Terahertz emission from laser-driven gas plasmas: a plasmonic point of view: supplementary material

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This document provides supplementary information to "Terahertz emission from laser-driven gas plasmas: a plasmonic point of view," https://doi.org/10.1364/OPTICA.5.001617. We briefly review the Maxwell-Fluid model describing the ionization current mechanism for terahertz generation in laser-induced gas plasmas. Then, we give details on the derivation of the simplified model describing the terahertz emission from a plasma slab which is used in the main article. Finally, we focus on simulation results with microplasmas and comment on the possibility of tuning the terahertz emission spectra by turning the linear polarization of the elliptically-shaped two-color driving laser pulses.

1. DEFINITIONS: FOURIER TRANSFORMS AND FAR-FIELD SPECTRA

In the following, we define the temporal Fourier transform $\hat{f}(\mathbf{r}, \omega)$ of a function $f(\mathbf{r}, t)$ by

$$\hat{f}(\mathbf{r}, \omega) = \frac{1}{2\pi} \int f(\mathbf{r}, t) e^{i\omega t} dt, \quad (S1)$$

and

$$f(\mathbf{r}, t) = \int \hat{f}(\mathbf{r}, \omega) e^{-i\omega t} d\omega. \quad (S2)$$

Furthermore, we define the longitudinal spatial Fourier transform $\hat{f}(\mathbf{r}_L, k_z, \omega)$ of a function $f(\mathbf{r}_L, z, \omega)$ by

$$\hat{f}(\mathbf{r}_L, k_z, \omega) = \frac{1}{2\pi} \int f(\mathbf{r}_L, z, \omega) e^{-ik_z z} dz, \quad (S3)$$

and

$$f(\mathbf{r}_L, z, \omega) = \int \hat{f}(\mathbf{r}_L, k_z, \omega) e^{ik_z z} dk_z. \quad (S4)$$

Note the difference in sign of the exponent for temporal and spatial transforms, which is common practice in the optical context.

We introduce the spectral poynting fluxes

$$\hat{S}(\mathbf{r}_L, k_z, \omega) = 2/\mu_0 \Re\{\mathbf{E}(\mathbf{r}_L, k_z, \omega) \times \mathbf{B}^*(\mathbf{r}_L, k_z, \omega)\}, \quad (S6)$$

where $\mu_0$ is the magnetic permeability and $\Re$ denotes the real part. The first expression is used to compute the angularly integrated far-field power spectrum in Maxwell-consistent 2D/3D simulations by integration over a closed surface around the plasma. To compute the angularly integrated far-field spectrum $\tilde{P}_L$ in Sec. 3 of the main article, we integrate $\hat{S}_L$ along $k_z$.

2. MODELING THE IONIZATION CURRENT MECHANISM FOR MICROPLASMAS

The THz generation by two-color laser pulses considered here is driven by the so-called ionization current (IC) mechanism [1]. A comprehensive model based on the fluid equations for electrons describing THz emission has been derived in [2]. In this framework, the IC mechanism naturally appears at the lowest order of a multiple-scale expansion. Besides the IC mechanism, this model is also able to treat THz generation driven by ponderomotive forces and others. They appear at higher orders of the multiple-scale expansion and are not considered here. In the
followed, we briefly summarize the equations describing the IC mechanism.

The electromagnetic fields \( \mathbf{E} \) and \( \mathbf{B} \) are governed by Maxwell’s equations in vacuum

\[
\nabla \times \mathbf{E} = -\partial_t \mathbf{B}, \tag{S7}
\]

\[
\nabla \times \mathbf{B} = \frac{1}{c^2} \partial_t \mathbf{E} + \mu_0 \mathbf{J} + \mu_0 \mathbf{J}_{\text{loss}}. \tag{S8}
\]

