Third and fifth harmonic generation by tightly focused femtosecond pulses at 2.2 μm wavelength in air

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Abstract: We report experiments on the generation of third and fifth harmonics of millijoule-level, tightly focused, femtosecond laser pulses at 2.2 μm wavelength in air. The measured ratio of yields of the third and fifth harmonics in our setup is found equal to $2 \cdot 10^{-4}$. This result contradicts the recent suggestion that the Kerr effect in air saturates and changes sign in ultra-intense optical fields.

OCIS codes: (190.2620) Harmonic generation and mixing; (190.3270) Kerr effect; (320.7110) Ultrafast nonlinear optics.

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1. Introduction

Linear and nearly instantaneous dependence of the refractive index of transparent media on the intensity of the optical field is the essence of the electronic Kerr effect, which is the cornerstone of nonlinear optics. In a recent publication, it has been suggested that the notion of the instantaneous electronic Kerr effect needs to be generalized [1, 2]. Measurements of the transient birefringence induced in various gases by ultraintense and ultrashort optical pulses have been interpreted as the saturation and sign reversal of the nonlinear refractive index, that occurs prior to the photoionization of the medium, at the optical intensity on the order of several tens of TW/cm^2. The common linear dependence of the index of refraction on the intensity of the optical field has been amended by the inclusion of new, non-perturbatively large terms proportional to second and higher powers of intensity.

The field of femtosecond laser filamentation is probably the one that would be affected the most, should the above suggestion be proven valid. For many years, it was believed that the intrinsically unstable balance between diffraction and self-focusing in ultraintense laser beams undergoing filamentation in gaseous media is stabilized by the weak defocusing action of plasma generated on the beam axis via multi-photon ionization [3–5]. The higher-order Kerr terms, if they did exist, could provide for an alternative stabilization mechanism [6]. The filament propagation would be much longer ranged, as the losses to ionization would be reduced compared to the established filamentation scenario based on plasma defocusing.

Effects associated with weak perturbative saturation of the electronic Kerr response have been considered in the context of laser filamentation previously [7–9]. In extremely intense optical fields above the onset of significant ionization, the Kerr effect can saturate simply due to the depletion of the population of neutral molecules in the gas [10]. Such intensities cannot

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be reached in femtosecond laser filaments under normal conditions because of the intensity clamping in filaments [11]. The suggestion that the Kerr effect may reverse sign and cause, among other things, beam self-defocusing, were too radical a departure from the established framework of laser filamentation and nonlinear optics in general. Almost immediately after the publication of [1,2], the results reported in that paper became the subject of intense debates that are still on-going at the time of this writing.

We will refer to the claim put forth in [1,2] as the Higher-Order Kerr Effect (HOKE) theory. Several independent experimental tests of HOKE in the context of laser filamentation have demonstrated that even if the higher-order Kerr terms do exist, they are not operative in common laser filaments in gases [12–14]. One experiment that contradicts that conclusion and supports HOKE has also been reported [15]. The question of where the non-perturbatively large HOKE terms exist, possibly masked by plasma effects, still remains open. It has been argued that the sign reversal of the Kerr effect may occur in a transient regime, while neutral molecules in the gas are undergoing multi-photon ionization [16, 17]. By direct numerical integration of the Schrödinger equation, it has been shown that the sign reversal of the polarization response of an isolated silver atom to an ultraintense laser field is completely dominated by plasma contribution and not by higher-order Kerr terms, as HOKE alleges [18].

Very recent experiments on direct profiling of the cross-phase modulation response in a thin gas target illuminated by an ultraintense and ultrashort laser pulse revealed no evidence of even a transient saturation of the Kerr effect, in complete contradiction with HOKE [19]. A plausible explanation of the original experiment [1, 2] in the framework of the conventional nonlinear optics has been also recently suggested [20].

