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Enhanced two-color high-harmonic generation achieved by adding an extra gas medium

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Abstract The high-harmonic generation in a two-color laser field, consisting of a 30-fs Ti:Sapphire laser pulse and its second harmonic, could be enhanced by installing an extra gas medium of Ar in front of the harmonic generation medium of He. The effects of the extra medium were examined by analyzing harmonic spectra taken under different experimental conditions. The observed characteristics of enhanced harmonic generation were explained in terms of the profile modification of the two-color laser beam. The 34th harmonic at 23.5 nm and 38th harmonic at 21.5 nm were enhanced 2.5 and 2 times, respectively.

1 Introduction

A high-harmonic light source possesses such unique properties as superb coherence and ultrashort pulse duration ranging from femtoseconds to attoseconds [1]. High-order harmonic generation (HHG) from atoms driven by a strong femtosecond laser field has been investigated for the development of ultrashort coherent extreme ultraviolet (XUV) sources for attosecond physics [2, 3], microscopic imaging [4], interferometry [5], and also for the exploration of atomic and molecular structures [6, 7]. This novel XUV source, however, has a significant drawback of low conversion efficiency, and various schemes have been developed to improve it. The employment of a loose focusing geometry to a long gas cell was successful in boosting the harmonic yield to a μJ level in the 40-nm region by increasing the interaction cross section and length [8, 9]. Spatial modification to a specific profile, such as flat-top shape, was also successful in expanding the effective harmonic generation volume [10, 11]. Methods using waveguide structures have been investigated in order to improve phase matching between a driving laser pulse and generated harmonics [12, 13]. These efforts made it possible to apply high-harmonic light source to specific applications such as XUV spectroscopy and microscopy, while it should be enhanced further to improve the applicability.

Another approach developed to significantly enhance harmonic efficiency is the use of a synthesized laser field. As a simple synthesized field, a two-color laser field, consisting of a fundamental laser field and its second harmonic (SH), was applied to HHG [14, 15]. It significantly improved the harmonic efficiency by generating short-path harmonics from atoms under a strongly ionizing condition, as compared to single-color harmonics generated under a weak ionization condition. We applied the strong harmonic by a two-color laser field to soft X-ray imaging experiments [4], and we found that the intensity of harmonic source should be improved further to enhance the spatial resolution of single-shot microscopy. More recently, two-gas schemes were proposed to enhance the harmonic efficiency using a mixed gas medium or two gas cells [16, 17]. In order to enhance the harmonic intensity further, the two-gas media approach can be employed in two-color HHG scheme.

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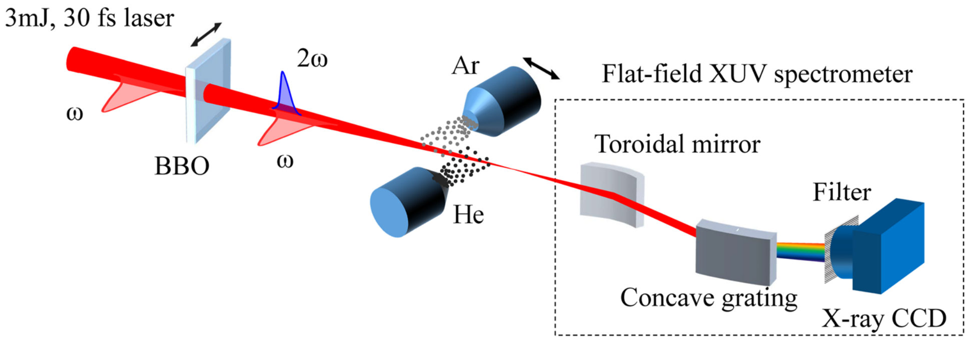


Fig. 1 Experimental setup for high-harmonic generation in a two-color laser field with two-gas media

In this paper, we present the experimental results of two-color high-harmonic generation performed using two-gas media. In addition to the harmonic-generating gas medium of helium, an extra gas medium of argon was installed in front of the harmonic generation medium. Under the optimized experimental conditions, the strong two-color harmonics at the 34th and 38th orders from helium were further enhanced by more than a factor of 2. From the characteristics of high harmonics obtained using the two-color field and two-gas media, we showed that the enhancement of harmonic yield originated mainly from the beam profile modification of the two-color laser field in the additional gas medium.