The plasma and electromagnetic fields are coupled via the conductive current density \( \mathbf{J} \) governed by

\[
\partial_t \mathbf{J} + \nu_{\text{ei}} \mathbf{J} = \frac{q^2}{m_e} n_e \mathbf{E}, \tag{S9}
\]

where electron-ion collisions lead to a damping of the current. The collision frequency is determined by [3]

\[
\nu_{\text{ei}} [s^{-1}] = \frac{3.9 \times 10^{-6} \sum Z^2 Z_{\text{ion}}^2}{E_{\text{elec}} (eV)^{3/2}}, \tag{S10}
\]

where \( Z_{\text{ion}} \) is the Coulomb logarithm and \( E_{\text{elec}} \) denotes the thermal and kinetic electron energy. The value \( Z_{\text{ion}} = 3.5 \) turned out to match the results obtained by more sophisticated calculations with Particle-In-Cell (PIC) codes in [2].

The densities of \( Z \) times charged ions are determined by a set of rate equations

\[
\partial_t n_{\text{ion}}^{(Z)} = W^{(Z)} n_{\text{ion}}^{(Z-1)} - W^{(Z+1)} n_{\text{ion}}^{(Z)} \tag{S11}
\]

\[
\partial_t n_{\text{ion}}^{(0)} = -W^{(1)} n_{\text{ion}}^{(0)}
\]

for \( Z = 1, 2, 3, \ldots, K \), and the initial neutral density is \( n_{\text{ion}}^{(0)}(t = -\infty) = n_a \). The tunnel ionization rate \( W^{(Z)} \) in quasi-static approximation creating ions with charge \( Z \) is taken from [4, 5].

Thus, \( W^{(Z)} \) is a function of the modulus of the electric field \( \mathbf{E} \). The atoms can be at most \( K \) times ionized and thus \( W^{(K+1)} = 0 \). The electron density is determined by the ion densities

\[
n_e = \sum_Z Z n_{\text{ion}}^{(Z)}. \tag{S12}
\]

The electron energy density \( \mathcal{E} = n_e \mathcal{E}_{\text{elec}} \) is governed by

\[
\partial_t \mathcal{E} = \mathbf{J} \cdot \mathbf{E}. \tag{S13}
\]

The loss current accounting for ionization losses in the laser field reads

\[
\mathbf{J}_{\text{loss}} = \frac{E}{|E|^2} \sum_Z Z^2 W^{(Z)} n_{\text{ion}}^{(Z-1)}, \tag{S14}
\]

where \( |E|^2 \) is the ionization potential for creation of an ion with charge \( Z \). Even though ionization losses as well as higher order ionization \( (Z = 2, 3, \ldots) \) are negligible in the framework of the present study, both are kept for completeness.

The model is implemented in the code ARCTIC, that solves Eqs. (S7)-(S8) by means of the Yee scheme [6]. We have previously benchmarked the code ARCTIC by the PIC code OCEAN [7] accounting for full kinetics of the plasma in [8].

3. PLASMONIC RESONANCES IN THE PLASMA SLAB MODEL

To elaborate the origin of the laser-polarization-dependence of the THz-emission spectra from elliptically shaped 2C-laser-induced plasmas, we consider a simplified system as sketched in Fig. 5(a) of the main article: A plasma slab of thickness \( d \) in \( x \) direction with time-invariant electron density \( n_0 \) and collision frequency \( \nu_{\text{ei}} \). Both quantities are translationally invariant in \( y \) and \( z \). Above and below the slab we assume semi-infinite vacuum with a constant (relative) permittivity \( \varepsilon^\nu = 1 \). By writing the total electric field as the sum of the laser field \( \mathbf{E}_L \) and the field due to laser-plasma interaction \( \mathbf{E} \), Eq. (2) in the main article reads as

\[
\partial_t \mathbf{J} + \nu_{\text{ei}} \mathbf{J} = \frac{q^2}{m_e} n_0 \mathbf{E} + \mathbf{t} \tag{S15}
\]

with the source term

\[
\mathbf{t} = \frac{\mu_0 q}{m_e} \mathbf{E}_L. \tag{S16}
\]

It is important to note that we assume that the source term \( \mathbf{t} \) depends on the product of the laser electric field and the time-dependent electron density \( n_e(t) \), as it is produced during the laser gas interaction. Only in the description of the response of induced plasmas, we consider a simplified system as sketched in Fig. 5(a) of the main article: A plasma slab of thickness \( d \) in \( x \) direction with time-invariant electron density \( n_0 \).

