High-order harmonic generation in crossed laser beams

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We investigate laser high-order harmonic generation in the presence of interfering light. A relatively weak interfering pulse intersects the primary harmonic-generating laser pulse at the focus. The influence on the harmonic-generation process is studied at near-counterpropagating and at perpendicular angles. The interfering beam creates a standing intensity and phase modulation, which disrupts microscopic phase matching and shuts down local high-harmonic production. Simple quasi-phase matching is demonstrated in which the interfering light disrupts harmonic generation in a portion of the laser focus. Under poor phase-matching conditions, perpendicularly propagating light is shown to enhance the 23rd harmonic generated in argon. © 2003 Optical Society of America

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1. INTRODUCTION

Harmonic generation in the presence of weak counterpropagating light was reported recently.[1] The interfering pulse, 60 times less intense, collided with the primary harmonic-generating laser pulse in the focus. Interference between the fields shut down the harmonic-generation process. The production of harmonics (orders in the range 20–30) was suppressed by 2 orders of magnitude when the counterpropagating light interfered with the primary pulse throughout a narrow jet of argon gas. Enhancement to harmonic emission was also demonstrated. Under poor phase-matching conditions, a single counterpropagating light pulse eliminated harmonic emission from a portion of a wide gas distribution, significantly boosting the emission to that of the 23rd harmonic.

The use of weak (nonionizing) interfering light represents a plausible approach for achieving quasi-phase matching.[2,3] A sequence of light pulses might be used to suppress out-of-phase harmonic emission in many selected zones of the generating volume, thus increasing the overall emission of particular harmonic orders. This approach compliments other methods of ameliorating phase mismatches, such as the use of waveguides, either hollow-core fibers[4,5] or self-guiding pulses,[6,7] which have been successfully applied to intermediate harmonic orders (as high as the 30th). Quasi-phase matching with an interfering light pulse may prove particularly useful for the high-order harmonics or for harmonics generated in ions for which phase mismatches can be severe and difficult to control.[8,9] We plan to attempt quasi-phase matching in these cases.

Figure 1 is a schematic of the setup used in our counterpropagating light experiments. Pulses from a 1-kHz repetition-rate Ti:sapphire laser system (800 nm) are split just before the temporal compression stage. A delay arm controls the relative timing of the two pulses, which can be compressed independently to different durations. The two beams enter the experimental chamber through different ports, one beam generating the high harmonics and the other providing the interfering light. Each beam is focused with a 30-cm focal-length lens. The diameter of the beams at each lens is 7.5 mm, providing $f/40$ focusing. The two beams have equal energies of 0.15 mJ. The duration of the primary (generating) laser pulse is 30 fs full width at half-maximum, producing a peak intensity of approximately 5×10^{14} W/cm^2 on a 50-μm-diameter focal spot (measured from $1/e^2$ intensity). The counterpropagating light reflects from a mirror with a hole drilled through its center. The hole provides an avenue for high harmonics to be measured while roughly half of the counterpropagating pulse energy is directed toward the focus.

In this paper we describe the results of a modified version of the experiment depicted in Fig. 1. Rather than sending the interfering light pulses into the focus in the exact counterpropagating direction, we directed the interfering light a few degrees off axis. We also sent the interfering light into the focus from the perpendicular direction. In both cases, the weak interfering light was effective in suppressing harmonic emission. An obvious advantage to working off axis is the avoidance of potentially damaging feedback to the laser amplifier system from the residual energy in counterpropagating beams.

