Phase-sensitive terahertz emission from gas targets irradiated by few-cycle laser pulses

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Abstract. The effect of the carrier envelope phase (CEP) of few-cycle laser pulses on terahertz (THz) emission from gas targets is investigated by analysis and two-dimensional particle-in-cell simulations. For linearly polarized (LP) light, the THz amplitude depends on the CEP phase sinusoidally. For circularly polarized (CP) light, the THz amplitude is independent of the phase, but its polarization plane rotates with the phase. By measuring the THz amplitude or polarization direction, one can determine the CEP of LP or CP laser pulses, respectively. We find that when the ionization degree of atoms is lower than 10\%, the phase dependence of the THz radiation is insensitive to intensity and duration of the laser pulse, which is preferable for the phase determination.
1. Introduction

Recently, terahertz (THz) emission was observed from ionized air when irradiated with a two-color (fundamental and second-harmonic) laser pulse [1]–[4] and also when hit by a single few-cycle laser pulse [5]. This THz emission is sensitive to the relative phase of the two-color waves and to the absolute phase of the few-cycle pulse, the so-called carrier envelope phase (CEP). Both the experiments were interpreted in terms of four-wave mixing (FWM). However, it was noted in [4] that the third-order susceptibility of air responsible for FWM is too small to generate the observed THz emission with field strength greater than kV cm$^{-1}$. Alternatively, ionization-induced current models [6, 7] have been proposed to explain this THz emission. These models predict that electrons released through tunneling ionization acquire transverse momenta while the laser pulse passes. The moving electrons form the THz radiating current.

The present work is motivated by the application of the THz emission to measurement of the absolute phase of few-cycle pulses. This method has been demonstrated experimentally in [5]. Absolute phase determination is crucial for attoscience [8]. The state-of-the-art approach of CEP measurement is stereo above threshold ionization (ATI) [9]. The THz-emission method is simpler than stereo ATI. Here, we investigate the THz emission generated by the few-cycle laser pulse interaction with gas targets, using two-dimensional particle-in-cell (2D-PIC) simulation and taking full account of tunneling ionization. We then compare the simulation results with the model of ionization-current-induced THz emission, which is developed in section 2 on a generalized basis, making use of constant transverse canonical momentum. The essential new point is that we can explore the phase-sensitive THz emission as a function of laser intensity, pulse duration and polarization, thereby testing the limits of the model description and identifying the best parameter regions for CEP measurements. These results are presented in section 3. For linearly polarized (LP) light, the THz amplitude depends on the CEP sinusoidally. For circularly polarized (CP) light, the THz amplitude is independent of CEP and its polarization plane rotates with the phase. By measuring the THz amplitude or polarization direction, we can determine the absolute phase of either LP or CP laser pulses.

The phase-sensitive THz emission depends critically on the ionization threshold. We find that for ionization degrees lower than 10%, the phase dependence of the THz radiation is insensitive to the intensity and duration of the laser pulse. This is favorable for the phase determination. The present simulations may also be used to study the best regimes for powerful generation of THz radiation [7].
The peak amplitude is proportional to the dipole acceleration

\[ \text{THz amplitude has a maximum along the light axis both in forward and backward directions.} \]

one then obtains the final momentum of the electron

\[ \text{pulse has passed, the field is} \]

\[ n \]

in a plane wave, we have the constant of motion \( -p_{\perp} \) transverse momentum of the electron, \( \text{ionization degree of atoms} \]

\[ \text{propagation can be neglected. The present calculation is similar to the quasi-static description} \]

\[ n \]

of ATI \[ \text{that the plasma density is sufficiently small so that the effect of plasma dispersion on light} \]

\[ \text{It builds up in a short time interval} \]

\[ \text{2. Model} \]

The mechanism of phase-sensitive THz emission under few-cycle laser irradiation can be divided into three steps. Firstly, electrons are ionized by tunneling. Secondly, the electrons get transverse momentum in the laser field. Finally, all moving electrons form an oscillating electric dipole, which emits THz radiation after the laser pulse has passed. The THz spectrum peaks at the plasma frequency of the ionized plasma volume.

