Towards a petawatt-class few-cycle infrared laser system via dual-chirped optical parametric amplification

Yuxi Fu, Katsumi Midorikawa & Eiji J. Takahashi

Expansion of the wavelength range for an ultrafast laser is an important ingredient for extending its range of applications. Conventionally, optical parametric amplification (OPA) has been employed to expand the laser wavelength to the infrared (IR) region. However, the achievable pulse energy and peak power have been limited to the mJ and the GW level, respectively. A major difficulty in the further energy scaling of OPA results from a lack of suitable large nonlinear crystals. Here, we circumvent this difficulty by employing a dual-chirped optical parametric amplification (DC-OPA) scheme. We successfully generate a multi-TW IR femtosecond laser pulse with an energy of 100 mJ order, which is higher than that reported in previous works. We also obtain excellent energy scaling ability, ultrashort pulses, flexible wavelength tunability, and high-energy stability, which prove that DC-OPA is a superior method for the energy scaling of IR pulses to the 10 J/PW level.

Since the invention of chirped pulse amplification (CPA) in 1985, which overcame the obstacles in amplifying an ultrashort laser pulse without damaging optical elements or crystals in a beam line, ultrahigh-intensity laser systems have progressed rapidly. By combining a Ti:sapphire crystal and CPA, laser systems with pulse energies of 100 J order and pulse durations of 10 fs order and a PW-order peak power have been achieved with wavelengths near 0.8 \( \mu \text{m} \). Owing to CPA technology, a new research field called *strong-field laser science* was born and has progressed rapidly. Recently, strong-field laser science has been experiencing a shift towards the use of infrared (IR) driving wavelengths. This is because they enable the demonstration of ponderomotive potential scaling of the electron energy in a high-intensity laser field. The ponderomotive potential is given by

\[
U_p[\text{eV}] = 9.337 \times 10^{-14} I[\text{W/cm}^2] (\lambda_0[\mu\text{m}])^2,
\]

which induces quiver motion in an electron from a laser field. This formula can be explained in terms of the so-called three-step model in a semiclassical framework. The ponderomotive potential is proportional to the square of the laser wavelength (\( \lambda_0 \)), thereby a long-wavelength laser is suitable for scaling up the electron quiver energy independently of the laser intensity (\( I_0 \)). One application of the ponderomotive potential scaling is to increase the photon energy in high-order harmonic generation (HHG). Indeed, increasing the photon energy in HHG up to the “water window” region was demonstrated using a 1.5 \( \mu \text{m} \) laser pulse in 2008. From the viewpoint of ponderomotive potential scaling, ultrafast lasers with long wavelengths are highly required not only for the generation of fully coherent ultrashort x-ray sources but also for attosecond pulse generation with high-order harmonics, obtaining a deeper understanding of photoionization, the time-resolved probing of molecular structures, the study of ultrafast solid physics, and so forth.

Normally, optical parametric amplification (OPA), which is a wavelength conversion method, is employed to generate ultrashort IR pulses. In OPA, a seed pulse is amplified by an ultrafast pump pulse in a nonlinear crystal through a parametric process. Owing to the limited sizes and damage thresholds of crystals, the output energy of an IR pulse via OPA is limited to the mJ level with pulse durations shorter than 50 fs. To overcome the difficulty in the energy scaling of OPA, optical parametric chirped-pulse amplification (OPCPA), which employs a separate high-energy pump with a long pulse duration, has been proposed and demonstrated. A high-energy pump pulse with a picosecond duration can be employed to avoid damaging nonlinear crystals in the OPA process. Up to now, the highest pulse energy ever reported for an IR OPCPA system has been 30 mJ with a pulse duration of 2 ns.

Extreme Photonics Research Group, RIKEN Center for Advanced Photonics, RIKEN, 2-1 Hirosawa, Wako, Saitama, 351-0198, Japan. Correspondence and requests for materials should be addressed to Y.F. (email: yxfu@riken.jp) or E.J.T. (email: ejtak@riken.jp)
IR femtosecond laser pulses. We demonstrate that DC-OPA is an ultimate method for providing ultrahigh-energy/peak-power energy scaling of IR pulses to 1 J order and even 10-J-order while maintaining a broad spectral bandwidth with multiple outputs with different spectral bandwidths and temporal chirps. In a word, DC-OPA is capable of the further obtaining high efficiency and a broad spectral bandwidth, in contrast to in standard OPCPA systems. By carefully choosing the chirp relations between seed and pump pulses, we achieve a high energy stability (per shot) of ~1% RMS. We also find that the chirp relations between seed and pump pulses play an important role in obtaining high efficiency and a broad spectral bandwidth, in contrast to in standard OPCPA systems. By carefully controlling the chirps of the seed and pump pulses, the DC-OPA system can be optimized to operate with multiple outputs with different spectral bandwidths and temporal chirps. In a word, DC-OPA is capable of the further energy scaling of isolated attosecond pulses (IAPs) by employing an IR two-color gating (IR-TCG) method for HHG. Furthermore, high-energy IR pulses also enable the production of ultrastrong light bullets with TW peak power over a distance of 100 m.