2 Two-color harmonic generation with two-gas media

The two-color high-harmonic generation was carried out by installing an additional gas medium to enhance further strong harmonics generated in the two-color laser field, consisting of the fundamental and the SH of a 3-mJ, 30-fs Ti:Sapphire laser at 820 nm, as shown in Fig. 1. A laser pulse was focused using a spherical mirror of 60-cm focal length into a helium gas jet with a 2-mm slit nozzle—the harmonic generation medium. The additional 2-mm gas jet of argon was installed in front of the helium jet to enhance harmonic generation. High harmonics were measured with a flat-field XUV spectrometer equipped with a toroidal mirror, a concave grating with 1200 grooves/mm, and an X-ray CCD (Princeton Instruments). In order to block co-propagating laser pulses, a set of aluminum filters was installed in the XUV spectrometer. Between the focusing mirror and the gas jets, a 200-μm-thick beta-barium borate (BBO) was used for the SH generation, converting about 20 % of the incident laser pulse energy to SH. The SH, generated using a type I BBO, was polarized orthogonally to the fundamental. The two-color harmonic generation from helium was examined to find out the optimum condition for maximum enhancement of harmonics.

The characteristics of HHG with two-gas media in two-color laser field were examined while controlling the positions and the backing pressures of the two gas jets. It was shown in our earlier works [14, 15] that in two-color laser field, the harmonics of $2(2n + 1)$th orders were generated to be very strong, while the cutoff order was reduced. In order to observe the effects of the additional gas medium, the optimization of two-color high harmonics, especially the 38th harmonic, from the helium gas jet alone was, firstly, carried out by controlling the density and the position of the helium jet. The 38th harmonic was strongest for the backing pressure of 2250 Torr and the jet position centered 5 mm before the laser focus. High-harmonic spectra were, then, examined while changing the backing pressure and the position of the argon gas jet placed before the helium gas jet. The position of the argon gas jet was adjusted with respect to the helium gas jet, and the argon backing pressure was varied from 0 to 600 Torr. The optimum position of the argon jet was found to be 2 mm away from the helium jet. While varying argon backing pressure, the enhancement of strong harmonics at the 34th and 38th orders was measured.

With the addition of the argon gas jet, the strong two-color harmonics could be enhanced further. As shown in Fig. 2a, c, these harmonics increased initially with argon pressure and, then, dropped after reaching a peak; the intensity of the 38th harmonic was doubled at the argon pressure of 240 Torr, while the 34th harmonic increased by a factor of 2.5 at the argon pressure of 280 Torr. The 34th harmonic formed a peaked region for the argon pressure between 240 and 280 Torr, but the 38th harmonic decreased quickly after reaching the peak at 240 Torr. In the case of the 42nd harmonic, its intensity was not enhanced with the addition of the argon jet, but decreased for the argon pressure higher than 240 Torr. For comparison, HHG only with the fundamental laser field was examined. The one-color harmonics from helium were obtained and are shown in Fig. 2b, d. Although the cutoff of these harmonics reached around the 79th order, the harmonics could not be enhanced with the addition of the argon jet. All the harmonics decreased with the increase in argon pressure after a slight increase

