function of time. The rate repeatedly peaks at the laser-field maxima and minima.

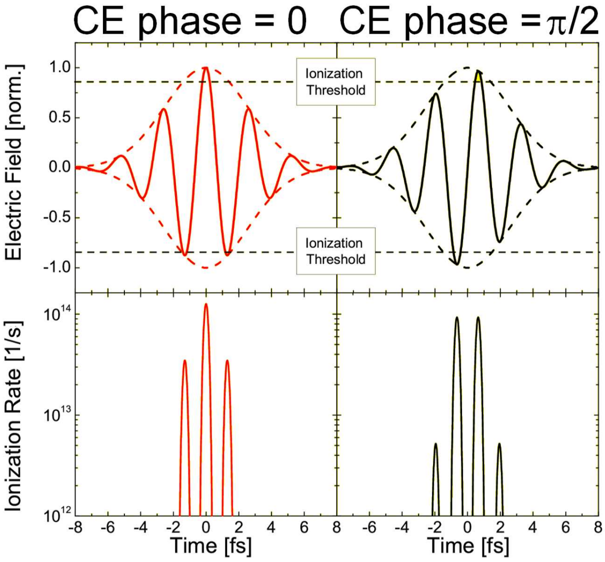

Fig. 1. Upper parts: Gaussian-shaped few-cycle optical pulses with different carrier envelope phases. Lower parts: Temporal dependence of the ionization rate for a pulse focussed into air. It is assumed that the air consists only of N_2 molecules.

While the rate as a function of time is symmetric with respect to $t = 0$ (defined as the time when the pulse envelope is maximal), there is a fundamental difference for the two CE phases if one divides the ionization processes into two groups, those occurring at positive, respectively negative field lobes (the signs indicating the sign of the electric field in Fig. 1). For CE phase $\varphi_{\text{CE}} = 0$, the total number of ionization processes for positive field lobes is larger than the total number of processes occurring for negative field lobes. For $\varphi_{\mathrm{CE}} = \pi/2$, the total numbers are equal.

Considering that the electrons are ejected in different directions for positive and negative field orientations, the total electron current calculated by temporal integration over the pulse duration turns out to depend on the CE phase. The time derivative of the current, however, determines the radiation field of the THz transient emitted from the plasma.

While the current depends also on the propagation of the electrons after ionization [9, 14], we can illustrate a basic CE phase dependence already by an evaluation of the ionization rates. We define an asymmetry parameter $R_\rho = \lim t_{t\to\infty}\left((\rho^+ - \rho^-)/(\rho^+ + \rho^-)\right)$, with ρ^+ and ρ^- being the densities of free electrons produced at positive and negative laser-field polarities. Figure 2 shows results of model calculations for R_ρ as a function of the CE phase for various pulse durations. R_ρ directly reflects the phase — in a cosine-like manner if the total plasma density is not too high. The contrast, however, rapidly decreases with rising pulse width, essentially vanishing for pulse durations above 10 fs.

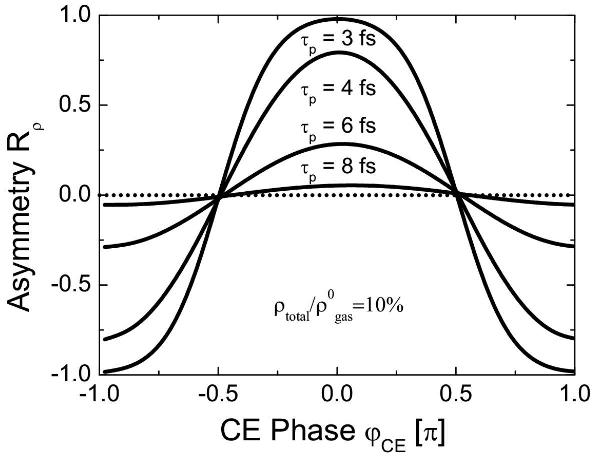

Fig. 2. Asymmetry parameter R_ρ of the plasma density vs. CE phase for several pulse durations. R_ρ is proportional to the macroscopic quasi-DC dipole moment which is the origin of the THz emission. The pulse intensity is chosen such that a plasma density ρ_{total} of 10% of the molecule density ρ^0_{gas} is obtained.

In Ref. [20], we have shown experimentally for the case of 8 fs pulses from a 2.8 kHz Ti:sapphire-amplifier-based laser system that the amplitude and polarity of the THz radiation emitted from a laser-induced air plasma indeed follows the prediction of a cosine-shaped CE-phase dependence.

