Phase-Matched High-Order Difference-Frequency Mixing in Plasmas

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The generation of coherent extreme ultraviolet radiation by phase-matched high-order difference-frequency mixing in plasmas is reported. The plasmas are produced by optical field ionization of atomic and molecular gases in an intense Ti:sapphire laser field. Phase matching for the high-order two-color $(\omega, 2\omega)$ mixing process, $9\omega = 6(2\omega) - 3\omega$, is demonstrated for the first time. [S0031-9007(96)00084-1]

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The generation of high-order harmonics of intense laser radiation has been intensively studied during the last few years [1]. It is of great importance to learn how to manipulate the high-order harmonic signals. The solution of this problem can provide a way to an efficient generation of coherent extreme ultraviolet (XUV) radiation.

So far, most of the efforts have been concentrated on studies of the nonlinear single-atom response. It has been shown that with increasing harmonic order the harmonic intensities, after an initial decrease, form a "plateau" region of fairly constant intensities. The plateau has a well-defined cutoff from which the harmonic intensities decrease rapidly. Recently, for more detailed studies of the single-atom response, a few high-order frequency mixing experiments [2,3] have been performed. Now the interest has been moved to more complicated problems related to propagation effects (collective medium response, phase matching, etc.). The motivation of the present Letter is to demonstrate the feasibility of phase-matched difference-frequency mixing in plasmas which could allow for some kind of "harmonic engineering." The goal is to replace the well-known plateau of harmonics by a single "peak" of one definite harmonic signal.

Unfortunately, available experimental and theoretical data have already led to a very disappointing conclusion [1] that in the strong laser field "the propagation effects play no role or they affect all the harmonics in the same way." Fortunately, this is only partially true. Two trivial arguments can help to understand this. First, the same laser field which is already "strong" for atoms (in comparison with the atomic field strength, E_{at}) is no longer strong for ions (in comparison with the ion field, $E_i \simeq Z^3 E_{\mathrm{at}}$, where Z is ion charge $+1$). Second, there are several relatively old tricks (see, for example, [4]) which allow one to improve phase-matching conditions and increase the role of propagation effects.

A very promising medium for high-order harmonic generation is a low-charged plasma. The nonlinear polarizability of low-charged ions is still high enough, and this plasma has a definite positive dispersion (which means that the index of refraction increases with increasing frequency) due to free electrons. It is well known [4] that in a medium with positive dispersion phase matching can be fulfilled for difference-frequency mixing processes. There are a few theoretical proposals [5–7] discussing this and other possibilities for phase matching and high-order harmonic generation in plasmas in detail. There also exists one experimental indication that noncollinear phase-matched difference-frequency mixing is probably responsible for the anomalous growth of the fifth harmonic of KrF laser radiation [8]. But, to our knowledge, there has been no experimental demonstration of this effect up to now.

The experimental setup is shown in Fig. 1. The pump laser is a 150 fs Ti:sapphire (BMI ALPHA 10A) operating at 770 nm. The laser radiation is sent through a 3 mm KDP crystal for frequency doubling. The energy of the second harmonic is 5.5 mJ, and the remaining energy of

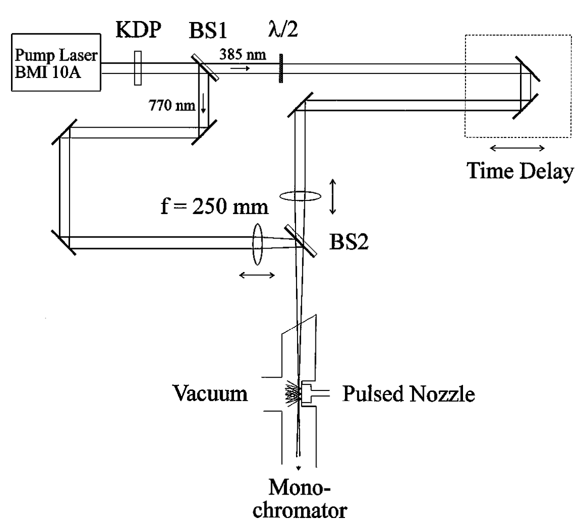

FIG. 1. Experimental setup. Beam splitters (BS) are highly reflective for 770 nm.