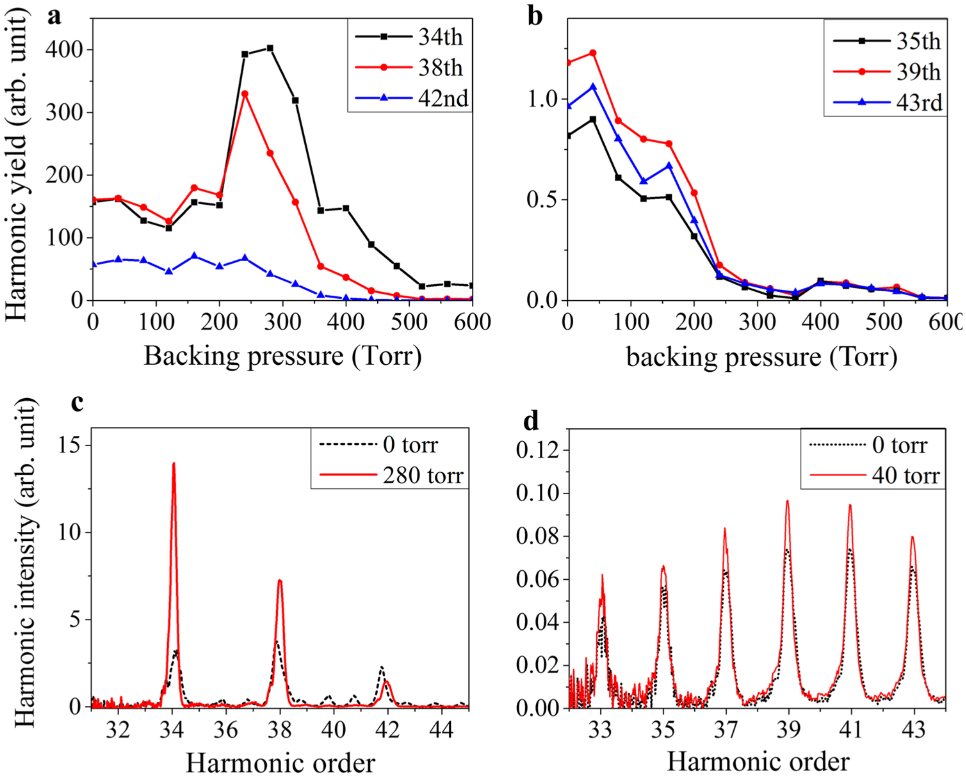


Fig. 2 **a** Yield of harmonics from helium in the two-color laser field measured with respect to the backing pressure of the additional gas jet of argon. **b** Yield of harmonics from helium, driven only by the fundamental laser field, measured with respect to argon backing pressure. **c** Harmonic spectra obtained from helium in the two-color laser field. The case of the argon gas jet with the backing pressure of 280 Torr is compared with the one without the argon jet. **d** Harmonic spectra obtained from helium in the fundamental laser field only. The case of the argon gas jet with the backing pressure of 40 Torr is compared with the one without the argon jet

near the argon pressure of 40 Torr. The observed difference indicates that the addition of the argon jet affected more sensitively the SH field than the fundamental laser field. Consequently, the installation of the argon gas jet in front of the harmonic generation medium of helium enhanced the strong two-color harmonics at the 34th and 38th orders.

3 Analysis of experimental results

The experimental results in Sect. 2 clearly showed the enhancement of the strong two-color harmonics from helium with the addition of the extra argon gas jet. For the understanding of the physical processes generating the enhancement, the contributions of low-order harmonics were examined. Takahashi et al. [16] reported that the harmonic enhancement was achieved due to the enhanced ionization of the harmonic generation medium of helium by low-order harmonics emitted from the medium of xenon, mixed with helium. On the other hand, Brizuela et al. [17] reported the enhancement of high harmonics due to strong subthreshold harmonics, particularly the 3rd harmonic, generated from the additional gas medium, interfering with the fundamental laser field. They showed a relation of the harmonic enhancement to the intensity and phase of the 3rd harmonic. We, thus, examined the effects of low-order harmonics in enhancing the two-color harmonics from helium.