2. BEAM INTERFERENCE

The suppression of high-harmonic generation by relatively weak counterpropagating light is explained in Ref. 3. The counterpropagating light induces standing amplitude and phase modulations on the generating laser field. The modulations repeat with a period of a half-laser wavelength. If the interfering beam enters at an angle other than counterpropagating, similar standing modulations result, albeit with different spatial geometry and characteristic period. Relatively weak interfering light (2 orders of magnitude less intense) can seriously disrupt phase mismatches in the process of high-harmonic gen-

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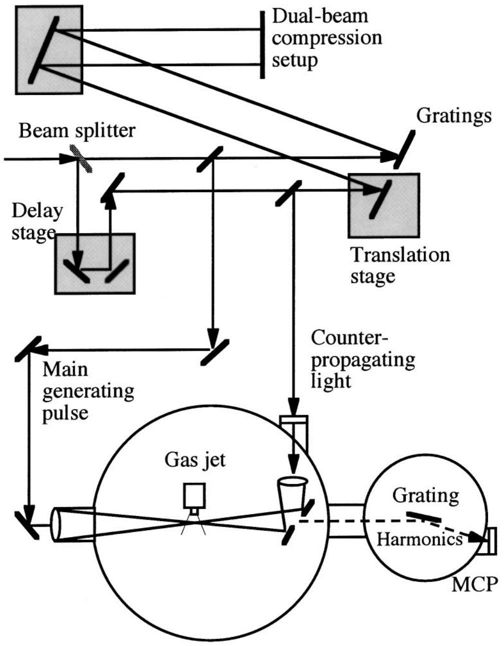

Fig. 1. Experimental setup.

eration, owing to the extreme nonlinearity of that process. A modest spatial variation in the phase of the laser field translates into a strong spatial variation in the phase of individual high harmonics, causing severe interference.

To appreciate the microscopic phase mismatches that occur in interfering laser beams, consider two intersecting plane waves, both with frequency ω. Let the primary wave, which has field strength E_1, travel in the z direction. Let the interfering wave, with field strength E_2, propagate at angle θ relative to the positive z axis. We take the polarizations of the two waves to be parallel, in a direction perpendicular to the plane containing both k-vectors.

If the primary field is much stronger than the interfering field, it is helpful to write the sum of the fields in a form that resembles the primary field by itself. The sum of the two plane waves is thus written as

$$\begin{aligned} &E_1 \exp[i(kz - \omega t)] + E_2 \\ &\quad \times \exp[i(kz \cos\theta + ky \sin\theta - \omega t)] \\ &\qquad = E_{\text{tot}}(y, z) \exp[i\phi(y, z)] \exp[i(kz - \omega t)], \end{aligned} \tag{1}$$

where

$$E_{\text{tot}}(y, z) = (E_1{}^2 + E_2{}^2 + 2E_1E_2 \cos\alpha)^{1/2}, \tag{2}$$

$$\phi(y, z) = \tan^{-1}\left[\frac{(E_2/E_1)\sin\alpha}{1 + (E_2/E_1)\cos\alpha}\right], \tag{3}$$

$$\alpha \equiv kz\cos\theta + ky\sin\theta - kz. \tag{4}$$

We see in this form the degree to which the net field deviates from that of a simple plane wave. Keep in mind that a single plane-wave field is the ideal for good harmonic phase matching (over a microscopic scale). Equations (1)–(4) reduce to the counterpropagating case[1] when $\theta = 180°$. The time-independent amplitude $E_{\text{tot}}(y, z)$ and phase $\phi(y, z)$ describe the standing modulations in the net field. These standing modulations, especially in phase, cause the disruption of phase matching and shut down the local harmonic production. In the exact counterpropagating case, the modulations depend only on z. In contrast, if the interfering beam is skewed in angle, the modulations depend on both y and z. In this off-axis case, each position y has a shifted intensity and phase modulation when it is observed along the z direction. Nevertheless, the modulations are always of same strength.

