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Compensation for phase mismatch of high harmonics by the group-velocity mismatch

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Abstract. **A mechanism providing an essential enhancement of the conversion efficiency of a single high harmonic in gaseous media is first proposed using an appropriate change in the phase mismatch and group-velocity mismatch in the vicinity of resonance.**

Keywords: *high harmonic generation, phase mismatch and group-velocity mismatch.*

1. The basic method for enhancing the harmonic generation efficiency (including high harmonics) is the phase-velocity matching of the fundamental radiation and the harmonic radiation. Phase matching (PM) for high harmonic generation (HHG) is hampered by relatively strong dispersion of refractive indices of atomic media, effect of free electrons, temporary variations in the particle concentration caused by ionisation of the medium, dependence of the harmonic phase on the fundamental radiation intensity. As a rule, PM is achieved using laser radiation beams with an appropriate phase shift and (or) compensating for the atomic dispersion by the dispersion of the electron gas. The phase matching in HHG is achieved in the waveguide regime [1] and in a capillary discharge [2], as well as by implementing quasi-synchronous PM in hollow waveguides [3] and by controlling the laser radiation wavefront [4, 5].

2. The present paper shows that the change in the phase mismatch and group-velocity mismatch during the HHG process in gaseous media near resonance may provide PM. The influence of group-velocity mismatch on the PM processes has been analysed mainly for frequency conversion processes in crystals (see, e.g., [6, 7]) and for the third harmonic generation in the waveguide regime [8]. For HHG in gaseous media the effect of the group-velocity mismatch is believed not to be essential. In the case of plasma media with the electron and ion density of 10^{18} cm^{-3} the coherence length of the qth harmonic $L_{cq} = \pi/\Delta k_q$ is ~0.01 cm. Here $\Delta k_q = qk_1 - k_q$ is the phase mismatch and k_i is the ith harmonic wave number. For the duration of a transform limited pulse $\tau = 50$ fs the characteristic quasi-static interaction length $L_{gq} = \tau/v_q$ in such media is about 5 cm. Here $v_q = 1/u_q - 1/u_1$ is the group-velocity mismatch and u_i is the group velocity of the ith pulse. Therefore, to equalise its effect with that of the phase mismatch, the quasi-static interaction length should be decreased by two or three orders of magnitude. The appropriate value of v_q corresponds to the group-velocity mismatch which is by nearly an order of magnitude smaller than its typical values for crystals in the optical range. However, using gaseous media with increased group-velocity mismatch, in particular, the media with autoionisation resonances, allows equalising the effects of group and phase mismatch. Note that since the group velocity is determined by the derivative of the wave number, the growth of the group-velocity mismatch may be more essential than the change in the wave number (and, therefore, the phase mismatch) and can be observed in a region lying far away from the resonance.

3. The analysis of wave propagation during the HHG was carried out using the fixed field approximation. Within the framework of the slowly varying amplitude approximation and the first-order dispersion theory, the envelope function of the generated pulse

$$E_q(z,t) = \tfrac{1}{2}A_q(z,t)\exp[\mathrm{i}(\omega_q t - k_q z)] + \text{c.c.}$$

in the moving coordinate system ($z = z$, $\mu = t - z/u_1$) may be written in the form (see, e.g., [6, 7])

$$\left(\frac{\partial}{\partial z} + v_q\frac{\partial}{\partial \mu}\right)A_q(z,\mu) = -\mathrm{i}\gamma_q d_q(\mu)\exp(-\mathrm{i}\Delta k_q z)$$

$$-\alpha_q A_q(z,\mu). \qquad (1)$$

Here $\gamma_q = 2\pi\omega_q^2 N/k_q c^2$; α_i is the absorption coefficient for the ith harmonic; N is the concentration of the atomic particles; $d_q(\mu)$ is the time-dependent dipole moment, oscillating at the frequency of the harmonic;

$$d(\mu) = \sum_m d_m(\mu)\exp[\mathrm{i}m(\omega_1 t + \phi_1)];$$

ϕ_1 is the initial phase of the fundamental radiation. In Eqn (1) the change in electron and ion concentrations during the pulse is not taken into account and the inequality $|\partial d(\mu)/\partial\mu < \omega_q|d(\mu)|$ is supposed to be valid.

