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OPTICS AND LASER PHYSICS

Optical Harmonics Generation under the Interaction of Intense (up to 10^{14} W/cm^2) Mid-Infrared Femtosecond Laser Radiation of a Fe:ZnSe Laser System with a Dense Laminar Gas Jet

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Low-order (fifth, seventh, and ninth) harmonics have been generated under the interaction of intense ($I \sim 10^{14}$ W/cm^2) femtosecond mid-infrared radiation of a laser system based on a Fe:ZnSe crystal (wavelength is 4.55 μm, pulse duration by the FWHM level of intensity is 160 fs, and the pulse energy is up to 3.5 mJ) with an argon jet (pressure is up to 10 bar) in the tunneling ionization regime (Keldysh parameter is $\gamma = 0.2$). The maximum energy efficiencies of the 5$^{\text{th}}$, 7$^{\text{th}}$, and 9$^{\text{th}}$ harmonic generation are 2×10^{-7}, 6×10^{-9}, and 3×10^{-10}, respectively. It has been established that nonlinear effects of propagation of generating radiation under an increase in the pressure of the gas jet begin to significantly affect the process of generation.

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INTRODUCTION

Low- and high-order harmonic generation is currently under active investigation. Radiation with the spectrum in the form of numerous individual odd harmonics of generating radiation was detected for the first time in 1987 by researchers from France and United States under the focusing of picosecond laser pulses into the volume of a gas-filled cell [1, 2]. The development of these studies continued with femtosecond lasers, which made it possible to increase the energy of generated radiation, to expand the range of generated harmonics toward high energies of photons [3], and to implement generation in the regime of few-cycle laser action on a medium [4]. The development of experimental methods of harmonic generation and theoretical approaches to the description of this process [5, 6] resulted in the fundamental understanding of harmonic generation as a complex effect whose description requires the use of laws of quantum mechanics, laser physics, and nonlinear optics [7].

Studies of high- and low-order harmonics generation are currently aimed at increasing the energy and average power of generated radiation [8], expansion toward the X-ray spectral range [9], effective formation of coherent attosecond pulses [10, 11] that can be used to study the dynamics of atomic systems at the attosecond time scale [12, 13], and the development of methods for controlling the polarization properties of generated radiation [14]. Harmonic generation was studied in gaseous and plasma media [15], in gas-filled capillaries [16], and in the bulk [17] and on the surface of solid targets [18, 19]. Although these methods are promising, the simplest experimental method is harmonic generation in gaseous media, in particular, because of the possibility of fine adjustment of the geometric and optical parameters of the gas jet in order to optimize the parameters of generated radiation.

The generation of short-wavelength radiation in the vacuum ultraviolet and X-ray spectral ranges owing to high-order harmonic generation opens the possibility for creating compact sources of coherent radiation in these spectral ranges [20]. According to experimental and theoretical studies, the short-wavelength edge of the spectrum of generated harmonics satisfies the law $\omega_{\max} \sim I\lambda^2$ [5], where I and λ are the intensity and wavelength of generating radiation, respectively. According to this law, the spectrum of generated harmonics can be expanded by increasing either the intensity or wavelength of generating radiation. An increase in the intensity results in the ionization of the medium and in an increase in the destructive contribution from free electrons, which violates the phase-matching condition, thus reducing the energy yield of generated harmonics. Thus, the most natural method of expanding the spectrum of generated harmonics is the increase of the wavelength of generating radiation.