An alternative test of HOKE has been proposed in [21]. The test is based on the dramatic effect that the inclusion of the HOKE terms would have on the efficiency of third and fifth harmonic generation by a femtosecond laser pulse with peak intensity above the alleged turnover point for the higher-order Kerr effect. As has been numerically shown in [21] for tightly focused femtosecond pulses at 1.3 μm wavelength, the ratio of the yields of the fifth and third harmonics generated by such pulses is a growing function of the pulse intensity. This ratio saturates at a value on the order of one if the HOKE terms are included in the model, and it remains very small ($\sim 10^{-4}$) when these terms are omitted.

The above qualitative conclusion is valid in a wide range of pump wavelengths, as the HOKE indices have been shown to depend very weakly on wavelength above 800 nm [22]. The weak dependence on wavelength is well known for the conventional n_2 index [23, 24]. There is no reason to expect the alleged higher-order Kerr terms to have qualitatively different wavelength dependence, as they are assumed to result from virtual transitions between bound states of the molecules in the medium, just like the n_2 term. The alleged turnover intensity at which the Kerr effect would turn to zero, provided that HOKE was correct, has been shown to be essentially independent on wavelength above 800 nm and, for air, equal to 35 TW/cm^2 [25].

The dramatic difference between harmonic yields in the two models is the consequence of the fact that in the established approach, the fifth harmonic generation is a cascade process involving the nonlinear mixing between the already generated third harmonic and the leftover pump, while in the new theory, the fifth harmonic is generated directly from the fundamental through the higher-order Kerr response.

In this paper, we report experimental results on the realization of the above proposal [21]. Our results show no evidence of the HOKE response.

We note that an experiment on the generation of third and fifth harmonics in an Argon cell using intense pump pulses at 800 nm wavelength has been reported much before the HOKE controversy had even emerged [26]. The results of that work have been recently used to argue in favor of the validity of HOKE [27]. In our view, [26] is not straightforwardly applicable to the

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resolution of the HOKE controversy for the following reasons. The generated fifth harmonic had a wavelength of 162 nm, which is in vacuum UV, thus making its reliable characterization difficult. Under the experimental conditions of [26], the predictions of the established and HOKE theories on the anticipated ratio of yields of the third and fifth harmonics differed by only about one order of magnitude [27], which is a quantitative, not qualitative difference. In order to reliably discriminate between the two theories in such a situation, the absolute energies of the generated harmonics had to be accurately measured. However, the accuracy of even approximately estimating the harmonic yields in [26] was severely impaired by the presence of as many as four apertures that allowed for the implementation of differential pumping in the setup. These apertures resulted in large and not easily quantifiable losses for the generated harmonics.

The experiment reported here is dedicated to the test of the HOKE hypothesis. It was designed to be free from the complications inherent to the setup used in [26]. In our case, third and fifth harmonics are generated using millijoule-level femtosecond laser pulses at 2.2 μm wavelength. Both third and fifth harmonics of the pump wavelength fall into the near UV – visible spectral range, making the precise and independent characterization of all optics used to route the harmonics straightforward and the harmonics themselves – easily measurable. The turnover intensity at 2.2μm wavelength, in the HOKE model, is about 35 TW/cm^2 [22]. The estimated intensity of the pump beam in the interaction zone of our setup is between 850 and 1,260 TW/cm^2, 24 to 36 times larger than that. Under these conditions, the ratio of the third to fifth harmonic yields, according to HOKE, should approach one.

The delayed (Raman) nonlinearity in the medium may contribute to the propagation dynamics in our experiment. However, the delayed response is too slow to have a significant influence on the yields of the generated third and fifth harmonics and thus cannot affect our conclusions.

We emphasize that our test is qualitative in nature. The four-orders of magnitude difference between the predictions of the established and HOKE theories, which persists across a wide wavelength range, makes the outcome of our test largely insensitive to the uncertainties in the material parameters and experimental conditions.