At a certain space point, the temporal evolution of the electron density \( n_e \) is

\[ \frac{dn_e(t)}{dt} = W_{i}(t)n_a(t), \]

where \( W_{i} \) is the tunneling ionization rate depending on the instantaneous electric field \( E \), and \( n_a \) is the density of neutral atoms. Equation (1) has the solution \( n_e(t) = n_0 - n_a(t) \) and \( n_a(t) = n_0 \exp(-\int_{-\infty}^{t} W_{i}(t') \, dt') \), where \( n_0 \) is the initial atom density. We define the final ionization degree of atoms \( W_{i} = n_e(t = \infty)/n_0 = 1 - \exp(-\int_{-\infty}^{t} W_{i}(t') \, dt') \).

Once the electron is released, its motion is governed by the laser field. For a free electron in a plane wave, we have the constant of motion \[ \mathbf{p}_{\perp}(t) - e\mathbf{A}_{\perp}(t) = \text{const.}, \]

\[ \mathbf{p}_{\perp} \]

is the transverse momentum of the electron, \( \mathbf{A}_{\perp} \) is the transverse vector potential of the laser pulse, and \( -e \) is the electron charge. At the electron’s birth time, one has \( \mathbf{p}_{\perp}(t = t_0) = 0 \). After the laser pulse has passed, the field is \( \mathbf{A}_{\perp}(t = \infty) = 0 \). Using the conservation of canonical momentum, one then obtains the final momentum of the electron

\[ \mathbf{p}_{\perp}(t = \infty) = -e\mathbf{A}_{\perp}(t = t_0). \]

It is determined by the vector potential felt by the electron at the birth time. Equation (2) is valid for laser duration \( T \ll \omega_p^{-1} \), where \( \omega_p = \sqrt{n_e e^2/m\varepsilon_0} \) is the plasma frequency. It is also assumed that the plasma density is sufficiently small so that the effect of plasma dispersion on light propagation can be neglected. The present calculation is similar to the quasi-static description of ATI [11].

Within the laser pulse, the current increment at time \( t \) is \( d\mathbf{J}_{\perp}(t) = -e(\mathbf{p}_{\perp}(t = \infty)/m) \)

\[ \frac{d\mathbf{n}_e(t)}{dt} = (e^2/m)W_{i}(t)n_a(t)\mathbf{A}_{\perp}(t) \, dt. \]

Integrating over the pulse duration \( 0 \leq t \leq T \), we obtain the current density at this space point

\[ \mathbf{J}_{\perp}(\mathbf{r}) = \frac{e^2}{m} \int_{0}^{T} W_{i}n_a\mathbf{A}_{\perp} \, dt. \]

Considering an ionized plasma volume with size much smaller than the plasma wavelength \( \lambda_p = 2\pi c/\omega_p \), we can integrate the current density \( \mathbf{J}_{\perp}(\mathbf{r}) \) over the whole plasma volume and obtain the initial dipole acceleration for THz emission

\[ \frac{d\mathbf{D}_{\perp}}{dt} = \frac{e^2}{m} \int d^3\mathbf{r} \int_{0}^{T} dt W_{i}n_a\mathbf{A}_{\perp}. \]

It builds up in a short time interval \( T \ll \omega_p^{-1} \). The dipole \( \mathbf{D}_{\perp} \) then oscillates due to the charge separation potential and radiates THz waves with the central frequency of \( \omega_p/2\pi \). The emitted THz amplitude has a maximum along the light axis both in forward and backward directions. The peak amplitude is proportional to the dipole acceleration

\[ E_{\text{THz}} \propto d\mathbf{D}_{\perp}/dt. \]
It is noted that the first peak electric field of the THz pulse is proportional to $-d\vec{D}_\perp/dt$. The dipole $\vec{D}_\perp$ obeys the periodicity $\vec{D}_\perp(\varphi + \pi) = -\vec{D}_\perp(\varphi)$, where $\varphi$ is the phase of the laser pulse.

The present model slightly differs from [7] in that we use the vector potential $\vec{A}_\perp$ to evaluate the final velocity of the electron and do not integrate over the electric field $\vec{E}_\perp$. Equation (4) permits a coarser time step in the numerical integration, since the electron momentum deduced from $\vec{A}_\perp$ is exact. Equation (2) is exact even for relativistic laser intensities and includes the effect of the magnetic fields. So the present model is expected to be correct also for higher intensities.

