Record Fifth-Harmonic–Generation Efficiency Producing 211-nm Pulses Using Cesium Lithium Borate

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Cesium lithium borate (CLBO) is a promising option for high-energy coherent-light generation in the UV region.

- High-energy coherent-light sources around 200 nm are necessary for diagnosing hot and dense plasmas.
- Wide-aperture fifth-harmonic generation (5HG) of Nd:YLF laser radiation has been realized with a cascade of deuterated potassium dihydrogen phosphate (DKDP), potassium dihydrogen phosphate (KDP), and CLBO crystals:
  - 275 mJ at 211 nm was reached with a 2.4-ns pulse
  - A conversion efficiency of 25% is the highest reported.
- The main limitations are two-photon absorption of fifth-harmonic radiation and a temperature gradient over the CLBO crystal.

High-energy, high-efficiency fifth-harmonic generation has been demonstrated with a large-aperture CLBO crystal.
Collaborators

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A 200-nm source is desirable to probe a high-density hot plasma

Frequency conversion of a 100-J, 1053-nm laser to the fifth harmonic is required to produce 10 J at 211 nm.

**TPD**: two-plasmon decay

**SRS**: stimulated Raman scattering

**SBS**: stimulated Brillouin scattering
Generating multiple joules at $5\omega$ reduces the crystal options to the KDP group and CLBO

- First 5HG in 1969*
- Wide-aperture, high-efficient 5HG in ammonium dihydrogen phosphate (ADP) at $-70^\circ\text{C}$**
- The CLBO crystal grew to $140 \times 110 \times 110 \text{ mm}^3$***

Second-harmonic generation (SHG):
DKDP, Type II, $30 \text{ mm} \times 30 \text{ mm} \times 27 \text{ mm}$

4HG: KDP, Type I, $30 \text{ mm} \times 30 \text{ mm} \times 15 \text{ mm}$

5HG: CLBO, Type I, $30 \text{ mm diam} \times 4 \text{ mm thick}$

The input polarization angle $\alpha$ was optimized.


o: ordinary
e: extraordinary
CLBO crystals in ovens were manufactured by Coherent, Inc.

The CLBO crystal is enclosed in an oven with dry nitrogen and held at 120°C to avoid hygroscopic damage to the surfaces.
The experiments were performed at LLE using the Multi-Terawatt (MTW) laser.

The narrowband mode of the MTW laser: \( E = 1.5 \text{ J (0.5\% rms)} \); beam size = \( 1.1 \times 1.1 \text{ cm}^2 \); \( \tau = 1 \text{ ns to 2.8 ns} \); \( I = 1.1 \text{ GW/cm}^2 \).
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The 5HG was performed with various flattop pulses.
Measured angular and temperature acceptances of $5\omega$ agree with simulations

**Angular acceptance**

- $\Delta\theta_{\text{FWHM}} = 1.7 \text{ mrad}$
- (calculated 1.65 mrad)

**Temperature acceptance**

- $\Delta T_{\text{FWHM}} = 7.1 ^\circ \text{C}$

- 4-mm long CLBO

![Graph showing angular and temperature acceptances with data points and curves]
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The maximum $5\omega$ conversion efficiency was reached with a 1-ns pulse.

The graph shows the relationship between input energy (mJ) and intensity (GW/cm²), with $5\omega$ efficiency and balance plotted on the y-axis. The formula $\eta (5\omega) = \frac{E_{5\omega}}{E_{\omega \text{ in}}}$ is also indicated in the graph.
The maximum $5\omega$ conversion efficiency was reached with a 1-ns pulse.

\begin{itemize}
  \item FR$_{5\omega}^* = E_{5\omega}/E_{\text{res (total)}} = 50\% \text{ (max).}$
  \item FR$_{4\omega} > 5\omega = 70\% \text{ (max).}$
\end{itemize}

\*FR: fraction
Two-photon absorption is the main fundamental limit for $5\omega$ generation in CLBO

- The energy balance (●) is decreased significantly by two-photon absorption.
- Two-photon absorption of $(4\omega + 4\omega)$ is relatively low.

$t = 1\text{ ns}$
Higher conversion efficiency would be possible if the $5\omega$ phase matching was uniform over the crystal.
The $5\omega$ beam nonuniformity comes from a temperature gradient over the CLBO crystal.

Crystal temperature ($^\circ$C)

$5\omega$ output

$5\omega$ tuned up

$5\omega$ tuned down

Fringe levels ($^\circ$C)
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Fifth-harmonic generation has been realized in a cascade frequency conversion

\[ E_\omega \text{ in} \]

\[ \eta_{2\omega} = \frac{E_{2\omega}}{E_\omega \text{ in}} \]

\[ \eta_{4\omega} = \frac{E_{4\omega}}{E_\omega \text{ in}} \]

\[ \eta_{5\omega} = \frac{E_{5\omega}}{E_\omega \text{ in}} \]

\[ FR_{2\omega} = \frac{E_{2\omega}}{E_\omega \text{ in}} \]

\[ FR_{2\omega} > 4\omega = \frac{E_{4\omega}}{E_\omega \text{ in}} \]

\[ FR_{2\omega} = \frac{E_{2\omega} \text{ res} + E_{4\omega} \text{ res} + 0.8 \times E_{5\omega}}{E_\omega \text{ in}} \]

\[ FR_{4\omega} > 5\omega = \frac{E_{5\omega}}{E_\omega \text{ in}} \]

\[ FR_{2\omega} > 4\omega = \frac{E_{4\omega} \text{ res} + 0.8 \times E_{5\omega}}{E_{2\omega} \text{ res} + E_{4\omega} \text{ res} + 0.8 \times E_{5\omega}} \]

\[ Balance = \frac{(E_\omega \text{ res} + E_{2\omega} \text{ res} + E_{4\omega} \text{ res} + E_{5\omega})}{E_\omega \text{ in}} = \frac{E_\omega \text{ res (total)}}{E_\omega \text{ in}} \]

*AR: antireflection coating
How efficient is the fifth-harmonic–generation process?

\[ \tau = 1 \text{ ns} \]

Half the optical output energy is at \(5\omega\).
Damages have been found on both input and output surfaces of CLBO, even in the area not exposed by the laser beam.

Damage thresholds*

- $\omega$: 1-on-1: $12.09 \pm 0.61$ J/cm$^2$, N-on-1: $16.36 \pm 3.26$ J/cm$^2$
  (1.4-ns Gaussian pulse)
- $2\omega$: 1-on-1: $5.11 \pm 0.26$ J/cm$^2$, N-on-1: $8.87 \pm 0.97$ J/cm$^2$
  (1.2-ns Gaussian pulse)

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*A. A. Kozlov and S. Papernov, tested in the LLE Damage Test Laboratory (2016).
The transmission response of the CLBO oven assembly

The CLBO oven input window is AR coated for $1\omega$ and $4\omega$; the crystal and output window are uncoated.