Let us now use Eq. (S15) and Maxwell’s equations to determine the response of the system. In frequency space (see Sec. 1 for definition), they can be rewritten for angular frequency \( \omega \neq 0 \) as

\[
\nabla \times \hat{\mathbf{E}} = i\omega \hat{\mathbf{B}}, \tag{S17}
\]

\[
\nabla \times \hat{\mathbf{B}} = -i\omega \varepsilon \hat{\mathbf{E}} + \hat{\mathbf{Q}}, \tag{S18}
\]

where for sake of brevity we introduced the source term

\[
\hat{\mathbf{Q}} = \frac{\mu_0 q}{i\omega + \nu_{\text{ei}}}. \tag{S19}
\]

The dielectric permittivity \( \varepsilon \) reads \( \varepsilon = \varepsilon^\nu + \varepsilon^p \) in the plasma slab \( (|x| \leq d/2) \), and \( \varepsilon = \varepsilon^\nu = 1 \) in the vacuum \( (|x| > d/2) \). The complex dielectric permittivity of the plasma is given by

\[
\varepsilon^p = 1 - \frac{\omega^2_0}{\omega^2 + i\omega\nu_{\text{ei}}}, \tag{S20}
\]

where the plasma frequency \( \omega_0^{p} = \sqrt{\frac{n_0 q^2}{m_e \varepsilon_0}} \) involves the time independent density \( n_0 \) of the slab. The time-independent collision frequency \( \nu_{\text{ei}} \) can be disregarded \( (\nu_{\text{ei}} = 0) \) and is kept here just for completeness.

Same as for \( \varepsilon \), we consider an excitation that is translational invariant in \( y \), that is, \( \partial_y \mathbf{E} = 0 \) and \( \partial_y \hat{\mathbf{Q}} = 0 \). Therefore, we can set all the \( y \)-derivatives to zero and Eqs. (S17)-(S18) separate into two sets of equations. The translational invariance of the slab in \( z \) allows to write down these two sets of equations in the spatial Fourier domain with respect to \( z \) \( (\partial_z \rightarrow ik_z) \) giving

\[
\partial_z \hat{E}_y = i\omega \hat{B}_z, \tag{S21}
\]

\[
-ik_z \hat{E}_y = i\omega \hat{B}_x, \tag{S22}
\]

\[
-ik_z \hat{B}_x - \partial_z \hat{B}_z = -i\omega \hat{E}_y + \hat{Q}_y \tag{S23}
\]
When then exciting the system by the source $\omega$, which will be highlighted in the coming derivation. In the waveguide context, it is also often termed the transverse electric (TE) mode, because the only electric field component $E_y$ is polarized in the transverse translational invariant direction. Here, the only fields different from zero are $(\tilde{B}_z, \tilde{E}_y, \tilde{E}_z)$. The 2nd set of equations (S24)-(S26) is associated with the so-called $S$ polarization. In the waveguide context, it is also often termed the transverse magnetic (TM) mode.

In the following, we will consider two different configurations:

(i) $S$ polarization with transverse excitation in $y$ ($\tilde{E}_y \neq 0 \neq Q_y$ and $\tilde{Q}_x = 0 = \tilde{Q}_z$)

(ii) $P$ polarization with transverse excitation in $x$ ($\tilde{E}_x \neq 0 \neq \tilde{Q}_x$ and $\tilde{Q}_y = 0 = \tilde{Q}_z$)

Note that (i) corresponds to the THz generation by $y$-polarized "elliptical beams" and (ii) by $x$-polarized "elliptical beams" as investigated in the main article.