2. Experimental setup and results

Our experimental setup is schematically shown in Fig. 1. Femtosecond pulses at 2.2 μm center wavelength are produced by a commercial high-energy Optical Parametric Amplifier (OPA, Palitra-HE model by Quantronix Corporation). The OPA is pumped by 40 fs-long, 20 mJ pulses at 800 nm wavelength generated by an ultrafast Ti:Sapphire amplifier chain operating at a 10 Hz pulse repetition frequency. The OPA is tuned to produce signal and idler pulses at 1.26 μm and 2.20 μm center wavelengths, respectively. The output beam from the OPA has a diameter of about 10 mm. Both signal and idler pulses have energies of about 1.5 mJ. The duration of the pulses is estimated by the OPA manufacturer at between 40 and 60 fs [28].

The co-propagating signal and idler beams are focused by a lens with 20 cm focal length. Immediately after the lens, the 1.26 μm signal is reflected by a dichroic beamsplitter operated at near normal incidence. This dichroic (CVI – Melles Griot Corporation, part number BBDS), is a dielectric low-pass filter. At normal incidence, it has a transmission cutoff at the wavelength of about 1.3 μm. The idler pulses at 2.2 μm are transmitted by this mirror with about 90% efficiency, while the transmission for the signal beam at 1.26 μm is less than 0.1%. For all wavelengths below 1 μm, transmission is less than 0.05%.

The energy of the focused 2.2 μm idler pulse, after the dichroic, is measured at (1.25 ± 0.05) mJ. Note that the dichroic is oriented with its substrate facing the incident beam. Thus visible and UV light possibly generated inside the substrate is reflected by the coating, and only the 2.2 μm idler pulses are passed into the interaction zone.

The length of the interaction zone in our setup is about 5 mm, which is much shorter

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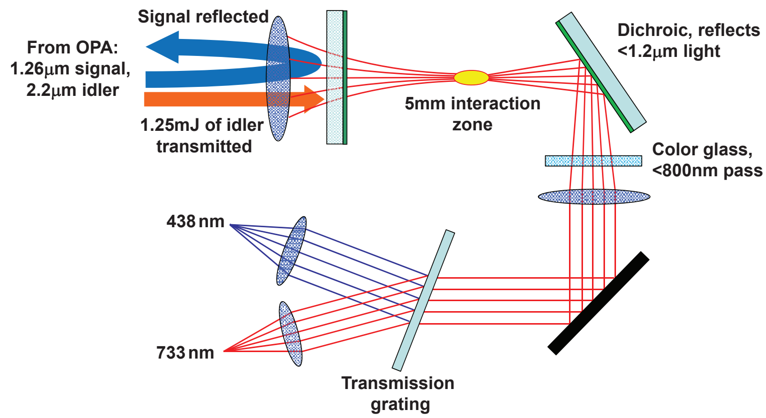

Fig. 1. Experimental setup.

than the phasematching lengths between the pump and both harmonics. Indeed, by extrapolating the available NIST data for the refractive index of air at 20°C temperature and 20% relative humidity [29], the value of the index at the pump wavelength can be estimated as $n(2.2\,\mu\mathrm{m}) = 1 + 2.6810 \cdot 10^{-4}$. The values of the index at the wavelengths of the harmonics are $n(733\,\mathrm{nm}) = 1 + 2.7064 \cdot 10^{-4}$ and $n(438\,\mathrm{nm}) = 1 + 2.7595 \cdot 10^{-4}$. These values translate into the estimated phasematching lengths of 14.4 cm and 2.8 cm between the pump and the third and fifth harmonics, respectively.

The third and fifth harmonics of the pump are reflected by another dichroic mirror, which is identical to the one used to block the signal pulse at 1.26 μm from the OPA. The second dichroic is operated with the coating facing the incident beam and at a 45° angle of incidence. This mirror efficiently reflects the generated harmonics, but blocks over 90% of the 2.2 μm pump. The residual pump is further attenuated by about five orders of magnitude by a 3 mm – thick color-glass filter plate (KG5 filter by Schott Glass) placed after the dichroic.

After being approximately collimated with another lens, the signals at third and fifth harmonics are spatially separated by a 300 lines per millimeter transmissive diffraction grating. The spectra of the two harmonics are recorded by a fiber-coupled spectrometer (model USB4000 by Ocean Optics Corporation), with the aid of two additional lenses that separately focus the harmonic beams after they are separated by the grating. These spectra are shown in Fig. 2. They are centered at 733 nm and 438 nm, very close to one-third and one-fifth of the pump wavelength of 2.20 μm.

