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Frequency-Synthesized Approach to High-Power Attosecond Pulse Generation and Applications: Applications

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Abstract

In part I of this work, we present the design, construction and diagnostics of a new scheme of generating high-power attosecond pulses and arbitrary waveforms by multicolor synthesis. In this chapter, we demonstrate selected applications of this novel source, such as coherently controlled harmonic generation as well as phase-sensitive two-color ablation of copper and stainless steel by this multicolor laser system.

Keywords: arbitrary waveform synthesis, attosecond, coherent control, perturbative nonlinear optics, optical harmonic generation, four-wave mixing, laser ionization and plasma, laser material processing, laser ablation

1. Introduction

In part I of this work [1], we reported a new high-power laser system for generating attosecond light pulses and arbitrary waveforms by frequency synthesis. The laser system can generate up to five amplitude and phase-controlled collinear beams with wavelengths from the fundamental output of the Nd:YAG laser ($\lambda = 1064$ nm) and its second ($\lambda = 532$ nm) through the fifth harmonic ($\lambda = 213$ nm). Sub-single-cycle (~ 0.37 cycle) sub-femtosecond (360 attosecond) pulses with carrier-envelope phase (CEP) control can be generated in this manner. The peak intensity of each pulse exceeds 10^{14} W/cm² with a focused spot size of 20 μ m. Stable square and sawtooth waveforms were also demonstrated [2].

The coherent control of nonlinear optical processes such as harmonic generation by waveform-controlled laser field is important for both fundamental science and technological applications. Previously, we have studied the influence of relative phases and intensities of the two-color pump (1064 and 532 nm) electric fields on the third-order nonlinear frequency conversion process in argon [2]. It was shown that the third-harmonic (TH) signal oscillates periodically with the relative phases of the two-color driving laser fields. The data are in good agreement with a perturbative nonlinear optical analysis of the TH signal, which consists of contribution of the direct third-harmonic-generation (THG), four-wave mixing (FWM) and the interference of the above two processes.

As an extension of this work, we have studied generation of harmonics by three-color synthesized waveform in inert gas systems. We will illustrate the physics involved by examining the case for fourth-harmonic generation (FHG) in Section 2.

Anomalous enhancement of the THz signal in the presence of the 532 nm beam was observed, however. In this work, we show that plasma generated through the ionization process during laser-matter interaction plays a significant role in the enhancement of the TH signal. We also demonstrated phase-sensitive two-color ablation of copper and stainless steel. Our results show that hole drilling is more efficient for optimized waveforms.

2. Nonlinear frequency conversion by coherently controlled three-color excitation of inert gases

In this section, we investigate the use of three-color laser fields as a source to generate harmonic signals in an isotropic media, for example, inert gases. With three-color pump and consider only the lowest order nonlinear processes in isotropic systems, that is, third-order nonlinear process, one can expect to generate 4th to 9th harmonics of the laser fundamental output. A richness of nonlinear effects and complicated quantum interference phenomena is predicted. This summarized in **Table 1**.

Using perturbative nonlinear formulism, we first derived the general formula of the harmonic electric field as well as the corresponding intensity. The coherent effect manifests itself through the interference of two frequency conversion pathways. In the following, we will use the case of FHG to illustrate the physical phenomena expected.

With three-color field (the fundamental ω_1 , second harmonic $\omega_2 = 2\omega_1$ and third harmonic $\omega_3 = 3\omega_1$) excitation, the fourth-harmonic signal can be generated by three nonlinear optical processes ($\omega_1 + \omega_1 + \omega_2 = \omega_4$, $-\omega_1 + \omega_2 + \omega_3 = \omega_4$, and $-\omega_2 + \omega_3 + \omega_3 = \omega_4$). The conversion efficiency for the fourth-harmonic signal can be modulated by the interference between each two of three FWM processes. As the relative phase between ω_1 , ω_2 and ω_3 vary, combinations of three sinusoidal modulation due to interference in the output intensity of the fourth harmonic at frequency ω_4 is predicted. We will also show that the relative amplitude of the fundamental, second-harmonic and third-harmonic driving laser field influences the fourth-harmonic signal.

Harmonic generation	Processes
Fourth	$\omega_1 + \omega_1 + \omega_2 \rightarrow \omega_4$ $-\omega_1 + \omega_2 + \omega_3 \rightarrow \omega_4$ $-\omega_2 + \omega_3 + \omega_3 \rightarrow \omega_4$
Fifth	$\omega_1 + \omega_2 + \omega_2 \rightarrow \omega_5$ $\omega_1 + \omega_1 + \omega_3 \rightarrow \omega_5$ $-\omega_1 + \omega_3 + \omega_3 \rightarrow \omega_5$
Sixth	$\omega_2 + \omega_2 + \omega_2 \rightarrow \omega_6$ $\omega_1 + \omega_2 + \omega_3 \rightarrow \omega_6$
Seventh	$\omega_2 + \omega_2 + \omega_3 \rightarrow \omega_7$ $\omega_1 + \omega_3 + \omega_3 \rightarrow \omega_7$
Eighth	$\omega_2 + \omega_3 + \omega_3 \rightarrow \omega_8$
Ninth	$\omega_3 + \omega_3 + \omega_3 \rightarrow \omega_9$

Table 1. Third-order nonlinear process ($\omega_n = \omega_i + \omega_j + \omega_k$, $\omega_n = \omega_i + \omega_j - \omega_k$, $n = 4-9$, $i, j, k = 1-3$) that can contribute to the generation of 4th to 9th harmonics of the laser fundamental output of a three-color field (the fundamental ω_1 , second harmonic $\omega_2 = 2\omega_1$ and third harmonic $\omega_3 = 3\omega_1$).