In this work, using DC-OPA, we successfully generate IR pulses with a total energy of over 210 mJ, which is 1–2 orders higher than previously reported works using OPA and OPCPA with pulse durations shorter than 50 fs. A high conversion efficiency of over 30% is obtained, which is higher than that of IR OPCPA systems and comparable with that of a standard OPA system. Our energy-scaling experiment shows that DC-OPA has excellent energy-scaling ability without sacrificing high conversion efficiency. Flexible wavelength tunability over an octave spectral range within 1–2 μm is obtained while the conversion efficiency is maintained. A short pulse duration of 44 fs (8.8 optical cycles) is achieved in our experiment. We also achieve a high energy stability (per shot) of ~1% RMS. We also find that the chirp relations between seed and pump pulses play an important role in obtaining high efficiency and a broad spectral bandwidth, in contrast to in standard OPCPA systems. By carefully controlling the chirps of the seed and pump pulses, the DC-OPA system can be optimized to operate with multiple outputs with different spectral bandwidths and temporal chirps. In a word, DC-OPA is capable of the further energy scaling of IR pulses to 1 J order and even 10-J-order while maintaining a broad spectral bandwidth with high efficiency. We demonstrate that DC-OPA is an ultimate method for providing ultrahigh-energy/peak-power IR femtosecond laser pulses.

**Results**

**Breaking the bottleneck in energy scaling by conventional OPA.** As mentioned above, OPA is the most widely employed method for obtaining intense IR femtosecond laser pulses. However, the achievable pulse energy is limited by the aperture sizes of IR nonlinear crystals, which is due to the capability of crystal growth in manufacturing. The DC-OPA scheme can break this bottleneck as shown by our experimental result in Fig. 1. In contrast to conventional OPA, which allows a maximum pump energy of up to 10 mJ order, we can easily apply pump energy of more than one order higher in OPA using a commercially available crystal size (20 mm × 20 mm × thickness mm). The key technique is to increase the pump pulse duration when its pulse energy increases. In such a manner, the laser intensity on a β-BaB$_2$O$_4$ (BBO) crystal, which is proportional to the pulse energy and inversely proportional to the pulse duration and the square of the beam diameter, is kept below its damage threshold value. Meanwhile, the duration of the seed pulse is also temporally increased with the aim of obtaining an optimal temporal overlap with the pump pulse and achieving an efficient energy conversion. Since both the broadband pump and seed pulses are stretched (temporally chirped), the method was named dual-chirped optical parametric amplification. Following the DC-OPA strategy, excellent energy scaling with 30.6%...
In Fig. 2(a). Owing to the high conversion efficiency, the pulse energies of the signal beams remain above 100 mJ.

Characteristics of OPA, OPCPA, FOPA, and DC-OPA for generating IR femtosecond laser in 1–2 μm region.

Table 1. Characteristics of OPA, OPCPA, FOPA, and DC-OPA for generating IR femtosecond laser in 1–2 μm region.

|                  | OPA15–17,45 | OPCPA18–20 | FOPA46,48 | DC-OPA25,26,36 |
|------------------|-------------|------------|-----------|----------------|
| Pump source      | fs Ti:sapphire CPA | ps laser (e.g. Yb:YAG) | fs Ti:sapphire CPA | fs Ti:sapphire CPA |
| Pump duration    | TL          | TL         | Chirped   | Chirped         |
| Pump spectrum    | Broad       | Narrow     | Broad     | Broad           |
| Maximum pump energy | 0.1 J      | 1 J        | 10 fs order | 100 fs order   |
| Seed duration    | TL          | Chirped    | TL        | Chirped         |
| Seed spectrum    | Broad       | Broad      | Broad     | Broad           |
| Synchronization  | Automatic   | Active stabilization | Automatic | Automatic       |
| Energy scaling   | Difficult   | Easy       | Possible  | Easy            |
| Output pulses    | Signal and idler | Signal and idler | Signal | Signal and idler |
| Conversion efficiency | 30–40%   | 10–30%     | 14%       | 30–40%          |
| Highest reported output energy | 10 mJ  | 30 mJ      | 30 mJ     | 100 mJ          |
| Few-cycle pulse generation? | Yes | Yes        | Yes       | Yes             |
| Preserve CEP stability of seed? | Yes | Possible   | No        | Possible        |
| Wavelength tunability | Excellent | Good       | Not reported | Excellent       |
| How a broad output spectrum is efficiently generated. | Thin crystal with broad phase-matched bandwidth. | Thin crystal with broad phase-matched bandwidth. | Pump duration on the 1 ps level. | Phase-matched bandwidth of crystal. |
| Compressor       | Not needed  | Needed     | Not needed| Most often but not always needed (See discussion of idler pulses in Fig. 4(b)). |
| Future energy scaling | 0.1 J order | Depends on pump laser | 1 J order | 10 J order |

Broad wavelength tunability of DC-OPA. The wavelength tunability of DC-OPA is confirmed in the next experiment, which is an important feature for extending the applications of a laser source. The experimental results are shown in Fig. 2, which shows a flexible wavelength-tuning capability in an one-octave spectral range of 1.2–2.4 μm. The total conversion efficiency of DC-OPA is maintained at ~30% at each wavelength as can be seen in Fig. 2(a). Owing to the high conversion efficiency, the pulse energies of the signal beams remain above 100 mJ in the entire tuning range. In contrast, the corresponding idler pulse energies are measured to be 100 mJ or less. This is because a longer-wavelength pulse has a lower pulse energy owing to its lower photon energy under a constant conversion efficiency. Moreover, the pulse energy is clearly lower at 2.1 μm and 2.3 μm, which is attributed to absorption by BBO crystals at wavelengths beyond 2 μm. The spectrum at each central wavelength is shown in Fig. 2(b). The spectral bandwidths at all these wavelengths support TL pulses of shorter than 10 optical cycles (FWHM).

Optimization and characterization: the output performance of DC-OPA. In this section, the optimization and characteristics of DC-OPA are demonstrated and discussed, respectively. For clarity, the signal wavelength is set at 1.5 μm unless specified.