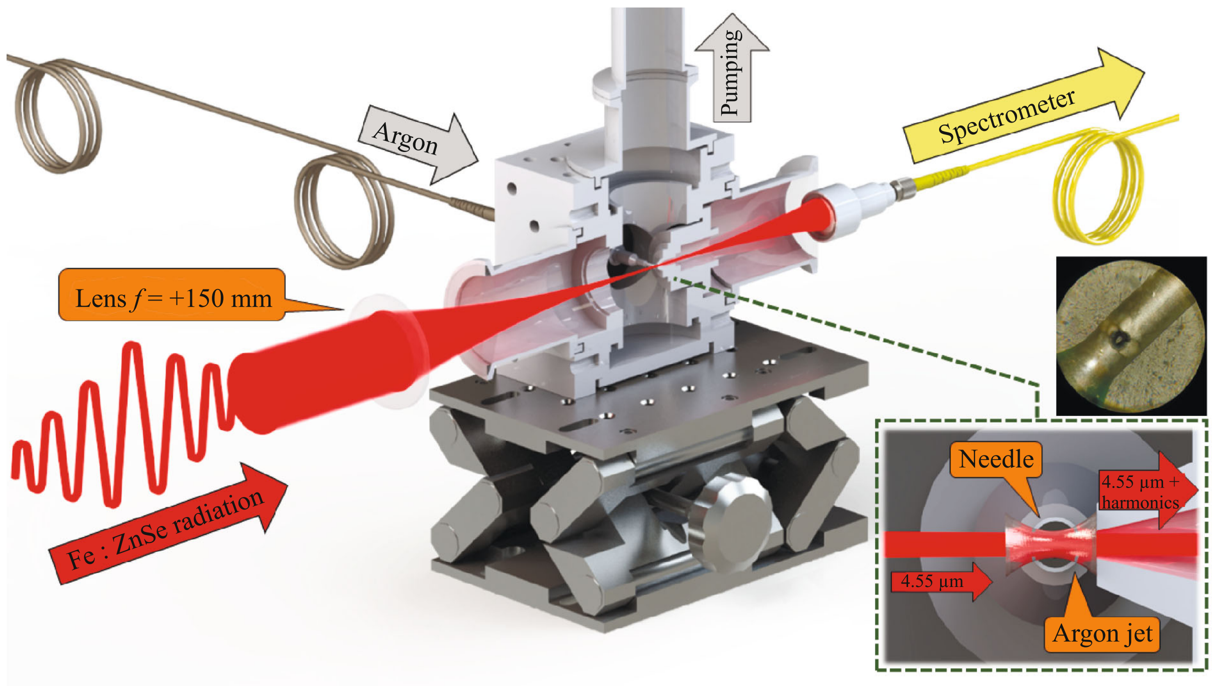

Fig. 1. (Color online) Layout of the experimental setup, the section of the interaction chamber, and (inset) the photograph of the orifice in the needle.

An increase in the wavelength of generating radiation reduces the efficiency of the photoemission response of a single atom owing to the diffusion of the freely propagating electron wave packet in the free motion stage, which reduces the energy response in a given spectral interval proportional to $1/\lambda^x$, where x = 5–6.5 according to theoretical and experimental results [21]. In this case, to conserve the energy of generated harmonics with an increase in the wavelength λ, it is necessary to increase the number of atoms involved in generation; this can be achieved by increasing the pressure of the gas jet and by the corresponding change in the phase-matching condition. One of the methods for the compensation of the increase in the wavelength of generating radiation can be the use of the medium for generation in the form of a dense (at a level of tens of bar) gas mixture of the atomic and molecular gases; the atomic gas provides a high nonlinearity, whereas the molecular gas allows one to control the phase-matching condition through the variation of its partial pressure [22].

Sources of the near-infrared (800 nm) [23] and mid-infrared (3.9 μm) [24, 25] ranges have already been used for harmonic generation. In this work, for harmonic generation, we used for the first time the unique femtosecond laser system based on the Fe:ZnSe crystal [26], which generates radiation at a wavelength of 4.55 μm, which is longer than wavelengths of sources previously used in harmonic generation experiments.

In this work, we detected low-order (5^{th}, 7^{th}, and 9^{th}) harmonics and studied the dependences of the generated harmonics energy on the pressure of the gas jet in the range of 0.25–10 bar. The measured dependences were compared to theoretical calculations within the nonperturbative approach to the description of radiation from a single atom [27] and the interference model, which allows the calculation of the response of an extended medium taking into account responses of atomic ensembles calculated quantum-mechanically [28]. The comparison of the experimental and theoretical results showed that an increase in the pressure of the gas jet significantly enhances the role of nonlinear propagation effects for laser radiation in the harmonic generation process; manifestation of these effects was detected in the performed experiment.

EXPERIMENTAL SETUP

The layout of the experimental setup is shown in Fig. 1. The femtosecond laser system based on a Fe:ZnSe crystal [26] (wavelength is 4.55 μm, pulse duration by the FWHM level of intensity is 160 fs, and the pulse energy is up to 3.5 mJ) was used as the radiation source. Laser radiation from this system was focused by a lens with a focal length of f = 150 mm inside a needle (inner diameter is 1 mm) through which argon (purity 6.0) was transmitted. The needle was placed in a chamber evacuated to a pressure is 0.01 Torr (Ebara EV-SA20, 1760 L/min). The input