Before computing the response of the plasma slab to an excitation by $\tilde{E}_x$, it is interesting to investigate the resonances of the homogeneous system ($\tilde{Q}_x = 0$). To this end, it is convenient to solve the reflection transmission problem and to consider the reflection coefficient. Singularities of the reflection coefficient describe resonant modes, i.e., modes which provide non-zero fields in the slab without an incoming wave and without a source term. Then when exciting the system by the source $\tilde{E}_x$, these modes are most likely excited and thus of great interest.

### A. Reflection coefficient of a slab

In the following, we consider the problem of computing the reflection coefficient for a plane wave arriving from one of the vacuum half-spaces and interacting with a plasma slab (see Fig. S1). To this end, we will operate in the $(\omega, k_z)$-space. We will detail the $P$ polarization case, because it is of most interest. The $S$ polarization case follows analogously with a small modification which will be highlighted in the coming derivation.

We denote by $\tilde{B}_i$ the amplitude of the magnetic field of the incident $P$ polarized wave and by $\tilde{B}_r$ the amplitude of the reflected wave. Without loss of generality, the incident wave arrives from the positive half-space. Then, the magnetic field in the vacuum for $x \geq d/2$ writes

$$B'_y(x) = B_\tau \exp \left[ -\Lambda^v \left( x - \frac{d}{2} \right) \right] + \tilde{B}_r \exp \left[ \Lambda^v \left( x - \frac{d}{2} \right) \right],$$

(S27)

where $\Lambda^v = \sqrt{k_z^2 - \omega^2/c^2}$. Denoting its $x$-derivative by $B''_y = \partial_x B'_y$, this implies the relation

$$\left( \begin{array}{c} \tilde{B}_y' \\ \tilde{B}_y'' \end{array} \right)_{x=\frac{d}{2}} = \left( \begin{array}{cc} 1 & 1 \\ -\Lambda^v & \Lambda^v \end{array} \right) \left( \begin{array}{c} \tilde{B}_i \\ \tilde{B}_r \end{array} \right),$$

(S28)

where we introduced the transformation matrix $K$.

For P polarization, only the transverse field $\tilde{B}_y$ is continuous at an interface while for the derivative we can only use that $e^{-\partial_x} \tilde{B}_y$ is continuous. The situation is easier for S polarization where both $\tilde{E}_y$ and $\tilde{E}_z$ are continuous at an interface. Thus, to handle the $P$ polarization and later on the $S$ polarization case in the same manner, we introduce $a_P = 1/e^\alpha$ and $a_S = 1$. Then, we relate the fields at the vacuum plasma interface at $x = d/2$ by

$$\left( \begin{array}{c} \tilde{B}_y' \\ \tilde{B}_y'' \end{array} \right)_{x=\frac{d}{2}} = \left( \begin{array}{cc} 1 & 0 \\ 0 & a_P^{-1} \end{array} \right) \left( \begin{array}{c} \tilde{B}_y' \\ \tilde{B}_y'' \end{array} \right)_{x=\frac{d}{2}},$$

(S29)

where we introduced the interface transition matrix $T$.

The magnetic field in the plasma ($|x| < d/2$) can be decomposed into a forward $\tilde{B}_+$ and a backward $\tilde{B}_-$ running wave as

$$\tilde{B}'_y(x) = \tilde{B}_i \exp (\Lambda^P x) + \tilde{B}_r \exp (-\Lambda^P x),$$

(S30)

where $\Lambda^P = \sqrt{k_z^2 - \epsilon_\tau \omega^2/c^2}$. Using this equation, a short computation relates fields at the two interfaces inside the plasma

$$\left( \begin{array}{c} \tilde{B}_y' \\ \tilde{B}_y'' \end{array} \right)_{x=-\frac{d}{2}} = M \left( \begin{array}{c} \tilde{B}_y' \\ \tilde{B}_y'' \end{array} \right)_{x=\frac{d}{2}},$$

