Abstract

Using the strong field approximation we calculate photoelectron momentum distributions generated in the interaction of low-frequency two-color laser fields with atomic gases. The field consists of an infrared linearly or circularly polarized pulse of intensity close to $10^{14}$ W/cm$^2$ and its second linearly polarized harmonic whose intensity does not exceed 10% of the fundamental. Our calculations aim to find a field configuration, which maximizes the photoelectron current left after the interaction. Such net currents result from asymmetries of photoelectron distributions in non-monochromatic coherent fields with fixed phases between the frequency components. We show that combining a circularly polarized intense pulse with a linearly polarized pulse of the second harmonic one could approach the highest possible asymmetry of the photoelectron distribution and therefore the highest value of the net current.

Keywords: terahertz radiation, strong-field ionization, photoelectron currents, strong field approximation

1. Introduction

Over the past two decades a new direction in the strong-field atomic and molecular physics has emerged which is the generation of light pulses of terahertz (THz) frequencies in the interaction of intense infrared laser radiation with atomic and molecular gases including ambient air. For the history and the state of the art of this research field, we direct the reader to [1–4] and the papers quoted there. Coherent light pulses with the carrier frequency $\nu \sim 1 \div 10$ THz are potentially useful for a variety of applications. In particular, they can be applied for excitation of spin waves, orientation of molecules and for terahertz spectroscopy of different materials, see [6–8] for details.

One of the promising schemes for the generation of THz radiation is based on ionization of gases by intense optical or infrared two-color (bichromatic) laser radiation.
a monochromatic electromagnetic field of nonrelativistic intensity the probability distribution \( w(p) \) in final photoelectron momenta \( p \) possesses inverse symmetry: \( w(-p) = w(p) \) that results from the respective symmetry of the electric field, \( E(t + \frac{T}{2}) = -E(t) \) of the laser, with \( T = \frac{2\pi}{\omega} \) being the optical period and \( \omega \) – the laser frequency. For a non-monochromatic field this symmetry does not hold, resulting to an asymmetric photoelectron distribution and a nonzero net photoelectron current.

An efficient way of breaking the symmetry of photoelectron distributions consists in the application of a two-color laser field, viz. a coherent superposition of an infrared laser pulse and its second harmonic generated after the propagation of the main laser pulse through a nonlinear crystal. It was shown both in experiments and theoretically [10–13] that such scheme leads to the generation of a considerable THz light signal even when only few percent of the fundamental pulse energy is converted into the second harmonic. By now, most of the studies explored the scheme with two linearly polarized pulses. It was shown in particular that the configuration with parallel polarizations leads to higher photoelectron currents and THz signals than the one where the polarization directions are orthogonal. The main disadvantage of the scheme with linear polarizations is connected to the fact that the asymmetry of photoelectron distributions is maximal when the electric fields of the two components are phase shifted by \( \frac{\pi}{2} \), so that the enhancement in the ionization rate due to the presence of the second harmonic is minimal (see, e.g., [9, 14] for details). As a result, the average photoelectron momentum \( p_0 \) which can be acquired within this scheme, appears proportional to the vector potential of the second field at the time instant when the first field has a maximum:

\[
p_0 \sim A_2(t_0) \approx \epsilon \frac{E_1}{\omega}.
\]

Here, \( E_1 \) is the amplitude of the fundamental field and \( \epsilon = E_2/E_1 \) is the ratio of the field amplitudes. For realistic experimental situations, this ratio is small, \( \epsilon \ll 1 \), corresponding to a few-percent efficiency of the nonlinear conversion. As a result, the net momentum can only be a relatively small fraction of the characteristic field momentum

\[
p_F = \frac{E_1}{\omega}.
\]

Such regime corresponds to the linear part of the plots shown on Figure 1.

A higher degree of asymmetry can only be achieved if both fields are of comparable amplitudes as it is also illustrated by Figure 1. However, the regime with \( \epsilon \approx 1 \) simultaneously with the fields coherently superimposed is difficult for experimental realizations.
Figure 1: Average (net) photoelectron momentum in arbitrary units calculated versus the parameter $\epsilon = E_2/E_1$ for the case when the field phases are shifted by $\pi/2$. The fundamental wavelength $\lambda = 800$ nm, and the intensities are shown on the inset in W/cm$^2$. Ionization of argon is considered. Sharp spikes on the curves result from the channel closing effect taking place with the growth of the total intensity; see [14] for details of the calculation.