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the fundamental is 25 mJ. The fundamental is separated from the second harmonic by beam splitter BS1. Both fields are linearly polarized in the same direction due to the $\lambda/2$ plate. After a variable delay the fundamental and the second harmonic are recombined by beam splitter BS2. Each of the beams is focused with $f/12$ lenses ($f = 250$ mm) into the gas jet of a pulsed nozzle close to the nozzle exit. With this setup a good temporal and spatial overlap of the two beams in the focus can be obtained. The focus diameter of the fundamental is ~ 40 μm, which corresponds to a laser intensity of $\sim 10^{16}$ W/cm^2.

As nonlinear media oxygen, argon, and xenon gases are used. The backing pressure can be varied in the range of 100–3000 mbar. Note that the pressure in a gas jet is 10–20 times lower than the backing pressure. The nozzle has three output holes, with a diameter of 300 μm each. The distance between the centers of these holes is 500 μm. The holes can be opened and closed with a mechanical shutter. This provides a simple possibility for a variation of the interaction length. Further details of the experimental setup can be found in [3,9].

The intensity of the fundamental laser radiation is well above the critical intensity necessary for fast optical field ionization of the used gases. Therefore, at the front of the laser pulse a plasma is formed which serves as a nonlinear medium for the rest of the laser radiation. The plasma production is clearly visible when the laser radiation is focused into the gas jet.

At the level of analysis intended here, we give only simple qualitative explanations of our results on the basis of perturbation theory. For sum-frequency mixing ($\omega_q = q\omega$) in the case of weak focusing ($b \gg L$, where b is the confocal parameter and L is the plasma length), the total phase mismatch between the generated q-order harmonic field and its driving polarization is $\Delta k_q = \Delta k_q^f + \Delta k_q^d$ (see [1,4]). The first positive term originates due to the focusing geometry and is equal to $\Delta k_q^f \simeq 2(q-1)/b$ for a Gaussian beam. The second term is a dispersive phase mismatch, which for sum-frequency mixing in a plasma is equal to

$$\Delta k_q^d = k_q - qk_1 = \frac{q^2-1}{q}\,\frac{\omega_p^2}{2\omega c}, \qquad \omega_p^2 = \frac{4\pi e^2 N_e}{m_e}, \tag{1}$$

where k_1 and k_q are the wave numbers of the fundamental and q-order harmonic, N_e is the electron density, and ω_p is the plasma frequency. This phase mismatch is positive and grows with the harmonic order. The transition to the tight-focusing geometry (for a beam focused into the center of the plasma) results in a reduction and then disappearance of high-order harmonic signals, because in the tight-focusing limit ($b \ll L$) the generation of high-order harmonics is possible only in media with negative dispersion ($\Delta k_q^d < 0$) [4].

Difference-frequency mixing in plasmas is much more favorable in terms of phase matching (see also [6–8]). The dispersive phase mismatch for the two-color (ω, 2ω) mixing process $\omega_q = q\omega = m(2\omega) - l\omega$ is given by

$$\Delta k_q^d = k_q - mk_2 + lk_1 = \left[\frac{m}{2} - l - \frac{1}{2m-l}\right]\frac{\omega_p^2}{2\omega c}, \tag{2}$$

where $q = 2m - l > 0$, $m, l = 0, \pm1, \pm2, \ldots$, and $m + l$ is odd. This formula describes all possible mixing processes; for example, when $m = 0$ and $l = -q$, Eq. (2) coincides with Eq. (1). The factor in square brackets is illustrated in Fig. 2 for high-order harmonics with $q = 7, \ldots, 17$. The harmonic order is constant along the solid lines. The transition along the dashed lines from one point to another corresponds to the transformation $q \to q + 1$, $m \to m + 1$, and $l \to l + 1$. As can be clearly seen in Fig. 2, the most favorable conditions in terms of phase matching occur when $m = 2l$. In this case the phase mismatch is negative and is given by

$$\Delta k_q^d = -\frac{1}{q}\,\frac{\omega_p^2}{2\omega c} = -\frac{e^2}{m_e c^2}\,N_e\,\frac{\lambda}{q}, \tag{3}$$

where λ is the wavelength of fundamental laser radiation. When $m = 2l$, the absolute value of the dispersive phase mismatch for difference-frequency mixing is reduced by a factor of q^{-2} in comparison to sum-frequency mixing.

