

## Supporting Information

### **Identification of Interstitial Oxygen Color Centers in CaGdAlO<sub>4</sub>**

#### **Single Crystals and their Elimination Strategy**

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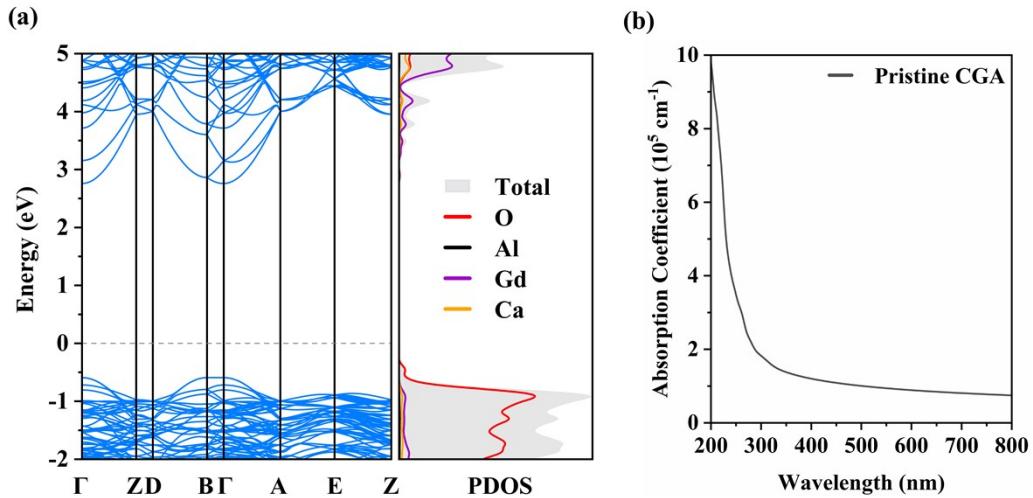
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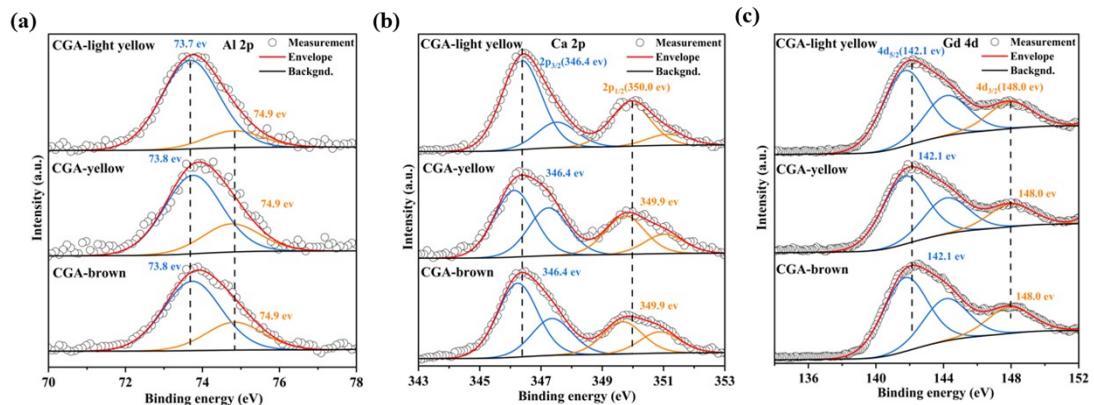
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**Table S1. System energy and O<sub>i</sub> doping energy**

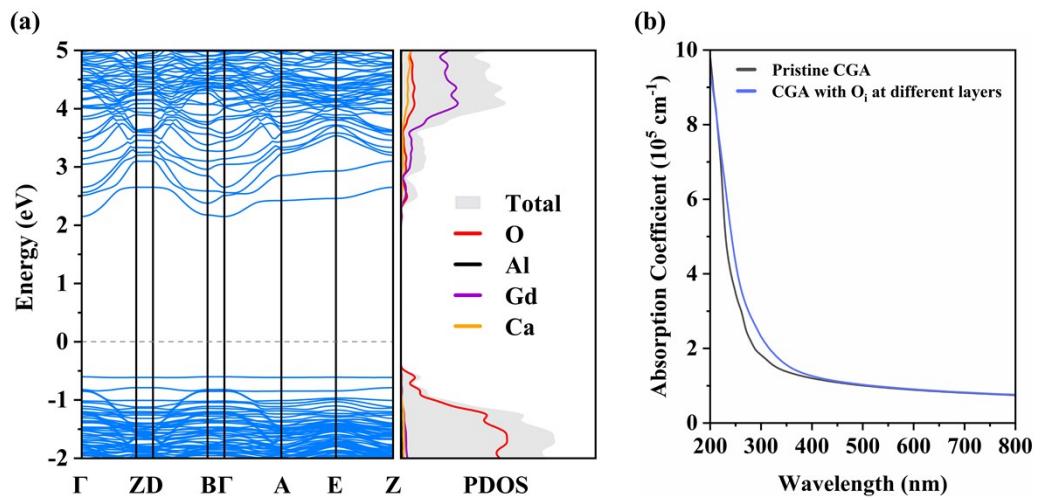
Interstice	Average energy of system (eV/atom)	E <sub>f</sub> (eV)
Non	-7.503	/
Al-O	-7.366	8.145
Gd/Ca-O	-7.448	-1.201



**Figure S1. (a) The electronic structure and (b) The absorption spectrum of pristine CGA**



**Figure S2. The XPS spectra of (a) Al 2p, (b) Ca 2p, and (c) Gd 4d for the CGA-light yellow, CGA-yellow, and CGA-brown samples.**



**Figure S3. (a) The electronic structure and (b) The absorption spectrum of CGA with  $\text{O}_i$  at different layers**