In previous papers [5]–[7], the static rate of tunneling ionization by Ammosov–Delone–Krainov (ADK) [12] for $W_i$ was used in equation (1). However, it is well known that the ADK formula is inaccurate for above-the-barrier ionization at higher laser intensities [13]. We therefore use empirical expressions for the ionization rates derived for the H atom by the numerical solution of the Schrödinger equation. They cover the two electric field regions $E < 0.16E_a$ [14] and $0.15E_a < E < 0.5E_a$ [15], where $E_a = 5.14 \times 10^{19}$ V cm$^{-1}$ is the atomic unit of the electric field. Joining the two regions at $E = 0.14E_a$, we have the following combined empirical ionization rate for H:

$$W_{i,H} = \begin{cases} 4.13\omega_a\exp(-2/3E' - 12E')/E', & E' < 0.14, \\ 2.4\omega_aE'^2, & 0.14 \leq E' \leq 0.5, \end{cases}$$

where $E' = E/E_a$ and $\omega_a = 4.13 \times 10^{16}$ Hz is the atomic unit of frequency. For the ionization of He $\rightarrow$ He$^+$, [13] gives ionization rates calculated from the Schrödinger equation. For the ionization of He$^+$ ions, one has an empirical formula similar to equation (6) [15].

3. PIC simulations and results

In order to test the model outlined above, we have conducted a series of 2D-PIC simulations for H and He gas. Our code follows the procedure of [16] to implement ionization, using the rates discussed above. Collisional ionization can be neglected because of the low densities considered here. For solving Maxwell’s equations, a numerical scheme is used free of dispersion in the laser propagation direction [17]. This point is found to be important for propagating few-cycle pulses with constant CEP. The simulation box is $200\lambda \times 200\lambda$ in the $x\gamma$-plane, and cell numbers per $\lambda$ are 20 and 16 in the $x$- and $y$-directions, respectively. We set the laser wavelength to $\lambda = 0.8\mu$m. The laser pulse propagates along the $x$-direction and has the temporal profile of $a_0\sin^2(\pi t/T)\cos[\varphi(t - T/2)]$ for $0 \leq t \leq T$. Here, $\varphi$ is the laser angular frequency and $\omega$ is the laser angular frequency and $\omega$ is the laser angular frequency and $\varphi$ is the CEP. $a_0 = \varepsilon E_0/m c$ is the normalized laser electric field, which is related to the light intensity through $I = a_0^2 \cdot 1.37 \times 10^{18} (\mu m \lambda^{-1})^2$ W cm$^{-2}$. The region of relativistic intensities is reached for $a_0 \geq 1$. The vector potential of the laser pulse can be obtained from $\vec{E} = -\partial \vec{A}/\partial t$. The laser pulse is focused on the center of the gas target (see below). The duration and focal radius of the laser pulse are $T = 4\tau$ (FWHM is 4.1 fs) and $R = 5\lambda$, respectively, where $\tau = \lambda/c$ is the light cycle time.

The initial gas target is rectangular and is located at the center of the simulation box. The gas target has length $L = 10\lambda$, and the width is chosen always making sure that it is larger than the width of the ionization-generated plasma. Atomic densities $n_0$ of H and He are 0.0025$n_c$ and 0.00125$n_c$, respectively, where $n_c = m_e\omega^2/e^2 = 1.1 \times 10^{21} (\mu m \lambda^{-1})^2$ cm$^{-3}$ is the critical plasma density beyond which the laser cannot propagate. After full ionization with $W_{i,H} = 1$, the plasma density is $n_e = 0.0025n_c$, corresponding to a plasma wavelength $\lambda_p = \lambda\sqrt{n_e/n} = 20\lambda$. 