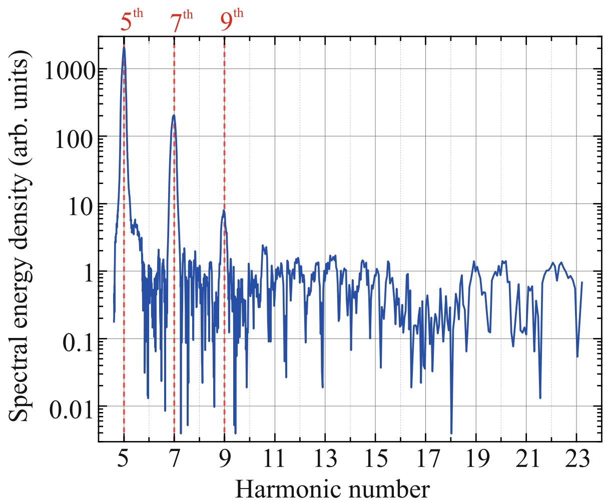

Fig. 2. (Color online) Spectrum of harmonics at an intensity of 1.13×10^{14} W/cm^2 on the gas jet, where the gas jet pressure is 10 bar. The 5$^{\text{th}}$, 7$^{\text{th}}$, and 9$^{\text{th}}$ harmonics are clearly seen.

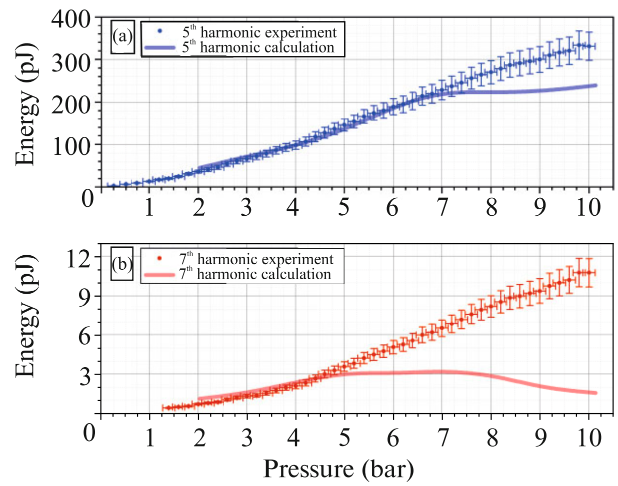

Fig. 3. (Color online) Dependences of the energies of the 5$^{\text{th}}$ (a) and 7$^{\text{th}}$ (b) harmonics on the pressure of the gas jet—experimental (points with errors) and calculated (lines) results.

and output orifices 400 and 160 μm in diameter, respectively, were made in the opposite ends of the needle, through which the laser beam passed ($1/e^2$ intensity diameter of the waist is (144 ± 17) μm; the corresponding Rayleigh vacuum length is 3.58 mm) and radiation of generated harmonics was emitted. The end orifice in the needle was closed. The energy of the generating pulse in the waist reached 1.57 mJ, which corresponds to the vacuum intensity at the laser beam waist of 1.13×10^{14} W/cm^2. Radiation of harmonics downstream of the interaction chamber was collected by a collimator to a fiber connected to an Ocean Optics QE Pro spectrometer with the detection range of 200–1000 nm. The argon pressure was controlled by means of a pressure regulator placed in the gas tract on the path to the needle.

RESULTS AND DISCUSSION

The radiation spectra of harmonics were obtained in the experiment for several pressures of the gas jet (Fig. 2). The maximum energy efficiencies of the 5$^{\text{th}}$, 7$^{\text{th}}$, and 9$^{\text{th}}$ harmonic generation are 2×10^{-7}, 6×10^{-9}, and 3×10^{-10}, respectively. Below, we discuss the dependences for the 5$^{\text{th}}$ and 7$^{\text{th}}$ harmonics because the 9$^{\text{th}}$ harmonic was detected only at the maximum pressure of 10 bar.

To experimentally determine the energy of each harmonic, we calibrated the energy spectrum taking into account the measured spectral response of the detection system and the calibration energy value. The energy of each harmonic was calculated as the integral of the spectrum near the spectral peak of the harmonic. Figure 3 shows the dependences of the experimental energies of the 5$^{\text{th}}$ and 7$^{\text{th}}$ harmonics on the pressure of the gas jet. Physical mechanisms determining the form of experimental dependences in Fig. 3 can be qualitatively described on the basis of the perturbative approximation and phase-matching effects. The perturbative approximation is applicable because the energies of the generated photons ($\hbar\omega_5 = 1.36$ eV, $\hbar\omega_7 = 1.91$ eV) are much lower than the ionization potential of the argon atom ($I_p^{\text{Ar}} = 15.76$ eV). According to [29], the pressure dependence of the energy of the q^{th} harmonic can be represented in the form