High efficiency and broadband spectrum. In the first stage of the DC-OPA system (see Methods section), the seed pulse is amplified to 6 mJ, corresponding to a parametric gain of ~3000. In the second stage, the signal pulse is further amplified to 120 mJ with a gain of 20. No fluorescence is observed from the BBO crystals when the seed pulse is blocked. Under the optimized conditions for a signal wavelength of 1.5 μm, the total output energy after the DC-OPA system is 218 mJ, which indicates an overall conversion efficiency of 31%. Considering only the second stage, the conversion efficiency is as high as 35%. Since our BBO crystals do not have any anti-reflection coatings, the conversion efficiencies of the whole DC-OPA system and the second stage are 34% and 38%, respectively, if the reflection of the pump pulse (~8%) on the crystal surfaces is taken into account. This high efficiency is similar to that of standard OPA31, which utilizes pump and seed pulses with a close to TL duration of 10 fs order.
Flexible control of spectral bandwidth and temporal chirp. To investigate the chirp effect, we vary the temporal chirp of the seed pulse by tuning its GDD and TOD values. The GDD and TOD values of a 530 mJ pump pulse are set at \(-25000 \text{ fs}^2\) and \(-48000 \text{ fs}^3\), respectively, which indicate a pulse duration of \(\sim 2.9 \text{ ps}\).

First, we only tune the GDD of a seed pulse, which affects the output energy as well as the spectra in DC-OPA as shown in Fig. 4(a,b), respectively. When the GDD of the seed pulse is set to \(0 \text{ fs}^2\), the output energy has a minimum value of \(21.5 \text{ mJ}\). In this case, only a small portion of the pump pulse is temporally overlapped with the seed for the shortest pulse duration. For the same reason, the conversion efficiency increases when the seed pulse has a longer duration at larger absolute GDD values. Another notable feature is that the conversion efficiency is higher when the seed and pump pulses have the same sign of the GDD (positive). For example, the output energy is \(112 \text{ mJ}\) for a seed GDD of \(30000 \text{ fs}^2\) but only \(61 \text{ mJ}\) for the same GDD value with the opposite sign. The reason for this can be understood from Fig. 3(c). Better phase matching between different pairs of wavelengths of the seed and pump pulses that overlap at different time slices is achieved when the seed and pump pulses have the same sign of the GDD. This is an interesting feature in DC-OPA, that is not found in OPCPA. Note that the optimum chirp signs of the seed and pump pulses observed experimentally may vary with the laser wavelengths, the type of crystals and their cutting angles.

Figure 4(b) shows the spectra of the signal (1500 nm) and idler (1730 nm) pulses corresponding to Fig. 4(a). The spectra show a complex characteristic with respect to the GDD of the seed pulse. For the signal pulse, positive GDD values are more favorable than negative values for creating a broader spectrum bandwidth. This is because a better phase matching can be realized, as shown in Fig. 4(a). When the GDD of the seed pulse increases from \(0 \text{ fs}^2\) to \(50000 \text{ fs}^2\), there is an optimum GDD value near \(40000 \text{ fs}^2\) at which the signal pulse has the broadest spectral bandwidth. Thus, it is confirmed that the best phase-matching condition is achieved at different...
wavelengths across the spectra of the seed and pump pulses. The above features are discussed in more detail in the Supplementary Information (see Supplementary S-Fig. 1 and discussion). It can also be observed that the narrowest spectrum bandwidth appears when the GDD of the seed pulse is 0 fs². This is because the signal pulse has the shortest duration. Then the temporal walk-off (group velocity mismatch) between the signal and idler pulses is comparable to their pulse durations. In other words, the walk-off becomes a dominant factor for determining the spectral bandwidth when the pulse duration is short, which is similar to in conventional OPA. For an idler pulse that is generated through difference frequency generation (DFG) between the pump and seed pulses, its spectral bandwidth is mainly determined by the frequency difference range between the temporally overlapped seed and pump pulses. When the GDD values of the pump (positive) and seed (negative) pulses have the opposite chirp sign, long-wavelength components of the seed pulse overlap with short-wavelength components of the pump pulse in the temporal domain and vice versa. Hence, the DFG between the pump and seed pulses has a broader bandwidth. In contrast, the idler pulse has the narrowest bandwidth when the seed GDD is near 26000 fs², where the frequency differences between the wavelengths of the pump and seed pulses that are temporally overlapped is small. In other words, the narrowest spectrum of the idler pulse is because the chirp rates of the seed and pump pulses are similar. Thus, it is expected that the idler pulse is almost chirp free even without compression. This is because chirps of the seed and pump pulses nearly cancel out in the DFG process.