Within the spectral approach the harmonic amplitude is presented as a Fourier integral

$$A_q(z,\mu) = \int_{-\infty}^{\infty}\Phi_q(z,\Omega)\exp(\mathrm{i}\Omega\mu)\mathrm{d}\Omega.$$

Then the right-hand side of Eqn (1) may be written as an integral that, alongside with the frequency-shifted amplitudes of the fundamental radiation, entering the dipole moment (the

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so-called convolution), contains also the phase mismatch for all spectral components (see, e.g., [9]). Under the near-resonance conditions at the frequency of the harmonic only the wave number of the harmonic changes essentially, while the fundamental radiation wave number is practically independent of the frequency within the line width (up to the ratio of the spectrum width to the radiation frequency).Then for the harmonic amplitude in the spectral representation we have

$$\frac{\partial \Psi_q(z,\Omega)}{\partial z} = -\,\mathrm{i}\gamma_q d(\Omega)\exp[\mathrm{i}\varphi_q(\Omega)z + \alpha_q(\Omega)z]. \qquad (2)$$

Here $\Psi_q(z,\Omega) = \Phi_q(z,\Omega)\exp(\mathrm{i}v_q\Omega z + \alpha_q z)$; $d(\Omega)$ is the Fourier transform of the time-dependent dipole moment; $\varphi_q(\Omega) = v_q\Omega - \Delta k_q(\Omega)$ is the generalised mismatch.

The dipole moment $d(\Omega)$ was determined using the technique, described in [10, 11]. The present solution is a set of odd harmonics, distributed in the plateau region. In the case of a relatively narrow spectrum ($\Delta\Omega_q \ll \omega_1$) we present the solution for the dipole moment in the form of a Gaussian function $d_q(\Omega) = d_{q0}\exp[-(\Omega/\Delta\Omega_q)^2]$. Then the spectral density of the harmonic radiation $s_q(z,\Omega) = |\Phi_q(z,\Omega)|^2$ is

$$s_q(z,\Omega) = |2\gamma_q d_{q0}|^2 \exp[-2(\Omega/\Delta\Omega_q)^2 - \alpha_q(\Omega)z]$$

$$\times \frac{\sin^2[\varphi_q(\Omega)z/2] + \sinh^2[\alpha_q(\Omega)z/2]}{\varphi_q^2(\Omega) + \alpha_q^2(\Omega)}. \qquad (3)$$

The analysis of Eqn (3) implies that the radiation spectral density of the harmonic is determined by the generalised mismatch φ_q, as well as by the value of $\Delta\Omega_q$.

4. For the analysis we have chosen the HHG process in laser plasma, consisting of indium ions, i.e., under the conditions, close to the experiment [12], in which a substantial increase (~200 times) was observed for the intensity of the 13th harmonic of Ti:sapphire laser radiation with the frequency near the strong transition into the autoionisation state In II $4d^{10}5s^2\,{}^1S_0 - 4d^9 5s^2 5p(^2D)\,{}^1P_1$. In the calculations the plasma density (i.e., the concentration of ions and electrons) was set to be 10^{18} cm^{-3}, the laser radiation intensity amounted to 2×10^{14} W cm^{-2}, the radiation wavelength was equal to 0.8 μm. Figure 1 shows a fragment of the time dependence of the dipole moment for a Gaussian laser pulse with the FWHM duration of 40 fs. In the calculation of $d(t)$ following the method of Refs [10, 11] the Slater wave functions [13] of the initial state were used. The inset in Fig. 1 shows the corresponding dependence of the dipole moment spectral distribution in the range of the 13th harmonic. From the figure it follows that $\Delta\Omega_{13} = 0.064\omega_1$.

The calculation of phase and group-velocity mismatch was accomplished using the values of the oscillator strengths and transition frequencies from [14]. The total phase mismatch of singly charged indium ions and free electrons was –129 cm^{-1}, the group-velocity mismatch was –970 fs cm^{-1}, the absorption coefficient was equal to 10^{-2} cm^{-1}. Figure 2 presents the dispersion dependence of the radiation spectral density of the 13th harmonic (solid curve) for the 1-mm-long plasma and the constant values of phase and group-velocity mismatches mentioned above. The dashed curve presents the dependence, obtained at the group-velocity mismatch two orders of magnitude smaller (i.e., typical for the absence of resonance). From Fig. 2 it is seen that the maximal spectral

