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LETTER

Coherent control of third-harmonic-generation by a waveform-controlled two-colour laser field

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Abstract
We investigate generation of the third harmonic (TH; $\lambda = 355$ nm) signal by two-colour excitation ($\lambda = 1064$ nm and its second harmonic, $\lambda = 532$ nm) in argon gas, with emphasis on the influence of relative phases and intensities of the two-colour pump on the third-order nonlinear frequency conversion process. Perturbative nonlinear optics predicts that the TH signal will oscillate periodically with the relative phases of the two-colour 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. For the first time, we show unequivocal experimental evidence of this effect. A modulation level as high as 0.35 is achieved by waveform control of the two-colour laser field. The modulation also offers a promising way to retrieve the relative phase value of the two-colour laser field.

(Some figures may appear in colour only in the online journal)

1. Introduction

Pulse shaping is an important method for coherent control of physical and chemical processes [1–3]. It is even now possible to control molecular modulation so as to modify the generation of ultraviolet (UV) sidebands in deuterium [4]. Following the development of attosecond photonics, the pulse shaping technique is no longer limited to control of the envelope shape of laser pulses. Scientists can manipulate the waveform of the electric field in the laser pulse envelope, as in [5–11]. This new technology has been dubbed lightwave electronics [12].

Waveform control of the electric field is essential for ultrafast nonlinear optical processes such as the generation of attosecond pulses by high-order harmonic generation (HHG). There have been many experimental and theoretical studies on approaches to increase the conversion efficiency and extend the cut-off limit of HHG. Popular schemes for excitation include the use of carrier-envelope phase (CEP) control of few-cycle pulses [13, 14], as well as the waveform synthesis of two-colour femtosecond pulses [15–21]. Surprisingly, there are few studies on the effects of coherent control in lower-order harmonic generation (LHG). The LHG technique is widely used for the generation of ultraviolet (UV) to vacuum ultraviolet (VUV) light. Such sources are essential in atomic and molecular spectroscopy, photochemistry and microfabrication. Earlier, Giammanco and co-workers [22–24] studied LHG by two-

colour excitation. Enhanced harmonic signals were observed for certain relative time delays between the exciting pulses. Modulation in the TH signal was recently reported by Xu *et al* [25] with a two-colour femtosecond laser field. The observed modulation depth was, however, quite small (∼0.06).

Typically, broadband mode-locked lasers were employed for waveform control of either HHG or LHG in the above cited works. From the point of view of Fourier-waveform synthesis, the ultimate waveform control scheme must have the ability to modulate both the amplitude and the phase of each frequency component. Chipperfield *et al* presented a proposal for optimizing the spectrum of HHG generated by a sawtooth-like waveform [26]. In this scheme, the exciting laser must have a coherent broadband spectrum with a bandwidth spanning more than two octaves. Because of the large number of modes present in a femtosecond pulse, it is difficult to control the amplitude and phase of each and every mode. As a result, synthesis of an arbitrary waveform, e.g. a sawtooth, is still beyond reach even though the mode spectrum of the laser can be close to two octaves [10, 11]. So far, just a few groups have used multi-colour single-frequency components spanning more than two octaves to synthesize arbitrary electric field waveforms. Notably, Shverdin *et al* [27] and Chan *et al* [5] used the frequency comb generated by high-order stimulated Raman scattering (HSRS) to synthesize sub-single-cycle and arbitrary waveforms. In our previous work [6, 7], we used the frequency comb formed by multiple cascaded harmonics of the same injection-seeded Q-switched Nd:YAG laser to synthesize the arbitrary waveform and sub-half-cycle pulses. Compared with the HSRS frequency comb, it is possible to generate higher peak power using the waveform synthesized by multiple cascaded harmonics. This feature is desirable for nonlinear optical processes, e.g. harmonic generation.