The emission bandwidths of the detected third and fifth harmonics are about 20 nm and 11 nm, respectively. Assuming that the harmonic pulses are both Gaussian and close to transform limit, the durations of the two pulses can be estimated as 40 fs and 25 fs. The ratio of these pulse durations is in the ballpark of $\sqrt{5/3}$, which would be expected in the case of the perfectly phase-matched harmonic generation with non-depleted pump.

The energies of the two harmonic pulses are measured by dedicated silicon detectors that were calibrated against a pyroelectric detector, using reference laser pulses at 800 nm wavelength. Losses that the generated harmonics experience on all optics used in the setup were independently verified by using a broadband UV-visible light source and a calibrated optical spectrum analyzer. Losses on propagation through the laboratory air on the scale of the experimental setup were completely negligible for all three wavelengths involved [30]. Accounting for all the losses, the energies of the third and fifth harmonic pulses immediately after the interaction zone are found equal to $(1.9 \pm 0.2)\,\mu$J and (0.40 ± 0.07) nJ, respectively. These pulse

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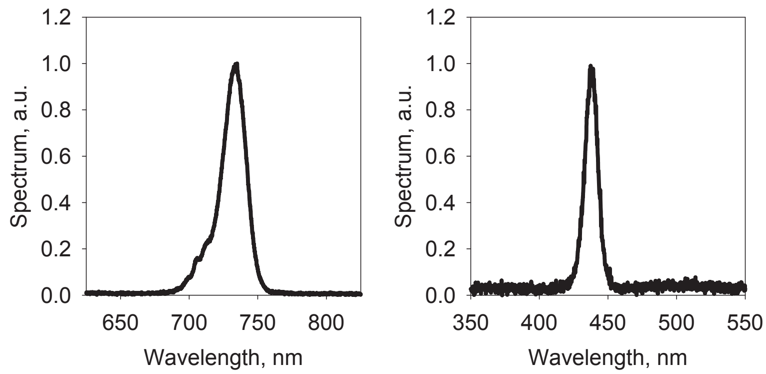

Fig. 2. Normalized spectra of the generated third and fifth harmonics on linear scale.

energies correspond to yields of $(1.50 \pm 0.15) \cdot 10^{-3}$ for third harmonic and $(3.0 \pm 0.5) \cdot 10^{-7}$ for fifth harmonic, with respect to the energy of the pump pulse at 2.2 μm wavelength in the interaction zone. The measured uncertainty of the harmonic yields is consistent with the level of the pulse-to-pulse energy fluctuations of the 2.2 μm pump. The ratio of yields for the two harmonics is $2 \cdot 10^{-4}$. Both the absolute conversion efficiencies and their ratio are close to the values obtained from the model based on the conventional Kerr effect [21].

No systematic study of the dependence of harmonic yields on the pump wavelength or pump pulse energy was conducted. We verified that reducing the pump power through the insertion of thin glass plates into the pump beam resulted in significant reduction of the yields.

To verify that the generated harmonics were in fact originating from the tightly focused interaction zone and not from any optics in the setup, we substituted the focusing lens with a glass plate with the same thickness as the lens and properly adjusted the collimation after the interaction zone. The intensities of both third and fifth harmonics of the pump fell below the detectability limit. By introducing obstructions into the harmonic beams near the edges of various optical elements used in our setup, we verified that the generated harmonics were not clipped or apertured anywhere on propagation from the interaction zone to the detectors.

3. Conclusion

To conclude, we have generated third and fifth harmonics of an ultraintense femtosecond optical pulses at 2.2 μm wavelength in ambient air. The ratio of yields of the two harmonics provides for a qualitative test of the recently proposed higher-order Kerr effect (HOKE) theory. Our results support the established paradigm of nonlinear optics that is based on linear and instantaneous dependence of the refractive index of transparent media on the optical intensity.

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