Generation of terahertz radiation from ionizing two-color laser pulses in Ar filled metallic hollow waveguides

I. Babushkin1, S. Skupin2,3, and J. Herrmann4

1 Weierstraß-Institut für Angewandte Analysis und Stochastik, 10117 Berlin, Germany
2 Max Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany
3 Friedrich Schiller University, Institute of Condensed Matter Theory and Optics, 07742 Jena, Germany
4 Max Born Institute for Nonlinear Optics and Short Time Spectroscopy, 12489, Berlin, Germany

Abstract: The generation of THz radiation from ionizing two-color femtosecond pulses propagating in metallic hollow waveguides filled with Ar is numerically studied. We observe a strong reshaping of the low-frequency part of the spectrum. More precisely, after several millimeters of propagation the spectrum is extended from hundreds of GHz up to \(\sim 150\) THz. For longer propagation distances, nearly single-cycle near-infrared pulses with wavelengths around 4.5 \(\mu\)m are obtained by appropriate spectral filtering, with an efficiency of 0.1–1%.

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References and links
diagnostic and imaging tools. Alternatively, THz radiation can also be emitted by few-cycle laser pulses. For several applications, coherent electromagnetic THz radiation is particularly interesting for a wide range of new applications, from probing complex molecules to various diagnostic and imaging tools. Alternatively, THz radiation can also be emitted by few-cycle laser pulses. For several applications, coherent electromagnetic THz radiation is particularly interesting for a wide range of new applications, from probing complex molecules to various diagnostic and imaging tools. Alternatively, THz radiation can also be emitted by few-cycle laser pulses.

In recent years, the range of wavelengths where coherent radiation can be generated has grown dramatically into both high and low frequency domain. Remarkably, most of the methods to obtain radiation at extreme frequencies use, in one or the other way, nonlinear processes in laser-induced plasma. One prominent example is high harmonic generation (HHG) where frequencies thousand times larger than the frequency of the pump pulse are excited, exploiting the recollision dynamics of electrons ionized by the intense light pulses. More recently it was demonstrated that a two-color fs beam allows generation of new frequencies just in the opposite part of the spectrum, namely in the THz range, hundreds times smaller than the optical pump frequency. To this end, a short two-color pulse of fundamental frequency and second harmonic is strongly focused into a plasma spot [1–12]. The observed THz emission generated in this scheme has been attributed to the laser-induced plasma current in the asymmetric two-color field [7, 8]. Using such scheme generation of strong THz radiation was reported, with a spectrum which can be as broad as 70 THz [8]. Such broad-band coherent radiation is very interesting for a wide range of new applications, from probing complex molecules to various diagnostic and imaging tools. Alternatively, THz radiation can also be emitted by few-cycle
pulses without second harmonics [13–17].

In this article we consider a modification of the above described focusing geometry in a bulk gas by using a metallic hollow waveguide with a cladding from aluminum to guide both THz and optical radiation [see Fig. 1(a)]. The use of a waveguide prevents the diffraction of light for all wavelengths involved. In addition, such setup allows to realize nearly single-mode operation for a wide range of frequencies. High threshold intensities for ionization-induced processes in the range larger than 100 TW/cm² have been already realized in the context of HHG schemes in hollow waveguides [18,19]. We will show that during propagation a dramatic broadening of the low-frequency part of the spectrum occurs, caused by only modest changes in the fundamental and second harmonic fields. In particular, ultrashort pulses of 10 fs duration lead to the formation of low-frequency spectrum extended from several hundreds of GHz to approximately 150 THz. Upon further propagation we report the generation of nearly single-cycle pulses with central wavelengths around 4.5 μm and efficiency of the order of 0.1–1%.