Assuming that the confocal parameters for all fields are equal (which is approximately fulfilled in our experiments), the value of the geometrical phase mismatch for the case of weak focusing can be estimated from $\Delta k_q^f \simeq 2(m - l - 1)/b$. This value is positive for $m = 2l$ and $l > 1$. Thus, the total phase mismatch, $\Delta k_q = \Delta k_q^f + \Delta k_q^d$, in the case of difference-frequency mixing depends on the plasma density [see Eq. (3)], and there exists an optimum electron density where the mismatch is equal to zero,

$$N_{\text{opt}} = \alpha^{-2}\,\Delta k_q^f\,\frac{q}{a_0\lambda} = q\,\frac{\Delta k_q^f}{r_e\lambda}, \tag{4}$$

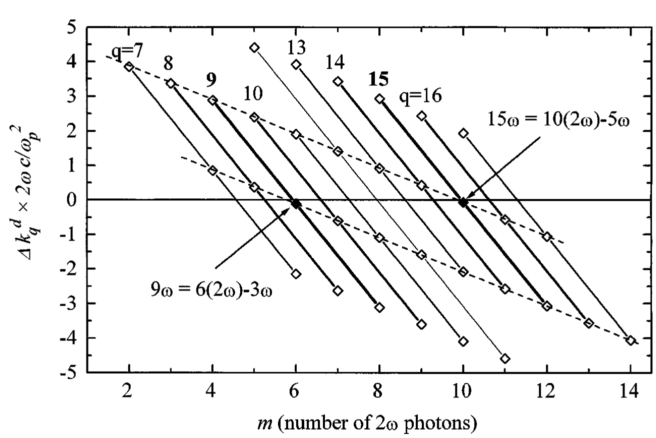

FIG. 2. Dispersive phase mismatch for two-color mixing process $\omega_q = m(2\omega) - l\omega$, where $q = 2m - l$. The harmonic order is constant along the solid lines. The transition from one point to another along the dashed lines corresponds to the transformation $q \to q + 1$, $m \to m + 1$, and $l \to l + 1$.

where $a_0 = 0.529 \times 10^{-8}$ cm is the Bohr radius, $\alpha = e^2/\hbar c \simeq 1/137$, and $r_e = e^2/m_e c^2 = 2.818 \times 10^{-13}$ cm is the classical electron radius. For this electron density the corresponding harmonic signal should dominate the spectrum.

In our experiments the measured confocal parameter is $b \simeq 1.1$ mm. These measurements were performed at atmospheric pressure with a strongly attenuated laser beam (see [9] for details). The length of the plasma for one open hole is $L \simeq 0.3$ mm. The total length for three open holes (taking into account the space between the holes) is $L \simeq 1.3$ mm. These parameters are somewhere in between the weak- and tight-focusing limits. The energy of the second harmonic radiation (2ω) is only 5.5 mJ. The highest harmonic which can be generated by this radiation (see Fig. 4 below) is the 7th (14ω) harmonic. Therefore, we are not able to observe very high-order difference-frequency mixing processes. We concentrate our attention on the generation of the 9-order harmonic due to the process $9\omega = 6(2\omega) - 3\omega$, where $m = 2l = 6$. Following the discussion above we expect that there exists an optimum electron density for the generation of this harmonic due to the difference-frequency mixing process. This density, estimated from Eq. (4), is $N_{\text{opt}} \sim 10^{19}$ cm^{-3} (weak focusing). In our experimental conditions (transient case) the value of the optimum electron density should be lower. One of the reasons for this is that the defocusing of laser radiation results in an effective increase of the confocal parameter and a corresponding reduction of the geometrical phase mismatch, Δk_q^f. To find the optimum density, we have studied the ratio of the 9-order harmonic signals generated in an O_2 gas jet by two-color (ω, 2ω) laser radiation and by the fundamental radiation alone as a function of the backing pressure. The real gas density in the gas jet and the corresponding electron and plasma densities (produced due to optical field ionization) vary approximately linearly with the backing pressure. Recall that the intensity of the fundamental laser radiation is $\sim 10^{16}$ W/cm^2 and is well above the optical field ionization threshold.