Next, we investigate the effect of the TOD when the GDD of the seed pulse is set at 36000 fs². The experimental result is shown in Fig. 5. As the TOD of the seed pulse changes from negative to positive, the conversion efficiency slightly decreases as shown in Fig. 5(a). However, the dependence of the output energy on the TOD is much weaker than that on the GDD even though a very wide range of TOD values from $-2 \times 10^5$ fs³ to $2.5 \times 10^5$ fs³ are assigned to the seed pulse. This is because the TOD does not significantly change the duration of a seed pulse as the GDD does. Thus, the seed pulse is still well overlapped with the pump pulse when different TOD values are assigned to the seed pulse in the experiment. This is also why the spectral bandwidth of the signal pulse does not change significantly with the TOD value as shown in Fig. 5(b). For the idler spectrum, there is an observable change with the TOD value. When the seed and pump pulses have a TOD with the same sign, the idler pulse has a slightly broader spectral bandwidth. This is because the spectrum of the idler pulse is determined by the temporally overlapped spectral components of both the seed and pump pulses. When the TOD of the seed pulse changes, the wavelength components across its spectrum are slightly modified in time domain. Hence, there is a slight change in the spectrum as the TOD change. A further discussion is given in the Supplementary Information (see Supplementary S-Fig. 2 and discussion).
shown in Fig. 6(c). The pulse is compressed to 49 fs (FWHM), with its temporal phase shown by the green solid line. Owing to the difference in the dispersion of the prism compressor between 1300 nm and 1500 nm, the separation of DC-OPA, as shown in Figs 4 and 5 and discussed above. Different types of compressors are needed or not necessary to compress idler pulses, which may have positive, negative, or zero chirps.

We evaluate the shot-to-shot pulse energy of signal (1.5 μm) and idler (1.73 μm) pulses. The results are shown in Fig. 7(a). Over 30 min, the energy fluctuations are 1.0% RMS and 1.1% RMS for the signal and idler pulses, respectively. High stability is achieved because the DC-OPA system is operated near a saturation condition. The near-field beam profile of the 1.5 μm signal pulses after the prism compressor is measured by an IR CCD camera (XC403, Xenics). Since the beam is too large for the IR camera to measure, we down-collimate the beam size to ~8 mm. The measured beam profile is shown in Fig. 7(b). We also check the focusability using an f/38 focusing geometry. The measured beam profile is shown in Fig. 7(c), and has a smooth intensity distribution. The focus size is measured to be ~140 μm (1/e²). This focusability is sufficient for strong-field physics research such as on HHG.

Figure 5. Dependences of (a) output energy and (b) spectra after DC-OPA on TOD of seed pulse with color scale indicating intensity (arb. units).

Characteristics of temporal pulse duration, energy stability, and beam quality. When the pump energy is ~700 mJ with the pulse duration increased to ~3.5 ps and the GDD is ~30000 fs², we obtain energies of 120 mJ (signal, 1500 nm) and 98 mJ (idler, 1730 nm) as shown in Fig. 2(a). Under these conditions, the seed pulse is stretched to ~3.7 ps with a GDD of ~50000 fs². The high-energy signal pulses with positive chirp are compressed by a prism compressor consisting of two large Brewster angle cut prisms (SK1300). To avoid damage or self-phase modulation inside the compressor, the beam size is increased to ~50 mm. The prism has high transmission for p-polarized pulses under Brewster angle incidence. Thus, the prism compressor can have a throughput of over 85% in our experiment. The calculated phase of the prism compressor with the opposite sign is applied to seed pulses using an acousto-optic programmable dispersive filter (AOPDF). The pulse is compressed to a near-TL duration by further precisely optimizing the spectral phase of the seed pulse. We employ spectral phase interferometry for direct electric-field reconstruction (SPIDER) to characterize the compressed pulse, which is shown in Fig. 6. Figure 6(a) shows the spectrum (blue solid line) and the measured phase (green solid line). The reconstructed temporal pulse (red solid line) and phase (green solid line) are plotted in Fig. 6(b). The pulse duration is evaluated to be 44 fs (FWHM) and consists of 8.8 optical cycles, which is close to its TL duration (black dashed line). In fact, for the seed pulse, the idler pulse can be compressed34,35. The shape of the idler pulse can be indirectly controlled by adjusting the seed phase using an AOPDF, which is helpful for compressing its pulse to the TL duration36, especially for a few-cycle duration. Note that the chirp of idler pulses is determined by the operating mode of DC-OPA, as shown in Figs 4 and 5 and discussed above. Different types of compressors are needed or not necessary to compress idler pulses, which may have positive, negative, or zero chirps.
Discussion
To obtain deeper insights into the features discussed in the previous section, advantages and future prospects of the DC-OPA scheme, we give a detailed discussion in this section.

**Generation of high-energy, few-cycle IR pulses by DC-OPA.** To achieve an even broader spectrum that supports a signal pulse with few-cycle TL duration (the seed spectrum should be sufficiently broad), two methods can be followed in DC-OPA. One method is to employ a pump laser with a broad spectrum. The other method is to employ other crystals with a broader phase-matching bandwidth. Note that the energy-scaling ability of DC-OPA is not restricted by the dimensions and type of crystals. Here we show three examples of calculation results of phase-matching conditions using BBO, BiB$_2$O$_6$ (BiBO), and YCa$_4$O$_3$(BO$_3$)$_3$ (YCOB) crystals with type-I cutting angles, as shown in Fig. 8. Pump and seed pulses propagate collinearly, by which the angular dispersion of the idler pulses can be avoided. In Fig. 8(a), signal pulse with a broad spectrum of 1.1–1.7 μm can be phase-matched using type-I BBO with our pump laser, which has a spectral range of 750–860 nm. From the feature of DC-OPA discussed above, the pump and seed pulses should have the same chirp signs. Note that the conversion efficiency from pump to IR pulses will be low because the amplified IR pulses will generate second harmonics, which have a similar phase-matching angle. For BiBO and YCOB crystals, as shown in Fig. 8(b,c), two phase-matching (PM) conditions, indicated by 'PM1' and 'PM2', can be employed, respectively. When the pump laser has a narrower spectrum bandwidth near 760 nm, PM1 phase-matching, which supports a very broad spectral bandwidth, become the better choice. In fact, a broad spectrum in the range of 1.0–2.0 μm can be expected under a good phase-matching condition. Of course, a seed pulse with the optimum chirp should be applied to obtain good conversion efficiency. 100-mJ-class or even joule-class IR pulses with a duration of two cycles can be expected when 1-J- to 10-J-class pump pulses are used.