$$E_q \sim |pF(p)|^2. \quad (1)$$

Here, p is the pressure of the medium proportional to the volume density N of atoms of the medium and $F(p) = \int_{-L/2}^{L/2} dz' \left(1 + i\frac{2z'}{b(p)}\right)^{1-q} \times e^{-i\Delta k_q(p,z')z'}$ is the phase-matching integral, where L is the length of the nonlinear medium, $b(p) = 2z_{\text{R}}(p)$ is the confocal parameter of the generating beam, $z_{\text{R}}(p)$ is its Rayleigh length, and $\Delta k_q(p,z)$ is the mismatch between the wave vectors of the generating wave and its q^{th} harmonic. Taking the material dispersion of the medium [30] and generated plasma [31], as well as the Gouy phase shift [21], the mismatch between the wave vectors can be expressed in the form

$$\Delta k_q(p,z) = \frac{2\pi q}{\lambda_1}\frac{p}{p_0}(n_q - n_1)(1-\eta) - q\frac{\lambda_1}{\pi a^2(p,z)} - q\frac{p}{p_0}\eta n_{\text{a}} r_e \lambda_1, \quad (2)$$

where λ_1 is the wavelength of generating radiation; p is the pressure of the gas jet; $p_0 = 1$ atm is the atmospheric pressure; n_q and n_1 are the refractive indices of the q^{th} and fundamental harmonics at a pressure of 1 atm, respectively; η is the degree of ionization; $a^2(p,z)$ is the square of the $1/e$-field radius of the Gaussian beam of fundamental radiation; n_a is the concentration of atoms in the gas jet; and r_e is the classical radius of the electron.

According to the above expressions, the quadratic increase in the energy of harmonics $E_q \sim p^2$ at low pressures is provided by an increase in the concentration of elementary emitters (atoms of the medium); in this case, the influence of phase mismatch Δk_q and, as a result, the phase matching integral $F(p)$ is small (see Eq. (1)). As the pressure of the gas jet increases, the influence of phase mismatch Δk_q becomes comparable with the effect of an increase in the number of atoms in the medium, which results in deviation from the dependence $E_q \sim p^2$.

For a more detailed and consistent description of the measured dependences, a model was used in the frame of which for description of the microscopic response of the medium a nonperturbative theoretical approach was applied to describe the radiation of a single atom [27], and the macroscopic response of the medium was described within an interference model [28]. The variations of the parameters of the laser beam propagating in the gas caused by dispersion and focusing by the lens, as well as the variation of the intensity of laser radiation over the waist and the Gouy phase shift, were taken into account. The effect of free electrons was taken into account within the classical Lorentz theory. The absorption of generated radiation was disregarded in the model. The results of the calculations within this model are shown in Fig. 3. The calculated dependence coincides with the experimental one at low pressures up to 7 and 5 bar for the 5$^{\text{th}}$ and 7$^{\text{th}}$ harmonics, respectively, whereas the calculated dependence differs from the experimental one at higher pressures. This difference can be explained by the effect of disregarded nonlinear propagation effects for generating radiation in the gaseous medium, which change the spatiotemporal structure of the field of the generating laser pulse. The influence of these effects on the generating pulse at low pressures of the gas jet is weak because the concentration of atoms in the medium is low; as a result, the calculated dependence coincides with the experimental one at low pressures. The concentration of atoms in the medium increases with the pressure, which results in the enhancement of nonlinear propagation effects on the generated pulse, leading to the deviation of the calculated dependence from the experimental one.

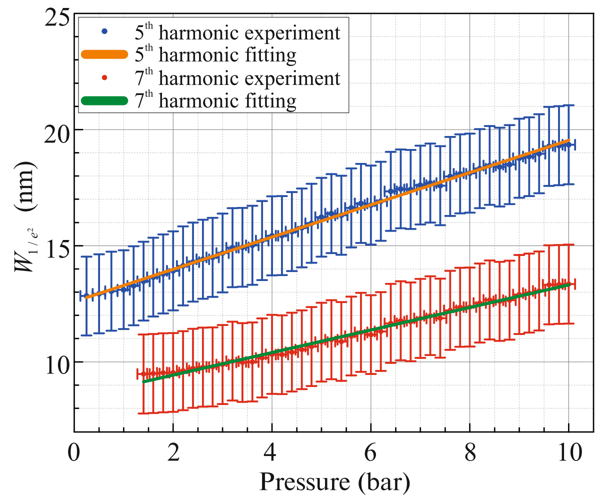

Fig. 4. (Color online) Dependence of the spectral width of the 5$^{\text{th}}$ and 7$^{\text{th}}$ harmonics on the pressure of the gas jet. Experimental dependences are indicated by dots, the result of linear approximation is indicated by lines. The approximation parameters are presented in Table 1.