We assume plane-waves propagating in the +z direction. The three-color field can be represented as:

$$\tilde{E}(z, t) = \frac{1}{2} \left[E_1 e^{i(k_1 z - \omega_1 t + \varphi_1)} + E_2 e^{i(k_2 z - \omega_2 t + \varphi_2)} + E_3 e^{i(k_3 z - \omega_3 t + \varphi_3)} + c.c. \right] \quad (1)$$

where φ_1 , φ_2 and φ_3 are the modulated phases of the three colors, respectively. As a source, the nonlinear polarization term in the medium induced by the three-color fields will generate several new frequency components. If we only consider third-order nonlinear optical processes only, assuming no pump depletion, the electric field of the fourth harmonic can then be rewritten as

$$E_4(z) = E_{4I}(z) + E_{4II}(z) + E_{4III}(z), \quad (2)$$

where

$$E_{4I}(z) = i \frac{12\pi^2}{n_4 \lambda_1} N \chi_{4I}^{(3)} E_1^2 E_2 L \sin c \left(\frac{\Delta k_{4I} L}{2} \right) e^{i\Delta k_{4I} L} e^{i(2\varphi_1 + \varphi_2)}, \quad (3)$$

$$E_{4II}(z) = i \frac{24\pi^2}{n_4 \lambda_1} N \chi_{4II}^{(3)} E_1^* E_2 E_3 L \sin c \left(\frac{\Delta k_{4II} L}{2} \right) e^{i\Delta k_{4II} L} e^{i(-\varphi_1 + \varphi_2 + \varphi_3)}, \quad (4)$$

$$E_{4III}(z) = i \frac{12\pi^2}{n_4 \lambda_1} N \chi_{4III}^{(3)} E_2^* E_3^2 L \sin c \left(\frac{\Delta k_{4III} L}{2} \right) e^{i\Delta k_{4III} L} e^{i(-\varphi_2 + 2\varphi_3)}, \quad (5)$$

With the phase or wave-vector mismatch given by $\Delta k_{4I} = k_4 - 2k_1 - k_2$, $\Delta k_{4II} = k_4 + k_1 - k_2 - k_3$ and $\Delta k_{4III} = k_4 + k_2 - 2k_3$. In this section, the symbol "I", "II", "III" represent the three possible four-wave mixing (FWM) processes with corresponding nonlinear susceptibilities: $\chi_{4I}^{(3)} = \chi^{(3)}(\omega_4; \omega_1, \omega_1, \omega_2)$, $\chi_{4II}^{(3)} = \chi^{(3)}(\omega_4; -\omega_1, \omega_2, \omega_3)$, $\chi_{4III}^{(3)} = \chi^{(3)}(\omega_4; -\omega_2, \omega_3, \omega_3)$, respectively. L stands for the nonlinear medium length. The intensity of the fourth-harmonic signal can then be written as

$$I_4(z) = \frac{cn_4}{8\pi} \left\langle \left| \tilde{E}_{4I}(z, t) + \tilde{E}_{4II}(z, t) + \tilde{E}_{4III}(z, t) \right|^2 \right\rangle \quad (6)$$

