Control of Laser High-Harmonic Generation with Counterpropagating Light

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(Received 30 March 2001; published 10 September 2001)

Relatively weak counterpropagating light is shown to disrupt the emission of laser high-harmonic generation. Harmonic orders ranging from the teens to the low thirties produced by a 30-femtosecond pulse in a narrow argon jet are "shut down" with a contrast as high as 2 orders of magnitude by a chirped 1-picosecond counterpropagating laser pulse (60 times less intense). Alternatively, under poor phase-matching conditions, the counterpropagating light boosts harmonic production by similar contrast through quasiphase matching where out-of-phase emission is suppressed.

DOI: 10.1103/PhysRevLett.87.133902 PACS numbers: 42.65.Ky

Aside from the atomic response, the conversion of laser energy into high-order harmonics is limited primarily by phase mismatches in the generation process [1,2] and by the reabsorption of harmonic light within the generating medium [3]. Phase mismatches arise from diffraction effects, refractive index mismatches, and intrinsic phase variations as atoms are subjected to differing laser intensities. Several avenues for reducing or eliminating phase mismatches have been and continue to be pursued. A notable advance has been the development of phase matching in wave guides, either hollow-core fibers [3,4] or self-guiding pulses [5,6]. This approach has been successful for intermediate harmonic orders, the output being limited primarily by reabsorption of the high harmonics in the medium. Other schemes include interplay between intrinsic and geometrical phase mismatches [7] and manipulation of the laser field, spatially [8] or temporally [9]. Self-phase matching in the nonadiabatic limit was predicted recently for extremely high-harmonic orders [10].

One obvious scheme for combating the problem of phase mismatches in high-harmonic generation is the method of quasiphase matching [10,11]. By suppressing the out-of-phase harmonic emission in selected zones of the generating volume the overall emission can be dramatically enhanced. This scheme, which has not previously been implemented for high-harmonic orders, might be pursued through a variety of methods: removing atomic population from out-of-phase zones, ionizing the atoms in the zones, or otherwise preventing harmonic emission in the zones. The first two approaches potentially introduce additional phase mismatches through the medium density, which must be taken into account. Alternatively, nonionizing counterpropagating light can be used to suppress the production of high harmonics through a microscopic or local phase mismatch.

The suppression of high-harmonic generation by relatively weak counterpropagating light was predicted and explained in Ref. [12]. Weak counterpropagating light induces not only a standing amplitude modulation on the laser field but, more importantly, also a standing phase modulation. Because of the extreme nonlinearity of the harmonic generation process, a modest spatial variation in the phase of the generating laser field translates into a strong spatial phase variation in the individual high harmonics. Even when the counterpropagating light is 2 orders of magnitude less intense than the main generating pulse, the resulting phase modulation is enough to cause severe phase mismatches for high harmonics over the spatial scale of a half laser wavelength. The phase mismatches on this microscopic scale essentially turn off the local high-harmonic production.

The use of counterpropagating light to suppress high-harmonic production is potentially more convenient than other conceivable quasi-phase-matching methods. A short laser pulse (e.g., 30 fs corresponding to a spatial extent of 10 μm) can be chirped and manipulated (or carved) into (longitudinal) spatial structures with repeated "on" and "off" zones with dimensions far smaller than the millimeter scale of a typical gas jet. Thus, one may feasibly produce many longitudinally selected zones within the typical scale of the generating medium. In addition, weak counterpropagating light does not harm or alter the medium as it passes through. Thus, the counterpropagating light can meet the main generating pulse at any point in the medium without affecting regions yet to be encountered by the main generating pulse. Quasiphase matching with counterpropagating light may prove useful when applied to harmonics with relatively high orders or harmonics generated from ions [13] where the wave-guide method cannot be used.

In this Letter we report on the effects of counterpropagating light on the production of high harmonics ranging from the teens to the thirties. Weak counterpropagating light is shown for the first time to substantially disrupt the high-harmonic generation process. We also demonstrate for the first time quasiphase matching with counterpropagating light. A single counterpropagating pulse is used to enhance the emission of the 23rd harmonic under conditions of poor phase matching.

