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Analytical treatment of quasi-phase matching of high-order harmonics in multijet laser plasmas: influence of free electrons between jets, intrinsic phase, and Gouy phase

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Abstract. Ab initio analytical model of quasi-phase matching in multijet laser plasmas is presented on the base of phase difference between harmonics. Phase mismatch compensation between jets is explained by the free electrons produced during laser ablation, while the Gouy phase shift of the driving pulse is considered insufficient for this purpose, as well as intrinsic phase variation at experimental conditions. Inverse proportionality of intensity of harmonics to square of harmonic order is derived from the suggested model of propagation and should be considered when analyzing high harmonic generation spectra by single-atom response even in the absence of propagation.

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1. Introduction

The increasing interest in quasi-phase matching (QPM) of high harmonics generation (HHG) in the multi-jet plasmas created by laser ablation of solid targets (laser plasmas) [1,2] relative to the well-studied QPM in hollow waveguides with variable diameters filled with noble gases is related with importance of phase-matching processes for production and control of attosecond pulses [3]. HHG QPM in multi-jet laser plasmas can be combined with other methods of enhancement of HHG in laser plasmas, such as nanoparticle – assisted HHG, resonant HHG, two-color HHG, HHG from fullerenes. The characteristic feature of laser plasmas for HHG is the presence of significant amount of free electrons compared to noble gases. Previously, phase mismatch compensation was be obtained by using counter-propagating driving beams [4-5], off-axial phase matching [6], phase matching in low-frequency fields [7] ,control of pressure in modulated waveguides [8], addition of buffer gas with anomalous dispersion [9] and self-focusing by Kerr nonlinearity [10]. One should note that gases used in HHG phase matching experiments are usually much denser than laser plasma. This can restrict conversion of plateau harmonics [11] due to absorption in noble gases, while for plasmas absorption is insignificant. The formation of phase matching conditions in the laser plasmas containing free electrons is more complicated due to low concentration of plasma for intensity-based optimization, and general problems related with precise control of concentration of ions and electrons in the interaction region. However, creation of multijet laser plasmas with QPM conditions has been considered a simple and reliable approach for strong increase of HHG conversion efficiency in laser plasma in different ranges of extreme ultraviolet (XUV).

In classical nonlinear optics of low-order harmonics the energy transfer from the driving pulse to its low-order harmonic is significant only when the phase mismatch induced by the dispersion of phase velocity does not lead to destructive interference of the harmonics generated in opposite phases. Perfect phase matching, when phase mismatch between harmonics is zero over entire nonlinear medium, can be achieved either in birefringence crystals [12] using difference of ordinary and extraordinary waves, or in specially designed photonic crystals [13, 14]. Unfortunately, in gas or plasma-based HHG the fulfillment of such conditions is possible only in the case of medium, where significant anomalous dispersion of buffer gas is present for certain harmonic. However, significant anomalous dispersion means significant absorption. In addition, concentrations of atoms in active medium are usually too low to manifest any notable atomic anomalous dispersion. In fact, matching of phase velocities of the driving pulse and the high harmonic is not required for HHG QPM, because generation of harmonics is based on recombination of accelerated electrons on parent ions, which is not simultaneous with the ionization process [15]. No direct transfer of energy from driving field to high harmonics field is possible, contrary to what is assumed in classical low-harmonics generation. HHG is single-atom process and the harmonics are generated independently along the propagation axis, but with fixed phase relative to driving field. Due to negative plasma dispersion the phase velocity difference of harmonics and driving field results in the fact that at some point in the generating medium the phase difference between the harmonic which arrived to that point and the harmonic generated in that point reaches π, after which further increase of phase difference leads to destructive interference of harmonics up to phase difference of 2π.

An alternative to perfect phase matching is classical QPM in periodically poled structures [16], where the nonlinear polarization axis is flipped at the distance, where the phase mismatch between the driving pulse and its propagating harmonic reaches π. So, instead of increasing from π to 2π, the phase mismatch increases from 0 to π once again. However, it is impossible in HHG to create similar periodically poled laser plasma for HHG because of the inversion symmetry of gaseous or plasma media, so alternative approach to QPM is required. HHG QPM in laser plasmas and gases by temporal, or equally, more easily achieved spatial [17] modulation of fields is based on compensation of phase mismatch $\Delta\phi$ accumulated between the two fields with different frequencies (in HHG this is the driving field and its certain harmonics) when propagating in the dispersive medium. Commonly accepted understanding of optimal HHG QPM is that multijet structure of gas or plasma changes $\Delta\phi$ from π at the exit from one jet to 2π at the beginning of another jet, so the separation between jets prohibits the destructive generation of harmonics with opposite directions of electric field when $\pi < \Delta\phi < 2\pi$. During HHG QPM the transition from plasma jets to no-plasma intervals is not as distinct, as , for example, the boundary between periodically poled crystal structures in classical QPM. The optical properties of no-plasma regions may also differ significantly from properties of plasma jets, so we consider the possibility for HHG QPM to be sub-optimal, fulfilling a less strict condition. Namely, the change of the phase at the exit of the jet is not necessarily π, but the total phase change of a harmonic after passing one jet and one no-plasma region is $\approx 2\pi$, which we will call 2π QPM condition. This 2π QPM condition is currently assumed to support the experimentally observed near-quadratic growth of HHG QPM efficiency with the number of jets, although we will show that it is necessary, but not sufficient condition for experimental HHG QPM reproduction.

HHG QPM in laser plasmas has been extensively studied both theoretically and experimentally during the last few years [18-22]. In [18] two one-dimensional propagation equations have been solved numerically for the driving pulse and its second harmonic acting as sources for HHG without use of slowly-varying envelope approximation. The major feature of HHG experiments using two-color pump, the appearance of strong even harmonics has been reproduced. However, very limited information on simulation parameters and, especially, on implementation of propagation of harmonics in [18] does not allow making any conclusions about the origin of HHG QPM in this case. Lack of information about the phase of harmonics due to their representation only as frequency-dependent nonlinear polarization in [18] does not allow the simulation of interference of harmonics. In addition, the Gouy phase and off-axis effects are not included in the model [18] that limits its further application to loose-focusing conditions only. In [19] the conditions for HHG QPM were presented for q^{th} harmonic taking into account phase velocity difference for the driving field and the harmonics of the laser plasma, nonlinear refraction, and Gouy phase shift. The theoretical calculations [19] claimed to explain the fulfillment of QPM conditions for only a limited group of harmonics by compensation of Gouy shift and dispersion-induced phase mismatch in every jet by Gouy shift alone in the interjet space. The good correspondence of theoretical predictions and experimental results in [19] is thus due to uncertainty in the choice of the ionization degree, as the Gouy phase shift over a single jet is very small.

