# Supplemental Information

## Optimizing Pulsed-Laser Ablation Production of AICI Molecules for Laser Cooling

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**Figure S1.** (A) Sample target with various AI:KCI molar ratios, as labelled in pink (1) 10:1, (2) 8:1, (3) 5:1, and (4) 1:4. (B) Target mappings for absorption of AICI, AI, and K. The color map on the right shows the variation in AICI absorption from 0 (dark blue) to 1 (yellow).



#### Simulated XCI<sub>n</sub> Sources

**Figure S2.** Simulated  $R_{mol}$  curves for different XCI<sub>n</sub> sources from Model A (A) and Model A' (B). With a fixed scaling factor ( $\gamma$ ), the optical density of each source in Model A is roughly 3 times larger than Model A' due to the aluminum recondensation.

**Table S1.** The  $R_{mol}^{max}$  found for each XCI<sub>n</sub> source from Model A and Model A'. These values are used to plot against the  $\rho_{Cl}$  in Figure 7B.

	Model A	Model A'
	$R_{mol}^{max}$	$R_{mol}^{max}$
KCI	3.75	1.55
NaCl	2.69	1.11
CaCl <sub>2</sub>	2.58	1.07
MgCl <sub>2</sub>	2.39	0.99
	1.79	0.74

#### Average Absorption from Time-Dependent Traces



**Figure S3.** Theory is compared to the experimental optical density signal from each chloride source at  $R_{mol} = 0.25$ . The experimental date is acquired by taking the peak optical density value from each time trace (A) and integrated over the time trace (B). Models A and A' are normalized to the AlCl<sub>3</sub> optical density.

#### Al Density Calculation

If we assume our ablation crater to be cone-shaped, we can calculate the volume of the ablation cone estimating a crater depth of 6  $\mu$ m<sup>1</sup> and our known ablation laser spot size of 80  $\mu$ m.

$$V = \frac{1}{3} \left[ \frac{1}{2} \pi r^2 \right] h$$

The resulting volume of sample ablated is  $5.03 \cdot 10^{-9}$  cm<sup>3</sup>. Then we can use this volume and stoichiometrically convert aluminum density to moles of aluminum in the gas phase after ablation,

$$2.70 \frac{g}{cm^3} \times 5.03 \cdot 10^{-9} cm^3 \div 26.98 \frac{g}{mol} \times \frac{6.022 \cdot 10^{23}}{mol}$$

to get  $3.03 \cdot 10^{14}$  atoms. We can then use our known volume of our cell (28 cm<sup>3</sup>) to calculate the maximum AI density after ablation,

$$\frac{3.03 \cdot 10^{14} atoms}{28 cm^3}$$
And our result is 
$$\frac{1.1 \cdot 10^{13} atoms}{cm^3}$$

### References

(1) U.P.B. Sci. Bull., Series A, Vol. 70, Iss. 4, 2008