(S31)

with

$$M = \left( \begin{array}{cc} \cosh (\Lambda^P d) & \sinh (\Lambda^P d) / \Lambda^P \\ \Lambda^P \sinh (\Lambda^P d) & \cosh (\Lambda^P d) \end{array} \right).$$

(S32)

In complete analogy to Eq. (S29), we obtain the fields in vacuum at the lower interface at $x = -d/2$ by

$$\left( \begin{array}{c} \tilde{B}_y' \\ \tilde{B}_y'' \end{array} \right)_{x=-\frac{d}{2}} = T^{-1} \left( \begin{array}{c} \tilde{B}_y' \\ \tilde{B}_y'' \end{array} \right)_{x=\frac{d}{2}}.$$

(S33)

We denote by $\tilde{B}_t$ the amplitude of the transmitted wave. Because there is no incoming wave from the negative half-space, the field in the vacuum for $x \leq d/2$ reads

$$\tilde{B}'_y(x) = \tilde{B}_t \exp \left[ -\Lambda^v \left( x - \frac{d}{2} \right) \right].$$

(S34)

In summary, we therefore obtain

$$\tilde{B}_t \left( \begin{array}{c} 1 \\ -\Lambda^v \end{array} \right) = \left( \begin{array}{c} \tilde{B}_y' \\ \tilde{B}_y'' \end{array} \right)_{x=-\frac{d}{2}} = T^{-1} M T K \left( \begin{array}{c} \tilde{B}_i \\ \tilde{B}_r \end{array} \right).$$

(S35)

After performing all the matrix multiplications, we end up two equations relating the field amplitudes $\tilde{B}_i$, $\tilde{B}_r$, and $\tilde{B}_t$. We can
These two conditions determine the yet unknown amplitudes \( \alpha \) and \( \beta \), respectively. The continuity of the transverse fields is used to determine the entire solution.

Equations (S24)-(S26) were used in Sec. 3 of the main article to compute the transverse Poynting fluxes via Eq. (S6).

C. P polarization case with transverse excitation in \( x \)

Next, case (ii) with \( \mathbf{I} = I_y \mathbf{e}_y \) is considered. Equations. (S24)-(S26) give the evolution equation for the transverse field \( \vec{B}_y \) in the plasma inside and neutral gas respectively,

\[
\frac{\partial^2}{\partial x^2} \vec{B}_y - \Lambda^2 \vec{B}_y = -ik \vec{Q}_x.
\]

In analogy to the S polarization case in the previous section, we obtain in the plasma

\[
\vec{B}_y^p = \Lambda^p \cosh (\Lambda^p x) - \frac{ik}{\Lambda^p} \vec{Q}_x \cosh (\Lambda^p x) - 1.
\]

and in upper vacuum

\[
\vec{B}_y^v = A^v \exp \left[ \pm \Lambda^v \left( x - \frac{d}{2} \right) \right].
\]

The difference to the S polarization case appears when applying the interface conditions at the plasma-air interface: \( \vec{B}_y \) and \( \vec{E}_y \) are continuous but according to Eq. (S26) \( \partial_x \vec{B}_y \) is not, because \( \mathbf{e}_x \) changes at the interface. Applying these P polarization interface conditions determines \( \Lambda^p \) and \( \Lambda^v \) to

\[
\Lambda^p = \frac{ik}{\Lambda^p} \frac{\Lambda^v}{\left( \Lambda^p \right)^2} \left( \frac{\Lambda^v}{\left( \Lambda^p \right)^2} + 1 \right),
\]

\[
\Lambda^v = \frac{ik}{\Lambda^v} \frac{\Lambda^v}{\left( \Lambda^p \right)^2} \cosh \left( \frac{\Lambda^p d}{2} \right) + 1,
\]

with common denominator

\[
D^p = \mp \Lambda^p \sinh \left( \frac{\Lambda^p d}{2} \right) - \Lambda^v \cosh \left( \frac{\Lambda^p d}{2} \right).
\]