Propagation of harmonics in gases and plasma using nonadiabatic paraxial approximation has been widely studied not only for HHG QPM in plasma [26] but also for HHG in gases [21] utilizing the approach initially developed for propagation of laser beams in atmosphere [23]. In all these calculations the driving pulse and harmonics are propagated separately, and only the equation for slowly-varying part of driving pulse is solved using finite-difference approaches. Although in principle this approach gives very precise results, the origin of inconsistence is the propagation of harmonics, as either their intrinsic phase is actually lost or harmonics are simply added incoherently. The latter approach allows one to get the L^2 dependence of HHG intensity yield with the growth of nonlinear medium length L, but such description is also incorrect due to absence of selectivity by harmonic order. In [24] it has been shown that "plasma lens" effect (which is actually influence of group velocity change in plasma simulated in paraxial approximation for infinite pulses) on driving radiation due to displacement current of free electrons is important even for simulation of ordinary HHG in noble gases, where concentrations of ions are relatively low. Self-modulation of propagating harmonics due to nonlinear polarization of atoms and ions which participate in HHG process was also suggested and tested in [19]. But the effect of plasma lensing on HHG QPM was never studied separately.

In [21] on the assumption of proportionality of HHG yield to $\sin^2(\Delta kL/2)$ the conditions for HHG QPM in gas jets were proposed based on gas density manipulation. The origin of that proportionality was, however, not addressed. It can be supposed that at typical gas-jet HHG QPM conditions the Gouy phase shift is negligible due to weak focusing conditions, intensity variation is insignificant to manifest the contributions of harmonic dipole phase variation, and the negative dispersion due to free electrons is compensated by positive dispersion of atoms in the given range of parameters. On the contrary, in plasma HHG QPM the ionization degree is relatively high and the atomic dispersion is negligible, so other mechanisms of HHG QPM can be important which are addressed in our article.

Recently a dependence of HHG QPM enhancement peak on the distance between laser plasma jets was experimentally discovered [25] using ablation of rotating disks on a rod parallel to disk surface as a solid target. The importance of such possibilities of fine-tuning and control of HHG QPM motivated us to investigate the influence of the spacing between plasma jets on HHG QPM in more detail, because no convincing explanation of the observed dependence was presented in [25].

To unify all the achievements in HHG QPM simulations we present below the approach for HHG QPM simulation based on analytical solution of three-dimensional propagation equation which includes all the factors influencing HHG QPM. The analysis of relative contribution to phase mismatch from different processes is presented.

2. Theory

Any focused beam, which passes the focus, experiences spatial confinement of the transverse momentum that results in modification of the phase called Gouy phase shift. Gouy phase shift is not quantum mechanical process, because it can be treated for a wave propagating along z axis as a change in the wave number k_z due to beam finiteness and momentum conservation [26]:

$$\overline{k}_z \equiv \frac{\left\langle k_z{}^2 \right\rangle}{k} = k - \frac{\left\langle k_x{}^2 \right\rangle}{k} - \frac{\left\langle k_y{}^2 \right\rangle}{k} \tag{1}$$

Transversal distribution for both driving field and its harmonics is considered Gaussian in this article for simplicity. For the monochromatic Gaussian beam the Gouy phase is expressed as:

$$\phi_G(z,\omega) = -\frac{1}{k}\int^z \left(\left\langle k_x{}^2 \right\rangle + \left\langle k_y{}^2 \right\rangle \right) = -\frac{X}{k}\int_{z_0}^{z_{\max}} dz \Bigg/ \left[W_0^2 \left(\frac{z_R^2 + z^2}{z_R^2} \right) \right] \tag{2}$$

SI units are used in the article. Here dimensionality of confinement $X=1$ for focusing by cylindrical lens and $X=2$ for spherical lens, z is the distance after the focus (pulse is propagating along z axis from $-\infty$ to ∞). Spherical lens ($X=1$) will be considered further. Here z_R is the Rayleigh length,

$$z_R = \frac{n(\omega) W_0^2 \omega}{2c}, \tag{3}$$

W_0^2 is the square of the beam waist radius, ω is the frequency of the focused radiation, $n(\omega)$ is the total refraction index of medium. All high harmonics generated in the medium also experience the Gouy shift because their wave vector is parallel to the wave vector of the incident driving radiation due to momentum and energy conservation laws. Frequency dependence actually implies dispersion of the Gouy phase in the near-field even in the absence of wavelength dependence of refraction, c is the speed of light. The integration over z gives us well-known generalized expression for a Gouy phase of a monochromatic Gaussian beam which passes through the focus [26]

$$\phi_G(z) = -\arctan(z / z_R) = -\arctan\left(\frac{2cz}{n(\omega) W_0^2 \omega} \right). \tag{4}$$

The maximal possible Gouy phase change is not wavelength-dependent and is achieved for a beam passing from far-field on one side to far-field on another side through the focus by integrating over z

$$\Delta\phi_{G_{-\infty,+\infty}}(z) = \int_{-\infty}^{\infty} \phi_G(z) dz = -\pi . \tag{5}$$

Propagation of both driving field and high harmonics in plasma consisting of free electrons and ions is governed by the nonlinear wave equation:

$$\nabla^2 E(x,y,z,t) - \frac{\partial^2 E(x,y,z,t)}{c^2 \partial t^2} = \frac{1}{c^2}\left[\frac{\partial}{\partial t}\left(\frac{i\omega_{pl}^2(t)}{\omega} E(x,y,z,t) \right) + \frac{\partial^2 P_{nl}(x,y,z,t)}{\partial t^2} \right]. \tag{6}$$

where plasma frequency in linear response is,

$$\omega_{pl}^2(t) = \sum_i \frac{N_i(t) q_i^2}{\varepsilon_0 m_i}, \tag{7}$$

$N_i(t) = N_{i_0} + \Delta N_i(t)$ is the concentration of charged particles, whose time dependence is due to ionization by the driving pulse, q_i is the charge and m_i is mass of the i^{th} type of species in plasma. Contribution of ions to plasma current is negligible compared to that of electrons due to much greater mass of ions. Only free electrons are considered for calculation of plasma frequency.