For our pump laser with a broad spectral bandwidth, PM2 has higher conversion efficiency for generating a broadband signal pulse when the pump and seed pulses have the same chirp sign. The spectrum is sufficiently broad (1.1–1.6 μm) to support a few-cycle pulse duration. It is also found that a BiBO crystal supports a slightly broader phase-matching bandwidth than a YCOB crystal for our pump laser. However, the much larger crystal aperture size of YCOB (80 mm) than that of BiBO (7 mm) makes YCOB a better choice for experiments. In an word, broad spectra supporting a few-cycle pulse duration can be obtained by the DC-OPA method using a variety of nonlinear crystals.

Figure 6. Pulse durations measured by SPIDER. Spectra (blue solid line) and phases (green solid line) of a compressed pulse at 1500 nm and 1300 nm are shown in (a,c), respectively. Reconstructed pulse envelopes (red solid line) and temporal phases (green solid line) are shown in (b,d), respectively. TL pulse envelopes are shown by black dashed lines.
Further energy scaling of DC-OPA. A typical OPCPA system requires a high-energy pump laser with an optimum pulse duration of 1 ps order to maintain the efficiency and spectral bandwidth\(^{23}\). This requirement is very challenging using current laser technology. In fact, the highest energy of a ps pump laser is 1 J with a pulse duration of ~5 ps, which greatly limits the further scaling up of the IR fs pulse energy to the joule class. In contrast, high-energy Ti:sapphire lasers with an output energy range from 1 J\(^{42}\) to 100 J order\(^{2,43}\) have been developed and utilized for applications. Hence, it is straightforward to further scale up the output energy of DC-OPA to 1 J\(^{26}\) and even 10 J-order by using high-energy Ti:sapphire lasers.

Here, we design experimental parameters for further energy scaling using a BBO crystal under type-II phase matching based on our experiment. Compared with our previous discussions\(^{26}\), we present more detailed parameters in this paper. Moreover, we present optimized chirps for seed and pump pulses, which are important for obtaining high conversion efficiency and a broad spectrum during energy scaling in DC-OPA.

The aperture size of the BBO crystal used is 22 mm × 22 mm, which is commercially available. The pump laser is a Ti:sapphire laser with a pulse energy of 15 J\(^{44}\) (such a laser system is also commercially available, e.g., Alpha XS 1 PW from Thales, France) and a TL pulse duration of shorter than 40 fs, and the pump beam diameters are 6 mm and 20 mm with pulse energies of 1.4 J and 13.6 J for the first and second stages, respectively. Note that a pump laser with a longer TL duration (narrower spectral bandwidth) can provide similar output efficiency for the DC-OPA system. However, the spectral bandwidth of the IR pulses will be narrower (longer TL pulse duration) in this design. Of course, the design parameters depend on the desired specifications of the IR pulses. The seed pulses have the same beam size. Under these conditions, the pump intensity in the first and second stages is ~6 GW/cm\(^2\) with ~550 ps pulse duration, which is below the damage threshold of the BBO crystal as estimated using the square-root scaling law\(^{20}\). The pump pulse is positively chirped with GDD and TOD values of 3.5 × 10\(^6\) fs\(^2\) and −6.5 × 10\(^6\) fs\(^3\), respectively. Simultaneously, the seed pulse with a pulse energy of ~10 μJ is temporally stretched to the same pulse duration of ~550 ps using an Öffner stretcher. The Öffner stretcher can provide a dispersion with GDD and TOD values of −8 × 10\(^6\) fs\(^2\) and −1 × 10\(^6\) fs\(^3\), respectively, which enable the chirp for the seed pulse to achieve an almost perfect phase-matching condition. More details are presented in the Supplementary Information (see Supplementary S-Fig. 3 and discussion). To achieve sufficient gain, the thicknesses of the crystals are 15 mm and 9 mm in the first and second stages, respectively. The temporal walk-offs between the pump, signal, and idler pulses are on the order of 1% of their pulse duration and thus can be neglected. The expected total output energy after DC-OPA is approximately 4.5 J with 30% conversion efficiency, which is estimated from the results of our experiment. The high-energy signal pulse is compressed to a duration of about 40 fs using a grating compressor. Thus, the peak power approaches 100 TW. Further energy scaling using a BBO crystal is also possible if a pump laser with even higher energy is employed by further increasing both the pump and seed pulse durations.

A YCOB crystal instead of BBO will be suitable for further energy scaling. There are three main considerations. First, a very large YCOB crystal with a size of up to φ80 × 200 mm\(^3\) can be fabricated\(^{41,45}\), which will
be convenient for experiments on further energy scaling. Second, a type-I YCOB crystal supports a broader phase-matching bandwidth than a type-II BBO crystal, as shown in Figs 3(c) and 8(c). Thus, it can be utilized to generate high-energy IR pulses with few-cycle pulse durations. Third, a YCOB crystal has a high laser damage threshold comparable to that of a BBO crystal. Using a large YCOB crystal as a nonlinear crystal for the DC-OPA system, a 5 PW Ti:sapphire laser with a pulse energy of ~150 J can be employed to pump the DC-OPA system by increasing the pump beam size to 24 mm and 80 mm in the first and second stages, respectively. The pump pulse duration is ~550 ps which is the same as above. A seed pulse with a optimum chirp with GDD and TOD values of $3.1 \times 10^6$ fs$^2$ and $-5.2 \times 10^6$ fs$^3$, respectively, is also provided by an Öffner stretcher. More details of the calculation and a discussion can be found in the Supplementary Information (see Supplementary S-Fig. 4 and discussion). The phase-matched bandwidth is sufficient to generate few-cycle IR pulses. Thus, IR pulses with ~45 J energy and few-cycle duration will be obtained using the DC-OPA method. The peak power reaches the PW level.

