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Selective generation of a higher harmonic in plasma

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***Abstract*. It is shown for the first time that the use of autoionisation states for phase matching leads to the efficient selection of a single harmonic generated in a plateau region in plasma. The selected harmonic frequency can be tuned by changing the relative concentration of plasma components and tuning the fundamental radiation frequency. It is shown that the contrast of the selected harmonic can exceed 10^4.**

Keywords: *higher harmonic generation, plasma, phase matching, autoionisation state*.

The higher harmonic generation (HHG) is a promising method for obtaining soft X-rays with a high spectral brightness and coherence and minimal pulse duration. However, practical applications of HHG involve considerable difficulties due to a low energy conversion efficiency and almost uniform radiation energy distribution in a comparatively broad spectral range (the plateau region) [1, 2]. The selection of a single harmonic or a group of harmonics is performed by 'passive' methods with the help of filters or dispersion elements, which introduce the energy loss (see, for example, [3]). Different methods for increasing the HHG efficiency, the selection of a single harmonic or a group of harmonics were proposed in a number of papers. These methods include the use of phase matching based on the compensation of the negative dispersion of a medium by employing the waveguide regime and ionisation of the medium [4], the realisation of quasi-synchronous interactions, the control of the wave front and spatial phase in hollow waveguides [5 – 7], and optimisation of the spectral and temporal distributions of the amplitude and phase [8, 9]. Nevertheless, the typical HHG efficiency remains at the level $10^{-6} - 10^{-8}$. In most papers, HHG was studied in inert gases (in jets or waveguides). Higher harmonic generation in plasma, which attracts recent interest, offers a number of advantages such as the possibility of using various elements with different physical properties, the control of the population of excited states, the preparation of extended media, etc. [10, 11].

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In this paper, we propose to use autoionisation states for HHG phase matching in plasma for separating a single harmonic in the plateau region. The response of atomic systems during HHG was analysed by solving approximately the nonstationary Schrödinger equation [12, 13]. In this approximation for a two-level atomic model, radiation is generated at frequencies $\Omega = \pm(2n+1)\omega \mp \Delta\omega_{kl}$ (here, $\Delta\omega_{kl}$ is the difference between the frequencies of the atomic states), whereas in an atomic model with one state, only odd radiation harmonics are generated.

The influence of phase conditions on HHG in a polyatomic system was analysed by using classical nonlinear optics equations [14, 15]. The solution of these equations in the case of a weak energy exchange is well known for different types of laser radiation focusing. When the diffraction length R_d of a Gaussian beam of fundamental radiation greatly exceeds the length L of a medium ($R_\mathrm{d} \gg L$), the intensity of the ith radiation harmonic is described by the expression

$$I_i = \left(\frac{2\pi\Omega^2 N d_n}{k_i c^2} L \frac{\sin \Delta_i}{\Delta_i}\right)^2. \quad (1)$$

Here, d_n are the Fourier components of the time-dependent dipole moment; $\Delta_i = (L/2R_\mathrm{d})(q-1-\alpha_i)$; $q = 2n+1$; N is the atomic medium density; and k is the wave number; $\alpha_i = R_\mathrm{d}\Delta k_i = R_\mathrm{d}[(2n+1)k - k_i]$ is the normalised phase mismatch. If $R_\mathrm{d} < L$, the consideration of diffraction leads to more complicated expressions. Nevertheless, in this case the solution can be obtained which is determined by the so-called phase integral [16]. The consideration of self-action effects complicates such solutions (see, for example, [17]).