For the analysis of ionization $\Delta N_i(t)$ by the driving pulse we use the ADK tunnel ionization rate [27], which is well justified for the considered strongly tunneling regime and long driving pulse. Note that the main contribution to the ionization by driving pulse is from neutral atoms.

For typical conditions of HHG QPM experiments the slowly varying envelope approximation (SVEA) is well satisfied, because the driving beam and harmonics are very directional, and the pulse is not a few-cycle pulse, so carrier-envelope phase effects are not significant, so we can use SVEA. Transversal intensity distribution of this pulse in *(x, y)* plane is also of Gaussian shape. The axial component of driving pulse is a monochromatic wave multiplied by Gaussian envelope in time. The resulting driving pulse is obtained as solution of (6) neglecting the dispersion of Gouy phase and group velocity dispersion, which are extremely small because the change of refraction index due to free electrons is much smaller than unity and is not significant if not the difference of refraction indices enters the calculation:

$$E_{\omega_0}(x,y,z,t) = E_{(peak)\omega_0} \frac{W_0}{W(z)} e^{-\left\{\frac{x^2+y^2}{W\ (z)} + \frac{\sigma^2}{2}\left[-\left(z+\frac{x^2+y^2}{2R(z)}\right)\frac{dk(\omega)}{d\omega}\Big|_{\omega 0}\right]^2 + \left(\frac{t-t_0}{t_{lifetime}}\right)^2\right\}} \times$$
$$\times e^{-i\left[\left(z+\frac{x^2+y^2}{2R(z)}\right)k(\omega)\big|_{\omega=\omega 0} - \omega_0 t + \phi_{G\omega 0}(z)\right]}, \tag{8}$$

where

$$W(z) = W_0 \sqrt{1+\left(\frac{z}{z_R}\right)^2}, \tag{9}$$

$$R(z) = z\left[1+\left(\frac{z_R}{z}\right)^2\right], \tag{10}$$

$$k(\omega)\big|_{\omega=\omega_0} = \frac{n(\omega)\omega}{2\pi c}\Big|_{\omega=\omega_0} = \sqrt{1-\frac{\omega_{pl}^2}{\omega_0^2}}\frac{\omega_0}{2\pi c}, \tag{11}$$

$$\frac{dk(\omega)}{d\omega}\Big|_{\omega=\omega_0} = \frac{\omega_0}{2\pi c}\sqrt{1+\frac{\omega_{pl}^2}{\omega_0^2}}, \tag{12}$$

$t_{lifetime}$ is the pulse envelope duration measured at 1/e level of the envelope magnitude, $E_{\omega_0}^{(peak)}$ is the maximal value of the driving field strength, σ is the width of Gaussian profile of radial intensity at 1/e level. The first exponent in (8) describes the slow evolution of the pulse envelope with space and time and contains no phase-related phenomena, except the difference of group velocities of the driving pulse and the harmonics. However, the second exponent in (8) describes the phase evolution of the driving field $E_{\omega_0}(x,y,z,t)$ as

$$e^{-i\left[\left(z+\frac{x^2+y^2}{2R(z)}\right)\left(1+\frac{\omega_{pl}^2}{2\omega_0^2}\right)\frac{\omega_0}{2\pi c} - \omega_0 t - \arctan\left(\frac{2cz}{\left(1+\frac{\omega_{pl}^2}{2\omega_0^2}\right)W_0^2\omega}\right)\right]}. \tag{13}$$

The amplitude of a harmonic in the single-atom response is determined by the energy conservation law $\hbar(q\omega_0) = q\hbar\omega_0$ and the relative conversion efficiency at frequency ω is determined by the induced dipole $d(\omega)$ in the frequency domain. It is often assumed that the phase shift between the driving pulse and the harmonic, that should be minimized or compensated, is given by the phase mismatch [19],

$$\Delta\vec{k} = \vec{k}_{q\omega_0} - q\vec{k}_{\omega_0}. \tag{14}$$

Despite the convenience of such an approach, it is evident that the harmonics cannot interfere with the driving pulse itself, so the phase mismatch should have another explanation. Namely, the momentum conservation law for radiative recombination (which should be fulfilled simultaneously with the energy conservation law for the real output of harmonics) implies that the wave vector $\vec{k}_{q\omega_0}$ of the emitted harmonic strictly equals the vector sum of wave vectors of the absorbed quanta of the driving pulse, which is for single-frequency driving pulse with the same polarization $\sum_q \vec{k}_{\omega_0}^{(q)}$. Otherwise, the inharmonic oscillation of the electron in laser-dressed states would result in symmetric radiation profile relative to the axis of oscillation. Here (q) denotes the propagation direction of the q-th photon and $\sum_q \vec{k}_{\omega_0}^{(q)} \neq q\vec{k}_{\omega_0}^{(1)}$, because some interacting photons can be propagating off-axis due to focusing by the lens and diffraction. As a result, the interference of the off-axial processes can take place for a small angle with the axis of propagation.

The existence of Gouy phase change of the driving pulse makes the correction to (14) as:

$$k_{q\omega_0} z = qk_{\omega_0} z = q\left(n(\omega_0)\frac{\omega_0}{2\pi c} + \phi_{G_{\omega 0}} \right) z = q\left(\sqrt{1-\frac{\omega_{pl}^2(t)}{\omega_0^2}}\frac{\omega_0}{2\pi c} z - \arctan\frac{z}{z_R} \right). \tag{15}$$