In this letter, we will show how waveform control by tuning the relative phase and amplitude of a two-colour laser field, consisting of the fundamental (ω_1) and second harmonic ($\omega_2 = 2\omega_1$) of a narrow-band pump laser, can affect third harmonic ($\omega_3 = 3\omega_1$) signal in argon. With two-colour excitation, the third harmonic signal can be modulated by the interference of direct third-harmonic generation (THG; $\omega_3 = \omega_1 + \omega_1 + \omega_1$) and four-wave mixing (FWM; $\omega_3 = \omega_2 + \omega_2 - \omega_1$) channels. As the relative phase between ω_1 and ω_2 varies, a sinusoidal modulation in output intensity at frequency ω_3 is expected and demonstrated. We will also show that the relative amplitude of the fundamental and second harmonic exciting light influences the contrast of the interference-induced modulation.

2. Basic principles of the third harmonic generated by two-colour excitation

We assume a plane wave propagating in the $+z$ direction. The applied fields can be represented as

$$\tilde{E}_i(z,t) = E_i e^{-i\omega_i t} + \text{c.c.} \tag{1}$$

with $E_i = A_i e^{i(k_i z+\phi_i)}$, $i = 1, 2$.

We consider the third-order nonlinear polarization induced by the two-colour laser pulses, which can be expressed as

$$\tilde{P}^{(3)}(z,t) = \varepsilon_0 \chi^{(3)} \tilde{E}^3(z,t) \tag{2}$$

with $\tilde{E}(z,t) = \tilde{E}_1(z,t) + \tilde{E}_2(z,t)$.

For the sake of simplicity, we set $\phi_1 = 0$. Therefore, $\Delta\phi = \phi_2 - 2\phi_1 = \phi_2$ is the relative phase between ω_1 and ω_2. As a source, the nonlinear polarization term will generate seven frequency components including the DC term, ω_1, $\omega_2 = 2\omega_1$, $\omega_3 = 3\omega_1$, $\omega_4 = 4\omega_1$, $\omega_5 = 5\omega_1$, and $\omega_6 = 6\omega_1$. We find that only the term with frequency ω_3 will exhibit modulation by varying the relative phase $\Delta\phi$. The nonlinear polarization for the frequency ω_3 can be written as

$$\tilde{P}_3^{(3)}(z,t) = \varepsilon_0(a\tilde{E}_1^3(z,t) + 3b\tilde{E}_2^2(z,t)\tilde{E}_1^*(z,t)) + \text{c.c.} \tag{3}$$

where $a = \chi^{(3)}(\omega_3;\omega_1,\omega_1,\omega_1)$ and $b = \chi^{(3)}(\omega_3;\omega_2,\omega_2,-\omega_1)$ denote the nonlinear optical susceptibilities for THG and FWM, respectively.

The electric field of the signal at frequency ω_3 can then be rewritten as

$$\begin{aligned}\tilde{E}_3(t) &= \varepsilon_0\left(a e^{-i3\omega_1 t}\int_{-L/2}^{L/2} A_1^3 e^{-i\Delta k_{13} z}dz + 3b e^{-i3\omega_1 t}\right.\\ &\quad\left.\times\int_{-L/2}^{L/2} A_2^2 A_1^* e^{-i(\Delta k_{123} z+2\Delta\phi)}dz\right) + \text{c.c.}\\ &= \varepsilon_0\left(aA_1^3\sin c\left(\frac{\Delta k_{13}L}{2}\right)\sin(3\omega_1 t) + 3bA_2^2A_1^*\right.\\ &\quad\left.\times\sin c\left(\frac{\Delta k_{123}L}{2}\right)\sin(3\omega_1 t+2\Delta\phi)\right)\end{aligned} \tag{4}$$

where $\Delta k_{13} = 3k_1 - k_3$, $\Delta k_{123} = 2k_2 - k_1 - k_3$ denote the phase mismatch, and L stands for the spatial extent of the laser pulse.