As an example, consider interfering light that intersects the primary wave at the perpendicular angle $\theta = 90°$. In this case Eqs. (2) and (3) reduce to

$$E_{\text{tot}}(y, z) = [E_1{}^2 + E_2{}^2 + 2E_1E_2\cos(ky - kz)]^{1/2}, \tag{5}$$

$$\phi(y, z) = \tan^{-1}\left[\frac{(E_2/E_1)\sin(ky - kz)}{1 + (E_2/E_1)\cos(ky - kz)}\right]. \tag{6}$$

The modulations in intensity and phase for perpendicularly propagating light are shown in Fig. 2. The gray scale is set in each case to the full range of variation, which depends on the ratio E_2/E_1. Lighter shades represent maximum values, and darker shades represent minimum values. When the perpendicularly propagating light is 100 times less intense than the generating light (i.e., when $E_2/E_1 = 0.1$) the intensity modulations fluctuate from $0.81I_1$ to $1.21I_1$. At the same time, the phase fluctuates from -0.03π to $+0.03\pi$. Again, this range of fluctuation is identical for any angle $\theta \neq 0$, including the counterpropagating case. This suggests that perpendicularly propagating light should be as effective in suppressing local harmonic emission as is counterpropagating light.

To appreciate how the interfering light disrupts the phase matching of harmonic emission, consider the emission of the qth harmonic along the z direction. To obtain the overall harmonic signal in that direction (i.e., the direction of the primary beam), one integrates (along z) harmonic emission from each location multiplied by the phase factor $\exp[iq\phi(y, z)]$. This periodic factor can cause serious disruption to the phase-matching integral if q is large enough that $q\phi(y, z)$ fluctuates through a range of π or more. This appended phase factor is thus able to disrupt harmonic emission within a microscopic length (λ in the perpendicularly propagating case; $\lambda/2$ in the counterpropagating case).

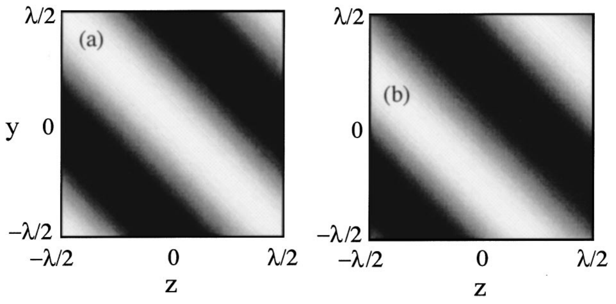

Fig. 2. (a) Intensity and (b) phase variations in the y–z plane for perpendicularly propagating light and $E_2/E_1 = 0.1$. MCP, microchannel plate.

3. NEAR COUNTERPROPAGATING INTERFERENCE

We investigated high-harmonic generation in the presence of interfering light shifted by 2° from the counterpropagating direction (i.e., $\theta = 178°$). The setup is the same as that in Fig. 1. The primary laser pulse was set to 30 fs, producing a peak intensity of approximately 5×10^{14} W/cm^2. Initially the two beams were aligned in the exact counterpropagating geometry, following the procedure described in Ref. 1. The influence of the interfering beam on high-harmonic generation was monitored as the angle of the beam was gradually shifted away from the counterpropagating direction. The electric fields in the two beams remained parallel because the offset of the interfering beam was in the vertical dimension while both beams were polarized in the horizontal dimension.

A 300-μm-diameter gas nozzle was positioned at the collision point of the two pulses. The argon pressure at the nozzle opening was estimated to be 4 Torr. The counterpropagating light pulse was chirped to 1 ps (FWHM), measured by cross correlation with the forward-propagating pulse. The gas nozzle was positioned close to the laser beam such that the gas distribution in the laser focus had a thickness similar to the size of the nozzle opening. In this case the primary pulse encountered the interfering light throughout the entire gas distribution.

When the two beams intersected properly at the focus, significant disruption to the harmonic signal was observed. The harmonics were detected through a small hole in the mirror from which the interfering light reflected. Adjustments were then made to the direction of the counterpropagating light by use of two mirrors. One mirror displaced the beam, and the other mirror redirected it to meet the primary beam in the focus. This was done iteratively while the suppression of the harmonic emission itself served as the alignment diagnostic. Because of the relatively small angular displacement, the intersecting beams remained well overlapped over the thickness of the gas jet.