The delay between ionization and recombination of an electron induces some additional shift of phase of a harmonic relative to the phase of the pump field due to intrinsic dipole phase, $\phi_{0q}\left(I_{\omega_0}, q\omega_0\right)$ [28]. The phase of the driving pulse is given by the expression $(k_{\omega_0} z U(\theta)\cos\theta - \omega_0 t)$. θ is angle between the wave vector of a photon of the driving field with the optical axis to account for noncollinear processes, $U(\theta)$ is the angular distribution of wave vectors of the driving pulse. For Gaussian pulses the corresponding distribution is governed by the change of radius of curvature of the wavefront. To simplify further analysis of the key properties of HHG QPM we will utilize the fact that for experimental conditions [2] HHG takes place in the region where $z << z_R$,so the curvature R(z) is much bigger than (x^2+y^2), and $R(z_2) \approx R(z_1)$, thus neglecting noncollinear processes. As a result, the phase difference between the high harmonic generated at the point z_2 and the harmonic that was generated along the same axis, but at z_1 point is determined by the phase difference between the driving pulse and the previously generated harmonic:

$$e^{-i\Delta\Phi} = e^{-i\left[\left(z_2+\frac{x^2+y^2}{2R(z_2)}-z_1+\frac{x^2+y^2}{2R(z_1)}\right)\left(\frac{\omega_{pl}^2}{2\omega_0^2}-\frac{\omega_{pl}^2}{2q^2\omega_0^2}\right)\frac{q\omega_0}{2\pi c}-q\arctan\left(\frac{2cz}{\left(1+\frac{\omega_{pl}^2}{2\omega_0^2}\right)W_0^2\omega_0}\right)+\arctan\left(\frac{2cz}{\left(1+\frac{\omega_{pl}^2}{2q^2\omega_0^2}\right)W_0^2 q\omega_0}\right)+\Delta\phi_{0q}\left(I_{\omega_0}, q\omega_0\right)\right]}. \tag{16}$$

$$1 >> \frac{\omega_{pl}^2}{2\omega_0^2} >> \frac{\omega_{pl}^2}{2q^2\omega_0^2}; \frac{\omega_{pl}^2}{2q^2\omega_0^2} \approx 0; \frac{\omega_{pl}^2}{2\omega_0^2} \neq 0. \tag{17}$$

The final expression of phase change for a harmonic propagated in uniform plasma jet from z_1 to z_2 is

$$e^{-i\Delta\Phi(z_2,z_1)} = e^{-i\left\{\left[(z_2-z_1)\left(\frac{\omega_{pl}^2}{2\omega_0^2}\right)\frac{q\omega_0}{2\pi c}\right]+\left[q\phi_{G_{\omega 0}}(z_2)-q\phi_{G_{\omega 0}}(z_1)-\Delta\phi_{G_{q\omega 0}}\right]+\left[\phi_{0q}\left(I_{\omega_0}, z_2\right)-\phi_{0q}\left(I_{\omega_0}, z_1\right)\right]\right\}}. \tag{18}$$

The term in the first square brackets determines the phase mismatch induced by free-electron dispersion. The term in the second square brackets determines the influence of the difference of Gouy phase of the driving pulse accumulated along the distance Δz and the change of Gouy phase of harmonics (note that at $\Delta z << qz_R$, $\Delta\phi_{G_{q\omega 0}} \approx 0$ and will be neglected in this article). The term in the third square brackets determines the intrinsic phase difference. According to [28], the intrinsic phase in the first approximation can be considered linear and dependent only on electron trajectories as

$$\Delta\phi_{0q}\left(I_{\omega_0}, q\omega_0\right) \approx -\alpha I(z) = -\alpha I_{peak_{\omega 0}} \frac{z_R^2}{z_R^2+z^2}. \tag{19}$$

Total HHG conversion efficiency is considered approximately the same in every point along z-direction in jet at the given distance from the optical axis $\sqrt{x^2+y^2}$ due to very small variation of average intensity of the driving pulse over jet length (1%-3%) with z in the considered experimental conditions of loose focusing [2], so the expression for onaxis HHG amplitude generated in the n^{th} jet, calculated at the end of this jet:

$$\Delta E_n = E_{q\omega_0}^{(atom,z=0)} \left(\frac{z_R^2}{z_R^2 + z^2} \right) \rho_{(atom)} \left(\delta S \right) \int\limits_{z_0+(n-1)(l_{jet}+d)}^{z_0+(n-1)(l_{jet}+d)+l_{jet}} e^{-i\Phi(z)} dz \tag{20}$$

$$\Phi(z) = \left(\frac{q\omega_{pl_0}^2}{4\omega_0 \pi c} \right) z - q \arctan\left(\frac{z}{z_R} \right) - \left(\frac{z_R^2}{z_R^2 + z^2} \right) \alpha I_{peak_{\omega 0}} \tag{21}$$

Here $E_{q\omega_0}^{(atom)}$ is the HHG output per atom calculated for intensity in the focus, $\rho_{(atom)}$ is the density of atoms in jet, δS is the elementary area surface, on which the intensity of driving pulse is considered constant, l_{jet} is jet length, d is the space between jets, z_0 is the position of the beginning of the first jet after the focus. Note that we did not calculate $E_{q\omega_0}^{(atom)}$,so the results are independent on the approximations of single-atom calculations. If we are only interested in relative enhancement of harmonics using the same parameters except different jet lengths, then $\rho_{(atom)}\left(\delta S\right)$ can be excluded from consideration. The single-atom response can be calculated in strong-field approximation [28], because the intrinsic phase is also calculated semiclassically [28, 29]. The expression (20) can be integrated analytically, if the expressions for Gouy phase and the intrinsic phase are approximated by Taylor expansion in the vicinity of z=0 (which is correct, because $z \ll z_R$).

$$\int e^{-i\Phi(z)} dz \approx \int \mathrm{e}^{-i\left[\left(\frac{\omega_{pl_0}^2}{4\omega_0 \pi c} - \frac{1}{z_R}\right) qz - \alpha I_{peak_{\omega 0}} + \frac{\alpha I_{peak_{\omega 0}}}{z_R^2} z^2\right]} dz = \sqrt{\frac{\pi}{4i\alpha I}} e^{i\left(\alpha I + \frac{q^2}{4\alpha I}\right)} z_R erf\left(\frac{\sqrt{i\alpha I} z}{z_R} + \frac{iq}{2\sqrt{i\alpha I}} \right) \tag{22}$$