or

$$I_4(z) = \frac{9216\pi^6 N^2 L^2}{c^2 \lambda_1^2 n_4} \left\{ \begin{aligned} & \frac{1}{n_1^2 n_2} \left(\chi_{4I}^{(3)} \right)^2 I_1^2 I_2 \text{sinc}^2 \left(\frac{\Delta k_{4I} L}{2} \right) \\ & + \frac{4}{n_1 n_2 n_3} \left(\chi_{4II}^{(3)} \right)^2 I_1 I_2 I_3 \text{sinc}^2 \left(\frac{\Delta k_{4II} L}{2} \right) \\ & + \frac{1}{n_2 n_3^2} \left(\chi_{4III}^{(3)} \right)^2 I_2 I_3^2 \text{sinc}^2 \left(\frac{\Delta k_{4III} L}{2} \right) \\ & + \frac{2}{\sqrt{n_1 n_3 n_1 n_3}} \chi_{4I}^{(3)} \chi_{4II}^{(3)} \sqrt{I_1 I_3} I_1 I_2 \text{sinc} \left(\frac{\Delta k_{4I} L}{2} \right) \\ & \times \text{sinc} \left(\frac{\Delta k_{4II} L}{2} \right) \cos \left(\frac{\Delta k_{4I} L}{2} - \frac{\Delta k_{4II} L}{2} + 3\varphi_1 - \varphi_3 \right) \\ & + \frac{2}{\sqrt{n_1 n_3 n_2 n_3}} \chi_{4II}^{(3)} \chi_{4III}^{(3)} \sqrt{I_1 I_3} I_2 I_3 \text{sinc} \left(\frac{\Delta k_{4II} L}{2} \right) \\ & \times \text{sinc} \left(\frac{\Delta k_{4III} L}{2} \right) \times \cos \left(\frac{\Delta k_{4II} L}{2} - \frac{\Delta k_{4III} L}{2} - \varphi_1 + 2\varphi_2 - \varphi_3 \right) \\ & + \frac{1}{n_1 n_2 n_3} \chi_{4III}^{(3)} \chi_{4I}^{(3)} I_1 I_2 I_3 \text{sinc} \left(\frac{\Delta k_{4III} L}{2} \right) \\ & \times \text{sinc} \left(\frac{\Delta k_{4I} L}{2} \right) \cos \left(\frac{\Delta k_{4III} L}{2} - \frac{\Delta k_{4I} L}{2} - 2\varphi_1 - 2\varphi_2 + 2\varphi_3 \right) \end{aligned} \right\} \quad (7)$$

In Eq. (7), the first, second and third terms are the three FWM processes, I, II and III, respectively. The last three terms are cross-terms due the interference of the optical fields generated by FWM processes I and II, II and III and III and I, in that order.

In the simulation, we used three-color laser fields (the fundamental, second harmonic, and third harmonic of the Nd:YAG laser) to generate fourth-harmonic signal in gaseous argon. For simplicity, we further assumed that the phase mismatch for all of FWM processes is equal and negligible. Further, the fundamental and second-harmonic power is the same and their sum is normalized.

In **Figure 1** we show the fourth-harmonic signal as function of the power ratio of the fundamental beam and that of the fundamental and second harmonic combined. The third-harmonic beam is held constant. Examining **Figure 1**, one can see clearly that much higher conversion efficiency of the fourth-harmonic signal would be generated if the normalized power ratio is around 0.8.

Figure 1. The fourth-harmonic signal versus the power ratio $P_1/P_1 + P_2$. Contributions by the three FWM processes and the cross-terms are shown as different colors.

Figure 2. The dependence of the fourth-harmonic signal on the phase of the fundamental beam.

The dependence of the fourth-harmonic signal on the phase of the fundamental beam is shown in **Figure 2**. Clearly, the modulation is more complex than the two-color case.

3. Third-harmonic generation by coherently controlled two-color excitation of inert gases: plasma effect

With two-color excitation, the third-harmonic signal is contributed by the direct THG ($\omega_3 = \omega_1 + \omega_1 + \omega_1$) and four-wave mixing (FWM, $\omega_3 = \omega_2 + \omega_2 - \omega_1$) processes and a cross term of the two. As the relative phase between ω_1 and ω_2 varies, a sinusoidal modulation in output intensity at frequency ω_3 is expected and was demonstrated in our previous work [2]. In intense laser field, plasma can be generated through the ionization of gases. Optical harmonic generation in plasmas has been studied for a long time. Recently, significant enhancement of the third-harmonic emission in plasma has been reported by Suntsov et al. [3]. More than two-order-of-magnitude increase of the efficiency of third-harmonic generation occurs due to the plasma-enhanced third-order susceptibility [5]. More specifically, the presence of charged species (free electrons and ions) can effectively increase the third-order nonlinear optical susceptibility [4, 5]. This indicates that the susceptibility can be expressed as a function of the plasma density N_e induced by laser field. Additionally, the refractive index of the target, for example, gases or solids, is also changed in the presence of the plasma. The wave-vector mismatch Δk , in plasma, between the fundamental and the third-harmonic signal can be derived by using the Drude model. Enhanced third-harmonic signal that eventually saturates at higher plasma density was predicted [6, 7]. In this chapter, we observed more than ten orders of magnitude enhancement of third-harmonic generation in argon plasma by employing the fundamental (1064 nm) and second-harmonic (532 nm) fields of an injection-seeded Q-switched Nd:YAG laser. Under the assumption that susceptibility and wave-vector mismatch depend on the plasma density, we show that plasma plays a significant role in the third-harmonic signal by an analysis based on the formalism of perturbative nonlinear optics. Significant enhancement of the TH signal is caused by the plasma-enhanced susceptibility of the dominant four-wave mixing process. When the plasma density is high enough, the TH signal becomes saturated and drops primarily due to the detrimental effect of the wave-vector mismatch.

