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0.35% THz pulse conversion efficiency achieved by Ti:sapphire femtosecond laser filamentation in argon at 1 kHz repetition rate

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Section 1: The walk-off distance of the focusing light beam and a plane light beam

The walk-off angle α between the fundamental and second harmonic laser beams can be calculated by the following equation^{S1}:

$$\tan\alpha = \frac{n_o^2 - n_e^2}{n_o^2 \sin^2\theta + n_e^2 \cos^2\theta} \sin\theta \cos\theta, \quad (\text{S1})$$

where n_o and n_e are the principal refractive indices of the index ellipsoid for the second harmonic laser beam, θ is the angle between the wave vector of the second harmonic laser beam and the optical axis of the β -BBO crystal. **Figure S1(a)** shows the schematic diagram of the focused fundamental laser incident onto the β -BBO crystal. The cross-sectional planes in yoz and xoz are respectively shown in **Fig. S1(b)** and **S1(c)**, presenting that the incident angle γ of the focused fundamental beam ranges from -0.86° to 0.86° . Using the incident angle, we can calculate the angle between the wave vector of the fundamental beam and the optical axis of β -BBO crystal, i.e. θ in **Eq. (S1)**. Therefore, we can calculate the variation of the walk-off angle α as a function of γ . The calculation results are shown in **Fig. S1(d)** and **S1(e)**. These two figures respectively show the calculation results in yoz plane and xoz plane. The two figures show that the focusing induced incident angle's variation causes only sub-micrometer scale's change in the walk-off distance, which is negligible compared with the millimeter scale's beam diameter. Therefore, we draw the conclusion that the difference between calculating the walk-off distance for a plane wavefront and a spherical wavefront is negligible in the β -BBO crystal.

Using similar procedures, the walk-off angle and distance between the fundamental and second harmonic laser beams in α -BBO and dual-wavelength plate (DWP) can be also calculated for different incident angles. The calculation results are shown in **Fig. S2**. It is seen that the incident angle, i.e. the focusing of the laser beam has little effect on the walk-off distance. Therefore, in experiments we draw the conclusion that the difference between calculating the walk-off distance for a plane wavefront and a spherical wavefront in α -BBO and DWP is negligible.

The temporal delay between the fundamental and the second harmonic laser beams is determined by their optical path difference. The optical path is determined by the real propagation distance and the refractive index. Since the walk-off angle and walk-off distance are nearly identical for the focusing light beam and the plane light beam based on the above analyses, the focusing light beam and the plane light beam have nearly identical beam paths, which can further induce identical refractive index even in the birefringent material. Therefore, it can deduce that the temporal delay calculated using the plane light beam can be considered as a good approximation of the focusing light beam.

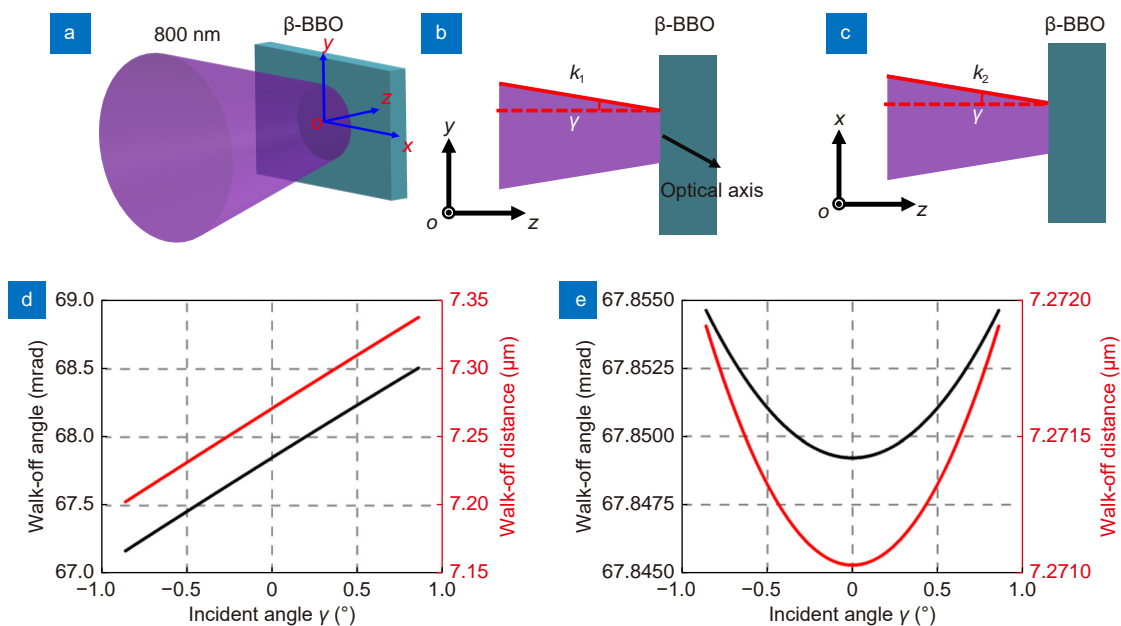


Fig. S1 | (a) Schematic diagram of the focused fundamental laser beam incident on the β -BBO crystal. (b) Schematic diagram of the cross section of (a) in yoz plane. (c) Schematic diagram of the cross section of (a) in xoz plane. (d) Dependences of the walk-off angle and distance on the incident angle in yoz plane. (e) Dependences of the walk-off angle and distance on the incident angle in xoz plane.

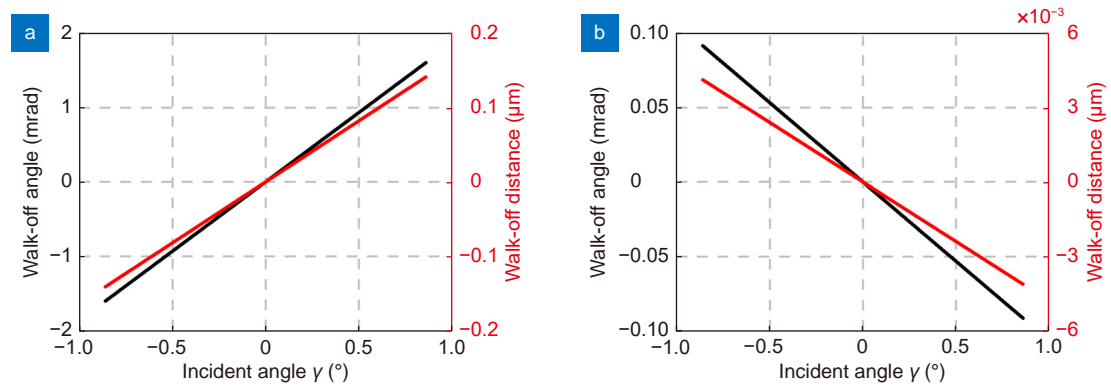


Fig. S2 | The walk-off angles and distances between fundamental and second harmonic laser beams in α -BBO (a) and dual-wavelength plate (DWP) (b) for different incident angles.

References

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