The phase mismatch of a plasma medium is determined by additive contributions of the atomic and electron components. The dispersion of the electron gas is positive, and the corresponding phase mismatch is

$$\Delta k_{q\mathrm{e}} = q k_{1\mathrm{e}} - k_{q\mathrm{e}} = \frac{e^2 N_\mathrm{e}}{m_\mathrm{e} c\omega}\left(\frac{1}{q} - q\right) < 0,$$

where N_e is the free electron density. The value and sign of the atomic component $\Delta k_q = q k_1 - k_q = q\omega c^{-1}(n_1 - n_q)$ (n_q is the refractive index at the qth radiation frequency) depend mainly on the refractive index at the fundamental radiation frequency due to monotonic and comparatively weak, as a rule, dispersion in the region located much higher than the atomic ionisation potential. For this reason, it is difficult to achieve phase matching for a single higher harmonic. In particular, simple estimates show that, when the phase matching $\Delta k_q \approx 0$ is realised for the 25th harmonic of a Ti : sapphire laser, the coherence length

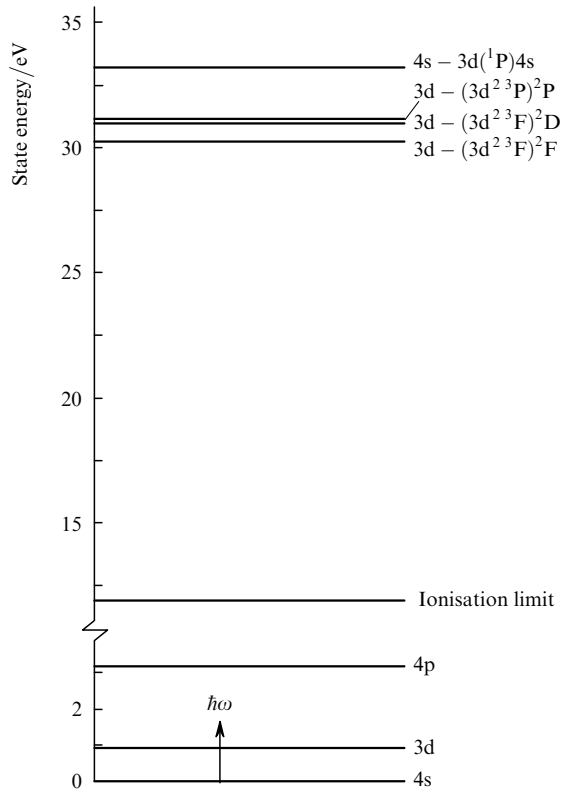

Figure 1. Strongest atomic transitions between the ground (4s) and metastable (3d) states and autoionisation states in Ca II ions.

$l_c = \pi/\Delta k_q < 1$ mm is achieved for adjacent harmonics when the medium density exceeds 10^{20} cm^{-3}. The consideration of the laser linewidth further complicates the selection of a single harmonic.

Consider the HHG model in calcium plasma containing singly charged ions and free electrons. Transitions between the ground (4s) and metastable (3d) states in calcium ions considerably enrich the absorption spectrum and change the optical properties of the atomic medium. Figure 1 presents the strongest atomic transitions between these and autoionisation states of Ca II ions. The energies and oscillator strengths of these transitions lie in the regions 29 – 33 eV and 0.2 – 2.1, respectively [18, 19]. These transitions correspond to the energies of the 19th – 23th harmonics of a Ti : sapphire laser and can change the phase mismatch to generate these harmonics.

By changing the relation between populations of the 4s and 3d states of Ca II, it is possible to change the phase mismatch and to achieve phase matching at different wavelengths of fundamental radiation and harmonics (Fig. 2). This figure presents the dependences of the phase mismatch on the fundamental radiation wavelength for different harmonics. The influence of free electrons leads to the appearance of a pedestal, which changes the optimal fundamental radiation wavelength at which phase matching is achieved, and also can result in the suppression or appearance of phase matching at certain wavelengths. It follows from Fig. 2 that the presence of autoionisation states allows one to achieve phase matching for a selected harmonic. In particular, when the phase matching is