As can be seen in Fig. 3, at backing pressures of 300–500 mbar, the ratio $I_9(\omega, 2\omega)/I_9(\omega)$ has a sharp maximum. The estimated electron density for these backing pressures is $N_e \sim 10^{18} - 5 \times 10^{18}$ cm^{-3}. For this estimate we take into account that the real pressure in a gas jet is 10–20 times lower than the backing pressure and assume 1–2 free electrons per molecule. Thus, the optimum density does really exist for the generation of the 9-order harmonic in the two-color laser field. This dependence on the backing pressure can be explained by the existence of the phase-matching mechanism for the 9-order harmonic generation $9\omega = 6(2\omega) - 3\omega$. We have been unable to identify any other explanation.

In Fig. 4 the high-order harmonic spectra generated in a single O_2 gas jet at a backing pressure of 330 mbar by fundamental radiation (ω), second harmonic radiation (2ω),

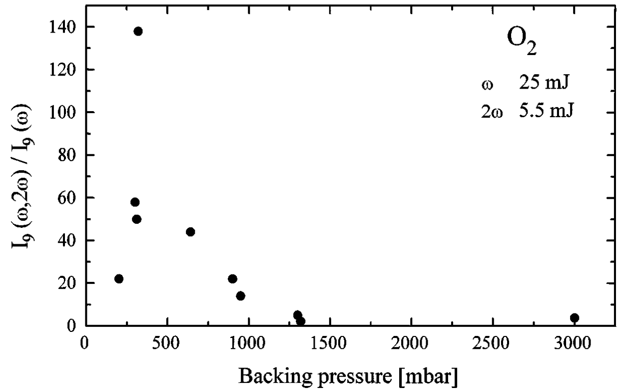

FIG. 3. Ratio of the 9-order harmonic signals generated in a single O_2 gas jet by two-color (ω, 2ω) laser radiation and by the fundamental radiation alone. Approximately 100 shots are used for each experimental point.

and two-color (ω, 2ω) laser radiation are compared. For the fundamental radiation alone, the plasma is produced and the sum-frequency mixing processes are suppressed due to an unfavorable phase mismatch. For the second harmonic the generation of the fifth harmonic (10ω) and