The intensity of the signal at frequency ω_3, as a function of relative phase $\Delta\phi$, can be written as

$$\begin{aligned}I_3(\Delta\phi) &\propto \int \tilde{E}_3^2(t)dt\\ &= a^2A_1^6\sin c^2\left(\frac{\Delta k_{13}L}{2}\right)\\ &\quad + 9b^2A_2^4A_1^2\sin c^2\left(\frac{\Delta k_{123}L}{2}\right)\\ &\quad + 6abA_1^4A_2^2\sin c\left(\frac{\Delta k_{13}L}{2}\right)\\ &\quad\times\sin c\left(\frac{\Delta k_{123}L}{2}\right)\cos(2\Delta\phi).\end{aligned} \tag{5}$$

The first and second terms are contributed by direct THG and FWM processes, respectively. The last term is the interference term controlled by the relative phase between the fundamental and second harmonic laser fields. The relative amplitude of the two will affect the contrast of interference modulation.

3. Relative phase measurement for waveform synthesis

The electric field waveform of a commensurate frequency comb can be expressed by $E(t) = \sum_n A_n \cos(n\omega t + \phi_n)$. The waveform is determined by the amplitudes, A_n, and the phases, ϕ_n, of the component waves. The amplitudes are easy to determine by measuring the energy of each frequency component. Measurement of the phase ϕ_n is more involved. In this letter, we use the interference of THG and FWM to determine the relative phase of fundamental and second harmonic beams.

The electric field waveform is invariant from a time shift of $\phi_1/\omega(t \to t - \phi_1/\omega)$. With such a time shift, the electric field becomes $E(t) = \sum_n A_n \cos(n\omega(t-\phi_1/\omega)+\phi_n) = \sum_n A_n \cos(n\omega t + \phi_n - n\phi_1)$, in which the phases are $\phi_1 \to 0, \phi_n \to \phi_n - n\phi_1$. This implies that, by measuring $n-1$ relative phases, we can fully determine the waveform. As shown in section 2, the relative phase $\Delta\phi$ can be deduced from modulation of the TH signal in an isotropic medium excited by a two-colour field.

For multi-colour cases, we can use the interference of FWM signals of higher harmonic frequency components to determine their relative phase, e.g. $\phi_n - \phi_{n-1} = \phi_{n-1} - \phi_{n-2} = \cdots = \phi_2 - \phi_1$. This method was employed by Chen *et al* [8] for adjusting the relative phase between Raman sidebands and reaches the condition of constant CEP. The absolute value of the relative phase of $\phi_2 - \phi_1$, however, cannot be determined. In comparison, the modulation of third harmonic intensity corresponds to the value of this relative phase (see equation (5)). Therefore, by conducting both experiments, i.e. the interference of FWMs of multi-colour fields and the modulation due to interference of FWM and THG for the fundamental and second harmonic beams, we can determine the relative phase and the synthesized electric field waveform.

4. The two-colour experiment

A schematic of the experimental setup is shown in figure 1. The two-colour laser system is a subset of our previously reported multi-colour laser system [6, 7]. The fundamental laser pulse is from an injection-seeded Q-switched Nd:YAG laser (Spectra Physic GCR-Pro 290). It can produce intense 1064 nm pulses with a pulse duration of 10 ns (full width at half maximum) and a transform-limited linewidth of $<0.003\ \mathrm{cm}^{-1}$. 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 and their relative phase is a constant.

We can adjust the relative phase and amplitudes of these two-colour laser fields independently. The fundamental and second harmonic beams are first dispersed by an equilateral prism. Next, the two angularly separated beams are made parallel to each other after passing through a larger equilateral prism. There are two amplitude modulators (AM) built by

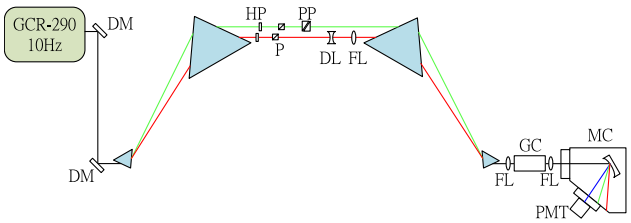

Figure 1. Experiment setup for the two-colour THG experiment: DM, dichroic mirror; HP, half-wave plate; P, polarizer; PP, prism pair; DL, defocusing lens; FL, focusing lens; GC, gas chamber; MC, monochromator; PMT, photomultiplier tube.