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Figure 1. THz emission from the H gas target for an LP laser pulse with $a_0 = 0.02$. (a) The backward THz pulse for the phase $\varphi = 0^\circ$. The circle marks the first peak $E_{y,\text{peak}}$ of this THz pulse. (b) The first THz peak $E_{y,\text{peak}}$ as a function of the phase $\varphi$. 2D (dotted line) and 3D (solid line) model calculations of $-dD_y/dt$ are based on equation (4). The circles are PIC results.

and a plasma frequency $\omega_p/2\pi \approx 18.7$ THz. Since the Rayleigh length of the laser pulse is $L_R \approx 78.5\lambda \gg L$, the dipole $\vec{D}_\perp$ is uniform in the $x$-direction.

In the following, we only analyze THz radiation emitted in the backward direction. In the forward direction, it is mixed with the low-frequency wing of the incident laser pulse. This problem arises because the few-cycle laser pulse used in the present simulation has a broad spectrum overlapping with the THz region. Realistic experimental pulses typically have a more narrow spectrum, well separated from THz frequencies so that in real experiments the THz emission can also be studied in the forward direction. Below, the electric field $E_y$ is traced on the light axis in the backward direction up to a position of $45\lambda$ off the target surface. There are still components of the laser pulse in the backward $E_y$, because of the reflections at the two boundaries of the gas target. These light-frequency components are weak and can be easily filtered numerically.

3.1. Linear polarization

Figure 1 shows THz radiation from a H gas target for a laser amplitude $a_0 = 0.02$, corresponding to an intensity of $I = 8.6 \times 10^{14}$ W cm$^{-2}$. At the end of the laser pulse, 96.7% of the H atoms close to the light axis are ionized. The radius of the ionized plasma column is $R_p \approx 4.5\lambda$. A typical THz pulse is shown in figure 1(a) for the phase $\varphi = 0^\circ$. This THz pulse is effectively single-cycle, and its magnitude approaches $0.12$ MV cm$^{-1}$. The circles in figure 1(b) give the first peak (also marked in figure 1(a) for $\varphi = 0^\circ$) as a function of $\varphi$ and are compared with model calculation ($-dD_y/dt$) based on equation (4). The dotted curve is obtained when evaluating the volume integral in equation (4) for 2D geometry, and the solid curve corresponds to 3D geometry. The difference between the 2D and 3D model calculations lies in the spatial integration in equation (4), which leads to $L \int_{-\infty}^{\infty} dy \, dz$ (2D) or $2\pi L \int_0^{\infty} r \, dr$ (3D, $r = \sqrt{y^2 + z^2}$), respectively. One finds that the peak amplitude $E_{y,\text{peak}}$ depends on $\varphi$ sinusoidally. The 2D
Figure 2. THz emission from the He gas target for an LP laser pulse with $a_0 = 0.2$. (a) The backward THz pulse for the phase $\varphi = 60^\circ$. (b) The first THz peak $E_{y,\text{peak}}$ as a function of the phase $\varphi$.

Simulation results naturally agree with the 2D model. There is a phase shift between the 2D and 3D model calculations, which is due to different intensity distributions of 2D and 3D laser beams in the $yz$-plane. One may notice that the space integration in equation (4) requires a focused laser pulse, even though equation (4) was derived from equation (2) for a plane wave. This is justified because equation (2) is a good approximation for a focused laser pulse at low intensities, since the oscillating amplitude of electrons is small and the electric field felt by electrons is approximately uniform in the transverse direction.

Next, let us consider a He gas target irradiated with a higher intensity laser with $a_0 = 0.2$ ($I = 8.6 \times 10^{16}$ W cm$^{-2}$). During the first 1.5 cycles of the laser pulse, both electrons of He atoms in the focal region have been released, and the transverse plasma radius is $R_p \approx 7\lambda$. Figure 2(a) shows the resulting THz pulse with an amplitude of $2.8$ MV cm$^{-1}$ for $\varphi = 60^\circ$. The THz peak $E_{y,\text{peak}}$ as a function of the phase $\varphi$ in figure 2(b) shows good agreement between simulations and model calculation. We also find that the amplitude ratios of THz pulses in figures 1(a) and 2(a) are in agreement with the model. From figure 2, we can learn that the model is at least correct for $a_0 \leq 0.2$.