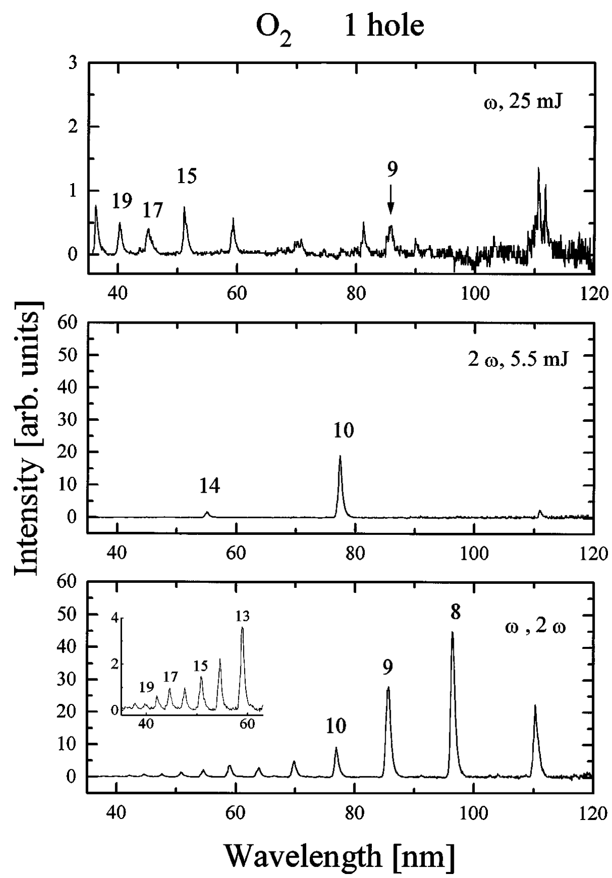

FIG. 4. Comparison of high-order harmonic spectra generated in a single O_2 gas jet at a backing pressure of 330 mbar by fundamental radiation (ω), by second harmonic (2ω), and by two-color (ω, 2ω) laser radiation. The spectral sensitivity of the detection system has been taken into account.

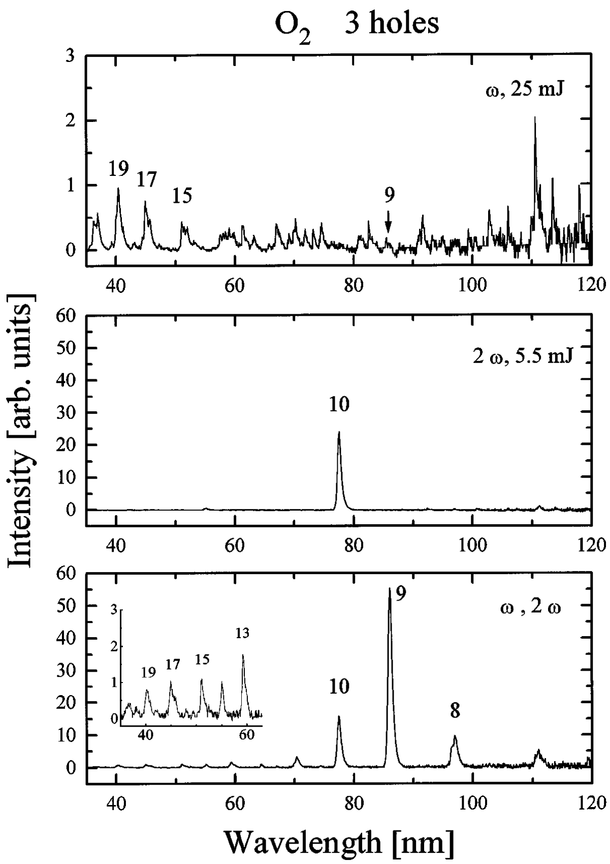

FIG. 5. Comparison of high-order harmonic spectra generated in three O_2 gas jets (three open holes) at a backing pressure of 330 mbar.

seventh harmonic (14ω) can be seen. With the two-color laser radiation, the intensity of the 9-order harmonic is much stronger than in the case of the fundamental radiation. The appearance of a strong 8-order harmonic signal in the two-color spectrum can be explained by the $8\omega = 5(2\omega) - 2\omega$ process, which has a small positive dispersion (see Fig. 2).

In Fig. 5 the same spectra as in Fig. 4 are shown for three open holes. Note that the harmonic intensities can be quantitatively compared. As can be seen, in the two-color case the 9-order harmonic is dominating the spectrum. This behavior of the 9-order harmonic with the plasma length provides additional evidence for phase-matched difference-frequency mixing, $9\omega = 6(2\omega) - 3\omega$, in the two-color laser field. The intensity of the 8-order harmonic signal reduces with the plasma length as should be expected for a process with a positive dispersion.

In conclusion, results presented in this Letter provide a first clear demonstration of phase-matched high-order difference-frequency mixing in plasmas. In the two-color (ω,2ω) laser field, the 9-order harmonic signal has a sharp maximum as a function of the backing pressure. The intensity of the 9-order harmonic grows with the plasma length and becomes dominating in the spectrum. This is evidence that in the two-color laser field the 9-order harmonic is generated by phase-matched difference-frequency mixing, $9\omega = 6(2\omega) - 3\omega$. We observe the analogous behavior of the 9-order harmonic signal with argon and xenon gases. We hope that the results demonstrated in this Letter can in the near future lead to a practical XUV radiation source.

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