The experimental setup for studying the effect of plasma formation on generation of third-harmonic signal by phase-controlled two-color excitation is shown in **Figure 3**. It is a simplified version of the multicolor laser system described in part I of this work and our previous papers [2, 3]. To reiterate, we employed a Q-switched Nd:YAG laser system (Spectra Physics GCR Pro-290) that generates intense 1064 nm pulses with a pulse duration of 10 ns (FWHM) and a line width of $<0.003 \text{ cm}^{-1}$. The laser pulse repetition rate is 10 Hz, and the maximum pulse energy is 1.9 J/pulse. The second-harmonic (532 nm) beam was generated by using the nonlinear optical crystal KD*P (type I phase matching). The maximum pulse energy of the second-harmonic signal is around 1 J/pulse. The fundamental and second-harmonic pulses propagate collinearly with a fixed relative phase. This two-color laser beams are separated by a prism pair into two arms. A power tunable two-color system can be generated with two amplitude modulators for each arm. The relative phase and amplitudes of these two-color laser fields can be timed independently by amplitude and phase modulators. The fundamental and second-harmonic beams are first angularly separated and then made parallel by a pair of

Figure 3. The experimental setup for studying the effect of plasma formation on generation of third-harmonic signal by phase-controlled two-color excitation.

prisms. With the desired amplitude ratio and relative phase, the two-color laser fields are recombined with an identical pair of prisms and then focused into a vacuum chamber filled with argon (10 Torr) by a 10-cm-focal lens to generate the third-harmonic (355 nm) signal. To overlap two foci of the fundamental and second-harmonic beam, the dispersion of the lens is compensated by a telescope in the fundamental arm. The third-harmonic generation is filtered by a monochromator (VM-502, Acton Research) and detected by a photomultiplier tubes (R11568, Hamamatsu).

With excitation by the two-color field (the fundamental ω_1 and second harmonic ω_2) of the Nd:YAG laser, the third-harmonic signal can be generated by two optical processes, i.e., $\omega_1 + \omega_1 + \omega_1 = \omega_3$ and $-\omega_1 + \omega_2 + \omega_2 = \omega_3$. We assume plane-waves propagating in the +z direction. The theoretical formulism is similar to the three-color case in Section 1. In the slow-varying envelope approximation and assume no pump depletion, the TH field can be written as

$$\hat{E}_3(z) = \hat{E}_{3I}(z) + \hat{E}_{3II}(z) = \left\{ \begin{array}{l} i \frac{3\pi^2}{n_3\lambda_1} \chi_I^{(3)} E_1^3 L \text{sinc}\left(\frac{\Delta k_{13}L}{2}\right) e^{i\frac{\Delta k_{13}L}{2}} e^{i3\varphi_1} \\ + i \frac{9\pi^2}{n_3\lambda_1} \chi_{II}^{(3)} E_1^* E_2^2 L \text{sinc}\left(\frac{\Delta k_{213}L}{2}\right) e^{i\frac{\Delta k_{213}L}{2}} e^{i(2\varphi_2 - \varphi_1)} \end{array} \right\} \quad (8)$$

where the subscripts "I" and "II" denote the two nonlinear processes, namely the direct THG and FWM; $\chi^{(3)}$ is the third-order nonlinear susceptibility, L is the length of the nonlinear material; φ_1 and φ_2 are the phases of the fundamental and second-harmonic beams, respectively. $\Delta k_{13} = 3k_1 - k_3$ and $\Delta k_{213} = 2k_2 - k_1 - k_3$ are wave-vector mismatch due to dispersion in the gaseous media. The refractive index of the gas can be calculated by using Sellmeier equation,

$$\begin{aligned}
n(\lambda) - 1 &= (n - 1)_{lines} + (n - 1)_{cont} \\
&= \frac{N_g r_e}{2\pi} \sum_i \frac{f_i}{\lambda_i^{-2} - \lambda^{-2}} + \frac{N_g}{2\pi^2} \int \frac{\sigma d\bar{\nu}_i}{\bar{\nu}_i^2 - \bar{\nu}^2} .
\end{aligned} \tag{9}$$

In Eq. (9), $N_g = P/k_B T$ is the gas density related to the pressure of the gas by ideal gas law N_g in which k_B is Planck constant; r_e is classical electron radius. The first term or $(n - 1)_{lines}$ refers to the contribution by discrete energy levels of the atom while the second term or $(n - 1)_{cont}$ is that by the continuum states. For the oscillator strengths f of argon, we used those listed in Ref. [8]. In addition, we take the photoionization cross-section from Ref. [9]. The intensity of TH signal, therefore, can be written as

$$\begin{aligned}
I_3(\varphi) &= \frac{cn_3}{8\pi} \left\langle \left| \hat{E}_{3I}(z, t) + \hat{E}_{3II}(z, t) \right|^2 \right\rangle = I_{3I} + I_{3II} + I_{3III} \\
&= \frac{576\pi^6 L^2 I_1}{c^2 n_3 n_1 \lambda_1^2} \left\{ \begin{aligned} &\frac{1}{n_1^2} \left(\chi_I^{(3)} \right)^2 I_1^2 \text{sinc}^2 \left(\frac{\Delta k_{13} L}{2} \right) \\ &+ \frac{9}{n_2^2} \left(\chi_{II}^{(3)} \right)^2 I_2^2 \text{sinc}^2 \left(\frac{\Delta k_{213} L}{2} \right) \\ &+ \frac{6}{n_1 n_2} \chi_I^{(3)} \chi_{II}^{(3)} I_1 I_2 \text{sinc} \left(\frac{\Delta k_{13} L}{2} \right) \text{sinc} \left(\frac{\Delta k_{213} L}{2} \right) \\ &\times \cos \left(\frac{\Delta k_{13} L}{2} - \frac{\Delta k_{213} L}{2} + 4\varphi_1 - 2\varphi_2 \right) \end{aligned} \right\} \tag{10}
\end{aligned}$$