Figure 1 shows a schematic of the setup used in the experiment. Pulses from a 1-kHz-repetition-rate Ti:sapphire laser system are split just before the pulse compression stage. A delay arm is used for one of the beams to control

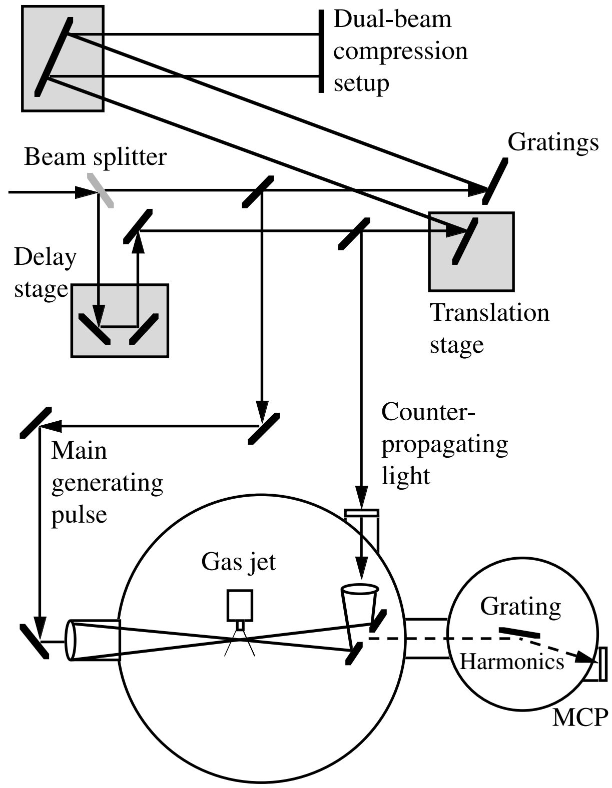

FIG. 1. Experimental setup.

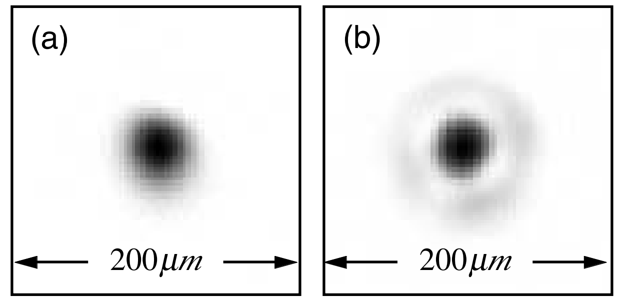

FIG. 2. Focal measurements of the (a) forward-propagating beam and (b) the counterpropagating beam.

the relative timing of the two pulses. The pulses are sent side by side through the compression grating pair. The first grating is split so that the compression of the two pulses can be controlled independently. In the experiment, one beam generates the high harmonics and the other provides the counterpropagating light. Each beam is focused using a 30 cm focal-length lens. The diameter of the beams at each lens is 7.5 mm, corresponding to $f/40$ focusing. The two beams have equal energies of 0.15 mJ. The duration of the generating laser pulse was measured by autocorrelation to be 30 fs full-width-at-half-maximum (FWHM), giving rise to a peak intensity of approximately 5×10^{14} W/cm^2 on a 50 μm diameter focal spot ($1/e^2$ intensity). The laser wavelength is centered on 800 nm.

The counterpropagating light is reflected from a flat mirror 25 cm before the focus. The mirror has a 2.5 mm hole drilled through its center. The hole provides an avenue for high harmonics to be measured while allowing roughly half of the counterpropagating pulse energy to be directed towards the focus. The hole in the mirror causes the counterpropagating beam profile to take on a doughnut shape in the near field which fills in at the focus [14]. Figure 2 shows an image of the focus of each beam, revealing a good spatial match between the two.

To accomplish the spatial and temporal alignment of the pulses, we rely on Rayleigh scattering from free electrons ionized by the laser. With the chamber backfilled and with both pulses temporally compressed to 30 fs, a distinctly higher amount of Rayleigh scattering occurs from the region where the two pulses interfere, owing to enhanced ionization. The excess Rayleigh scattering from the collision point is monitored with a charge-coupled device (CCD) camera. This assures both spatial and temporal alignment of the two pulses. When the chamber is evacuated, the excess Rayleigh scattering from the collision point can also be observed in the gas jet.

The 300 μm diameter gas nozzle was positioned at the collision point of the two pulses. The duration of the counterpropagating light was adjusted by increasing the grating separation and thereby introducing a negative chirp. The pulse duration was lengthened to 1 ps (FWHM), measured by cross correlation with the forward-propagating pulse. A delay stage compensates for the displacement of the grating. When the counterpropagating pulse is strongly chirped, the collision of the two pulses cannot be discerned through Rayleigh scattering from ionized electrons. However, the temporal alignment is assured by monitoring the cross correlation of the two beams. The gas nozzle was positioned just at the edge of the colliding laser beams where the thickness of the gas flow is similar to the thickness of the nozzle opening. The nozzle consists of a 3 mm section of a syringe needle. The gas flowed continuously during the experiment.

The high harmonics are detected using a single microchannel plate (MCP) coupled to a phosphor screen and recorded using a CCD camera. The high-harmonic light passes through two apertures on the way to the MCP. Differential pumping maintains a pressure of 10^{-6} torr near the detector. The harmonics are spectrally resolved using a 600 line/mm tungsten-coated grazing incidence grating (radius of curvature $R = 1$ m). The highest harmonic produced in argon observed under our laser conditions was the 31st.