However, we used expressions (20-21) in our calculations to avoid errors due to Taylor expansion. Only two factors can contribute to compensation of the phase mismatch from π to 2π in the free space between plasma jets according to the accepted models: Gouy phase shift influence and difference of intrinsic phase. Note that in the absence of intrinsic phase change and low initial ionization of ablated media, Gouy phase can give nearly perfect phase matching even in continuous media if $z_R = 4\omega_0 \pi c / \omega_{pl_0}^2$, especially for very high orders of harmonics, but for low-order harmonics the influence of the intrinsic phase becomes comparable with the Gouy phase very early. The sign of Gouy phase and intrinsic phase change is also important and means that these phase contributions cannot increase the phase mismatch induced by the dispersion to 2π . In this case only two evident possibilities of phase mismatch compensation exist, which depend on initial concentration of free electrons. In the case of low initial concentration of free electrons the only thing to be phase -matched is the negative dispersion due to ionization by driving pulse. This ionization is also very low in typical gas HHG experiments and can be matched by positive dispersion of ions and waveguide geometry [21]. On the contrary, we assume that in plasma HHG the initial concentration of free electrons is much higher than expected from the estimate of multi-photon ionization yield at the intensities of the heating pulses that is due to Coulomb explosion during laser ablation. There were no on-the-fly measurements of concentration of free electrons in given region during laser ablation for HHG studies. The concentration in the interaction area of free electrons produced during laser ablation cannot be reliably calculated due to uncertainty of their velocity distribution, but the estimates using equilibrium Saha equation give us the concentration in the range of 10^{17}-10^{18} cm^{-3} [30]. Anyway, nothing can be said about the angular distribution of velocity of these electrons. So, for the purposes of our article we will assume that the initial concentration of free electrons between jets is very close to their concentration inside the jets, but the analytical description of phase matching is also applicable when there are no electrons between jets. Absence of neutral atoms or ions between jets excludes generation of harmonics between jets in counter-phase to the harmonics generated earlier.

The total HHG QPM yield can be obtained by propagating of harmonics amplitudes (20-21) to the end of the remaining laser plasma jet and summation of their amplitudes after the exit of the last jet,

$$I_{tot} = \frac{c\varepsilon_0}{2} \left[\sum_{n=1}^{N_{jets}} \left(\Delta E_n e^{-i\left(\frac{q\omega_{pl_0}^2}{4\omega_0 \pi c}(N_{jets}-n)(l_{jet}+Fd) - q\Delta\phi_{G_{\omega 0}}(n) - \Delta\phi_I(n) \right)} \right) \right]^2 , \tag{23}$$

$$\Delta\phi_{G_{\omega 0}}(n) = \arctan\left(\frac{z_0 + N_{jets}(l_{jet}+d) - d}{z_R} \right) - \arctan\left(\frac{z_0 + (n-1)(l_{jet}+d) + l_{jet}}{z_R} \right), \tag{24}$$

$$\Delta\phi_I(n)=\left(\frac{z_R^2}{z_R^2+\left[z_0+N_{jets}(l_{jet}+d)-d\right]^2}-\frac{z_R^2}{z_R^2+\left[z_0+(n-1)(l_{jet}+d)+l_{jet}\right]^2}\right)\alpha I_{peak_{\omega_0}}. \quad (25)$$

Here F=0 if we consider no free electrons between jets and F=1 if there are free electrons between jets. The enhancement given by (23-25) is almost independent on the single-atom response if the intensity dependence of intrinsic phase is reproduced correctly by the considered single-atom response calculation. Note that function

$$f(b)-f(a)=\int_a^b e^{-iKz}dz=\frac{ie^{-iKz}}{K}\Big|_a^b=\frac{i\cos(Kb)}{K}-\frac{\sin(Kb)}{K}-\frac{i\cos(Ka)}{K}+\frac{\sin(Ka)}{K}, \quad (26)$$

which is the most important part of our consideration of HHG QPM, is sensitive to the values of boundaries a and b, even if $K(b-a)=\pi$ and its real part varies from 0 to 2. If we consider only the real part of (20), the maximal value of (20) is achieved if, in addition to HHG QPM optimality condition of π phase difference over a jet,

$$\frac{q\omega_{pl}^2}{4\omega_0\pi c}\left(z_0+l_{jet}\right)-\arctan\left(\frac{z_0+l_{jet}}{z_R}\right)-\left(\frac{z_R^2}{z_R^2+\left(z_0+l_{jet}\right)^2}\right)\alpha I_{peak_{\omega_0}}-$$
$$-\left[\frac{q\omega_{pl}^2}{4\omega_0\pi c}z_0-\arctan\left(\frac{z_0}{z_R}\right)-\left(\frac{z_R^2}{z_R^2+z_0{}^2}\right)\alpha I_{peak_{\omega_0}}\right]=\pi, \quad (27)$$

the condition on z_0 is fulfilled in the assumption that phase change over a jet is π:

$$\frac{q\omega_{pl}^2}{4\omega_0\pi c}z_0-\arctan\left(\frac{z_0}{z_R}\right)-\left(\frac{z_R^2}{z_R^2+z_0{}^2}\right)\alpha I_{peak_{\omega_0}}=\pm\frac{\pi}{2}\pm 2\pi N. \quad (28)$$

The relation (28) determines the influence of the initial value of the intrinsic phase on optimal focusing conditions. In experiments the optimal initial position of the target relative to focus is determined by trial-and -error method, but is never varied with the jet length.

3. Results and discussion

The relative importance of different phase-matching processes in laser plasma can be estimated from parameters of the most efficient HHG QPM experiments. For the analysis of relative importance of different contributions to phase HHG QPM in multi-jet laser plasma we considered the experimental conditions [2] for silver plasma, where total HHG intensity was much higher than in typical gas HHG experiments due to higher intensity of the driving pulse. Driving Gaussian pulse with central wavelength of 804 nm, intensity of 5×10^{14} W cm^{-2} and duration of 64 fs (at FWHM level) is used for calculations. The distinctive feature of the experiments [2] is that Rayleigh length of the radiation focused by spherical lens is very large, 2.4 cm. The total length of the plasma media does not exceed 0.6 cm. Values of $\alpha_q=3\times10^{-14}$ rad cm^2 W^{-1} for short trajectories and $\alpha_q=25\times10^{-14}$ rad cm^2 W^{-1} for long trajectories [29] give us intrinsic phase at z=0 for the short and the long trajectories correspondingly as -15 and -125 radians.

It is seen from the dependence on distance after focus of Gouy phase of the driving pulse multiplied by 43 and 35 correspondingly, as well as the change of intrinsic phase for short and long trajectories (Fig. 1) that both the change of the intrinsic phase for long trajectories and the influence of the change of Gouy phase of the driving field are significant, but even if added together, cannot compensate the 8π phase change in the case of 8-jet laser plasma. In fact, both Gouy phase and intrinsic phase change are harmful for the experimental conditions [2] and their influence was minimized by adjustment of the position of plasma relative to the focus.