2. The model

In the previous works, THz emission by two-color fields in gases has been interpreted by four-wave mixing rectification [1–5]. In contrast, later studies attributed THz emission to the laser-induced plasma-current in the asymmetric two-color field [6–10] described by a local quasiclassical model of the electron current without taking into account propagation effects. As an alternative approach, particle-in-cell simulations were used [11,12]. Those methods are, however, computationally very expensive, and propagation effects have been taken into account over distances of a few micrometers only. In such models the microscopic current is described as a sum over contributions from all electrons, born at discrete times $t_n$. The "continuous" analog of this approach [7–9] is given by:

$$J(t) = q \int_{-\infty}^{t} \mathbf{v}(t,t_0) \rho(t_0) dt_0,$$

where $q$ is the electron charge, $\mathbf{v}(t,t_0)$ is the velocity of electrons at time $t$ which were born at time $t_0$, and $\rho(t)$ is the electron density. In this work we use the following differential equation for the macroscopic plasma current $\mathbf{J}$ [20,21]:

$$\frac{d}{dt} J(t) + \nu_e J(t) = \frac{q^2}{m_e} \mathbf{E}(t) \rho(t),$$

where $\nu_e$ is the current decay rate and $m_e$ is the electron mass. Remarkably, Eq. (1) can be obtained from the integral equation mentioned above by taking into account that (in the presence
of decay \( v(t, t_0) = \frac{q}{m_e} \int_{t_0}^{t} E(\tau) e^{\nu_e(\tau-t)} d\tau \). This shows equivalence of the “microscopic current” approach and Eq. (1).

In the present article, the static model for the tunneling ionization [22] is used:

\[
\rho(t) = W_{ST}[E(t)][\rho_n - \rho(t)], \quad W_{ST}[E(t)] = \alpha \frac{E_n}{|E(t)|} \exp \left\{ -\beta \frac{E_n}{|E(t)|} \right\}. \tag{2}
\]

Here, \( \rho_n \) is the neutral atomic density, and \( E_n \approx 5.14 \times 10^{11} \text{ Vm}^{-1} \) the atomic field; the coefficients \( \alpha = 4a_0r_{H}^{5/2}/c \) and \( \beta = (2/3)r_{H}^{5/2}/c \) are defined through the ratio of ionization potentials of argon and hydrogen atoms \( r_{H} = U_{Ar}/U_{H}, U_{Ar} = 15.6 \text{ eV}, U_{H} = 13.6 \text{ eV}, \) and \( a_0 \approx 4.13 \times 10^{16} \text{ s}^{-1} \) [6, 22]. For 1 atm pressure, \( \rho_n = 2.7 \times 10^{19} \text{ cm}^{-3}, \) \( 1/\nu_e = 190 \text{ fs} \) (see [20]).

To describe the light propagation in the waveguide with the optical axis along \( z \) direction, we start from the nonlinear Helmholtz equation for the fast oscillating optical field \( E(r, z, t) \):

\[
\frac{\partial^2 \tilde{E}(r, z, \omega)}{\partial z^2} + k^2(\omega)\tilde{E}(r, z, \omega) + \Delta_{\perp} \tilde{E}(r, z, \omega) = -\mu_0 \omega^2 \tilde{P}_m(r, z, \omega), \tag{3}
\]

where \( r = \{x, y\}, \Delta_{\perp} = \partial_x^2 + \partial_y^2, n(\omega) \) is the refractive index of Ar, \( k(\omega) = n(\omega)\omega/c, \tilde{E}(r, z, \omega) \) and \( \tilde{P}_m(r, z, \omega) \) are the Fourier transforms of \( E(r, z, t), \) \( P_m(r, z, t) \) with respect to time, \( P_m \) is the nonlinear polarization, which includes both the Kerr \( (P_{Kerr} = \epsilon_0 \chi^{(3)}[|E|^2]E) \) nonlinearity and the plasma contribution: \( P_m = P_{Kerr} + \tilde{J}/\omega + \tilde{J}_{loss}/\omega, \) where \( J_{loss} = W_{ST}(\rho_n - \rho)U_{Ar}/E_n \) is a loss term accounting for photon absorption during ionization; “division by a vector” means the component-wise division [22]. Decomposing the field \( E(r, z, t) \) into a series of linear eigenmodes of the waveguide \( \{F_j(r), j = 1, \ldots, \infty\} \) with corresponding propagation constants \( \beta_j(\omega) \) and neglecting backward reflection [23], we obtain the following set of equations for the amplitudes of the eigenmodes \( E_j, \) coupled through the nonlinear polarization term \( P_{nl}^{(j)} = \int F_j \tilde{P}_m dr: \)

\[
\frac{\partial E_j(z, \omega)}{\partial z} = i\beta_j(\omega)E_j(z, \omega) + i\frac{\mu_0 \omega^2}{2\beta_j(\omega)} P_{nl}^{(j)}(z, \omega). \tag{4}
\]

