Ultrafast high-power lasers are employed in a wide variety of applications in science and industry. Thin-disk oscillators can offer compelling performance for these applications. However, because of the high intracavity peak power, a large amount of self-phase modulation (SPM) is picked up in the intracavity environment. Consequently, the highest performance oscillators have been operated in a vacuum environment. Here, we introduce a new concept to overcome this hurdle. We cancel the SPM picked up in air by introducing an intracavity phase-mismatched second-harmonic-generation crystal. The resulting cascaded $\chi^{(2)}$ processes provide a large SPM with a sign opposite the one originating from the air. This enables laser operation in air at 210 W average output power with 780 fs, 19 $\mu$J pulses, the highest output power of any semiconductor saturable absorber mirror (SESAM) modelocked laser operated in air to date, to the best of our knowledge. This result paves the way to a novel approach for nonlinearity management in high-power lasers.

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Ultrafast laser technologies are a crucial tool for a wide variety of applications ranging from science, such as time-resolved studies and XUV generation, to industry, for instance, in high-precision material processing. During the last decade, high-power sources based on Yb-doped gain materials, shaped in the thin-disk [1], fiber [2], and slab geometry [3] have had an impressive development, leading to ultrafast amplifier systems exceeding the kW-level average power milestone. Using thin-disk laser (TDL) technology, oscillators delivering multi-100-W average power and tens-of-$\mu$J pulse energy at MHz repetition rate have been demonstrated [4–6]. This approach enables us to use a table-top and comparatively cost-effective TDL oscillator as an ultrafast high-power laser source. Hence, TDL oscillators, due to their excellent beam quality and low-noise properties [7], are a highly attractive alternative to multi-stage amplifier systems composed of a low-power oscillator, pulse stretcher, amplification stages, and pulse compressor [2,8]. In fact, TDL oscillators are being used for extra- and intra-cavity XUV generation and high-power frequency conversion to the mid-IR, and are potential sources for high-power THz generation [9].

A significant challenge in these TDL oscillators is the high intracavity peak power, which can exceed 100 MW. At such peak powers, the phase accumulated because of the nonlinear refractive index of the intracavity air represents a major contribution to the overall self-phase modulation (SPM). Since the modelocking process relies on soliton pulse formation, which requires a balance between group-delay dispersion (GDD) and SPM [10], this very large amount of SPM ultimately hinders pulse formation. Different methods have been developed so far to overcome this challenge. One is to compensate this large SPM with a corresponding amount of GDD obtained through dispersive mirrors. This creates a tradeoff between the amount of GDD in the cavity and the output pulse energy of the laser (“Standard TDL” in Fig. 1). However, dispersive mirrors have substantially worse thermal behavior compared to Bragg mirrors, making it very challenging to add a large number of them in a high-power oscillator [8,11]. A different approach consists of operating the oscillator in vacuum or helium environment so that the air contribution to the SPM is almost removed (“Vacuum/He TDL” in Fig. 1) [8]. This approach led to the record results in average power and pulse energy. However, the advantages in performance offered by operation of the TDL in vacuum are offset by the significantly increased cost and complexity of such a system. For many scientific and industrial applications, a simpler solution would be required.

Here, we present a new and much simpler technique to cancel the intracavity SPM picked up in air by exploiting cascaded quadratic nonlinearities (CQN) [12]. In CQN, a second-harmonic-generation (SHG) crystal yields an effective nonlinear refractive index that is tunable in magnitude and sign. CQN have been successfully employed for modelocking of lasers in both the positive and negative dispersion regimes [13–17], pulse compression [18,19], for nonlinear-mirror-type modelocking schemes in TDLs [20,21], and in regenerative amplifiers [22]. Here, we introduce a CQN crystal inside the laser cavity in a phase-mismatched, low-loss configuration. This allows us to cancel up to 80% of the total SPM of air. We balance the remaining SPM through just five dispersive mirrors, enabling soliton pulse formation. We obtain 210-W average power at 780-fs pulse duration, 10.96-MHz repetition rate, and 19.2-$\mu$J pulse energy using $-16$, 800 fs$^2$ of GDD (“This result” in Fig. 1). This result
In order to modelock the oscillator, we used an in-house 
grown SESAM as an end mirror, where the beam radius is 
\( \approx 850 \, \mu m \). The SESAM consists of a distributed 
AlAs/GaAs 

