

## Supporting Information

### **K<sub>3</sub>Sr<sub>3</sub>Li<sub>2</sub>Al<sub>4</sub>B<sub>6</sub>O<sub>20</sub>F: A Competitive Nonlinear Optical Crystal for Generation of 266 nm Laser**

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**Table S1.** Comparison of the refractive indices between the experimental and calculated values for  $\text{K}_3\text{Sr}_3\text{Li}_2\text{Al}_4\text{B}_6\text{O}_{20}\text{F}$ .

| wavelength(nm) | $n_e$<br>(exp.) | $n_e$ (cal.) | error                  | $n_o$<br>(exp.) | $n_o$<br>(cal.) | error                  | $\Delta n$ |
|----------------|-----------------|--------------|------------------------|-----------------|-----------------|------------------------|------------|
| 363.0          | 1.5480607       | 1.5480964    | $2.11 \times 10^{-6}$  | 1.6140911       | 1.614131        | $8.41 \times 10^{-6}$  | 0.0660304  |
| 404.7          | 1.5413516       | 1.5412506    | $-2.26 \times 10^{-6}$ | 1.6058339       | 1.6057219       | $-9.10 \times 10^{-6}$ | 0.0644823  |
| 435.8          | 1.5374093       | 1.5373977    | $4.68 \times 10^{-6}$  | 1.6009877       | 1.6009927       | $-7.18 \times 10^{-6}$ | 0.0635784  |
| 480.0          | 1.5330321       | 1.5331759    | $1.83 \times 10^{-5}$  | 1.5957001       | 1.5958045       | $3.08 \times 10^{-6}$  | 0.062668   |
| 546.1          | 1.5287139       | 1.5286895    | $3.59 \times 10^{-5}$  | 1.5902523       | 1.5902645       | $1.99 \times 10^{-5}$  | 0.0615384  |
| 587.5          | 1.5266605       | 1.5266215    | $4.27 \times 10^{-5}$  | 1.5877042       | 1.5876893       | $2.76 \times 10^{-5}$  | 0.0610437  |
| 643.8          | 1.5244593       | 1.5244239    | $4.60 \times 10^{-5}$  | 1.5849692       | 1.5849227       | $3.32 \times 10^{-5}$  | 0.0605099  |
| 706.5          | 1.5225345       | 1.5225647    | $4.21 \times 10^{-5}$  | 1.5825412       | 1.58254         | $3.28 \times 10^{-5}$  | 0.0600067  |
| 852.1          | 1.5196966       | 1.5196984    | $6.86 \times 10^{-6}$  | 1.5786911       | 1.5787041       | $8.05 \times 10^{-6}$  | 0.0589945  |

$$\Delta n = n_o - n_e$$

**Table S2.** Sellmeier coefficients derived from the measured refractive indices.

$$n^2 = a + \frac{b}{\lambda^2 - c} - d\lambda^2 \quad (1)$$

|   | n <sub>o</sub> | n <sub>e</sub> |
|---|----------------|----------------|
| a | 2.474073       | 2.291630       |
| b | 0.016601       | 0.013379       |
| c | 0.006212       | 0.004471       |
| d | 0.006649       | 0.0009463      |

**Table S3.** Comparison of NLO properties among BBO, CLBO, and  $\text{K}_3\text{Sr}_3\text{Li}_2\text{Al}_4\text{B}_6\text{O}_{20}\text{F}$  at 532 nm.

| Crystal                                             | BBO   | CLBO  | $\text{K}_3\text{Sr}_3\text{Li}_2\text{Al}_4\text{B}_6\text{O}_{20}\text{F}$ |
|-----------------------------------------------------|-------|-------|------------------------------------------------------------------------------|
| Birefringence                                       | 0.120 | 0.053 | 0.063                                                                        |
| Phase-matching angle ( $\theta/^\circ$ )            | 47.5  | 61.5  | 64.8                                                                         |
| Walk-off angle ( $\theta/^\circ$ )                  | 4.8   | 1.83  | 1.92                                                                         |
| Acceptance angle ( $\text{mrad} \times \text{mm}$ ) | 0.95  | 2.68  | 2.49                                                                         |

The formulas for the walk-off angle and acceptance angle are as follows:

$$\alpha = \arctan \left[ \frac{1}{2} \frac{(n_e^2 - n_o^2)}{n_o^2 \sin^2 \theta + n_e^2 \cos^2 \theta} \sin(2\theta) \right] \quad (2)$$

$$\Delta\theta(\text{I}) = \pm \left| \frac{\pi L}{\frac{\omega_1}{c} n_e^3(2\omega_1, \theta_m) [n_e^{-2}(2\omega_1) \sin(2\theta_m)]} \right| \quad (3)$$

**Table S4.** Surface laser damage threshold values of  $\text{K}_3\text{Sr}_3\text{Li}_2\text{Al}_4\text{B}_6\text{O}_{20}\text{F}$  (KSLABF) before and after humidity (50 %) exposure in the air and  $\beta\text{-BaB}_2\text{O}_4$ .

| Compound                       | Laser beam<br>(nm) | Pulse width<br>(ns) | LIDT<br>(J/cm <sup>2</sup> ) | IDT<br>(GW/cm <sup>2</sup> ) |
|--------------------------------|--------------------|---------------------|------------------------------|------------------------------|
| $\beta\text{-BaB}_2\text{O}_4$ | 1064               | 5                   | 19.24                        | 3.85                         |
| KSLABF                         | 1064               | 5                   | 21.80                        | 4.36                         |
| KSLABF (after four months)     | 1064               | 5                   | 22.05                        | 4.41                         |

**Figure S1.** Optical path about laser output.