In Eq. (10), the first, second and third term corresponds to THG, FWM and a cross-term due to the interference of the former two processes. For the sake of simplicity, we can set $\phi_1 = 0$. Therefore, $\Delta\phi = \phi_2 - 2\phi_1 = \phi_2$ is the relative phase between ω_1 and ω_2 . In media with normal dispersion, for example, the non-resonant excitation of room-temperature argon gas, the relative magnitude of the wave vectors is $k_1 < k_2 < k_3$. Accordingly, the phase mismatch, $|\Delta k_{13}| > |\Delta k_{123}| \approx 0$, is negligible. A sinusoidal dependence of the TH signal on the relative phase is thus expected. An example is shown in **Figure 4**. The pulse energy of the 1064 nm and 532 nm beams were 70 and 1 mJ, respectively. The pressure of the argon gas was 100 Torr. As the beams are slightly elliptical, we measured the TH signal in two transverse directions. The percentile errors in the X- and Y-directions are shown. Note that the TH signal is very weak if only the fundamental beam is used for excitation.

It was found that the TH signal can be enhanced by more than one order of magnitude with two-color excitation. In **Table 2**, we summarize the phase modulation and enhancement of the TH signal with two-color excitation for several ratios of fundamental and second-harmonic pulse energies. The fluctuations of the TH signal when the relative phase of the fundamental and second-harmonic beams is a constant is also shown.

We observed the enhancement of the TH signal is substantial for two-color excitation. Plasma emission was found to be visible to the naked eye in such cases. It is reasonable to assume that

Figure 4. Typical trace of the TH signal plotted as a function of the relative phase between the fundamental and second-harmonic beams. The system noise level corresponding to situation in which the slit of the monochrometer was closed is also shown.

Excitation Source	The modulation of phase or contrast (Normalized)	The fluctuation in TH power without phase delay (normalized)	Enhancement ratio (two-colour/one color)
(1) 1064 (70mJ/ pulse) +532 (1mJ/ pulse)	0.45	0.065	9.02~16.38
(2) 1064 (110mJ/ pulse) +532 (1mJ/ pulse)	0.6258	0.166	2.28~5.52
(3) 1064 (70mJ/ pulse) +532 (20mJ/ pulse)	0.3446	0.1457	13.41~20.58

Table 2. The phase modulation and enhancement of the TH signal with two-color excitation for several ratios of fundamental and second-harmonic pulse energies.

laser-induced ionization in the inert gases, for example, argon. With our experimental conditions, the ionization process is in the multiphoton ionization regime, which occurs when an atoms simultaneously absorbing several photons. The multiphoton ionization rate $w(\omega, F)$ can

be calculated by the Perelomov-Popov-Terent'ev (PPT) model, where F is the laser fluence. The rate is a function of the laser oscillation frequency and laser field strength. For the two-color case, we assume an effective frequency which is calculated from the power distribution of laser frequency to describe the influence of the two-color electric field on the ionization rate.

$$\omega_{eff} = \frac{\int_0^\infty \omega |E(\omega)|^2 d\omega}{\int_0^\infty |E(\omega)|^2 d\omega} \quad (11)$$

Besides, for our nanosecond pulse, there are several million cycles inside the pulse envelope for fundamental beam in the near infrared. The cycle-averaged ionization rate is thus used in this work [10]. That is,

$$\bar{w}_{PPT}(F_a) = \frac{1}{T_0} \int_0^{T_0} w_{PPT}(t) dt \quad (12)$$

or

$$\bar{w}_{PPT}(F_a) = \sqrt{\frac{2}{\pi}} \sqrt{\frac{3F_a}{2F_0}} w_{PPT}(F_a) \quad (13)$$

The ionization probability of the atoms by the laser pulse can be calculated by solving the rate equation

$$p = 1 - e^{-\int_{-\infty}^{\infty} w_{PPT}(t) dt}. \quad (14)$$

This allows us to calculate the plasma density in terms of the density of the neutral gas.

$$N_e = p \times N_g \quad (15)$$

The step-like behavior for the ionization probability as shown in **Figure 5** is caused by the increase of the effective frequency when the number of the second-harmonic photons increases. That is, there are new absorption processes occurring when the effective photon energy of the pulse reaches the threshold of the ionization process.