combining a half-wave plate and a polarizer which is used to modulate the energies of fundamental and second harmonic pulses, respectively. A phase modulator (PM) for second harmonic pulses is employed to adjust the relative phase, $\Delta\phi$. The PM is made by a pair of right-triangle prisms. By sliding the relative position of the two prisms along their hypotenuse, we can change the effective path length travelled by the second harmonic pulses. It will vary the relative phase, but will not shift the beam alignment. The separated fundamental and second harmonic beams will be recombined and collimated with another pair of prisms set out symmetrically to the first pair. This setup of amplitude and phase modulation is similar to the '4f-system' widely used in image processing and pulse shaping. We then focus the collinearly propagating phase-controlled two-colour laser pulses into the centre of a gas chamber with a simple lens ($f = 10$ cm). The laser spot size is around 20 μm at focus. For overlapping the foci of the fundamental and second harmonic beams, we set up a lens system that consists of a defocusing lens and a focusing lens just after the amplitude modulator for the fundamental beam. By adjusting the relative distance of these two lenses, we can control the relative position and overlap of the focal points of these two-colour laser pulses. The gas chamber is filled with argon gas. The gas pressure is around 100 Torr. After the exit window of the gas chamber, we used another lens ($f = 15$ cm) to focus the TH signal into the entrance slit of monochromator (VM-502, Acton Research). The two-colour laser field and the signal at ω_3 are dispersed by this monochromator. Finally, the TH signal is detected by a photomultiplier tube (R11568, Hamamatsu).

5. Results and discussions

Equation (5) predicts that the relative amplitude and the relative phase between the fundamental and second harmonic laser pulses will influence the intensity of the TH signal. Figures 2(a)–(c) show the experimental and theoretically simulated results. The fundamental and second harmonic pulse energies for the three different cases are (a) 70 and 1 mJ, (b) 110 and 1 mJ and (c) 70 and 20 mJ, respectively. The relative phase $\Delta\phi$ was varied over a range of about 4π. For comparison, TH signals by the fundamental pulses only are also shown. The solid curve in figures 2(a)–(c) is the theoretically predicted sinusoidal variation of the TH signal by two-colour excitation. The experimentally measured TH