Figure 3 shows the emission of the 23rd harmonic with the interfering light shifted 2° from the counterpropagating direction. The intensity of the counterpropagating light was lower than that of the main generating pulse by roughly a factor of 30 (instead of 60, because the light was reflected from a part of the mirror with no hole). The intensity of the interfering light was insufficient to ionize the argon gas significantly. The relative arrival time of the counterpropagating pulse was scanned (plotted as a function of delay length). As is evident from Fig. 3, when the arrival time of the interfering light was synchronized such that it interfered with the primary pulse throughout the gas distribution, the emission of the 23rd harmonic was reduced by 2 orders of magnitude. Similar behavior was observed for the neighboring harmonics. This behavior closely matches the results that were obtained previously in the counterpropagating case.[1]

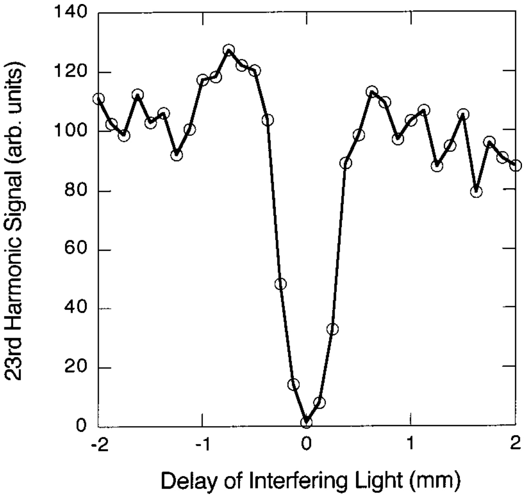

Fig. 3. Suppression of the 23rd harmonic generated in a narrow gas distribution when interfering light intersects 2° from counterpropagating.

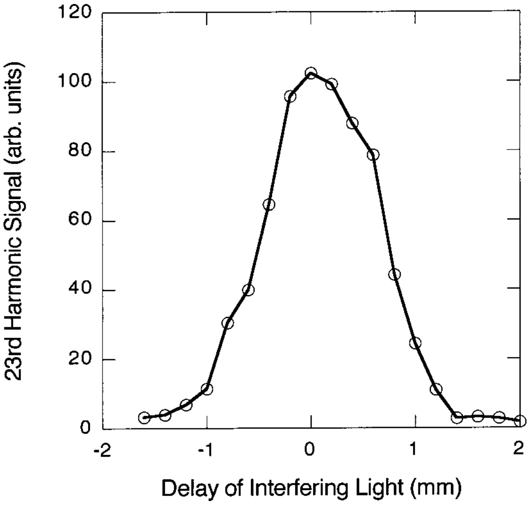

Fig. 4. Enhancement of the 23rd harmonic generated in a wide gas distribution when interfering light intersects 2° from counterpropagating.

We created poor phase-matching conditions by moving the gas jet a distance of 1 mm away from the laser focus. We estimate that this produced an ~1-mm-thick a gas distribution in the laser focus. The pressure in the wider gas distribution was adjusted to 2–4 Torr (with increased backing pressure). Under these conditions there was more than one phase zone within the gas distribution, which resulted in poor emission for the 23rd harmonic. Figure 4 shows the emission of the 23rd harmonic as a function of delay of the interfering pulse. With the thicker gas distribution, the 1-ps interfering pulse interacted with the primary pulse in only approximately one third of the gas distribution. In Fig. 4 the harmonic emission shows a large enhancement as the interfering light suppressed out-of-phase harmonic emission in a part of the gas jet. The enhancement restored the harmonic signal to a level similar to that achieved under good phase-matching conditions (i.e., with the narrower gas distribution).