Bragg reflector grown at 580°C and three InGaAs quantum wells 
as absorber grown at 280°C in an antiresonant configuration [24,25]. We measured our SESAM to have a saturation fluence 
\( F_{\text{sat}} = 50 \mu J/cm^2 \), a modulation depth \( \Delta R = 2.7% \), and non-
saturable losses \( \Delta R_{\text{ns}} = 0.35% \) [26]. The SESAM was contacted 
by TRUMPF on a polished copper heatsink (cold radius of 

curvature >500 m [25]).

We use only five Gires–Tournois interferometer (GTI)-type 
dispersive mirrors, yielding a total GDD of \( D = -16,800 \, \text{fs}^2 \)

per round trip. Achieving 210-W output power with 780-fs pulses 
without CQN would require \( \approx 5 \) times more negative GDD. 
Thus, the use of CQN critically helps the balance between

SPM and GDD. CQN offer a large effective nonlinear refractive 
index contribution \( n_{\text{eff}}^{(2)} \), which depends on the second-
order nonlinear coefficient \( d_{\text{eff}} \) and the phase mismatch 
\( \Delta k = k_{\text{SH}} - 2k_{\text{FW}} \), where SH stands for second harmonic 
and FW for fundamental wave. This \( n_{\text{eff}}^{(2)} \) can be tuned in sign 
and magnitude via \( kL \) [12]. In this laser experiment, we exploit 
a negative \( n_{\text{eff}}^{(2)} \) from a SHG crystal in order to pick up a nega-
tive nonlinear phase shift, which counteracts the positive one 
picked up in air. A potential drawback of this technique is the 
loss caused by the SH generated in the cascading processes, since 
the SH light is not resonant in the laser cavity. The SHG 
efficiency scales with the peak intensity; hence, it represents an 
inverse saturable loss. On the other hand, if such losses are small 
compared to the modulation depth of the SESAM \( AR \), this prop-
erty can stabilize the modelocking process [16,27]. In order to 
minimize the second-harmonic losses, we operate the crystal near 
the SHG minima, which correspond to \( \Delta kL \approx 2\pi n_{\text{min}} \), where \( L \) 
is the length of the crystal and \( n_{\text{min}} \) is an integer. Experimentally, 
we monitor the SHG losses measuring the power of a cavity green 
leakage (“Photodiode” in Fig. 2) and adjust the crystal’s tilt angle 
\( \theta \) through a piezo-controlled mount. In this way, we can operate 
the crystal in the SHG minima.

To quantify the losses and the phase shift introduced by the 
CQN device, let us consider a pulse with peak intensity \( I_{pk} \), pro-
gressing through the SHG crystal. We call the phase shift intro-
duced for the peak of the pulse \( B_{\text{CQN},p} \) and the efficiency of the 
SHG process \( \eta_{\text{CQN},p} \):

\[
B_{\text{CQN},p} \approx \frac{-\xi L I_{pk}}{\Delta k}, \quad (1a)
\]

\[
\eta_{\text{CQN},p} \approx 0.835(\delta L)^2 I_{pk}/(\Delta k \tau_p)^2, \quad (1b)
\]

where we define a group-velocity mismatch parameter 
\( \delta = 1/\nu_{\text{SH}} - 1/\nu_{\text{FW}}, \quad \xi = 2(\alpha_{\text{nr}} d_{\text{eff}}^2)/\nu_{\text{cr}}^2 e^\xi (\nu_{\text{cr}}^3/\gamma n_{\text{abs}})^n_{\text{abs}} \), and 
\( \tau_p \) is the full-width-at-half-maximum (FWHM) 
duration of the pulse, assuming a sech^2 shape. These equations assume the 
cascading regime, where the phase mismatch is large and the 
transfer of energy from the fundamental to the second harmonic 
is small. The phase shift presented in Eq. (1a) has a well-known 
expression in literature [28]. We obtain Eq. (1b) in the supple-
mentary material assuming a short crystal fulfilling \( \tau_p > 2\delta L \), 
together with a large enough \( \Delta k \), and operation in a SHG 
minimum (i.e., \( \Delta kL = 2\pi n_{\text{min}} \)). In this short-crystal regime, 
the phase mismatch \( \Delta k(\lambda) \) is close to \( 2\pi n_{\text{min}} \) across the whole 
pulse spectrum, allowing for very low SHG losses for the
intracavity pulse. Hence, the ratio between nonlinear phase shift [Eq. (1a)] and nonlinear losses [Eq. (1b)] is lower than in the long-crystal limit ($\tau_p < \delta L$) [13]. Additionally, short crystals are beneficial in high-power applications in order to minimize thermal lensing.