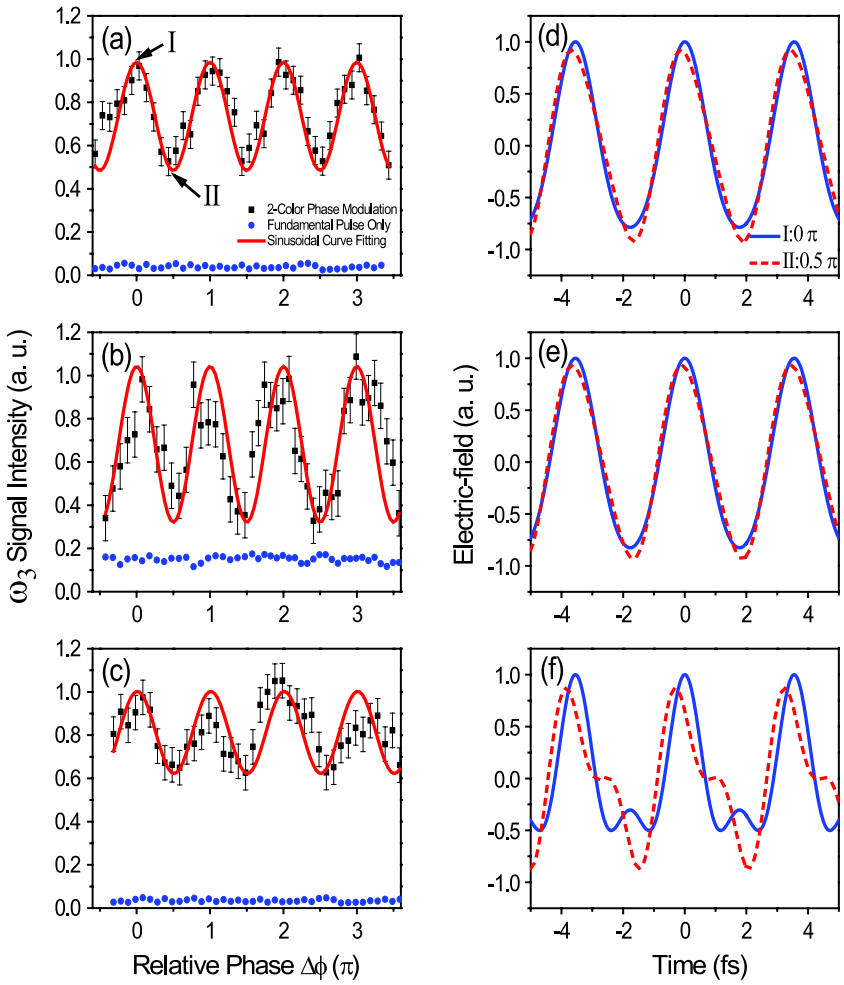

Figure 2. The TH signal is measured as a function of the relative phase, $\Delta\phi$. The ratio of pulse energies for the fundamental and second harmonic are: (a) 70 mJ:1 mJ, (b) 110 mJ:1 mJ, (c) 70 mJ:20 mJ, respectively. The solid squares are the TH signals generated by two-colour excitation. The discs are the TH signal generated by the fundamental pulse only. The solid curve is the theoretically predicted sinusoidal modulation of the TH signal by two-colour excitation. The deviations are: (a) 0.06, (b) 0.1, (c) 0.08, respectively. (d)–(f) The electrical-field waveforms of the two-colour exciting laser for the corresponding data in (a)–(c), respectively. The solid curve shows the two-colour electric field waveform when the relative phase is 0π (I). The dashed curve shows the two-colour electric field waveform when the relative phase is 0.5π (II).

signals all show a modulation with a period of 0.89 fs or a phase change of π. These data have been normalized to the maxima of the TH signals.

When the frequencies of laser fields are far off any resonances of argon, magnitudes of the nonlinear susceptibilities for THG and FWM will be comparable. That means $a \approx b$. Equation (5) can then be rewritten as

$$\begin{aligned} I_3(\Delta\phi) \propto A_1^6 \sin c^2\left(\frac{\Delta k_{13}L}{2}\right) + 9A_2^4A_1^2 \sin c^2\left(\frac{\Delta k_{123}L}{2}\right) \\ + 6A_1^4A_2^2 \sin c\left(\frac{\Delta k_{13}L}{2}\right) \\ \times \sin c\left(\frac{\Delta k_{123}L}{2}\right)\cos(2\Delta\phi) \\ = \alpha^2A_1^6 + 9\beta^2A_2^4A_1^2 + 6\alpha\beta A_1^4A_2^2\cos(2\Delta\phi) \end{aligned} \quad (6)$$

where $\alpha = \sin c\left(\frac{\Delta k_{13}L}{2}\right)$, $\beta = \sin c\left(\frac{\Delta k_{123}L}{2}\right)$.

The intensity of the TH signal is thus influenced by three factors: relative phase, $\Delta\phi$; relative amplitude, A_1/A_2; and the phase mismatch ratio of Δk or (α/β) of the fundamental and second harmonic light pulses.