The significant difference of phase change for long trajectories is very hard to be compensated. We suppose that long trajectories are significantly suppressed in HHG QPM experiments [2] and make all calculations for short electron trajectories only. However, if intensity variation along the plasma is suppressed (by either focusing with greater Rayleigh length or using less divergent beams), both trajectories can be QPM enhanced. The Gouy phase changes almost linearly with z in the considered focusing area, so the contribution of the Gouy phase shift is that the actual phase mismatch due to other factors can be constant at every jet and slightly less than multiple of 2π before any subsequent jet. In the experimental conditions of very efficient harmonic generation [2] there is no physical mechanism except the existence of free electrons between jets, which could explain the observed compensation of phase mismatch using widely accepted assumption that the space between jets is absolutely free.

Regarding the optimal position of focusing due to initial value of intrinsic phase, it is quite sufficient to maximize the HHG output at the first jet, where the driving field intensity is maximal. Then the value of

$z_0 = -0.72 l_{jet}$ was found to be the most efficient for single-jet HHG output for all jets according to (28) and well-reproducing the experimental results where the optimal focusing conditions were not changed over all the experiments.

As the maximally enhanced harmonic depended on jet length at the same ablation and driving pulse conditions, we assume that ionization degree after ablation is the same for any number of jets. It is widely accepted that ionization, which occurs during interaction with the driving pulse, influences plasma oscillation frequency. On the contrary, we assume that oscillations of these electrons are not plasma oscillations, but contribution to HHG or above-threshold ionization.

First, density of free electrons equal to 4.96×10^{16} cm^{-3} both inside and outside jets was used for HHG QPM simulations using (26-25). This gives us modified coherent length l_{coh} equal to 0.4 mm, 0.5 mm and 0.9 mm for the 43th, the 35th and the 19th harmonics correspondingly. Determination of coherent length as $l_{coh} \approx 1.4\times10^{18} / qN_e$ cm^{-3} [2] is not practical as it does not include the influence of Gouy phase change of the driving pulse. The results of HHG QPM simulations are presented in Fig.2 for all plasma media configurations used in [2]. Note that the single-atom response gives us q^2 multiplier to Fourier transform of the induced dipole *d(t)*, which is effectively consumed by propagation effects responsible for the plateau-like structure of HHG yield. It is seen in Fig.2 that although the harmonics reported in the experiment [2] are QPM-enhanced, there are some other strongly enhanced harmonics. In addition, the 25th harmonic reported to be enhanced in experiment with 3 0.9-mm plasma jets is not so strongly enhanced in the simulation. It is possible that the concentration of electrons in the jets is not uniform, but either smoothly varying on the whole plasma length, or is smaller between jets. The addition of off-axis contributions to the 8 0.4-mm jet plasma structure allowed getting similar enhancement of neighboring harmonics due to variation of intrinsic phase over the radial direction, which is close to experiments [2].

To check the reproducibility of the unexpectedly enhanced harmonics we present the dependence of the total enhancement of the most strongly enhanced harmonics in eight-jet simulation as a function of plasma jet count (Fig. 3). It is seen that the harmonics other than the 43th and the 21th are strongly oscillating with the number of jets. So, their generation is in fact too dependent on the combination of intrinsic phase, Gouy phase and exact positions and boundaries of the jet to be significantly enhanced in real experiments. The square growth of the intensity of the 21th harmonic in the case, when the electrons between jets were compensating the phase of the 21th harmonic to π at the beginning of the following jet, is in direct contradiction with the 2π QPM condition, which shows that HHG 2π QPM condition on phase velocity is not sufficient to reproduce the experiments correctly. However, the phase change of the 21th harmonic over the nonlinear medium can be compensated by Gouy phase influence. The enhancement of 21th harmonic is also confirmed experimentally in [2].

We performed a different set of simulations, when there were no free electrons between jets. In this case the 2π QPM condition is not fulfilled before every jet, and no HHG enhancement was observed which scaled with number of jets. So, the existence of free electrons between jets is currently the only mechanism that can explain the HHG QPM in the case of high initial concentration of free electrons.

The analysis of the influence of z_0 on the presented data of QPM enhancement (Fig.4) has shown that this enhancement is almost periodic by z_0. The sensitivity is explained by significant variation of the initial value of the intrinsic phase with the focus position even in the case of loose focusing due to relation (26). In experimental studies there is indeed a dependence of HHG QPM yield on the position of the jets relative to focusing. However, this position is now not controlled with high precision, so the experimental verification of this dependence will give interesting method to check the validity of our QPM approach. In addition, depending on z_0, different neighboring harmonics can be enhanced more strongly. This result can be explanation of the fact that in real experiments several neighboring harmonics are enhanced almost equally. The analysis of the influence of concentration of free electrons between jets has shown that the HHG QPM is quite sensitive to relative concentration of electrons in jets and in the interjet space. Setting in the simulation the concentration of free electrons between jets lower than 90% of the concentration of electrons inside jets resulted in disappearance of HHG QPM for all harmonics. This is possibly due to the fact that spectral broadening of harmonics was not considered.

The efficient concentration of free electrons in laser jets can be easily estimated from the experimental measurement of the dependence of intensity enhancement of a single high harmonic in short plasma with the length of the plasma. Then the first maximum will give the concentration of free electrons, which is necessary for phase change of $\Delta\Phi(\Delta z = l_{jet}) = \pi$ over the jet. Unfortunately, this measurement has not been performed yet for relatively highly ionized plasmas. On the other hand, for extremely low-ionized laser plasmas the intensity of the 11th and the 17th harmonics on the length of single jet was measured to grow as square of the plasma length as the length varied from 0.1 to 10 mm [31]. But in that case the plasma was much shorter than the coherence length for given concentration of free electrons.