Now, we consider influence of the plasma on the third-harmonic signal. We assume that the third-order optical susceptibility is a sum of the susceptibilities for the neutral and ionized gas atoms.

$$\chi_{I,p}^{(3)} = \chi_{I,g}^{(3)} + \gamma_{I,p} N_e \quad (16)$$

$$\chi_{II,p}^{(3)} = \chi_{II,g}^{(3)} + \gamma_{II,p} N_e \quad (17)$$

In the above two equations, the ratios $\gamma_{I,p}$ and $\gamma_{II,p}$ are values determined by the experiment. Here, we assume $\gamma_{I,p} = \gamma_{II,p} = 4 \times 10^{-49}$ and $\chi_{I,g}^{(3)} = \chi_{II,g}^{(3)} = \chi_{II,Ar}^{(3)} = 3.8 \times 10^{-26} \text{ m}^2/\text{V}^2$ [11].

Figure 5. Ionization probability calculated by PPT model for argon at 100 Torr, excited by two-color field with fundamental pulse energies at 150 mJ and 200 mJ.

Additionally, we note that the refractive index of the media would also be changed when the plasma is generated. This can be calculated, in the first approximation, by using the Drude model.

$$n_{\omega,p} = n_{\omega,g} - \frac{N_e}{2N_c} \quad (18)$$

where $N_c = \epsilon_0 m_e \omega^2 / e^2$ is the critical plasma density when the laser and plasma frequencies are equal. The subscripts p and g represent plasma and neutral gas, respectively. For argon, $n_{\omega,g} = n_{\omega,Ar}$. The wave-vector mismatch becomes $\Delta k_{13,p} = 3k_{1,p} - k_{3,p}$ and $\Delta k_{213,p} = 2k_{2,p} - k_{1,p} - k_{3,p}$. The intensity of third-harmonic signal can then be calculated using Eq. (10). This is plotted as a function of the energy of the second-harmonic pulse for two values of the pulse energies for the fundamental beam in **Figure 6**. The experimental data are in good agreements with the simulated values using the above theoretical formulism.

The four-wave mixing process is dominant in the third-harmonic signal. For our experimental conditions, the THG component is approximately 10^{-4} that of the FWM process. In the low plasma density limit, the FWM term can be written as $I_{3II} \propto (\chi^{(3)}_{II,p})^2 \propto N_e^2$. This indicates that the enhancement of the third-harmonic signal is due to the plasma-enhanced susceptibility for the FWM process. On the other hand, when the plasma density is high enough, the wave-vector mismatch Δk becomes significant due to the plasma-induced refractive index change, which is linearly proportional to the plasma density.

Thus, in the high plasma density limit, the four-wave mixing term becomes $I_{3II} \propto \text{sinc}^2(\Delta k_{213,p} L/2)$. This is one of the reasons why the third-harmonic signal saturates at high plasma density. As a result, the TH signal is higher when the pulse energy of the

Figure 6. The comparison of the experimental and simulation results for two-color excited third-harmonic signal in argon as a function of the second-harmonic pulse energy.

fundamental output was lower. This is in good agreement with the experimental results for fundamental pulse energies of 150 and 200 mJ. However, the theoretically predicted threshold for plasma enhancement does not match that of the experimental data. This may be explained by the dependence of the threshold on the step-like enhanced ionization probability. The step-like behavior caused by new absorption processes becomes dominant when the effective photon energy (effective frequency) reaches the threshold of this process. However, in reality, there actually exist many quantum processes involving the absorption of several photons at frequencies of ω and 2ω . The different quantum processes have different ionization rates. When the power ratio of the two-color field is changed, the ionization probability of the different quantum process is also changed. It could be argued that the variation of ionization rate with the second-harmonic pulse energy is continuous rather than step-like when the plasma density increases. This in turn should shift the threshold pulse energy.

4. Laser-material processing with multi-color synthesized light field

Lately, high-energy laser beams have been increasingly used for processing and fabrication of material and devices. These include the fabrication of micro electro mechanical systems, optoelectronic components, biomedical micro fluid chips and silicon chip processing, electronic packages and drilling of circuit boards, to name just a few.

There are two kinds of mechanisms occurring during laser processing of materials: a photo-thermal one and a photo-chemical one. In the photo-thermal mechanism, laser beams with

high-power density are used as a thermal source which is focused on an object for a period of time. The energy absorbed on the surface of the object is transferred into the bulk of the object via thermal conduction. Thereafter, a part of the object is melted or vaporized by the deposited thermal energy. The laser spot is moved to another part of the work piece ready for further processing. In the photo-chemical mechanism, the bonding of molecules in the material to be processed is broken after absorption of one or more photons, which make electrons hop between energy levels and molecular bonds in the material can be broken as a result [12, 13].

In laser processing, the laser is chosen according to characteristics such as energy absorption, thermal diffusion and melting point of the material. For example, ablation is performed on various materials using lasers with appropriate wavelength. It is interesting, therefore, to investigate whether synthesized waveforms proposed and demonstrated in our work could be advantageous for laser processing.