The free parameters in the design of the CQN device are the crystal length $L$, the intensity on the crystal $I_{pk}$, adjustable through the laser spot size on the SHG crystal, and the phase mismatch $\Delta k$. The goal is to get a large amount of negative phase shift and as little as possible SHG losses, i.e., to maximize $B_{\text{CQN,avg}}/\eta_{\text{CQN,avg}} \sim \Delta k/L$. Thus, our formulas suggest to use short crystals operated at large phase-mismatch angles. We employed an AR-coated type-I LBO crystal (Cristal Laser) with a length $L = 5$ mm, in a position where the $1/e^2$ beam radius is $\approx 850$ $\mu$m. In this way, we have a peak intensity on the crystal below $5$ GW/cm$^2$.

We next consider the balance of the different sources contributing to the cavity SPM. The total phase shift $B_{\text{CQN,avg}}$ and losses $\eta_{\text{CQN,avg}}$ per round trip due to the SHG crystal are obtained multiplying the single-pass values [Eqs. (1a) and (1b)] for $(1 + R_{\text{OC}})$ where $R_{\text{OC}} = 60\%$ is the reflectivity of the OC. A convenient way to express the phase shift is to introduce the SPM coefficient $\gamma = B/P_{\text{pk,IC}}$, where $P_{\text{pk,IC}}$ is the intracavity peak power immediately before the OC. Regarding the air, we integrate the peak intensity in a cavity round trip to obtain the total SPM, denoted $B_{\text{air,avg}}$ (Supplement 1), and we obtain $\gamma_{\text{air}} \approx 10.6$ mrad/MW. In Fig. 3, we plot the expected losses $\eta_{\text{CQN,avg}}$ and the SPM coefficients $\gamma_{\text{CQN}}$ for the CQN device, according to our analytical model (green) and a numerical simulation (blue). We use $d_{\text{eff}} = 0.83$ pm/V for LBO [29]. We obtained the numerical solution by directly solving the pulsed coupled-wave equations for the laser parameters at the maximum output power ($\tau_p = 780$ fs, $P_{\text{pk,IC}} = 54$ MW). For the intrinsic nonlinear refractive index of the LBO, we use $2 \times 10^{-16}$ cm$^2$/W [30]. The analytical model accurately predicts the SHG losses in the minima and the phase shift. The positive contribution to the phase shift from the crystal’s intrinsic $n_2$ leads to a slightly less negative SPM coefficient $\gamma_{\text{CQN}}$ in the numerical model compared to the analytical solution, since this term is not included in the latter. The other sources of SPM, e.g., the disk, contribute only by few percent and so have been neglected.