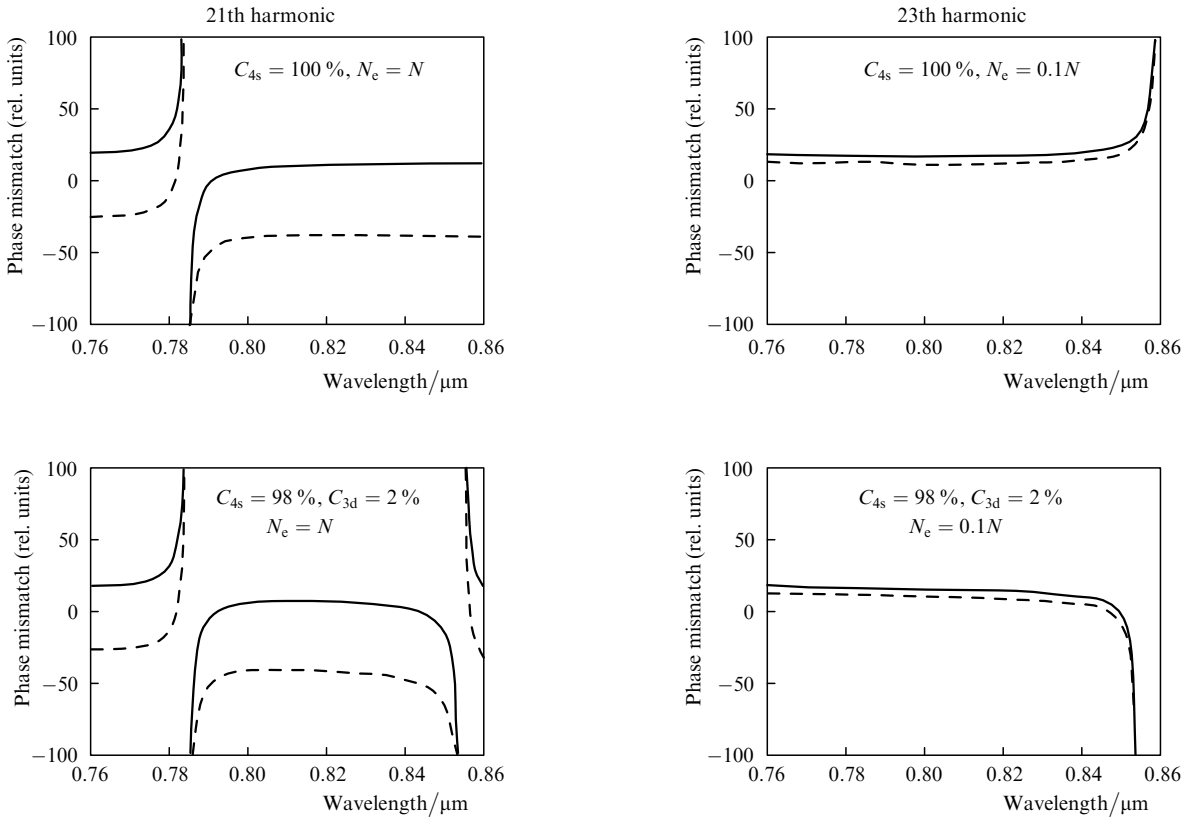

Figure 2. Dependences of the phase mismatch for the 21th and 23th harmonics of the fundamental radiation at the concentrations of Ca II ions in different states $C_{4s} = 100\,\%$, $C_{4s} = 98\,\%$, and $C_{3d} = 2\,\%$. The dependences obtained taking into account the influence of free electrons are shown by dashed curves for $N_e = N$ and $N_e = 0.1N$.

achieved for the 21th harmonic of a 0.78-μm laser, the coherence length smaller than 1 mm is achieved for adjacent harmonics at the ion density $\sim 10^{17}$ cm^{-3}.