As is seen from the plot, the linear regime, when the effect of the second field on the average momentum is perturbative extends up to $\epsilon \approx 0.2$, which corresponds to 4% of the laser intensity converted into the second harmonic, the value close to typical experimental numbers.

In this work, we discuss a similar two-color setup, but with the fundamental field circularly polarized. This discussion is stimulated by a recent paper [15] where the generation of THz light by a superposition of two circularly polarized pulses was considered and a gross enhancement in the THz signal compared to the case of linear polarizations was observed at all other parameters but the field polarizations fixed. Basing on the strong field approximation [16–19] commonly used for description of laser-atom interaction in the strong-field regime, we show that a setup with a superposition of circularly polarized fundamental and linearly polarized second harmonic field can generate photoelectron spectra with an exceptional degree of asymmetry, making the net momentum close to the field momentum (2).
2. Photoelectron Distributions in the Presence of a Circularly Polarized Fundamental Field

In order to justify the idea of using at least one circularly polarized pulse, we describe the ionization dynamics within the strong field approximation (SFA) [16–19], which allows a very simple qualitative analysis. We assume the fields in the form

\[ E_1(\varphi) = E_0(\cos \varphi, \sin \varphi), \quad (3) \]

\[ E_2(\varphi) = \epsilon E_0(\cos (\varphi + \alpha), 0) \quad (4) \]

and calculate the SFA ionization amplitude and probability along the standard expressions:

\[ M(p) = \int dt \langle \Psi_p | \hat{V}(t) | \Psi_0 \rangle, \quad (5) \]

\[ dw(p) = |M(p)|^2 dp^3. \quad (6) \]

Here, \( \Psi_0 \) and \( \Psi_p \) are the initial and the final state wave functions correspondingly, the latter is approximated by the Volkov wave; and \( \hat{V}(t) \) is the laser–atom interaction operator. For a detailed discussion of Equations (5) and (6) and of approximations made there, we send the reader to review [19].

The time integral in the amplitude (5) is usually calculated by the saddle-point method. In the strong field limit we are considering here saddle points with real parts close to that time instants where the field is maximal give an exponentially dominant contribution into the amplitude. At the same time, the corresponding photoelectron momentum is close to the minus vector potential of the laser field taken at this time instants,

\[ p \approx -A(t_0). \quad (7) \]

This qualitative picture is summarized in Figure 2.

Given that, one could easily estimate the net momentum as \( -A(t_m) \) with \( t_m \) being the time instant where the field has its absolute maximum. The resulting momentum distribution calculated along the SFA and shown in Figure 3 fully confirms this qualitative picture: It consists on a single pronounced maximum at the momentum close to the one of the fundamental field vector potential and of two secondary maxima that gives a minor contribution into the net photoelectron momentum.
3. Discussion

The obtained results show that a two-color laser field consisted of a strong circularly polarized fundamental harmonic and of a linearly polarized second harmonic of infrared radiation generate photoelectron distributions highly asymmetric in the polarization plane. This asymmetry almost maximizes the net momentum of photoelectrons after the interaction with the laser pulse is over. Besides, the distribution near the maximum appears quite narrow in the momentum space, making the photoelectron current almost monoenergetic.

As far as emission of THz radiation caused by this current is concerned, the absolute value of the net momentum determines the radiation intensity. Thus, the optimal choice of the laser parameters should be dictated by maximization of the vector potential amplitude. At the same time, the laser intensity cannot be infinitely increased
3D plot of the photoelectron momentum distribution (in arbitrary units) calculated from Equations (5) and (6) for ionization of hydrogen by a laser pulse consisted of an 800-nm circularly polarized fundamental harmonic of intensity $10^{14}$W/cm$^2$ and a linearly polarized second harmonic with intensity equal to 5% of the fundamental ($\epsilon = 0.22$). The relative phase of the fields $\alpha = 0$. Horizontal axes show the projections of the photoelectron momenta in the polarization plane measured in atomic units.

because of the fast saturation of ionization at intensities exceeding $10^{14}$W/cm$^2$. As a result, the natural way for increasing the THz signal lies in the application of mid-infrared laser fields with wavelengths $\lambda \sim 2 \div 4 \mu m$ that should be possible with the presently existing laser sources.