 Femtosecond SESAM-modelocked lasers rely on soliton pulse formation. In this regime of SESAM modelocking, pulse duration and intracavity pulse energy $E_{\text{IC}} = E_{\text{out}}/T_{\text{OC}}$ depend mostly on the GDD versus SPM balance and only marginally on the parameters of the saturable absorber [10]. Their relation is governed by the so-called soliton formula, $\tau_p \approx 1.76(2|D|)/(\gamma^{\text{soliton}}E_{\text{IC}})$, where $\gamma^{\text{soliton}} = \gamma_c/4$ takes into account the effective phase shift for a pulse with a Gaussian spatial profile compared to the phase shift for the peak of the pulse [16,31]. By tuning the phase mismatch $\Delta k$, we can adjust the net SPM coefficient $\gamma$ [Fig. 3(b)]. Thanks to the straightforward tunability of $\Delta k$ by adapting the crystal’s tilt during live laser operation, we obtain the shortest pulse duration for several values of the output power (cfr. Fig. 4 and Table 1). In contrast, a standard TDL, having a fixed amount of GDD and SPM, operates only over a fixed power range and has the shortest pulses only at the maximum output power. In Fig. 4 we present the laser output power versus pump power for three phase-matching configurations. The blue and red curves are obtained operating the SHG crystal, respectively, in the fourth ($\Delta kL \approx 8\pi$) and third ($\Delta kL \approx 6\pi$) SHG minimum. The slope in yellow is obtained starting from the third SHG minimum and gradually decreasing the $\Delta k$ as the pump power is increased, in order to reduce the net SPM coefficient $\gamma$. Like this, we keep the pulse duration equal to the minimum achievable for our laser, but at increased output power. At the maximum output power ($210$ W, $780$ fs), we measured a SHG efficiency $\approx 1.8$ times the one we had in the third SHG minimum. This suggests a shift in $\Delta kL$ from the third SHG minimum of $\approx -0.2\pi$, i.e., $\Delta kL \approx 5.8\pi$. For this value of $\Delta kL$, we have $\gamma_{\text{CQN}} \approx -2.1 \times 10^{-15}$ cm$^2$/W [28].

Next, in Table 1, we quantify the SPM cancellation effect occurring in the laser for several operating points. Except for the point at $210$-W output power, we experimentally optimized

![Fig. 3. Round-trip SHG losses (a) and SPM cancellation (b) due to the CQN device. By operating the crystal in a SHG minimum, few-0.1% losses can be obtained while canceling most of the SPM from air.](image-url)

![Fig. 4. Laser slopes: output power (a) and pulse duration (b) as a function of the pump power. Different colors refer to different phase mismatch values $\Delta k$ of the SHG crystal.](image-url)

| $\Delta kL$ | $P_{\text{out}}$ (W) | $\tau_p$ (fs) | $\gamma_{\text{soliton}}$ (mrad/MW) | $\gamma_{\text{air}}$ (mrad/MW) | $\gamma_{\text{CQN}}$ (mrad/MW) | $T_{\text{OC}}$ (fs) |
|------------|----------------------|---------------|-----------------------------------|-----------------------------|-----------------------------|--------------------|
| $8\pi$     | 210                  | 782           | 2.1                               | $-8.5$                      | $-8.6$                      | 81%                |
| $6\pi$     | 162                  | 805           | 2.6                               | $-8.0$                      | $-8.3$                      | 78%                |
| $8\pi$     | 112                  | 741           | 4.2                               | $-6.4$                      | $-6.2$                      | 59%                |
| $10\pi$    | 85                   | 789           | 5.5                               | $-5.1$                      | $-5.0$                      | 47%                |
| $12\pi$    | 72                   | 782           | 6.2                               | $-4.4$                      | $-4.1$                      | 39%                |
| $14\pi$    | 61                   | 865           | 6.6                               | $-4.0$                      | $-3.6$                      | 33%                |

$\gamma_{\text{soliton}}$ is obtained from the soliton formula $\gamma_{\text{air}} = 10.6$ mrad/MW, and $\gamma_{\text{CQN}}$ is the expected negative SPM coefficient from the CQN device. The last column represents the fraction of SPM from air canceled by CQN.
We expect terms to this SPM coefficient are the intracavity air.

In the presented laser, the output power was limited by diffraction-limited beam quality ($M^2 < 1.05$) in all configurations. In the presented laser, the output power was limited by the pump intensity on the disk, already close to the safety limit.

In conclusion, we demonstrated a novel concept to cancel the SPM picked up in air in the context of high-power ultrafast oscillators. This allowed us to obtain laser performance in line with best-in-class TDLs using, instead of a complex vacuum system, an inexpensive and easy-to-set-up nonlinear crystal. Next to SESAM-modelocked TDL, this technique can be applied to high-power KLM oscillators. Additionally, we prove here that self-defocusing nonlinearities can be used at unprecedented power levels of up to 500 W intracavity power, hence offering a new toolset for high-average-power lasers.

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See Supplement 1 for supporting content.

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