By measuring the function $E_{y,\text{peak}}(\varphi)$ shown in figures 1(b) and 2(b), the absolute phase of the few-cycle pulse can be determined, as was done in [5]. This function depends on the intensity, duration and shape of the laser pulse. For determining the CEP, it is necessary to find a region where the function $E_{y,\text{peak}}(\varphi)$ is weakly dependent on these laser parameters. Figure 3(a) shows 3D model calculations of $-dD_y/dt$ for H gas at different laser intensities. We find that $E_{y,\text{peak}}(\varphi) \propto -\sin(\varphi + \varphi_0)$, where the offset $\varphi_0$ is plotted as a function of laser amplitude $a_0$ in figure 3(b). One observes $E_{y,\text{peak}}(\varphi) \propto -\sin(\varphi)$ with offset $\varphi_0 = 0$ for $a_0 \leq 0.01$. We find that this constant $\varphi_0$ occurs when the final ionization degree $W_\text{fi}$ is smaller than 10% (see figure 3(a)). As shown in figure 3(b), the critical field strength for the constant $\varphi_0$ region decreases with the laser duration, since more atoms are ionized for the longer pulse under the same intensity. To easily understand the distribution $E_{y,\text{peak}}(\varphi) \propto -\sin(\varphi)$ with $\varphi_0 = 0$, we show the temporal evolution of laser vector potential and ionized electron density for $\varphi = 0^\circ$ and 90$^\circ$ in figures 3(c) and (d), respectively. Here, the laser pulse has $a_0 = 0.008$ and $T = 4\tau$, and the final ionization
Figure 3. 3D model calculation of THz emission from the H gas target for an LP laser pulse. (a) $-dD_y/dt$ for different laser amplitudes. The values in square brackets are the laser amplitudes and corresponding ionization degrees $W_{fi}$ (in percent) of H atoms near the laser axis after the laser pulse has passed. The laser pulses have $T = 4\tau$. (b) $\varphi_0$ as a function of laser amplitude. Two cases of $T = 4\tau$ and $5\tau$ are plotted. (c) and (d) The vector potential $|A_y|$ (dashed lines for positive branches and dotted lines for negative ones) and the instant ionization ratio $dn_e/dt$ (solid lines) for absolute phase (c) $\varphi = 0^\circ$ and (d) $\varphi = 90^\circ$. The laser pulse has $a_0 = 0.008$ and $T = 4\tau$. In all frames, we take $R = 5\lambda$.

of H atoms close to the laser axis is $W_{fi} = 1.4\%$. Because ionization is small, the ionization peak is the same as for the field peak of the laser pulse and is symmetric around the field peak. For the cosine pulse in figure 3(c), one finds that the integration over $A_y dn_e$ is exactly zero. For the sine pulse in figure 3(d), however, the integration over the two main peaks of ionization is nonzero and negative. For higher laser intensities, ionization mainly occurs at an earlier stage of the laser pulse. For example, the third ionization peak in figure 3(c) will become weaker and even disappear, which will lead to an effective dipole and cause $E_{y,\text{peak}}(\varphi)$ to depart from the distribution of $-\sin \varphi$.

3.2. Circular polarization

For the left-handed CP laser pulse with electric fields $e_y = a_0 \sin^2 (\pi t/T) \cos [\omega(t - T/2) + \varphi]$ and $e_z = a_0 \sin^2 (\pi t/T) \sin [\omega(t - T/2) + \varphi]$, the absolute phase $\varphi$ rotates the whole laser pulse in the transverse space. Due to rotation invariance, one readily obtains that the THz amplitude is independent of the phase and that its polarization plane rotates with $\varphi$. We also carried out PIC simulations on THz emission from a CP pulse, and again found good agreement between the PIC result and 2D model. However, due to the fact that 2D simulation has no rotation...
Figure 4. 3D model calculation of THz emission from the H gas target for a CP laser pulse. (a) THz polarization angle $\theta$ as a function of the laser amplitude $a_0$. The phase is $\varphi = 0^\circ$. Two cases of $T = 4\tau$ and $5\tau$ are plotted. (b) The vector potentials $A_y$ (dashed line) and $A_z$ (dotted line) of the laser pulse and the instant ionization ratio $dn_e/dt$ (solid line) of the electron density for the phase $\varphi = 0^\circ$. The laser pulse has $a_0 = 0.008$ and $T = 4\tau$. In both frames, we take $R = 5\lambda$.