4. PERPENDICULARLY PROPAGATING INTERFERENCE

We modified the setup in Fig. 1 to accommodate an interfering beam propagating from the perpendicular direction. The arrangement ensures that the polarization of the two beams remains parallel. For this perpendicular propagation, the extent to which the overall harmonic signal can be extinguished is limited by the lateral width of the interfering beam, assuming a sufficiently long pulse duration. To avoid wasting laser energy, the length (duration) of the interfering light pulse should match the width of the pulse in the focus. The duration of the interfering pulse need be long enough only to remain present while the primary pulse crosses side to side through it. To increase the lateral width we tilted the focusing lens of the interfering beam to introduce astigmatism. A CCD image of the astigmatic beam profile taken at the focus is shown in Fig. 5. The astigmatism produced a beam approximately twice as wide as it was tall. Unfortunately, we were unable to match the full thickness of the gas jet (300 μm) with the astigmatism because of the need to have high intensity available in both beams for alignment purposes.

The spatial and temporal overlap of the colliding pulses must be carefully aligned. To accomplish the initial alignment we relied on Rayleigh scattering from free electrons ionized by the laser pulses. With the chamber backfilled with air (at atmospheric pressure) and with both pulses temporally compressed to 30 fs, a distinctly higher amount of Rayleigh scattering occurred from the region where the two pulses collide. We attribute the increased scattering to enhanced ionization owing to the interference between the two fields. The excess Rayleigh scattering from the collision point was monitored with a CCD camera, as shown in Fig. 6. The bright collision point ensures both spatial and temporal alignment of the two pulses.

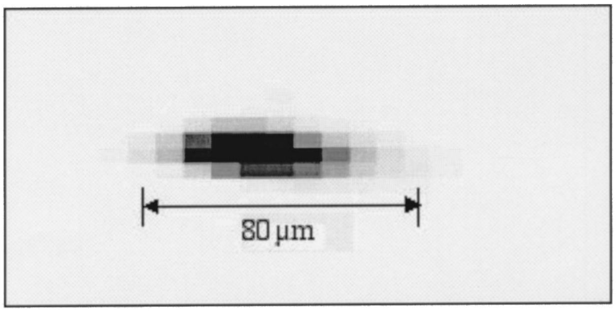

Fig. 5. Astigmatic focus of the interfering light.

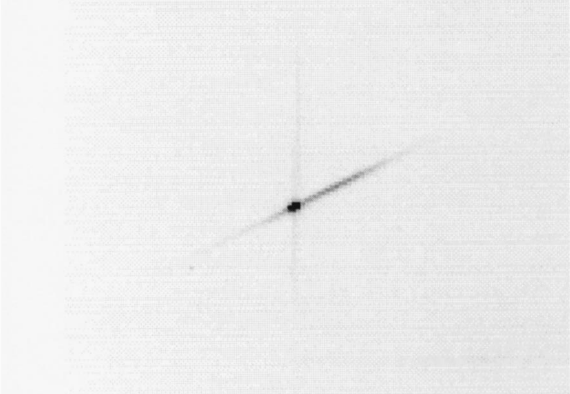

Fig. 6. Rayleigh scattering produced by free electrons in the focus of intersecting pulses.

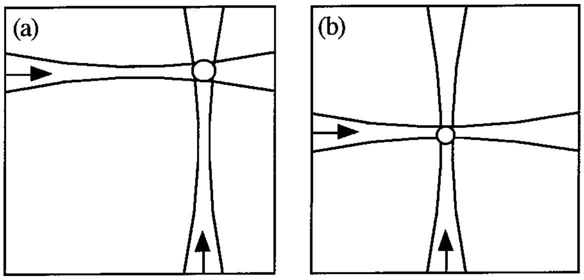

Fig. 7. Alignment of beams (a) in the atmosphere and (b) in vacuum.