This hypothesis is confirmed by the experimentally detected influence of the self-phase modulation (SPM) on the spectrum of generated harmonics. In particular, the spectral width of generated harmonics in the experiment changes under the variation of the pressure of the gas jet. The measured dependences of the spectral width of harmonics on the pressure of the gas jet are shown in Fig. 4.

The effect of the self-phase modulation is manifested in the linear dependences in Fig. 4. In particular, the broadening of the spectrum under the self-phase modulation due to the third-order nonlinearity $\chi^{(3)}$ can be represented in the form [32]

$$\Delta\omega(t) = -kn_2\int_0^L \frac{\partial I(t,z')}{\partial t}dz', \qquad (3)$$

where $k = 2\pi/\lambda$ is the wavenumber of the pulse at the central wavelength, n_2 is the nonlinearity coefficient determining the nonlinear addition to the refractive index $\Delta n = n_2 I$, t is the time in the running coordinate system, and L is the length of the medium. Since the nonlinear index n_2 of argon is linearly proportional to the gas pressure [33], according to Eq. (3), the broadening of the spectrum should also be proportional to the gas pressure, which is indeed observed in the experiment (Fig. 4). Thus, the effect of the pressure of the gas medium on the spectral width of harmonics can be attributed to self-phase modulation. According to the approximation of dependences in Fig. 4, the rate of increase in the spectral width of the fifth harmonic with the pressure is higher than that for the seventh harmonic because the narrowing of the spectrum of the q^{th} harmonic compared to the spectrum of generating radiation is proportional to q. In particular, the

Table 1. Parameters of the approximation of the dependences in Fig. 4

Function	$y = a + bx$	
Dependence	5th harmonic	7th harmonic
a	12.595 ± 0.035	8.459 ± 0.049
b	6.942 ± 0.060	4.854 ± 0.079
R^2	0.997	0.989

spectral energy density of the generated second harmonic is [34]

$$S_2(\omega) \sim \mathrm{sinc}^2\left(-\frac{\Delta k_2 z}{2}\right) S_1(\omega). \tag{4}$$

Generalizing Eq. (4) to the case of the q^{th} harmonic and taking into account that $\Delta k_q \sim q$ (see Eq. (2)), we conclude that the narrowing of the spectrum of the q^{th} harmonic is described by the function

$$S_q(\omega) \sim \mathrm{sinc}^2\left(-\frac{\mathrm{const} \cdot z}{2 \times 1/q}\right). \tag{5}$$

The width of this function is $\sim 1/q$. Consequently, the ratio of the spectral widths for the 5^{th} and 7^{th} harmonics, as well as the ratio of the rates of their increase with the gas pressure growth, should be equal to the inverse ratio of the numbers of the harmonics, i.e. $w_5/w_7 = q_5/q_7 = 5/7 \approx 0.71$. The ratio of the increase rates in the dependences in Fig. 4 (see Table 1) is (4.854 nm/(10 bar))/(6.942 nm/(10 bar)) = 0.699 ± 0.012. Thus, both ratios coincide within the experimental error. Therefore, the difference between the increase rates in the spectral widths of the fifth and seventh harmonics is explained by the dependence of the narrowing of the harmonic spectrum on its number.

CONCLUSIONS

The low-order (5^{th}, 7^{th}, and 9^{th}) harmonics have been generated under the excitation of the argon jet with the pressure up to 10 bar by the intense (1.13×10^{14} W/cm^2) femtosecond mid-IR (4.55 μm) laser radiation. The maximum energy efficiencies for the 5^{th}, 7^{th}, and 9^{th} harmonic generation are 2×10^{-7}, 6×10^{-9}, and 3×10^{-10}, respectively.

Comparison of experimental results and theoretical calculations indicates a noticeable manifestation of nonlinear propagation effects influencing the spatiotemporal distribution of the laser pulse field at high (above 5 bar) pressures of the gas jet. In particular, as shown in the experiment, an increase in the pressure of the gas jet leads to the broadening of the spectrum of each generated harmonic due to the effect of self-phase modulation of generating radiation.

Thus, since effective harmonic generation involving long-wavelength radiation is associated with an increase in the pressure of the gas jet, a significant influence of nonlinear propagation effects on the energy yield of generated radiation is observed in the case of the use of mid-IR radiation for harmonic generation in contrast to near-IR radiation.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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