From equation (6), the phase mismatch term, Δk, for the direct THG process is $\Delta k_{13} = 3k_1 - k_3$. The corresponding one for FWM is $\Delta k_{123} = 2k_2 - k_1 - k_3$. The phase mismatch term for the interference of THG and FWM processes depends on both Δk_{13} and Δk_{123}. In media with normal dispersion, e.g. the non-resonant excitation of room-temperature argon gas in our experiments, the relative magnitude of the wave vectors is $k_1 < k_2 < k_3$. Accordingly, we expect $|\Delta k_{13}| > |\Delta k_{123}| \approx 0$ and $\alpha < \beta$. Indeed, by substituting the amplitude ratios and relative phases of the two-colour exciting fields for data shown in figures 2(a) and (b) in equation (6), we find that the phase mismatch ratio (α/β) is around 8×10^{-3} for the two cases. This also explains why the conversion efficiency of the TH signal by FWM is higher than that by THG. The phase

matching condition can be readily controlled by varying the gas density and/or the focusing beam configuration [28].

With a fixed phase mismatch ratio, (α/β), the modulation of the TH signal can be adjusted by altering the relative amplitude (square root of intensity) between A_1 and A_2 and/or the relative phase, $\Delta\phi$. For the data shown in figures 2(a) and (b), the input pulse energy of the second harmonic is fixed at 1 mJ/pulse, while the fundamental pulse energy is increased from 70 to 110 mJ/pulse. The effective ratios of relative amplitude of the fields are 1/0.12 and 1/0.095, respectively. The experimental results show that the modulation contrast of the TH signals increases from 0.25 to 0.35. In order to achieve a high-contrast modulation of the TH signal, the control of the relative amplitude is clearly important.

In another experiment, when the input pulse energy of the fundamental is set at 70 mJ/pulse while the second harmonic pulse energy is increased from 1 to 20 mJ/pulse (see figures 2(a) and (c)). The ratios of relative amplitude of the two cases are 1/0.12 and 1/0.53, respectively. We find that the modulation of the TH signal decreases from 0.25 to 0.2. If we use the same ratio of phase mismatch (α/β) as in the cases of figures 2(a) and (b), the modulation contrast should reduce to 0.02 in figure 2(c). This is a difference of about one order of magnitude for modulation. The total energy of two-colour pulses for the data in figure 2(c) is 90 mJ, which is smaller than that for figure 2(b), i.e. 111 mJ. For the former, we observed apparent plasma fluorescence emitted near the focus point in the gas chamber. That is, an increase in the second harmonic pulse energy is more effective in inducing plasma generation than corresponding increases of the fundamental light. The laser-induced plasma not only induces a more intense TH signal but also affects the modulation of the interference channel. The plasma effect should be an important issue when the pulse energy of the driving laser increases, such that the nonlinear optical interaction progresses from the perturbative to the non-perturbative regime. Nonetheless, as shown in figure 2(c), the same signal modulation behaviour means that we still can retrieve the relative phase and obtain the waveform shown in figure 2(f). The change of modulation depth is currently under investigation.

The agreement between equation (6) and the experimental data confirms that modulation of the TH signal depends on the relative phase. In reverse, the modulation can be used to retrieve the value of relative phase. Together with measured amplitudes of the optical fields of the two-colour pump beams, we can then determine the synthesized waveforms corresponding to various modulation positions in figures 2(a)–(c). Figures 2(d)–(f) show the electric field waveforms synthesized by the two-colour pump corresponding to the maxima and minima of the TH signal in figures 2(a)–(c). The solid curves correspond to a relative phase of zero (maxima of TH signals). The dashed curves are for the case whereby the relative phase is equal to 0.5π (minima of the TH signal in figures 2(a)–(c)).

For data in figures 2(d) and (e), the amplitude ratios of fundamental to second harmonic are as large as 1/0.12 and 1/0.095, respectively. As a result, the differences between the dashed and solid curves are small. If the ratio is nearly one, the differences will be considerably larger. Although the difference is small, the modulation depth is large. That means the modulation of the interference signal is very sensitive to the relative phase. The waveform retrieved in figure 2(f) represents the electric field waveform measured in figure 2(c), corresponding to the case of zero and 0.5π of relative phase between the two-colour fields. For this experiment, there are ions present during the experiment. The TH signal was much higher than that predicted by the simple theory used in this letter. To explain the enhancement, we do need a more elaborate theoretical background. Nonetheless, the data in figure 2(c) show as large a modulation depth as in figures 2(a) and (b) when we scan the phase modulator. From the modulation signal, we can reliably retrieve the relative phase and deduce the waveform as shown in figure 2(f).