Such a phase matching achieved due to the influence of autoionisation states leads to the increase in the generation efficiency of an individual harmonic. Figure 3 presents the spectral distribution of higher harmonic intensities calculated for Ca II ions with the populated 4s state. The wavelength, linewidth, and intensity of the fundamental radiation used in calculations were 0.79 μm, 8 nm, and 3.5×10^{14} W cm^{-2}, respectively. Note that the linewidth of atomic transitions was neglected in calculations. The spectral distribution of harmonic intensities related to the response of individual ions is denoted by circles. One can see that this dependence is of a plateau-like type – the intensity distribution of higher harmonics is virtually uniform in the region covering ~ 20 odd harmonics. The intensity distribution for higher harmonics obtained for the collective response of the medium of length 1 cm at the concentration of Ca II ions of 10^{19} cm^{-3} is shown by triangles. One can see that the phase-matching condition leads to the selection of the 21th harmonic with the contrast (the intensity ratio of the selected and adjacent harmonics) over 10^4. The estimated conversion efficiency of radiation intensity to the selected harmonic was $\sim 10^{-2}$.

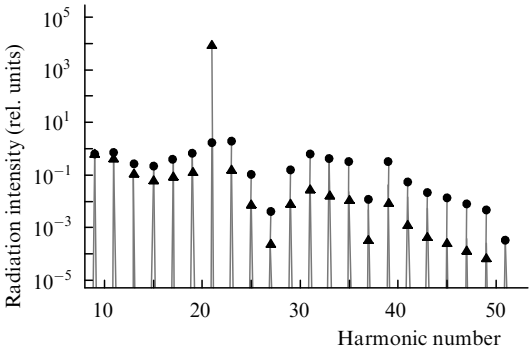

Figure 3. Intensity distribution for higher harmonics. The responses of single ions and ion medium are shown by circles and triangles, respectively.

It follows from (1) that a decrease in the medium density reduces the relative phase mismatch for adjacent harmonics and the contrast of the selected harmonic decreases. As the concentration of Cs II was decreased down to 10^{18} cm^{-3}, the contrast decreased to $\sim 5 \times 10^3$, whereas at the ion concentration of 10^{17} cm^{-3} the contrast remained at the level 10^3. As the spectral width of fundamental radiation was increased up to 32 nm, the contrast decreased approximately by four times. Note that variations in the spectral width almost did not change relations between the intensities of other higher harmonics.

According to the results presented in Fig. 2, a change in the relative concentration of plasma components leads to the shift of the radiation wavelength at which a single harmonic can be selected, and also to the change in the number of the selected harmonic. When the concentration of Ca II ions in the initial 3d state is equal to 10 %, the contrast of the selected 23th harmonic of the 0.8-μm radiation is $\sim 5 \times 10^2$ for other parameters corresponding to the dependence in Fig. 3.

It was found experimentally in [20] that the contrast of the 13th harmonic of a Ti : sapphire laser generated on the indium surface in laser plasma was ~ 200. Our calculations show that the influence of the strong $4d^{10}5s^2\,{}^1S_0 - 4d^9 5s^2\,{}^5p\,({}^2D)^1P_1$ transition to the autoionisation state of indium ions leads to the achievement of phase matching for the 13th harmonic and provides a similar contrast (we used in calculations the level energies and oscillator strengths from [21]). However, this contrast was obtained in calculations for the fundamental radiation wavelength that was greater than in experiments. The consideration of the Stark shift or Kerr nonlinearity leads to a better agreement with experimental data. Note that the increase in the harmonic intensity can be explained by the achievement of resonance conditions. However, the intensity of adjacent harmonics also increases under resonance conditions, whereas the realisation of phase-matching conditions can provide the selection of a single harmonic, in particular, in regions located far from resonance regions.

Thus, we have shown that the use of autoionisation states leads to the selective generation of a single harmonic in plasma. Our calculations have shown that the phase matching achieved for the HHG of radiation from a Ti : sapphire laser in the Ca II plasma due to the influence of autoionisation states leads to the selection of a single harmonic, the contrast for the 21th harmonic achieving 10^4. The selection contrast depends on the medium density and laser linewidth. The frequency of the selected harmonic can be tuned in the plateau region by changing the relative concentration of plasma components and the fundamental radiation wavelength. Due to the influence of the Stark shift, Kerr nonlinearity or generation of free electrons, the optimal laser radiation frequency at which a single harmonic is efficiently selected is shifted.

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