Finally, Eqs. (S21)-(S22) determine the magnetic field components as

\[
\vec{B}_x = -\frac{k \omega}{\omega^2} \vec{E}_y
\]

\[
\vec{B}_z = \frac{k \Lambda^v}{\Lambda^p} \sinh (\Lambda^p x)
\]

Equations (S40) and (S46) were used in Sec. 3 of the main article to compute the transverse Poynting fluxes via Eq. (S6).
Fig. S2. Angularly integrated far-field spectra for the elliptical beams from Sec. 4 of the main article and corresponding results from 2D simulations assuming translational invariance in y (dashed lines). The solid black line specifies the emission spectrum from a 3D simulation with laser polarization at 45° in the xy plane. The black dash-dotted line is computed from the superimposed fields obtained with an x- and a y-polarized laser in 3D.

4. THZ EMISSION FROM ELLIPTICALLY SHAPED MICROPLASMAS

In the main article we investigate terahertz (THz) emission from elliptically shaped two-color (2C)-laser-induced gas-plasmas. In such a configuration, the free electron density profile with a small transverse size along x and a large transverse size along y is created. For the considered microplasmas, the transverse plasma profile is strongly elliptical, that is, along x direction the plasma size is less than 1 μm, whereas along y direction the plasma is approximately 10 μm wide. Thus, by rotating the linear laser electric field polarization with respect to the transverse plasma profile, we can select the strength of the electron density gradients along the excitation direction.

As it can be seen in the 3D angularly integrated far-field spectra presented in Fig. S2, when exciting the plasma along strong electron density gradients (x-polarized laser associated with P polarization), the THz spectrum is broadened up to about 50 THz (dark red solid line), which corresponds to the maximum plasma frequency \( \nu_{\text{max}} \). In contrast, when exciting the plasma along the weak electron density gradients (y-polarized laser associated with S polarization) no such broadening is found (light gray solid line).

Results of corresponding 2D simulations (\( \partial_y = 0 \)) are shown as dashed lines in Fig. S2. Here, we find a similar behavior as in 3D: no broadening if the laser electric field is oriented in the now translationally invariant y direction (S), and broadening up to \( \nu_{\text{max}} \) if the laser electric field points in the direction of the strong electron density gradient, that is, along the x direction (P). Treating the problem in 2D geometry, i.e., assuming translational invariance in one transverse direction (here y), the electromagnetic fields separate into two cases: S polarization case that governs the fields \( B_x, E_y, B_z \) for a y-polarized driving laser pulse and the P polarization case that governs the fields \( E_x, B_y, E_z \) for an x-polarized driving laser pulse. Any other polarization state in 2D can be written as the superposition of these two cases, if coupling through the laser generated plasma is ignored.

For example, an incoming laser pulse that is linearly polarized under 45° in the xy plane will give an electric field solution that can be written as \( E = E_0^{\parallel} / \sqrt{2} + E_0^{\perp} / \sqrt{2} \), where \( E_0^{\parallel} \) and \( E_0^{\perp} \) are the solutions for an S and a P polarized driving laser pulse, respectively. Therefore, if the 3D elliptical beam can be indeed approximated by the idealized 2D case, and no detrimental non-linear coupling occurs, this property should hold. We checked this by comparing the angularly integrated THz-far-field power spectrum for a simulation with polarization at 45° (black solid line) and the result for the superposed fields (black dashed line) in Fig. S2. Both overlap almost perfectly, which further justifies our analysis of the 2D configuration. Moreover, the possibility of superposing the THz fields which are produced by an x- and y-polarized laser pulse could be important for applications, because it implies that the THz emission spectrum can be tuned by rotating the linear polarization of the incoming elliptical laser pulse.

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