The variations of TH signals in figures 2(a)–(c) are 0.06, 0.1 and 0.08 in arbitrary units, respectively. These include all the uncertainties of amplitudes and phases of the driving laser fields as well as the fluctuation in generation of the third-order nonlinear signals and the data acquiring system. Therefore, the uncertainty of synthesized electric field waveform should be smaller than 6%.

The absolute values of relative phases of the component waves are essential for waveform synthesis. We have shown that the modulation of the TH signal corresponds to the relative phase between the fundamental and second harmonic of the two-colour pumping laser field. The present work thus suggests a promising way for *in situ* determination of the relative phases in multi-colour-pumped HHG. Previously, we used second-order nonlinear processes from multiple BBO crystals to retrieve the relative phases of five-colour fields and then determine the synthesized waveform [6]. The system is somewhat elaborate, and crystals are prone to damage in strong laser fields. Further, HHG is almost exclusively conducted in rare gases. By measuring *in situ* the interference of FWMs of multi-colour fields in the HHG chamber, together with the modulation due to interference of FWM and THG for the fundamental and second harmonic beams, we can determine unequivocally the relative phase and the synthesized electric field waveform.

It is interesting to consider the effect of the bandwidth of each colour in the two-colour laser fields on the TH signal. With two-colour femtosecond laser pulses, Xu *et al* [25] observed a modulation of about 0.06 for the TH signal. In our experiments, the modulation is as high as 0.35. This can be explained by noting that the modulation depth depends on the pulse duration. For broadband excitation, an additional factor, $\exp(-4t^2/3\tau_\omega^2)$, should be multiplied with the FWM intensity and interference terms of equation (6). In this factor, the parameter t $(=\Delta\phi/\omega_2)$ is the relative time delay of these two-colour pulses and τ_ω is the pulse duration. For a broadband femtosecond laser field, the duration of τ_ω is small and the exponential term will become small. As a result, the modulation effect is not so apparent. For a two-colour narrow-band laser, such as we employed, the pulse duration is much longer than the time delay t and the exponential term is close to unity. This explains the more prominent modulation effect.

6. Conclusion

We have investigated the generation of TH signal (λ = 355 nm) by two-colour excitation (1064 and 532 nm) in argon gas, with emphasis on the influence of relative phases and amplitudes of the two-colour fields on the third-order nonlinear frequency conversion process. Using perturbative nonlinear optics, we predict that the TH signal will show sinusoidal modulation as the relative phases of the two-colour driving laser fields are varied. This is due to the interference from a direct THG channel and a four-wave mixing (FWM) channel. For the first time, we show unequivocal experimental evidence of this effect. A modulation depth as high as 0.35 has been observed. Further, plasma induced by the two-colour laser field is shown to have a significant effect on generation of the harmonic signal [24, 29–31]. The present work thus also suggests a promising way for *in situ* determination of the relative phases in multi-colour-pumped HHG. By measuring *in situ* the interference of FWMs of multi-colour fields in the HHG chamber, together with the modulation due to interference of FWM and THG for the fundamental and second harmonic beams, we can determine unequivocally the relative phase and the synthesized electric field waveform. This is especially advantageous for experiments using a multi-colour quasi-single-frequency laser source, because measurement of the relative phase by interference modulation is direct and does not need any iterative algorithm like the FROG technique [32]. Finally, the present experiment suggests that waveform synthesis in the VUV spectral range by higher harmonic generation using waveform-controlled multi-colour quasi-single-frequency laser fields is feasible. Using our five-colour quasi-single-frequency Nd:YAG laser system [6], the same theoretical derivation predicts generation of controllable VUV radiation up to the 15th harmonic of the fundamental. Such a waveform synthesized VUV waveform is very useful for the coherent control of atomic and molecular dynamics.

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