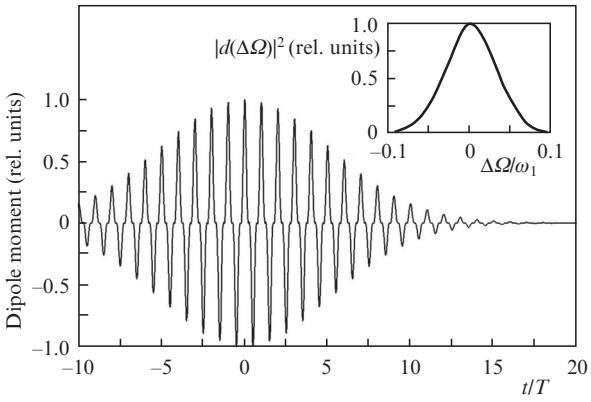

Figure 1. Fragment of the normalised time dependence of the dipole moment for a Gaussian fundamental beam (T is the oscillation period). The inset shows the corresponding dependence of the square of dipole moment on the oscillation frequency in the region of the 13th harmonic.

density of the harmonic at larger group-velocity mismatch is essentially higher (nearly by 340 times), and the spectrum of the main maximum is nearly 3.3 times narrower. The radiation energy of the harmonic in this case is ~120 times greater. The values of the phase and group-velocity mismatches and their ratio are essentially changed within the linewidth of laser radiation, which requires the analysis of partial spectral components. In the inset of Fig. 2 the dispersion dependences of the spectral density of the 13th and 15th harmonics are presented, which were calculated with the changes of phase and group-velocity mismatches taken into account. It is seen that the spectrum of the 13th harmonic acquired an indented shape which is determined by the relation between the dispersion terms of different orders. The maximal spectral density of the 13th harmonic is 277 times higher than the spectral density of the 15th harmonic, and the radiation energy of the 13th harmonic is nearly 65 times greater than that of the 15th harmonic.

Note, that in the experiment [12] the central line of the 13th harmonic was also shifted; however, its spectrum was almost two times narrower that that of adjacent harmonics,

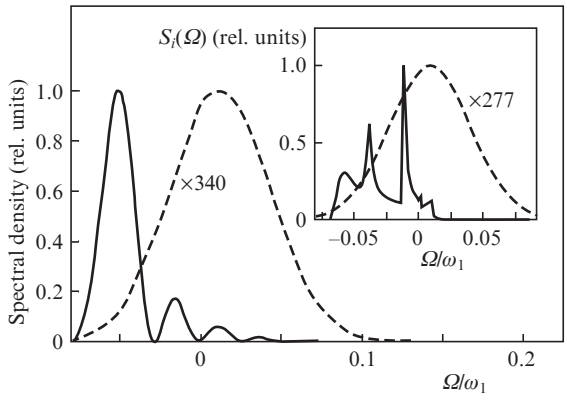

Figure 2. Spectral density of the 13th harmonic s_{13} versus frequency far from the resonance (dashed curve) and near the resonance (solid curve). The inset shows the frequency dependence of the spectral density of the 13th harmonic (solid curve) and the 15th one (dashed curve) within the linewidth of laser radiation with the change of the phase and group detunings taken into account.

and the spectral broadening of all harmonics was greater than the calculated one. The change in the spectra of harmonics may be explained as follows. In the calculations the dependence of the harmonic phase on the laser radiation intensity was not taken into account. When the gradients of the temporal and spatial intensity distributions are significant, this circumstance may change the spectrum of the harmonic. The change in the concentration of the indium ions was also not taken into account, which is valid at relatively low intensities. Preliminary analysis of the effect of the concentration change on the nonlinearity coefficient has shown that the spectral width of the harmonic increases nearly by 1.2 times. In the estimates the change in the ion concentration as a function of time was approximated by the function $1 - a\{1 + \tanh[b(t - \tau_d)/\tau]\}$, where $a = 0.3$, $\tau_d = 0$, and $b = 4$ are coefficients determined from agreement with the formulae of the ADK-theory [15]. However, at the laser radiation intensities greater by an order of magnitude than the one involved in the calculations, the spectral broadening of high harmonics due to the change in the plasma composition during the pulse agreed with the experiment.

5. Thus, a mechanism for enhancing the efficiency of high harmonic generation and selecting a single harmonic in the plateau region is proposed by changing the phase and group-velocity mismatches near the resonance. In this case under the phase mismatch the difference in group velocities provides the PM regime at the frequency, changing due to the medium dispersion with the radiation linewidth. As a result the width and position of the central frequency component of the harmonic are changed. The demonstrated possibility of a substantial intensity increase of a separate harmonic agrees with the known experimental data.

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