Characteristics of standard OPA, OPCPA, FOPA, and DC-OPA. Strictly speaking, OPCPA and DC-OPA are different types of OPA. Here, we consider another OPA method called frequency domain OPA (FOPA)\(^4\). FOPA is derived from OPA and has the ability to generate high-energy few-cycle IR laser pulses. To help readers understand the different techniques, the characteristics of standard OPA, OPCPA, FOPA, and DC-OPA are shown in Table 1, which are based on previous studies. OPA, FOPA, and DC-OPA employ broadband femtosecond lasers as pump sources, while OPCPA employs a narrow-band laser as a pump (Table 1 lists the commonly employed pump lasers). The energy-scaling capability of standard OPA is determined by the dimension scaling of nonlinear crystals. OPCPA, FOPA, and DC-OPA can scale up the output energy by employing pump pulse with long durations, and thus are not restricted by the manufacturing capability of crystals. Compared with OPCPA and FOPA, DC-OPA has higher efficiency, achieving an IR femtosecond laser energy in the 100 mJ class in an experiment for the first time. For OPA, OPCPA, and DC-OPA, the signal (amplified seed) and idler (DFG between pump and seed) pulses can be obtained as output pulses and utilized for applications. However, for FOPA, idler pulses are difficult to use because they cannot be recovered spatially and compressed temporally by the grating, which is only designed for the signal pulses. Table 1 also lists the carrier-envelope phase (CEP) stability of the output pulses when seeded by a seed pulse with a stable CEP. OPCPA and DC-OPA can maintain the same CEP stability as that of the standard OPA if bulk and chirp mirror compressors are employed. In contrast, FOPA tends to deteriorate the CEP stability owing to the employment of gratings, which are key elements in FOPA but one of the main noise sources for the CEP\(^27,47\). In addition, the spectrum bandwidth and efficiency after FOPA are strongly determined by the gratings, which are very difficult to fabricate with high efficiency over a broad spectrum (e.g., higher than 90% over a spectral range of one octave). To efficiently generate IR pulses with a broad spectral bandwidth, OPA and OPCPA require thin crystals with a broad phase-matched bandwidth. FOPA and DC-OPA each have their own options given as below. By increasing the number of crystals in the Fourier plane (spatial), which are tuned to be phase-matched at different wavelengths, a broad spectrum can be obtained in FOPA. For DC-OPA, by optimizing the temporal chirps of pump and seed pulses, a broad spectrum with high efficiency can be obtained, which does not suffer a loss of spectrum bandwidth when thicker crystals are used to increase the efficiency of energy scaling. Considering all the aspects listed in Table 1, it can be concluded that DC-OPA is a superior method for generating high-energy IR fs pulses with high efficiency and excellent energy-scaling ability.

**Figure 9.** Schematic of the experimental setup. PM: prism (SK1300); BS: beam splitter; DM: dichroic mirror; DL: delay line; Lc: concave lens; Lx: convex lens; CX: convex mirror; CC: concave mirror; RM: roof mirror.
**Methods**

**Experimental setup.** The experimental setup is shown in Fig. 9. The laser system starts from a pre-amplifier (Lambert Elite Duo, Ti:sapphire CPA from Coherent Inc.), which has a central wavelength of ~805 nm and a repetition rate of 1 kHz. Before the laser beam is compressed by the compressor inside the pre-amplifier, it is split into two beams by a beam splitter. The uncompressed beam with a pulse energy of 1.5 mJ and a pulse duration of ~150 ps, whose beam pointing is actively stabilized by a beam stabilizer (Aligna 4D, TEM Messtechnik), is further amplified to a pulse energy of 1 J by a multipass power amplifier (MPA). The MPA is pumped by two flash-lamp-pumped lasers (Precision-II, Continuum) which are operated at a 10 Hz repetition rate. After the MPA, a grating compressor, which is built inside a vacuum chamber, is employed to manipulate the pulse duration as well as the temporal chirp. Then these pulses with an energy of 700 mJ and a beam diameter of 20 mm are utilized to pump a two-stage DC-OPA system. Another beam from the pre-amplifier is compressed to a 25 fs pulse and an energy of 3–4 mJ by a grating compressor. This pulse is employed to pump an OPA system (TOPAS Prime, Coherent Inc.), which provides wavelength-tunable IR pulses in the range of 1.2–1.6 μm. An IR pulse with an energy of ~15 μJ and a beam size of ~2.5 mm passes through an AOPDF (Dazzler, Fastlite), which is programmed to increase the pulse duration with a precisely manipulated temporal chirp. Then, the temporally chirped IR pulse with a pulse energy of ~2 μJ is employed as a seed pulse for the DC-OPA system. The beam diameter of seed pulses is adjusted to match that of the pump beam using concave and convex lens pairs. Type-II BBO crystals (θ = 27°) without anti-reflection coatings are employed as nonlinear crystals in both stages. The crystal dimensions are 10 × 10 × 5 mm3 and 20 × 20 × 3 mm3 for the first and second stages, respectively. In the first stage, the pumping energy is ~70 mJ, corresponding to a laser intensity of approximately 60 GW/cm² when the pulse duration is ~3.5 ps. A non-collinear configuration (3–6.5° depending on the central wavelength) between the pump and seed pulses is employed for two main reasons. One is that the signal, idler and residual pump pulses can be easily separated in space. The other is that signal pulses with a broader spectrum bandwidth might be obtained under a non-collinear configuration. In the first stage, the parametric amplification gain is 2000–5000. Only the signal pulse is selected and sent to the second stage of the DC-OPA system for further amplification. The second stage is also constructed in a non-collinear configuration with a small angle of ~1° between the pump and signal pulses. The purpose of this configuration is to separate high-energy pump, signal and idler pulses, which are not obviously helpful for increasing the phase-matched bandwidth. The pumping energy is ~630 mJ corresponding to a laser intensity of approximately 50 GW/cm² on the BBO crystal when the pulse duration is ~3.5 ps. The amplification gain in the second stage is 20–30. The high-energy signal pulse with a beam diameter of ~50 mm is sent to a Brewster angle cut prism compressor (SK 1300) with a throughput of over 85%. The pulse duration and spectral phase of the compressed signal pulse are characterized by the SPIDER method.