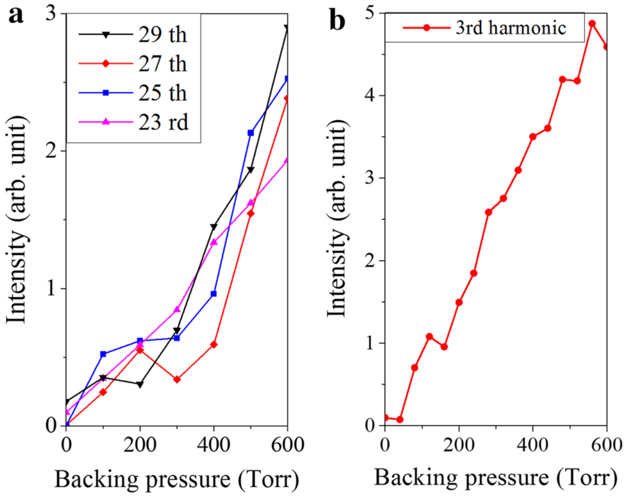


Fig. 3 Harmonic intensity variation of **a** the 23rd–29th harmonics and **b** the 3rd harmonic generated from argon with respect to backing pressure

In order to find out the contribution of low-order harmonics to the enhancement, we measured low-order harmonics in two parts. The first part of measurement was carried out for the harmonics in the range of 23rd–29th harmonics with respect to argon backing pressure. As shown in Fig. 3a, these harmonics increased with argon pressure in our experimental pressure range of up to 600 Torr. Though the harmonics from argon might boost the HHG in helium by assisting the ionization, as observed in the work by Takahashi et al., this mechanism cannot explain our case because we could not observe the enhancement of the harmonics in our one-color result. In our case, the harmonics generated from argon were significantly weaker than those from xenon used in the previous work. In the second part, the 3rd harmonic was measured while varying the argon pressure. We found again that the 3rd harmonic increased monotonically with argon pressure, as shown in Fig. 3b. The intensity ratio between the fundamental and the third harmonic was found to be an order of magnitude smaller than that used in the work by Brizuela et al. In our case, the 3rd harmonic was too weak to enhance the HHG in the helium medium by increasing ionization probability. Thus, the harmonic enhancement could not be explained with the contribution from low-order harmonics in our case, and other factor must be considered in order to explain the observed enhancement.

Furthermore, the possible change in the relative phase between the fundamental and the SH fields in the argon medium can be crucial for the two-color HHG. The relative phase shift was estimated from the refractive index modification by ionization, calculated with the ADK model. When we verified the relative phase effect at the argon position of $z = -9$ mm, the periodic modulation of the harmonics with respect to argon pressure was well estimated to the variation by the phase shift as shown in Fig. 4. At $z = -7$ mm, with the laser intensity above the saturation intensity of argon, the periodic modulation disappeared in the pressure range above 200 Torr, in which we observed harmonic enhancement. We can, thus, exclude the effect of relative phase as well in explaining the origin of HHG enhancement.

The other factor considered was the beam profile modification experienced during the propagation of the two-color laser pulse through the argon medium. In our experimental conditions, the laser intensity reached 3×10^{15} W/cm^2 at the focus, and the argon medium could be easily ionized even at the position 7 mm before the focus. When an intense laser pulse propagates through a gas medium ionized by the laser pulse itself, the ionized medium, or plasma, defocuses the laser pulse and, as a result, the laser beam profile can be modified [18]. Since HHG is sensitive to the beam profile of a driving laser pulse, the beam profiles of the fundamental and the SH beams should be examined after propagating through the argon jet.

We measured the beam profiles of the fundamental and the SH to investigate the relation with the measured high harmonics. The beam profiles of the fundamental and the SH at the entrance of the helium gas jet were measured by relaying the beam images using a concave mirror placed 70 cm after the laser focus, while changing the argon pressure up to 600 Torr, as shown in Fig. 5. The results show that the laser intensity decreased gradually while the beam size increased with argon pressure. It is noted that the modifications imposed to the fundamental and the SH are not the same. As the argon jet was located in the converging laser beam, the inner part of the beam, propagating through a higher electron density region, experienced strong defocusing, while the outer part still converged. As a result, the beam profile of the propagating laser beam became flattened. The reduction in peak intensity and the increased beam size agreed with the observation that the harmonic enhancement, shown in Fig. 2b, was more significant for the 34th harmonic than the 38th or the 42nd harmonic. For comparison, the beam profile modification in the neon gas jet, instead of the argon gas jet, was also measured. The beam profile modification was found to be minimal, and the two-color harmonics were almost unaffected. Due to the large ionization energy, 21.56 eV for neon, the ionization of neon, estimated using the ADK formula, was 6.7 % at the cutoff intensity for the 34th harmonic. On the contrary, the ionization of argon was 99.8 % for the same intensity. Thus, the profile modification imposed by the addition of the argon gas jet can be responsible for the enhancement of the two-color harmonics.