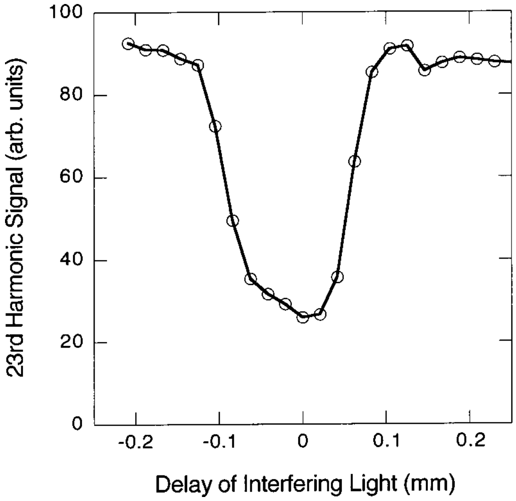

Fig. 8. Suppressed harmonic emission with the interfering perpendicularly directed light.

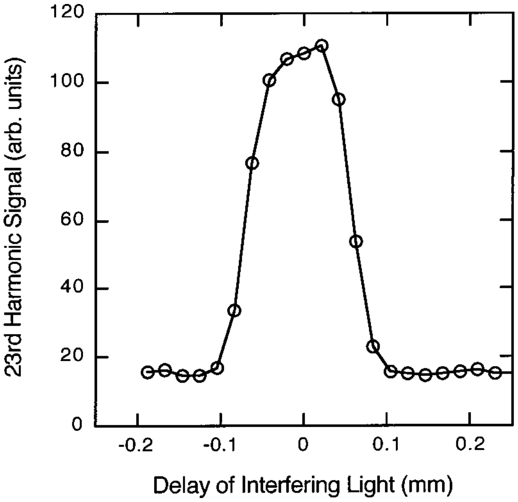

Fig. 9. Enhanced harmonic emission with the interfering perpendicularly directed light.

Alignment of the two beams is aided by choice of a camera angle in which the two beams appear to be parallel (i.e., the camera is in the plane of the crossing beams). The delay in the interfering pulse is then repeatedly scanned while subtle adjustments are made to the pointing of the beam. The bright dot blinks on for just a moment during the temporal scan when good spatial alignment is achieved. Then the optimal temporal alignment is found. In vacuum, the longitudinal locations of the foci differ from those in atmosphere that are due to self-focusing or self-defocusing effects or both. Therefore, at atmospheric pressure the intersection point had to be set outside best focus such that in vacuum the foci shifted to the intersection point, as depicted in Fig. 7.

The duration of the interfering pulse was set to 270 fs, which corresponds to the time that it takes light to travel the width of the 80-μm-wide astigmatic beam waist. This width is narrower than the gas distribution, ~200 μm (~4 Torr). Thus the interfering light interacted with the primary pulse in only approximately half of the gas distribution. Figure 8 shows the emission of the 23rd harmonic as a function of delay of the interfering pulse. As shown in Fig. 8, with appropriate timing the emission is extinguished by as much as a factor of 3. This amount of suppression is consistent with the fact that the harmonic emission could not be turned off throughout the entire gas distribution.

As we had done before, we created poor phase-matching conditions by backing the gas jet 1 mm away from the two beams. The backing pressure was increased to achieve similar pressure in the laser focus. As shown in Fig. 9, harmonic emission was enhanced when the interfering light eliminated out-of-phase harmonic emission in a small portion of the interaction region. Again, the narrowness of the interfering focus probably limited the enhancement to approximately a factor of 6; the harmonic signal was not fully restored to its level with the narrower gas jet.

5. CONCLUSIONS

In summary, we have demonstrated that relatively weak interfering light substantially turns off laser high-harmonic generation. The interfering light may intersect the primary laser pulse at virtually any angle (other than nearly copropagating with the primary laser pulse). This variance of angle eliminates the potential problem of unintended laser feedback into the amplifier system. The use of interfering light promises to be an effective tool for quasi-phase matching of high-order harmonic generation. Because the interfering light can be weak enough not to harm the generating medium, it may be employed in cooperation with other phase-matching approaches. We plan to use multiple interfering pulses in future experiments to deal with severe phase mismatches that arise from interference of many phase zones in the focus. In particular, we shall attempt quasi-phase matching for high-harmonic orders generated from both neutrals and ions.

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