Figure 3 shows the relative emission of the 23rd harmonic produced in argon with a jet backing pressure of 20 torr, which we estimate resulted in a pressure of 4 torr at the nozzle. Each data point represents an average of 1700 shots read from selected pixels on the CCD camera. The relative arrival time of the 1 ps counterpropagating pulse was scanned (plotted as a function of delay length). We checked that when the counterpropagating pulse

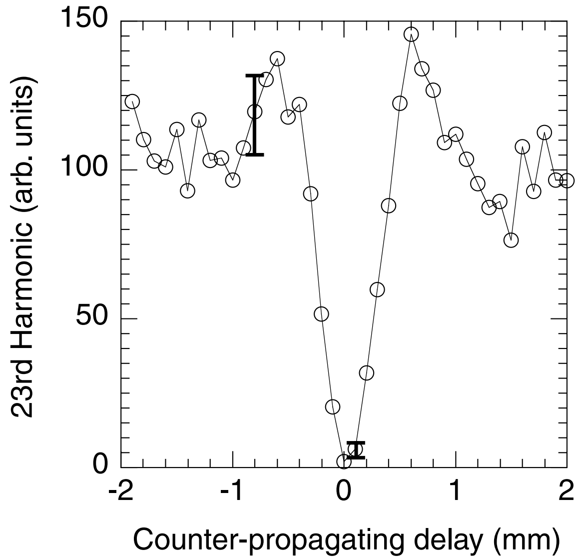

FIG. 3. Harmonic signal as a function of counterpropagating pulse delay with a narrow gas distribution.

arrived either well before or after the main generating pulse, the harmonic production was the same with or without the counterpropagating light. This rules out the possibility that the counterpropagating light harms the medium (e.g., through ionization). The intensity of the counterpropagating light was lower than the main generating pulse by roughly a factor of 60. Its intensity was thus insufficient to ionize the medium significantly.

As is evident in Fig. 3, when the timing of the counterpropagating pulse is aligned such that the main generating pulse is continually in the presence of the counterpropagating light while in the medium, the harmonic generation process is turned off, nearly to the point of extinction. The contrast for the 23rd harmonic is approximately 2 orders of magnitude. Similar behavior is seen for the neighboring harmonics (up to the highest ones visible), although the lower harmonic orders (i.e., the teens) are not as strongly extinguished. This is expected since a stronger standing phase modulation on the laser field is required to suppress the lower harmonic orders.

To understand why relatively weak counterpropagating light dramatically suppresses high-harmonic production, consider the sum of strong and weak oppositely traveling plane wave fields (with respective field strengths E_1 and E_2). The sum can be written with the form of the stronger field as follows [14]:

$$E_1 e^{i(kz-wt)} + E_2 e^{-i(kz+wt)} = E_t(z) e^{i[kz-wt+\phi(z)]}, \quad (1)$$

where

$$E_t(z) = \sqrt{E_1^2 + E_2^2 + 2E_1E_2\cos 2kz}, \quad (2)$$

and

$$\phi(z) = -\tan^{-1}\frac{E_2 \sin 2kz}{E_1 + E_2 \cos 2kz}. \quad (3)$$

Note that $E_t(z)$ and $\phi(z)$ are time independent and provide standing amplitude and phase modulations. Figure 4 shows the relative intensity $[E_t(z)/E_1]^2$ and phase $\phi(z)$ when the weak field is 100 times less intense (i.e., $E_2/E_1 = 0.1$). As seen in Fig. 4, both the intensity and phase modulations are periodic over a half-laser wavelength. The standing phase variation fluctuates over a total range of 0.06π, which translates into a phase variation of more than π for harmonics beyond the 15th order.

Consider phase matching of the qth harmonic over a distance of a half-laser wavelength. To obtain the harmonic emission in the direction of the strong beam, one integrates (along z) the emission from the atoms multiplied by the standing phase factor $\exp[iq\phi(z)]$. This appended phase factor is enough to dramatically disrupt the phase-matching integral over this microscopic scale.

In the actual experiment, the envelope of each laser pulse varies in time (especially the compressed pulse), in contrast with plane waves. Moreover, the counterpropagating pulse used in the experiment was chirped so that the frequencies of the colliding pulses did not necessarily match everywhere in the collision region. Nevertheless, as evidenced by the experimental results, the fact that the pulses vary somewhat from true plane waves is not critical to achieving strong harmonic suppression. We verified that a positive chirp suppresses the harmonic production similar to the negative chirp. It is possible that intensity-dependent intrinsic phases also play a role [15]. As seen in Fig. 4, the maximum intensity in the standing modulation is approximately 50% above the minimum. An intensity-dependent intrinsic phase variation in the qth harmonic is therefore likely to add further disruption to the phase-matching integral (or conceivably undo disruption under very specific circumstances).