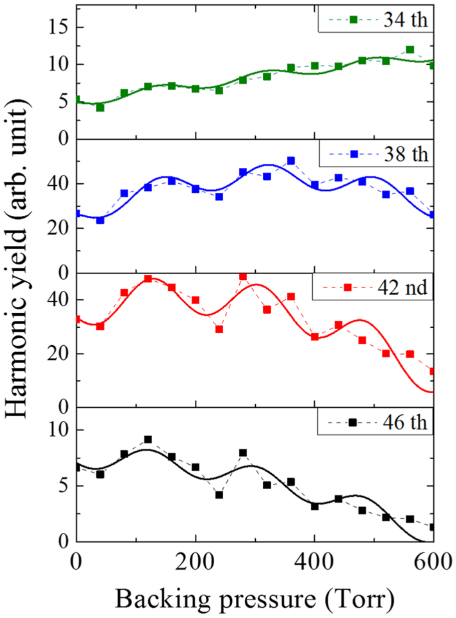


Fig. 4 Harmonic yield (*dot*) and fitting result by the estimated relative phase (*solid line*) at the argon position of $z = -9$ in our two-gas two-color scheme

To clarify the relation between the profile modification and the enhancement, we examined the images of the fundamental and the SH components of the two-color laser field. In Fig. 6, the fundamental and the SH images are shown for the argon pressure of 0, 280, and 400 Torr. It is seen in Fig. 6 that the beam profile is flattened after passing

Fig. 5 Modification of **a** peak intensity and **b** beam diameter ($1/e^2$) of the fundamental and the SH beams of the two-color laser field, measured while changing the backing pressure of the argon gas jet