Ablation of materials with multiple lasers, for example, lasers with dual colors were reported recently [14–18]. Incoherent or coherent summation of multi-color beams can be implemented. With incoherent summation of two femtosecond and nanosecond class pulsed lasers, an enhancement of volume of the vaporized material was observed by Théberge and Chin [14]. In this work, the free electrons and defect states induced by intense fs pulses were exploited by the ns pulses. In another work, Okoshi and Inoue [15] demonstrated that superimposed fs pulses at the fundamental (ω) and small fraction of the second-harmonics (2ω) output of the Ti: sapphire laser with the relative fluence ratio 1/39 was able to etch polyethylene (PE) much deeper and faster. They attributed the observe phenomena by the higher photon energy of 2ω pulses which can cut the chemical bonds of PE to form a modified layer of PE on the ablated surface. However, this article did not discuss about the temporal dynamics of the laser ablation process. On the other hand, the enhancement of absorption/reflection was observed in fused silica with coherent summation of dual-color pulses at zero delay [16]. This is because of defect states formation or free electron plasma generated in the material this way. For silicon, the ablation process was reported in the case of nanosecond and picosecond laser pulses where a small portion of the (2ω) beam can excite electrons into the conduction band [17]. For femtosecond pulses, this effect became insignificant because a sufficient population in the conduction band is created by multiphoton absorption in silicon. However, on the scale of carrier lifetime, all of the above-mentioned works consider relatively long time delays between the beams of two colors ($\sim$ picoseconds).

We note that tunable relative-phase control between the two dual-color exciting laser was applied in order to study the physical mechanism of intense-field photoionization in the gas phase [19–21]. Schumacher and Bucksbaum [19] reported that number of photoelectrons created in a regime that both multiphoton and tunneling ionization mechanisms are present is indeed dependent on the relative phase of the dual-colors. Later, Gao et al. [20] showed that the observed phase-dependence represents a quantum interference (QI) between the different channels corresponding to different number of photons involved. Recently, in comparison with monochromatic excitation, the threshold of plasma creation in the material to be ablated has been identified to be significantly reduced with the use of a ns infrared laser pulses and its second-harmonic one [21]. The observed phenomenon was attributed to the field-dependence

of the ionization cross section. In this work, we focus on the ablation study of metals under phase-controlled dual color ns pulses with the relative delay between the two color being less than one oscillation period.

Results of preliminary experiments on drilling of copper and stainless steel with the multi-color laser system used in this work (see **Figure 7**). The nonlinear optical crystals for harmonic generation are arranged in a cascaded layout. The crystals are KD*P type II for the second harmonic, ω_2 , KD*P type I for the third harmonic, ω_3 , BBO type I for the fourth harmonic, ω_4 , and BBO type I for the fifth harmonic, ω_5 , respectively. Therefore, the spectra of the five-color frequency components spans from near infrared (1064 nm) to the ultraviolet (213 nm). The cascaded harmonic generation setup was adopted to ensure that the second-order nonlinear optical process all occurred collinearly so that fundamental and harmonics overlapped spatially with each other. The amplitude and relative phase of each harmonics can be adjusted independently.

We studied two-color laser ablation of cooper and steel to demonstrate the feasibility of the approach. In the plane-wave approximation, the synthesized dual-color laser field can be written as,

$$E_{\text{Total}}(t) = E_{\omega}(t) \times \cos[\omega t] + E_{2\omega}(t) \times \cos[2\omega t + \theta], \quad (19)$$

where $E_{\omega}(t) = E_{0,\omega} \exp[(-2\ln 2)t^2/\tau_{\omega}^2]$ and $E_{2\omega}(t) = E_{0,2\omega} \exp[(-2\ln 2)t^2/\tau_{2\omega}^2]$ are the amplitudes of fundamental and second-harmonic optical fields with pulse durations, τ_{ω} , and $\tau_{2\omega}$, both assumed to be around 15 ns; where ω , and θ are the angular frequency and relative phase between two fields, respectively. Because the ns pulse duration we can neglect a group velocity mismatch when dual wavelength waveform propagates through a media. Therefore, the first and second-harmonic pulses are fully overlapped and the joint pulse has the same pulse duration around 15 ns. **Figure 8(a)** and **(b)** illustrate the simulated synthesized electric-field waveforms and the instantaneous intensities using Eq. (19) for the relative phases of $\theta = 0, \pi/2, \pi$, and $3\pi/2$, respectively, and $E_{2\omega}/E_{\omega} \approx 0.68$ (they are selected to match one of the experimental parameters corresponded to the intensity ratio $P_{2\omega}/P_{\omega} \approx 0.47$). It is clear that the waveform of combined ω and 2ω fields are very sensitive to the relative phase between the two. As shown in **Figure 8(a)**, the synthesized waveform E_{Total} has a symmetric shape for $\theta = 0$ and $\theta = \pi$. It exhibits, however, an asymmetric profile with enhanced electric field during one-half cycle of its oscillation for θ equal to $\pi/2$ and $3\pi/2$. In comparison **Figure 8(b)**, the instantaneous intensity of the two-color field for $\theta = 0$ and π are quite similar to the cases where $\theta = \pi/2$ and $3\pi/2$ except for a shift in the time domain, like for. In other words, the period of the change of instantaneous intensity can be considered to be π .

Figure 7. A schematic view showing a multi-color harmonic synthesized laser system for laser processing.