**Funding**

This work was supported by the Russian Foundation of Basic Research through [Grant No. 16-02-00963a].

**References**

[1] Cook, D. J. and Hochstrasser, R. M. (2000). Intense terahertz pulses by four-wave rectification in air. *Optics Letters*, vol. 25, p. 1210.
[2] Clough, B., Dai, J., and Zhang, X.-C. (2012). Laser air photonics: beyond the terahertz gap. *Materialstoday*, vol. 15, p. 50.

[3] Kim, K. Y., Glownia, J. H., Taylor, A. J., et al. (2007). Terahertz emission from ultrafast ionizing air in symmetry-broken laser fields. *Optics Express*, vol. 15, p. 4577.

[4] Oh, T. I., You, Y. S., Jhajj, N., et al. (2013). Scaling and saturation of high-power terahertz radiation generation in two-color laser filamentation. *Applied Physics Letters*, vol. 102, p. 201113.

[5] Kampfrath, T., Sell, A., Klatt, G., et al. (2011). Coherent terahertz control of antiferromagnetic spin waves. *Nature Photonics*, vol. 5, p. 31.

[6] Fleischer, S., Zhou, Y., Field, R. W., et al. (2011). Molecular orientation and alignment by intense single-cycle THz pulses. *Physical Review Letters*, vol. 107, p. 163603.

[7] Lu, M., Shen, J., Li, N., et al. (2006). Detection and identification of illicit drugs using terahertz imaging. *Journal of Applied Physics*, vol. 100, p. 103104.

[8] Daigle, J. F., Théberge, F., Henriksson, M., et al. (2012). Remote THz generation from two-color filamentation: long distance dependence. *Optics Express*, vol. 20, p. 6825.

[9] Kotelnikov, I. A., Borodin, A. V., Shkurinov, A. P. (2011). *Journal of Experimental and Theoretical Physics*, vol. 139, p. 1081.

[10] You, Y. S., Oh, T. I., and Kim, K. Y. (2012). Off-axis phase-matched terahertz emission from two-color laser-induced plasma filaments. *Physical Review Letters*, vol. 109, p. 183902.

[11] Bagulov, D. S., Kotelnikov, I. A. (2013). *Journal of Experimental and Theoretical Physics*, vol. 143, p. 26.

[12] Johnson, L. A., Palastro, J. P., Antonsen, T. M., et al. (2013). THz generation by optical Cherenkov emission from ionizing two-color laser pulses. *Physical Review A*, vol. 88, p. 063804.

[13] Vvedenskii, N. V., Korytin, A. I., Kostin, V. A., et al. (2014). Two-color laser-plasma generation of terahertz radiation using a frequency-tunable half harmonic of a femtosecond pulse. *Physical Review Letters*, vol. 112, p. 055004.

[14] S.V. Popruzhenko, V.A. Tulsky. (2015). Control of terahertz photoelectron currents generated by intense two-color laser radiation interacting with atoms. *Physical Review A*, vol. 92, p. 033414.

[15] Meng, C., Chen, W., Wang, X., et al. (2016). *Applied Physics Letters*, vol. 109, p. 131105.

[16] Keldysh, L. V. (1965). Ionization in the field of a strong electromagnetic wave. *Soviet Physics—JETP*, vol. 20, p. 1307.

[17] Faisal, F. H. M. (1973). *Journal of Physics B*, vol. 6, p. L89.
[18] Reiss, H. R. (1980). Effect of an intense electromagnetic field on a weekly bound system. *Physical Review A*, vol. 22, p. 1786.

[19] Popruzhenko, S. V. (2014). Keldysh theory of strong field ionization: history, applications, difficulties and perspectives. *Journal of Physics B*, vol. 47, p. 204001.