We created poor phase-matching conditions by moving the gas jet away from the laser focus a distance of 1 mm.

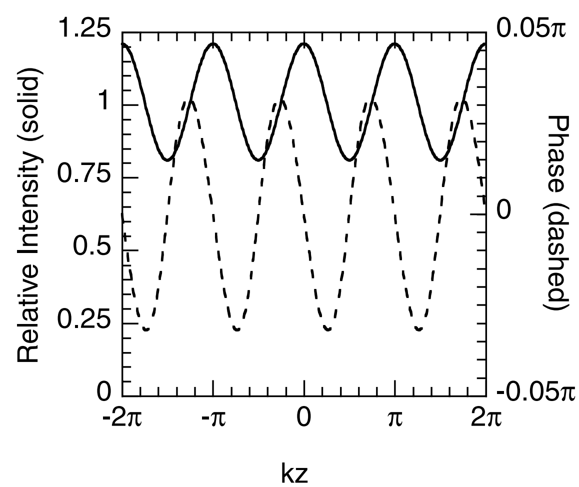

FIG. 4. Intensity and phase variation when a plane wave is met by a counterpropagating plane wave with one-hundredth the intensity.

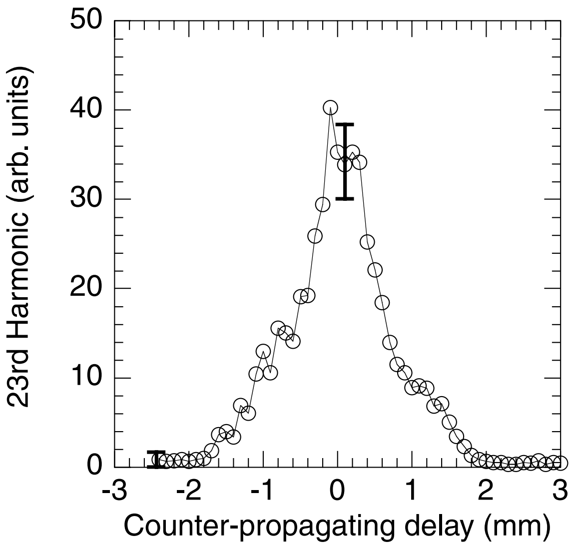

FIG. 5. Harmonic signal as a function of counterpropagating pulse delay with a wide gas distribution.

The backing pressure was increased to 140 torr. We estimate that this produced a gas distribution in the laser focus approximately 1 mm thick with a pressure of 2–4 torr. In the absence of counterpropagating light, the 23rd harmonic emission with the wider gas distribution was very low due to a severe phase mismatch, arising mainly from geometrical effects owing to the Gouy shift. The Rayleigh range of the laser beams is approximately $z_0 = 2$ mm. Near the focus, the corresponding phase mismatch over a longitudinal distance of Δz is then on the order of $23 \tan^{-1}(\Delta z/2 \text{ mm})$. This corresponds to a π phase mismatch over a distance of roughly $\Delta z = 0.3$ mm. Therefore, we estimate there to be approximately three phase zones within the gas distribution, the outer two zones having the same phase and differing from the center zone by π. The result was very poor emission of the 23rd harmonic because the center zone with its higher gas density presumably offset the emission from the other zones.

Figure 5 shows the emission of the 23rd harmonic as a function of the counterpropagating pulse delay. Since the gas distribution is much thicker, the 1 ps counterpropagating pulse is able to interact only with the generating pulse over a fraction of the gas distribution (i.e., approximately one phase zone). As seen in Fig. 5, the harmonic emission shows a nearly one-hundred-fold increase as the counterpropagating light is able to suppress harmonic emission in a portion of the gas jet. The curve also gives direct evidence for the thickness of the gas distribution. Keep in mind that the relative speed between the colliding pulses is $2c$, so the thickness of the gas distribution is half of the width suggested by the profile plotted against the delay of the counterpropagating pulse.

In summary, we have demonstrated that relatively weak counterpropagating light turns off laser high-harmonic generation and is an effective tool for quasiphase matching. Because the counterpropagating light does not harm the generating medium, this method might be employed in cooperation with other phase-matching approaches. In future work, we plan to apply this technique with multiple counterpropagating pulses to deal with severe and difficult-to-address phase mismatches (e.g., higher harmonic orders generated from both neutrals and ions) where there are many phase zones in the interaction volume.

This work was supported by the National Science Foundation under Grant No. PHY-9985080.

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