**References**

1. Strickland, D. & Mourou, G. Compression of amplified chirped optical pulses. Optics Communications 56, 219–221 (1985).
2. Chu, Y. et al. High-energy large-aperture Ti:sapphire amplifier for 5 PW laser pulses. Optics Letters 40, 5011–5014 (2015).
3. Danson, C., Hillier, N., Hoppes, D. & Neely, D. Petawatt class lasers worldwide. High Power Laser Science and Engineering 3, 1–14 (2015).
4. Agostini, P. & DiMauro, L. F. The physics of attosecond light pulses. Reports on Progress in Physics. 67, 813–855 (2004).
5. Krausz, F. & Ivanov, M. Attosecond physics. Reviews of Modern Physics. 81, 163–234 (2009).
6. Umstadter, D. Relativistic laser–plasma interactions. J. Phys. D: Appl. Phys. 36, R151–R165 (2003).
7. Corkum, P. B. Plasma perspective on strong-field multiphoton ionization. Phys. Rev. Lett. 71, 1994–1997 (1993).
8. Takahashi, E. J., Kanai, T., Ishikawa, K. K., Nabekawa, Y. & Midorikawa, K. Coherent water window X ray by phase-matched high-order harmonic generation in neutral media. Physical Review Letters 101, 253901 (2008).
9. Wolter, B. et al. Strong-field physics with mid-IR fields. Physical Review X 5, 021034 (2015).
10. Ishii, N. et al. Carrier-envelope phase-dependent high harmonic generation in the water window using few-cycle infrared pulses. Nature Communications 5, 3331 (2014).
11. Heyl, C. M., Arnold, C. L., Cosairon, A. & L’Huillier, A. Introduction to macroscopic power scaling principles for high-order harmonic generation. Journal of Physics B: At. Mol. Opt. Phys. 50, 013001 (2017).
12. Pazourek, R., Nagle, S. & Burgdorfer, J. Attosecond chronoscopy of photoemission. Review of Modern Physics 87, 765–802 (2015).
13. Todd, M. W. et al. Application of mid-infrared cavity-ringdown spectroscopy totrace explosives vapor detection using a broadly tunable (6–8 μm) optical parametric oscillator. Applied Physics B 75, 367–376 (2002).
14. Crosson, E. R. et al. Stable isotope ratios using cavity ring-down spectroscopy: Determination of 13C/12C for carbon dioxide in human breath. Analytical Chemistry 74, 2003–2007 (2002).
15. Cerullo, G. & Silvestri, S. D. Ultrastable optical parametric amplifiers. Review of Scientific Instruments 74, 1–18 (2003).
16. Nisoli, M. et al. Highly efficient parametric conversion of femtosecond Ti:sapphire laser pulses at 1 kHz. Optics Letters 19, 1973–1975 (1994).
17. Théri, N. et al. 10 mJ 5-cycle pulses at 1.8 μm through optical parametric amplification. Applied Physics Letters 106, 091110 (2015).
18. Ross, I. N., Matousek, P., New, G. H. C. & Osyry, K. Analysis and optimization of optical parametric chirped pulse amplification. Journal of the Optical Society of America B 19, 2945–2956 (2002).
19. Mücke, O. D. et al. Self-compression of millijoule 1.5 μm pulses. Optics Letters 34, 2498–2500 (2009).
20. Vayuel, A., Bodnar, N., Webb, B., Shah, L. & Richardson, M. Concepts, performance review, and prospects of table-top, few-cycle optical parametric chirped-pulse amplification. Optical Engineering 53, 051507 (2014).
21. Riddi, J. V., Law, R. J., Luk, T. S. & Cameron, S. M. High-power optical parametric chirped-pulse amplifier system with a 1.55 μm signal and a 1.064 μm pump. Optics Letters 30, 1974–1976 (2005).
22. Moses, I., Manzoni, C., Huang, S.-W., Cerullo, G. & Kärtner, F. X. Temporal optimization of ultrabroadband high-energy opcpa. Optics Express 17, 5540–5555 (2009).
23. Fattahi, H. et al. Third-generation femtosecond technology. Optica 1, 45–63 (2014).
24. Zhang, Q., Takahashi, E. J., Mücke, O. D., Lu, P. & Midorikawa, K. Dual-chirped optical parametric amplification for generating few hundred mJ infrared pulses. Optics Express 19, 7190–7212 (2011).
25. Fu, Y., Takahashi, E. J. & Midorikawa, K. High-energy infrared femtosecond pulses generated by dual-chirped optical parametric amplification. Optics Letters 40, 5082–5085 (2015).
26. Fu, Y., Takahashi, E. J., Zhang, Q., Lu, P. & Midorikawa, K. Optimization and characterization of dual-chirped optical parametric amplification. Journal of Optics 17, 124001 (2015).
27. Yin, Y. et al. High-energy two-cycle pulses at 3.2 μm by a broadband-pumped dual-chirped optical parametric amplifier. Optics Express 24, 24989–24998 (2016).