In many experiments with few-cycle laser pulses, especially those of the attosecond kind, where phenomena occurring on time scales of less than an optical cycle are studied, the CE phase plays a major role. Its knowledge and control are of prime importance for the correct performance and interpretation of the experiments [20]. A number of techniques exist for the measurement of the CE phase, the most well-known ones being f-to-$2f$ interferometry and ATI (above-threshold ionization) [21]. THz emission from plasmas could develop into an alternative approach. For it to develop into a practical tool for CE-phase determination, however, the low THz signal strength which we have obtained in the measurements of Ref. [20] must be improved considerably. Experiments are under way aiming at a strong signal enhancement by measures such as THz detection with a much broader bandwidth, and transition to solid-state targets in vacuum instead of air for plasma generation.

We want to address now another question, namely that relating to the distinction between *absolute* and *relative* CE phase determination. If a technique is said to determine the absolute phase, this implies that the CE phase at the location of an experiment can be determined at least in principle by CE-phase-probing measurements occurring typically at a different location. A relative measurement only allows to determine changes of the phase, without the possibility to determine the absolute value of the CE phase at the experiment. ATI is often considered a technique which allows absolute phase determination (whereas f-to-$2f$ interferometry is not) [21]. ATI measures the directional asymmetry of ionization events occurring in an atom jet[*]. It fulfills two requirements: one has (i) a well-defined spot where the measured physical process occurs, and (ii) has only linear optical components in the beam path to the experiment. Both aspects together allow to principally calculate the CE phase at the location of the experiment from the measured data. Strictly speaking, the absolute phase determination should also occur on a pulse-to-pulse basis in order to capture any abrupt changes of the CE phase. This is not, however, how the techniques generally work in practice because the signal-to-noise ratios are not good enough.

With respect to the THz-emisison approach, it is not clear at present whether it could develop into a technique for absolute CE phase determination. At present, both requirements mentioned in the preceeding paragraph are challenging. First, the size of the plasma is not negligible with the consequence that the locality of the measured effect is not well defined. Second, THz phase distortions by nonlinear phase distortions of the incoming optical beam cannot be excluded with certainty yet. It is known that self-focusing and defocusing effects occur in air, respectively air plasmas, and their existence raises doubt with respect to the CE phase traceability from the measurement location to that of the targeted experiment.

Another challenge remains to be addressed, namely the assignment of the polarity and amplitude of the THz signal to the CE phase of the incoming laser

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[*] In this asymmetry aspect, there is a considerable conceptual similarity between ATI and THz emission from a plasma with the main practical difference that ATI works in vacuum and with gas jets whereas THz emission requires (at least in its present implementation) a denser medium to generate sufficient signal strength.

pulse generating the plasma. While Fig. 2 has linked the asymmetry factor R_ρ to the CE phase, this relationship will differ from that between the measured THz signal and the CE phase. The challenge for a correct assignment is illustrated with the help of Fig. 3 which displays result of more extensive model calculations where not only the asymmetry of the ionization is taken into account but also the subsequent trajectories of the ejected electrons under the influence of the optical electric field [14, 22]. The maximal polarization does not occur for $\varphi_{\mathrm{CE}} = 0$ any more but is shifted by nearly $\pi/2$. In summary, CE-phase-dependent THz emission from laser-induced plasmas has a good chance to become a versatile tool for CE-phase determination if the sensitivity can be enhanced significantly. It is an approach which is comparatively easy to implement. The capability to determine the absolute CE phase should exist at least for plasmas generated from solid targets in vacuum.

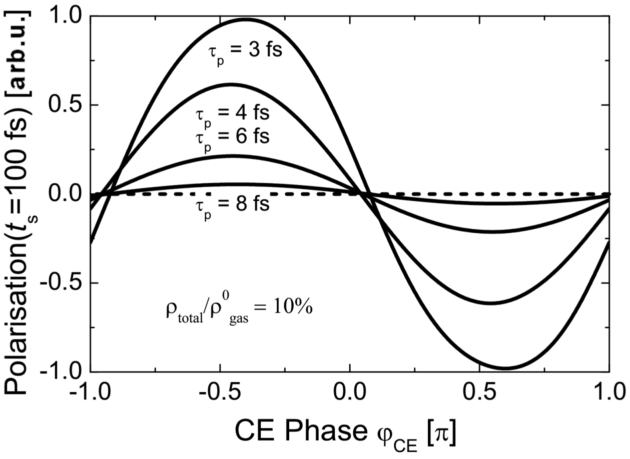

Fig. 3. Plasma polarization as a function of the CE phase calculated with the same optical beam parameters as the data shown in Fig. 2. The trajectories of the electrons after the ionization events are taken into account.

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