Equations (1), (2) and (4) comprise a closed set of equations which are solved by a standard split-step method for first five transverse modes \( (E_j \) quickly decays with \( j; \) for typical simulation parameters the energy in the fifth mode is already less than 1%). The spatial field distribution \( \tilde{E} \) can be reconstructed as \( \tilde{E} = \sum_j \tilde{E}_j F_j, \) It is important to note that Eq. (4) is derived without using the slowly varying envelope approximation and is valid for all frequencies. In particular, the computed THz signal is the low-frequency part of \( \tilde{E} \) (that is, we assume that the detector is placed just at the output of the waveguide).

3. Numerical simulations

In our simulations, we assume a dielectric waveguide with aluminum coating of the inner walls and diameter \( d = 100 \mu \text{m} \) filled with Ar at 1 atm pressure. Such waveguides can be routinely produced (see, e.g., [24]). The main purpose of the waveguide is not to modify the dispersion relation but to confine both pump and THz radiation (especially the latter, because it quickly spreads out otherwise as a result of diffraction [25]). Taking into account that THz as well as fundamental and second harmonic frequencies undergo strong absorption due to plasma generation we restrict our simulations to distances \( \leq 1 \text{ cm} \).

The linearly-polarized waveguide mode \( EH_{11} \) has no cut-off at low frequencies, and the fundamental and second-harmonic frequencies can be coupled into the waveguide with almost 100% efficiency from the free-space propagating laser mode. The dispersion and losses for the mode \( EH_{11} \) of the 100-\( \mu \text{m} \) waveguide are shown in Fig. 1(b) in low-frequency range. Both
Fig. 2. (Color online). Evolution of the THz spectrum upon propagation in a hollow waveguide at 1 atm gas pressure with diameter $d = 100 \mu m$, assuming Gaussian pulses with intensities $I_\omega = 10^{14}$ W/cm$^2$, $I_{2\omega} = 2 \times 10^{13}$ W/cm$^2$, equal pulse durations (10 fs) and phase difference $\theta = \pi/2$ at the input. (a) The spectrum and (b) low-frequency part of spectrum [note the logarithmic scale of y-axis in (a) and of both axes in (b)] for $z = 0.5$, 2, and 10 mm of propagation (black dashed, green dotted and red solid lines). (c) Pulses obtained by filtering the long-wavelength part of the spectrum below 250 THz. Propagation distances, color coding and line styles are the same as in (a).

quantities are calculated using the direct solution of the corresponding boundary value problem [26], Drude model for the dispersion in aluminum [27] as well as Sellmeier formula for the dispersion in argon [28]. One can see that the losses are relatively large only in the small frequency range corresponding to wavelengths of the order of the waveguide diameter. The aluminum coating allows the modeling of linear dispersion and losses in the whole frequency range from sub-THz to sub-PHz by the Drude model [27].

Results of simulations for different propagation lengths are shown in Fig. 2. We assume two Gaussian Fourier-limited pulses with equal duration (10 fs full width at half maximum) launched simultaneously into the waveguide, frequencies $\nu_0 = 375$ THz (corresponding to $\lambda = 800$ nm) and $2\nu_0$ and intensities $I_\omega = 10^{14}$ W/cm$^2$ and $I_{2\omega} = 2 \times 10^{13}$ W/cm$^2$ (at the center of the capillary). The initial phase difference between the two pulses is $\theta = \pi/2$.

At the initial stage of propagation, broadband THz radiation is generated from approximately 0.5 THz to 80 THz [see Fig. 2(a) and Fig. 2(b)]. With further propagation (up to 2 mm) the low-frequency part of the spectrum grows in the direction to higher frequencies (up to 150 THz). At the same time, the spectral intensity around 10 THz degrades because of the strong losses of the waveguide mode. From 2 mm up to ~7 mm the long-wavelength part of the spectrum shifts continuously towards higher frequencies. Finally, starting from 7 mm the spectrum does not change considerably anymore, except for its decrease due to losses. At this propagation distance, the long-wavelength part of the spectrum is localized around 60-80 THz, with the center around 66 THz (corresponding to a wavelength $\sim 4.5 \mu m$). The observed efficiency of this "frequency down-conversion" reaches 0.25 percent.

