Supplementary Information

# Defect Induced Photoluminescence and Triboluminescence in Layered $CaLaAl_3O_7$

Yezhou Li,[a,b] Igor Koskin,[c] Zhidong Ma,[a] Enrico Benassi,[*a,c,d] Zhaofeng Wang[*a]

[a] *State Key Laboratory of Solid Lubrication, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, P. R. China. Email: ebenassi3@gmail.com (EB), zhfwang@licp.cas.cn(ZW)*

[b] *School of Information Science & Engineering, Gansu University of Chinese Medicine, Lanzhou 730000, China*

[c] *Faculty of Natural Sciences, Novosibirsk State University, Novosbirsk 630090, Russian Federation*

[d] *Public technical service centre, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, China*

## 1. Methods

### 1.1 Synthesis of $CaLaAl_3O_7$ powders

$CaLaAl_3O_7$ powders were prepared by a solid-state reaction method. First, stoichiometric raw materials $Ca_2CO_3$ (99.99%), $La_2O_3$ (99.99%), $Al_2O_3$ (AR) were homogeneously mixed in a agate mortar. Then, the mixture was transferred in a corundum crucible and sintered at 1600 $^oC$ for 4 h in a tube furnace (GSL-1600X) under air or reducing atmosphere (90% $N_2$, 10% $H_2$). Finally, the samples were obtained after naturally cooling to room temperature.

### 1.2 Fabrication of $CaLaAl_3O_7$/PDMS elastomer

Polydimethylsiloxane (PDMS, Sylgard 184, Dow Corning) was employed as the elastic matrix for $CaLaAl_3O_7$. First, 1.5 g of PDMS base resin and 0.15 g of curing agent were mixed in a petri dish with a diameter of 60 mm. Then, 3 g of $CaLaAl_3O_7$ powders were dispersed in the above mixture by mechanical stirring for 10 min. After curing at 60 ˚C for 4 h, the $CaLaAl_3O_7$/PDMS elastomer was obtained.

### 1.3 Computational details

The influence of compression on the crystal structure of $CaLaAl_3O_7$ was theoretically investigated to fully explain the observed TL. The crystal structure of $CaLaAl_3O_7$ was fully relaxed (both atomic positions and unit cell parameters) utilizing the Plane Wave Self-Consistent Field (PW-SCF) approach coupled with the Generalized Gradient Approximation (GGA) PBE[1,2] functional and with Projector Augmented Wave (PAW). After full relaxation of $CaLaAl_3O_7$ cell, isotopically compressed by multiplier (from 0.99 to 0.85 with 0.01 step) was build and then the atomic positions were relaxed. In

order to simulate anisotropic compression of $CaLaAl_3O_7$ cell compressed in each crystallographic direction by 0.95, 0.90 and 0.85 multipliers and then relaxed.

Further post processing of electron density required additional SCF calculation of relaxed cell with high-density grid (~0.01 Å in each direction). Atomic charges were computed by integrating atomic basins (evaluated from high-density grid SCF calculations) by Yu and Trinkle integration scheme. Projected DOS evaluations for $CaLaAl_3O_7$ cell were performed based on high-density grid SCF calculation.

Same kinetic energy cut-off (60 Ry), electron density energy cut-off (600 Ry) were maintained throughout whole computational study. All geometry optimizations were made in three consecutive steps with 2×2×2, 4×4×4 and 6×6×6 K-points grid in order to aid troubled SCF convergence.