28. Yin, Y. et al. Towards terawatt sub-cycle long-wave infrared pulses via chirped optical parametric amplification and indirect pulse shaping. Scientific Reports 7, 45794 (2017).

29. Takahashi, E. J., Lan, P., Mücke, O. D., Nabekawa, Y. & Midorikawa, K. Attosecond nonlinear optics using gigawatt-scale isolated attosecond pulses. Nature Communications 4, 2691 (2013).

30. Panagiotopoulos, P., Whalen, P., Kolesik, M. & Moloney, J. V. Super high power mid-infrared femtosecond light bullet. Nature Photonics 9, 543 (2015).

31. Takahashi, E. J., Kanai, T., Nabekawa, Y. & Midorikawa, K. 10 mJ class femtosecond optical parametric amplifier for generating soft x-ray harmonics. Applied Physics Letters 93, 041111 (2008).

32. Limpert, J. et al. Ultra-broad bandwidth parametric amplification at degeneracy. Optics Express 13, 7386–7392 (2005).

33. Tang, Y. et al. Optical parametric chirped-pulse amplification source suitable for seeding high-energy systems. Optics Letters 33, 2386–2388 (2008).

34. Cheriaux, G. et al. Aberration-free stretcher design for ultrashort-pulse amplification. Optics Letters 21, 414–416 (1996).

35. Danilevicius, R., Zaukevicius, A., Budriunas, R., Michailovas, A. & Rusteika, N. Femtosecond wavelength-tunable OPCA system based on picosecond fiber laser seed and picosecond DSPS laser pump. Optics Express 24, 17532–17540 (2016).

36. Fu, Y., Takahashi, E. & Midorikawa, K. Energy scaling of infrared femtosecond pulses by dual-chirped optical parametric amplification. IEEE Photonics Journal 9, 1503108 (2017).

37. Herrmann, D. et al. Generation of sub-three-cycle, 16 TW light pulses by using nonlinear optical parametric chirped-pulse amplification. Optics Letters 34, 2459–2461 (2009).

38. Nabekawa, Y. et al. Multi-terawatt laser system generating 12-fs pulses at 100 Hz repetition rate. Applied Physics B 101, 523–534 (2010).

39. Cunningham, E., Wu, Y. & Chang, Z. Carrier-envelope phase control of a 10 Hz, 25 TW laser for high-flux extreme ultraviolet quasi-continuum generation. Applied Physics Letters 107, 201108 (2015).

40. Ishii, N. et al. Sub-two-cycle, carrier-envelope phase-stable, intense optical pulses at 1.6 μm from a BiB3O6 optical parametric chirped-pulse amplifier. Optics Letters 37, 4182–4184 (2012).

41. Guo, X. et al. Experimental demonstration of broadband femtosecond optical parametric amplification based on ycb crystal at near critical wavelength degeneracy. Optics Communications 370, 98–102 (2016).

42. Leblanc, A., Monchoè, S., Bourassin-Bouchet, C., Kahaly, S. & Quéré, F. Ptychographic measurements of ultrahigh-intensity laser-plasma interactions. Nature Physics 12, 301–305 (2016).

43. Wang, X. et al. Quasi-monoenergetic laser-plasma acceleration of electrons to 2 GeV. Nature Communications 4, 1988 (2013).

44. Kim, H. et al. Enhancement of electron energy to the multi gev regime by a dual-stage laser-wakefield accelerator pumped by petawatt laser pulses. Physical Review Letters 111, 165002 (2013).

45. Guo, X., Tokita, S., Tu, X., Zheng, Y. & Kawanaka, J. Prospects of obtaining terawatt class infrared pulses using standard optical parametric amplification. Laser Physics 27, 025403 (2017).

46. Schmidt, B. E. et al. Frequency domain optical parametric amplification. Nature Communications 5, 3643 (2014).

47. Gruson, V. et al. 2.5 TW, two-cycle IR laser pulses via frequency domain optical parametric amplification. Optics Express 25, 27706 (2017).

Acknowledgements
The authors would like to acknowledge the technical support from Dr. Bing Xue. This work was sponsored by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT) through a grant for Extreme Photonics Research, by the Japan Society for the Promotion of Science through a Grant-in-Aid for Scientific Research, and by MEXT through Grant-in-Aid Nos 26220606, 16K13704, and 17H01067. This work was also part of the Advanced Photon Science Alliance Project commissioned by MEXT. Y. Fu also acknowledges financial support from Incentive Research Projects of RIKEN.

Author Contributions
Y.F. designed the experimental setup and performed the experiments and simulations. K.M. directed the research in accordance with the Extreme Photonics Research Project of RIKEN. E.J.T. conceived the experimental idea and supervised this project. Y.F. and E.J.T. discussed all the experimental results and wrote the manuscript, which was modified by all the authors.

Additional Information
Supplementary information accompanies this paper at https://doi.org/10.1038/s41598-018-25783-0.

Competing Interests: The authors declare no competing interests.

Publisher’s note: Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2018