The electric field obtained by filtering the long-wavelength part of the spectrum for different propagation distances is shown in Fig. 2(c). One can see that despite the strong reshaping of the spectrum, the envelope of the obtained THz pulses does not change significantly. The amplitude of the pulse grows considerably during propagation, from approximately 1% of the pump amplitude at $z = 0.5$ mm (which correspond to the conversion efficiency $\sim 10^{-4}$) to about 5% at $z = 10$ mm (which correspond to the conversion efficiency $\sim 2.5 \times 10^{-3}$).

The behavior observed is very robust to the change of system parameters [see Fig. 3]. It is qualitatively similar for different pulse durations, initial phase differences $\theta$, intensity ratios $r = I_{2\omega}/I_\omega$, and gas pressures. However, THz generation is more efficient for shorter pulses and larger intensity ratios $r$ [up to 1% in Fig. 3(b)], which can be explained by the resulting additional field asymmetry.
Fig. 3. (Color online). Spectra for various parameters modified as compared to Fig. 2 (1 cm of propagation). (a) Different input pulse durations: 25 fs (red solid line), 100 fs (green dashed line); blue dotted line shows the case of different fundamental (10 fs) and second harmonic (7 fs) pulse durations. (b) Red solid line: pressure 0.5 atm; green dashed line: $I_{2\omega} = 60$ TW/cm$^2$; blue dotted line: $\theta = 0$. Note that the spectra are normalized to their intensity maxima (at $\Omega \approx \nu_0$) to compare efficiencies.

4. Discussion and Conclusion

In all simulations presented in the previous section the dominant contribution to THz emission is coming from the electron current $J$. We verified that the direct contribution of the Kerr non-linearity is at least two orders of magnitude smaller, and the impact of $\sim J_{\text{loss}}$ is also negligible. Thus, our results support the mechanism of ionization-induced THz emission [7–12]. The key ingredients for this generation mechanism are the temporal asymmetry of the pump field and a field dependent (tunnel) ionization rate. Such ionization rate produces isolated ionization events and therefore a stepwise increase of the free electron density $\rho$ in time, which is responsible for the observed THz emission [25]. As revealed by Fig. 2(a), the spectrum of the pump remains almost unchanged upon propagation. Nevertheless, it experiences a slight broadening. Moreover, due to the plasma-induced refraction index change the maximum frequency of the fundamental pulse is slightly shifted to the blue [21, 29, 30]. In our case the shift is small, only about 10 THz. However, as elaborated in [25], even small changes in the pump fields can affect dramatically the spectral shape of the generated low-frequency radiation, because they influence significantly the temporal positions of the ionization events: Contributions from electrons born in different ionization events are superimposed, and interference effects lead to a reshaping the THz spectra.

The observed mechanism is rather general and insensitive to the particular choice of parameters (see also Fig. 3) as well as to the details of the modeling approach. In particular, for intensities $\sim 100$ TW/cm$^2$ the ionization rate Eq. (2) is only a rough estimate [15–17,31], but the resulting electron densities are reasonable [31]. Therefore, Eq. (2) leads to rather good agreement with experimental data for comparable intensities (albeit different geometry) [25,32]. In order to corroborate further our claim of generality we made simulations analogous to Fig. 2 but with hydrogen instead of argon (not shown), which allows us to use a verified tunneling rate [15,17]. These simulations produce results similar to presented in Fig. 2.

In conclusion, we studied ionization-induced THz emission in a noble gas ionized by strong asymmetric laser fields. We have shown that the mechanism allows a relatively simple generation of an extremely wide range of frequencies, starting from THz to near infrared. By using hollow waveguides we overcome the strong diffraction of the generated low-frequencies. We report that depending on the length of the waveguide either THz supercontinuum or almost single-cycle near-infrared pulses can be produced at output with 0.1–1% efficiency. We believe that our findings may open new intriguing possibilities to control the spectral and temporal shape of low-frequency radiation generated by the ionization dynamics in intense optical fields.