In the simplest form of on-axis interaction of a monochromatic pulse with laser plasma our model with constant concentration of free electrons was able to reproduce the strong enhancement of the 43th harmonic and the 21st harmonic [2]. However, the neighboring harmonics were not enhanced correctly, so we added dynamical ionization by the driving pulse. At the same time, single-atom HHG yield was considered proportional to instantaneous pulse intensity. We also included the on-axis contributions for various distances from optical axis, which were separated by dr=0.1 mkm up to 1/e level of intensity and the contribution of n-th slice in (x,y) plane to (n-1)th slice was weighted recursively as $n^2/(n-1)^2$. The resulting intensity enhancement over the eight-jet plasma is presented in Fig. 5 together with the experimental results from [2]. The good correspondence with the experimental data reveals the importance of the ionization by the driving pulse to the total QPM enhancement of HHG. The enhancement of harmonics close to the 21st harmonic is explained by partial compensation of the mismatch by Gouy and intrinsic phase change, which is not confirmed experimentally due to uncertainty of the exact boundaries of the plasma jets. As a consequence, only the model of HHG QPM, which assumes free electrons between jets, gives the results, which actually include the main properties of HHG QPM experiments - quadratic growth of intensity of a small group of neighboring harmonics with the number of jets without any modification of intensity and focusing of the driving field. It is important that the concentration of free electrons cannot be determined directly from the coherent length defined by phase velocity dispersion only. Interferometry approaches can be used to determine the profile of concentration of free electrons [32]. However, we know no HHG studies, which included measurement of concentration of free electrons using interferometry methods due to complication of experimental set-up and non – obvious demand for such studies. Alternatively, one can minimize the initial ionization of detached atoms using non-intense and longer pulses (on the order of several nanoseconds) using electronic delay line. The developed model is a bit approximate and cannot be used for detailed numerical prediction for small number of jets, partly due to some uncertainty about the actual plasma jet lengths. However, the QPM enhancement of multiple harmonics is predicted due to the additional ionization of the plasma by the driving pulse and the corresponding change of the coherence length.

Manipulation of concentration of free electrons between jets can be used as a promising way to increase the conversion efficiency by more than an order of magnitude for several harmonics. Although direct injection of free electrons without ions is problematic, it is in principle possible to create periodically modulated plasmas by laser ablation so that in one set of jets the ionization degree is too high for any harmonic generation, and in the second set of jets the ionization degree is not so low. The equal concentration of electrons or ions is not as important as precise control of ionization degree and focusing conditions, which are nevertheless easily achievable using current experimental approaches for table-top HHG sources.

5. Conclusions

Analytical on-axis description of phase shift of harmonics along plasma was presented on the base of nonlinear wave equation. It is shown that away from resonances two regimes of phase matching exist - compensation of dispersion-induced phase mismatch by Gouy phase in extended plasmas and HHG QPM using phase change due to free electrons in the space between plasma jets. The analytical model of on-axis HHG QPM by compensation of phase mismatch to 2π using free electrons between jets supports all experimental features of HHG QPM if the ionization by driving pulse is included. The intensity-dependent intrinsic phase of harmonics determines the optimal position of multijet targets relative to focusing point. Influence of Gouy phase shift of the driving pulse was found to be insufficient to compensate phase mismatch in the case of focusing with high value of Rayleigh length for eight-jet and five-jet plasma. Intrinsic phase variation was also insufficient for QPM in given experimental conditions. Suppression of HHG QPM at high intensities of driving field or due to focusing of the driving pulse by a lens with small Rayleigh length is the result of strong increase of contribution of intrinsic phase, which is highly nonlinear and cannot be phase matched in such systems. Further increase of Rayleigh length of focusing lens as well as manipulation of concentration of free electrons between jets using ablation of complex targets with different ionization energies can increase HHG conversion efficiency in multijet plasmas.

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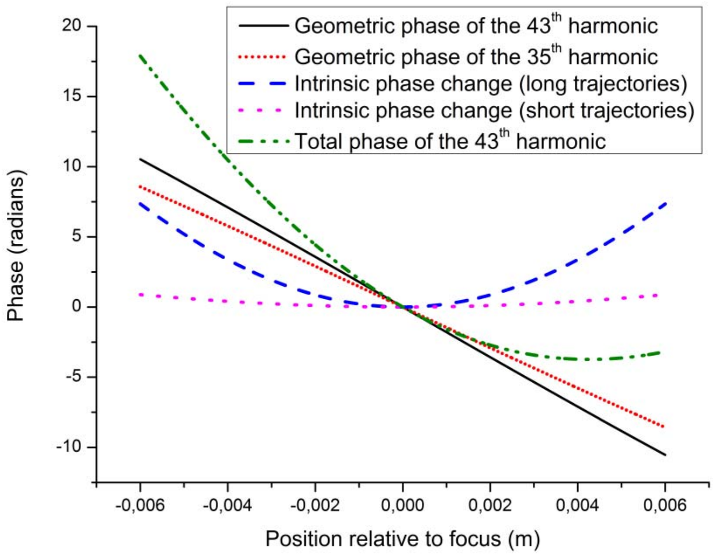

Figure 1. (Color online) The dependence on distance from focusing point of non-plasma contributions to phase change of harmonics. a) (black solid) Gouy phase of the driving pulse multiplied by 43; b) (red short dot) Gouy phase of the driving pulse multiplied by 35; c) (blue dash) intrinsic phase change for long trajectory; d) (magenta short dash) intrinsic phase change for short trajectory. e) (green dash dot dot) total phase for the long electronic trajectory of the 43th harmonic.

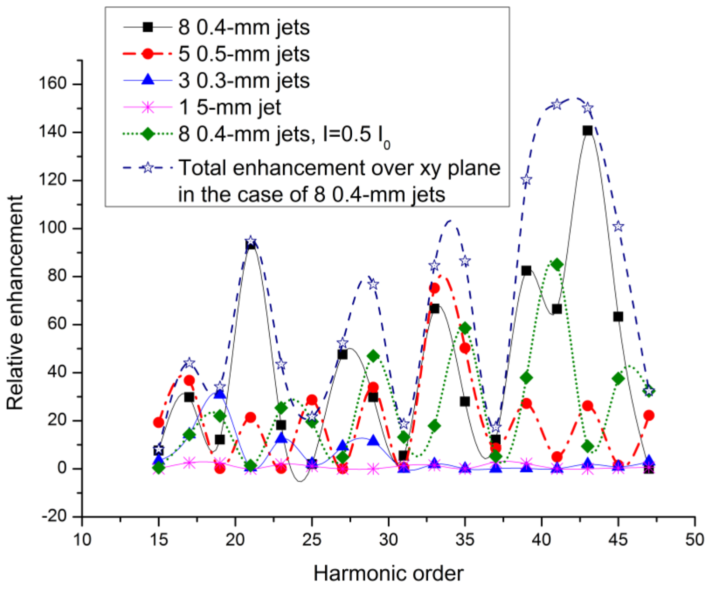

Figure 2. (Color online) The HHG QPM enhancement for different plasma multijet structure.