Invariance in the $yz$-plane, we do not show PIC results and only give the 3D model calculation as followed.

The fact that the THz amplitude is independent of the absolute phase for CP pulses makes them suitable for serving as a THz source. For a LP pulse, one has to adjust and stabilize the phase to generate the strongest THz signal. For the CP pulse, the CEP information can be extracted from the THz polarization direction. Figure 4(a) shows the polarization angle $\theta$ of the THz pulse radiated from the H gas target as a function of the laser amplitude $a_0$ for the fixed phase $\varphi = 0^\circ$. The polarization angle $\theta$ is defined as the angle between the direction of the first peak of the THz electric field and the $y$-direction. For $a_0 \leq 0.008$, we find that the polarization angle remains at $\theta = 90^\circ$ and is insensitive to the pulse duration. In this low-intensity region, one has $\theta = \varphi + \pi/2$, since $\theta$ rotates with the phase. For $a_0 = 0.008$, about 5.6% of H atoms are ionized after the laser pulse with $T = 4\tau$ has passed. Similar to the LP light case, ionization degrees $W_i \leq 10\%$ are needed for an intensity-insensitive $\theta(\varphi)$. For CP light, the ionization event is only determined by the pulse envelope, so that there is only one ionization peak. For low laser intensities, the ionization peak is located at the center of the pulse, as shown in figure 4(b), where the laser amplitude is $a_0 = 0.008$. The vector potential always has a maximum at the pulse center. For the special case of $\varphi = 0^\circ$ in figure 4(b), $A_z$ is maximum at $t = 2\tau$ and symmetric with respect to the central point, but $A_y$ is antisymmetric. So the deduced dipole is along the $z$-direction, i.e. the polarization angle $\theta = 90^\circ$. With increasing laser intensity, ionization occurs at the rising edge of the laser pulse and also in the transverse wings of the radial profile at somewhat later times. In this case, the THz polarization direction cannot be found in a simple way, but depends sensitively on the laser intensity as shown in figure 4(a). These properties are similar to the photoelectron CEP determination proposed in [18].
4. Summary and conclusions

The central topic of the present paper is THz emission from gases when hit by few-cycle laser pulses. The motivation was the application of the THz wave to determination of the absolute phase of the incident laser pulse. The THz emission discussed here is related to ionization. We therefore consider laser intensities ranging from onset of ionization to levels at which gas in the focal volume is just fully ionized, i.e. typically $10^{15} - 10^{17} \text{W cm}^{-2}$ for gases like H and He. At higher intensities, in particular beyond $10^{18} \text{W cm}^{-2}$, THz radiation is also generated, but due to different dynamics (e.g. wake-fields) [19]–[22]. The mechanism discussed here should be distinguished from the linear mode conversion of laser to THz radiation occurring in plasma ramps [19] and THz emission from a THz-wavelength-scale plasma oscillator [22].

Here, we have studied the generation of THz dipoles by tunneling and above-barrier ionization. The photoelectrons perform space-charge oscillations and radiate THz waves at the local plasma frequency, depending on gas density. For CEP measurements, it is best to keep the radiating volume smaller than the plasma wavelength so that the THz radiation emerges essentially from a point dipole. For a typical THz emission with frequency 1 THz (i.e. wavelength 300 $\mu$m), such configurations can be achieved by using thin gas layers having a thickness of 100 $\mu$m or less. We developed 2D and 3D model analyses and confirmed the model results of the 2D case by means of 2D-PIC simulation. Both linear and circular polarizations have been treated. For linear polarization, the laser CEP is directly mapped into the electric field amplitude of the THz wave from which the absolute phase can be measured easily. At larger intensities causing higher ionization, the relative dependence between the laser and THz wave becomes intensity-dependent. This effect is quantitatively described by the model in agreement with the simulations. For circular polarization, the laser CEP can be determined from the polarization angle of the THz wave. The corresponding dependence was calculated.

In conclusion, the present paper advances our understanding of photoionization-induced THz emission in the interaction of few-cycle laser pulses with gases, in particular, when used to determine the absolute phase of the incident pulse. This may help to make this method more accessible for laser control and attosecond experiments.

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