Figure 8. (a) Comparison of synthesized laser fields, and (b) instantaneous intensity of the fundamental wave (ω) and its second-harmonic (2ω) field with that of a ratio, $E_{2\omega}/E_{\omega} \approx 0.68$; shown are the cases of different relative phases $\theta = 0, \pi/2, \pi$, and $3\pi/2$, respectively.

During the experiment, we applied the dual color ns pulses with the same total energy (~ 100 mJ) to 150 μm thick copper and stainless steel foil. We adjusted the phase modulators only, so, varied the relative phase between the harmonics. Then we measured the time required to make a pass through hole in a foil and estimated the ablation rate. In **Figure 9(a)**, we have plotted diameters of holes drilled in copper sheets as a function of relative phases between the fundamental (ω_1) and second (ω_2) harmonics of the synthesizing laser. Pictures of the drilled holes are also presented. Similar results for stainless steel are shown in **Figure 9(b)**. Clearly, there is an optimal phase relationship between the two colors where higher instantaneous intensity causes higher ablation rate.

Figure 10(a)–(d) shows simulated results the peak strengths of the synthesized laser field with various relative phases ($\Delta\varphi = 0, 0.5\pi, \pi$, and 1.5π). As can be seen in **Figure 10(b)** and **(d)**, the synthesized laser field is expected to exhibit the higher peak strength at relative phases of $\Delta\varphi = 0.5\pi$, and 1.5π . Therefore, ablation is expected to be more efficient for these waveforms.

Figure 9. The diameters holes drilled in (a) copper, and (b) stainless steel by synthesized laser fields with different relative phases between the fundamental (ω_1) and second (ω_2) harmonics of the Q-switched laser. Pictures of drilled holes are also shown.

Figure 10. Numerical simulation of the peak strength of the laser waveform synthesized by two-color laser fields with various relative phases between the fundamental and the second harmonic (a) $\Delta\varphi = 0$, (b) $\Delta\varphi = \pi/2$, (c) $\Delta\varphi = \pi$, and (d) $\Delta\varphi = 3\pi/2$, respectively.

In another experiment, we fixed the exposure time at 10 s, varied the relative phase of the two-color beams and examined the ablated holes afterwards. In **Figure 11(a)**, we have plotted diameters of holes drilled in copper sheets as a function of relative phases between the fundamental (ω_1) and second harmonics (ω_2) of the synthesizing laser. Pictures of the drilled holes are also presented. Similar results for stainless steel are shown in **Figure 11(b)**. These data clearly show the dependence of ablation rate on the synthesized waveform, that is, relative phase of the fundamental (ω_1) and second harmonic (ω_2) of the single-frequency Nd:YAG laser.

5. Summary

As an application of the high-power laser system based on synthesized waveforms, we studied harmonic generation by three-color waveform synthesis in inert gas systems. In third-order nonlinear optics, the interaction between three-color beam and inert gases can be used to generate fourth to ninth harmonics of the laser fundamental output. For fourth-harmonic generation, there are three kinds of four-wave mixing processes: $\omega_4 = \omega_i + \omega_j + \omega_k$, $\omega_4 = \omega_i + \omega_j - \omega_k$, where $i, j, k = 1, 2, 3$. For fifth-harmonic generation, there are three possible processes:

Figure 11. Diameters of holes drilled in (a) copper, and (b) stainless steel by synthesized laser fields with different relative phases between the fundamental (ω_1) and second (ω_2) harmonics of the Q-switched laser. Pictures of drilled holes are also shown.

$\omega_5 = \omega_1 + \omega_2 + \omega_2$, $\omega_5 = \omega_1 + \omega_1 + \omega_3$, $\omega_5 = \omega_3 + \omega_3 - \omega_1$. For the sixth and seventh harmonic, there are two kinds of four-wave mixing processes, and so on. To illustrate, we present in detail the simulation results for fourth-harmonic generation using three-order nonlinear processes. It is shown that the fourth-harmonic signal varies with the phase of the fundamental beam.

Previously, we studied the influence of relative phases and intensities of the two-color pump on the third-order nonlinear frequency conversion process. It is shown that the third-harmonic (TH) signal oscillates periodically with the relative phases of the two-color driving laser fields due to the interference of TH signals from a direct third-harmonic-generation (THG) channel and a four-wave mixing (FWM) channel. In intense laser field, however, plasma can be generated through the ionization process. In the multiphoton ionization region, the plasma density was estimated by the Perelomov, Popov, and Terent'ev (PPT) model where the instantaneous laser field and frequency of laser are taken into account. Under the assumption that susceptibility and wave-vector mismatch depend on the plasma density, we show that plasma plays a significant role in the generated third-harmonic signal. The simulation results are in good agreement with the experiments.

Finally, we showed preliminary data indicating that the synthesized two-color laser fields are powerful in enhancing the conversion efficiency of HHG and VUV spectra. We also demonstrated phase-sensitive two-color ablation of copper and stainless steel. Our results show that hole drilling is more efficient with the use of optimized waveforms.

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