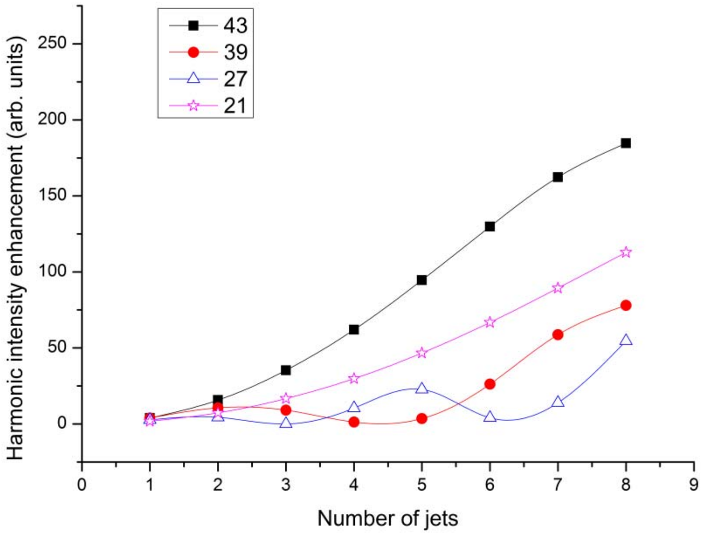

Figure 3. (Color online) Variation of the QPM-enhanced intensity of different harmonics with the number of 0.4-mm plasma jets. Lines are drawn to guide the eye. Inset: notation of harmonic orders: 43th (black filled squares) , 39th (red filled circles), 27th (blue empty triangles up) 21th (empty stars)

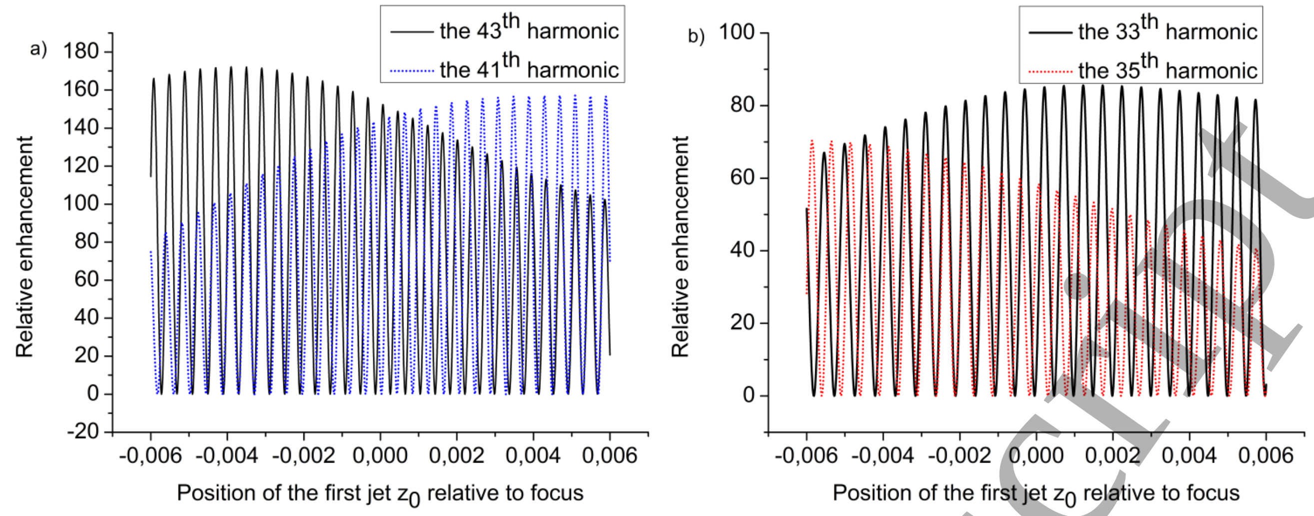

Figure 4. (Color online) The dependence of maximally enhanced harmonic on the position of the beginning of the first jet z_0 relative to focus. a) the 43th harmonic in the case of eight 0.4-mm jets, dotted line- the 41th harmonic b) the 33th harmonic in the case of five 0.5-mm jets, dotted line- the 35th harmonic

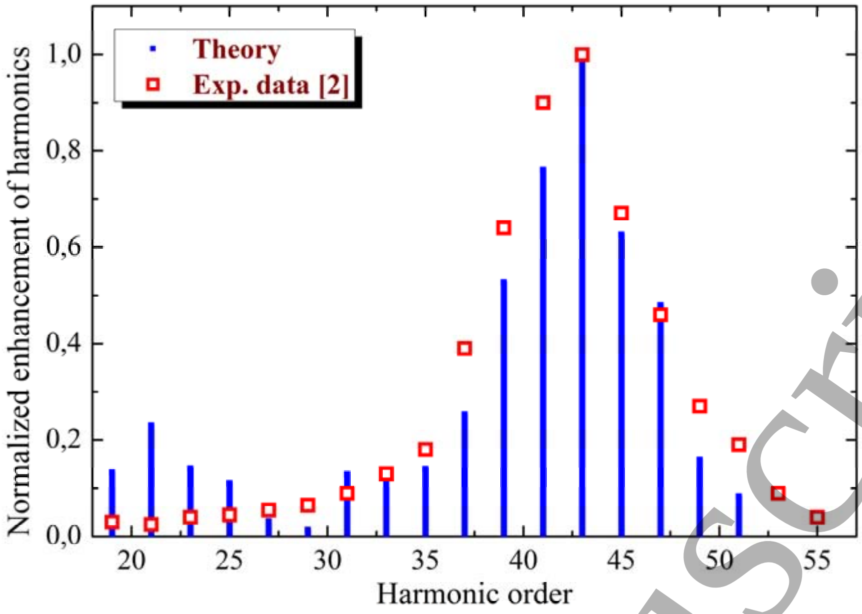

Figure 5. (Color online) Normalized HHG QPM enhancement in the case of 8 0.4 mm-jet plasma (vertical lines), which assume the same intensities of all the harmonics. Squares are the experimental results.