All geometry optimizations, SCF and PDOS analysis were performed with the Quantum Espresso v6.4 computational package.[3] Electron density post processing was performed using CRITIC2 code.[4] The results of cells optimizations were visualized using VESTA program.[5]

**1.4 Characterizations**

A Rigaku D/Max-2400 X-ray diffractometer was used to measure the X-ray diffraction (XRD) patterns of $CaLaAl_3O_7$ powders. The morphology and microstructure of the samples were examined by SEM (S-340, Hitachi, Japan). The absorption spectra were collected by a UV-visible (UV-vis) spectrophotometer (lamda950; PerkinElmer Inc.) using $BaSO_4$ as a reference. The PL spectra were obtained by using a FLS-920T fluorescence spectrophotometer with a Xe 900 lamp (450 W) under the scanning step

of 1 nm. To avoid the light source interference of 350 nm, a 372 nm filter was employed on the detection side. TL signals were in situ collected via an optical fiber connected with a collimator (BFC-441; Zolix Instruments Co., Ltd.). The collected signals were then transferred into a spectrometer (Omni-λ300i; Zolix Instruments Co., Ltd.) equipped with a CCD camera (iVac-316; Edmund Optics Ltd.). During TL test, the applied normal load and rubbing speed are 5N and 200 rpm, respectively. The ThL experiments were performed using a FJ-427A ThL detector with the temperature range from 20 to 400 °C and a heating rate of 1 K/s, and the detected signals were in range from 300 to 850 nm.