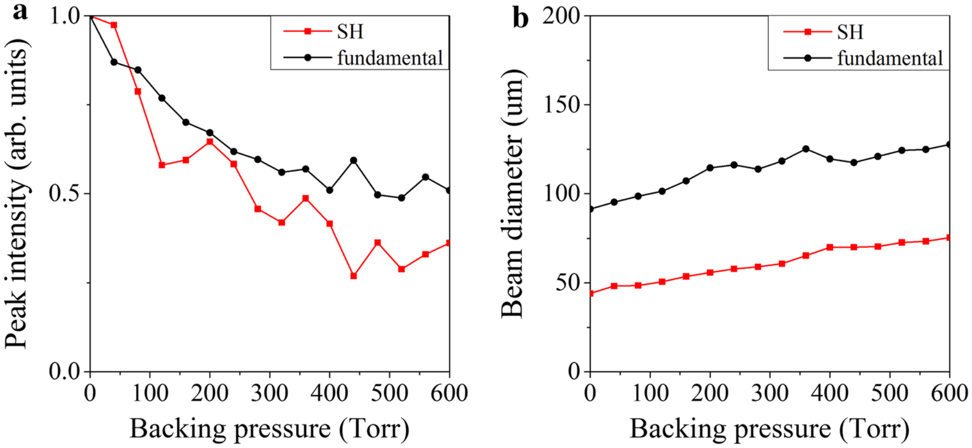
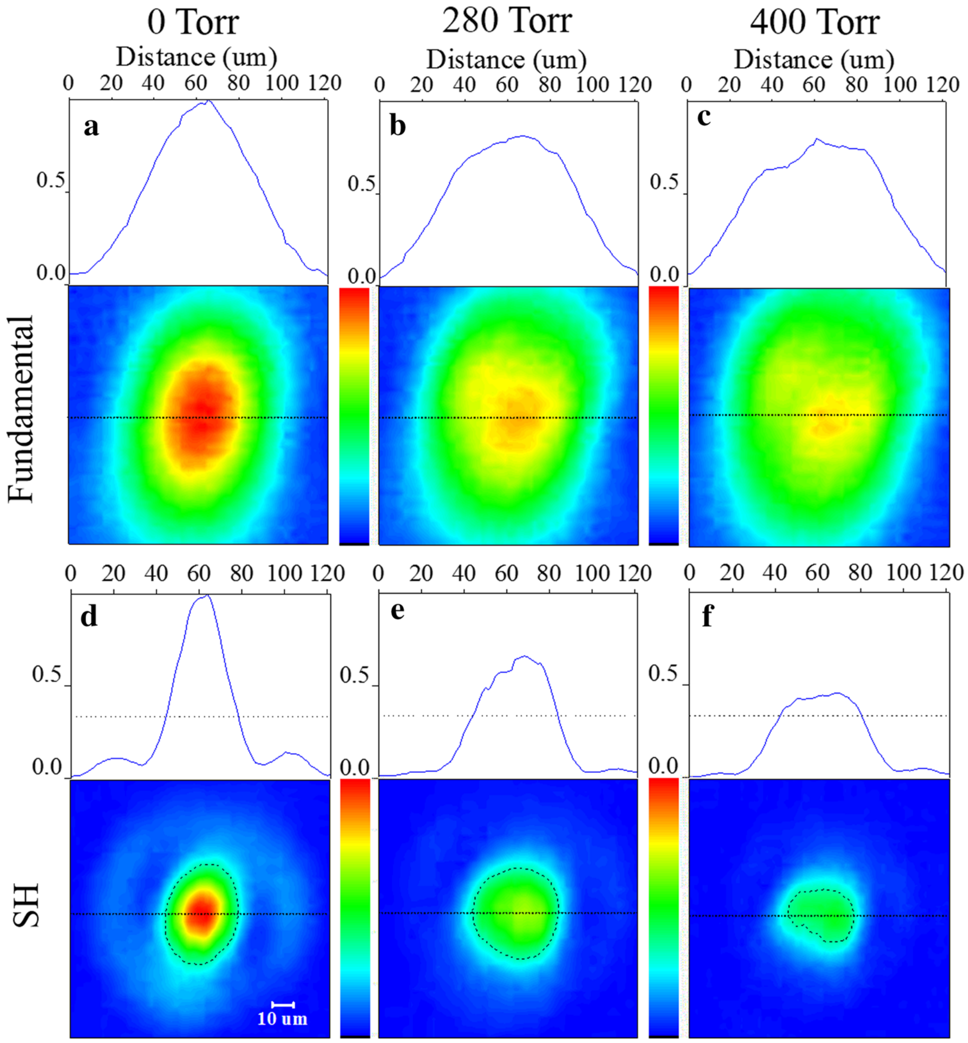


Fig. 6 Measured profiles of the fundamental **a** without argon gas, **b** with argon gas for the backing pressure of 280 Torr, and **c** of 400 Torr. Fundamental profiles **d** without argon gas and **e**, **f** with the same argon gas conditions as that in **b**, **c**, respectively. The *dotted contour lines* in **d**, **e**, **f** indicate the cutoff intensity, 5.9×10^{14} W/cm^2, for the 34th order



through the argon medium and that the profile modification is more significant for the SH images than for the fundamental images. The profile flattening can be understood by considering the plasma defocusing effect of a converging beam. As the central portion of the argon medium was ionized more than the outer region, the defocusing was stronger in the central region, but the outer part was still converging due to the weaker defocusing. The dotted line in the SH image shows the contour line corresponding to the cutoff intensity of the 34th order. The cross section of

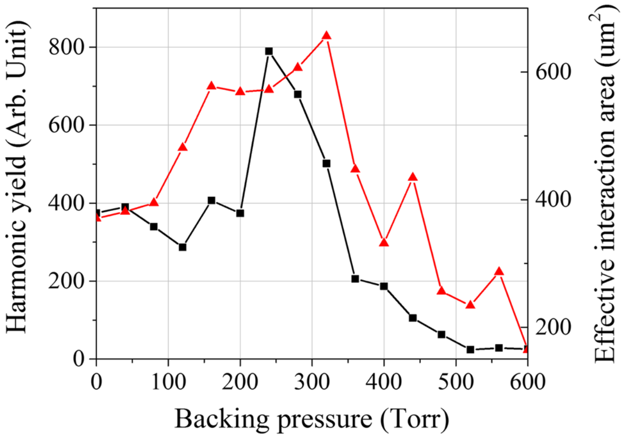


Fig. 7 Effective interaction area of the SH (*red*) from the measured SH beam profile after adding the argon medium, and total yield of the harmonics from the 34th to the 42nd order (*black*)

the helium medium for generating the 34th harmonic was clearly expanded in the case of 280 Torr and then reduced at 400 Torr, agreeing qualitatively with the observed enhancement of the 34th harmonic. The smaller enhancement for the 38th and 42nd harmonics could be understood by considering that the peak laser intensity decreased during the propagation through the argon medium. The difference of beam profile flattening between the SH and the fundamental can also be understood by considering the time delay between the two laser fields that let the SH meet higher electron density. In addition, the stronger beam profile flattening for the SH than for the fundamental increased the spatially overlapped volume between the fundamental and the SH, which also helped the enhancement of the two-color harmonic generation.

In order to verify further the effect of beam profile modification, we compared the effective interaction area of the SH in the helium medium and the harmonic yield. The effective area profile with intensity higher than the cutoff intensity of the 34th harmonic was estimated from the measured SH beam profile, as marked by the dotted contour lines in Fig. 6d–f. Figure 7 shows the effective interaction area with respect to argon backing pressure and the total harmonic yield from the 34th to the 42nd order. The enhancement of harmonic signal is consistent with the increase in the effective interaction area. Consequently, our experimental results confirmed that the harmonic enhancement originated mainly from the beam profile modification of the SH in the argon medium.

Another interesting spectral feature observed with the installation of the additional gas jet of argon was the spectral bandwidth of two-color harmonics. As seen in Fig. 2, the spectral bandwidth of the strong harmonics was significantly improved. The spectral bandwidth, measured by the relative bandwidth ($\lambda/\Delta\lambda$), was improved by 1.8 times from 78 to 140 for the 34th harmonic and by 1.3 times for the 38th harmonic. In the case of one-color HHG, the spectral bandwidth can be modified by controlling laser chirp [19]. In the case of two-color HHG, the coherent control method of HHG, however, cannot be applied because the SH conversion efficiency is strongly affected by laser chirp. Only near-chirp-free laser pulses were applied for the most efficient SH generation to achieve the strongest two-color HHG. The improved spectral bandwidth is advantageous in obtaining high-resolution images in X-ray microscopy applications, such as an X-ray transmission microscope using Fresnel zone plates [4], X-ray coherent diffraction imaging, and Fourier transform holography [20]. The improvement in spectral bandwidth gained by installing the additional gas jet of argon, thus, provided an additional advantage in applications.

4 Conclusion

We investigated the enhancement of the HHG from helium in a two-color laser field by installing an extra argon gas medium. It was found that the beam profile modification, gained during the propagation through the argon medium, was responsible for the enhanced two-color harmonic generation at the 34th and 38th orders. The profile flattening of the laser beams, especially the SH, increased the effective harmonic generation volume, enhancing the two-color HHG. In addition, the spectral bandwidth of strong two-color harmonics was improved, which is advantageous for imaging and spectroscopic applications. The addition of an extra gas medium, thus, can provide another useful optimization knob, along with laser parameters of two-color laser fields, for harmonic enhancement and spectral bandwidth control needed for versatile applications.

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