## 2. Additional experimental data

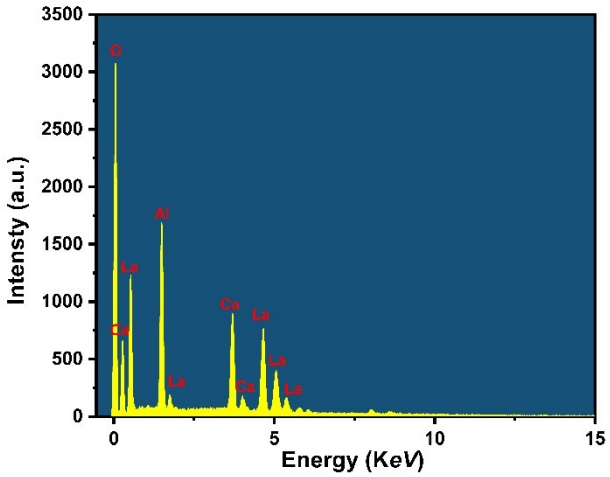


**Figure S1.** EDS spectrum of the as-prepared $CaLaAl_3O_7$.

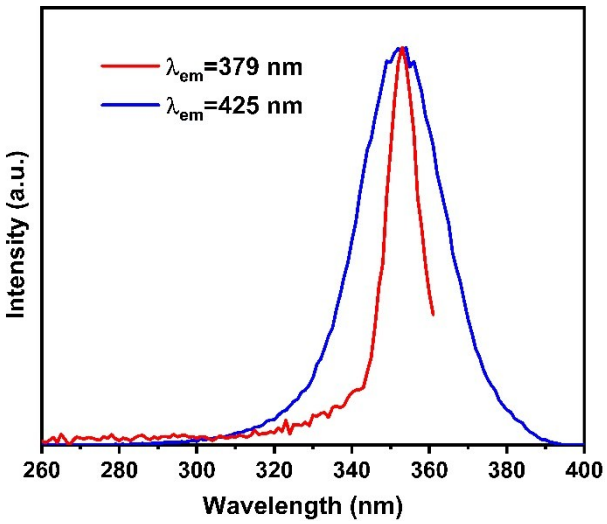


**Figure S2.** Excitation spectra of $CaLaAl_3O_7$ monitored at 379 and 425 nm.

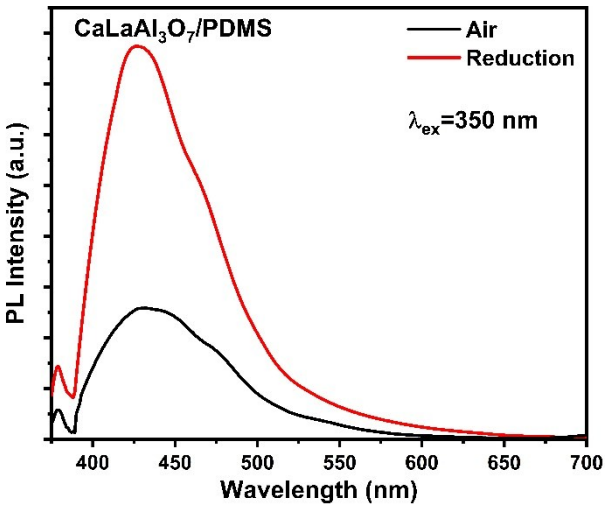


**Figure S3.** PL spectra of $CaLaAl_3O_7$/PDMS elastomer synthesized at air and reduction atmosphere, respectively. It suggests that PDMS has no interference on the luminescent properties of $CaLaAl_3O_7$.

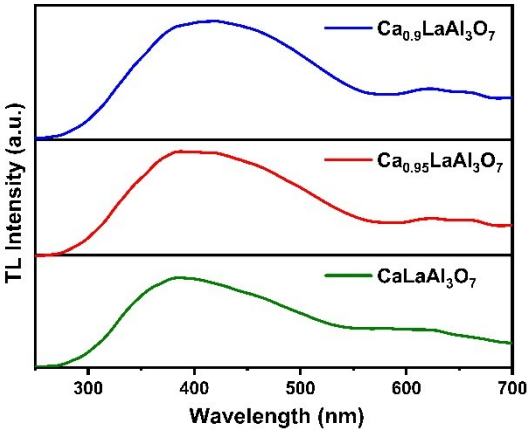


Figure S4. TL spectra of $Ca_{1-x}LaAl_3O_7$ ($x$=0, 0.05, 0.10) synthesized under air atmosphere. It is observed that with the increase of Ca vacancies, the red TL emission of $CaLaAl_3O_7$ gradually increases. This directly supports that the newly observed red emission band in the TL spectra of $CaLaAl_3O_7$ should come from the Ca vacancies in structure.

## 2. Computational data

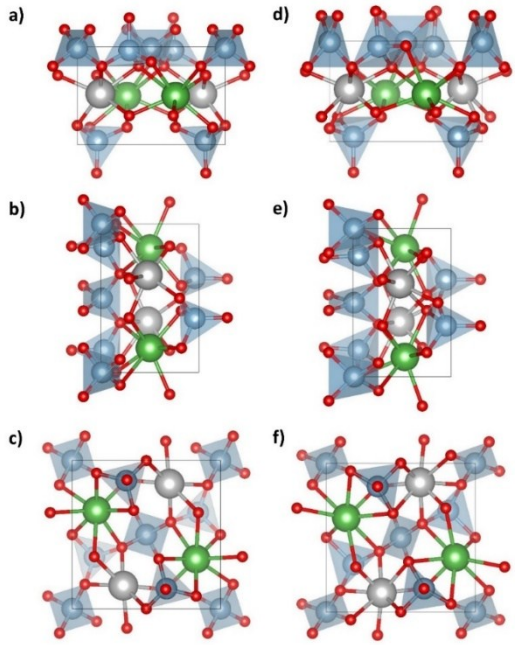


**Figure S5**. Fully optimised $CaLaAl_3O_7$ cell (a-c) and relaxed isotropically compressed (0.85 multiplier) cell (d-e) shown alongside a-axis (a, d); alongside b-axis (b,e); alongside c-axis (c,f). Red color stands for O atoms, blue – Al, silver – Ca, green – La. Cell volume is kept the same for comparison clarity.

**Table S1**. Atomic positions of fully relaxed $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.164098530 | 0.664098530 | 0.501909253 |
| Ca | 0.344079287 | 0.155920713 | 0.478851824 |
| Ca | 0.655920713 | 0.844079287 | 0.478851824 |
| La | 0.835901470 | 0.335901470 | 0.501909253 |
| Al | 0.000000000 | 0.000000000 | 0.012962363 |
| Al | 0.500000000 | 0.500000000 | 0.012962363 |
| Al | 0.364241380 | 0.864241380 | 0.959469564 |
| Al | 0.150135081 | 0.349864919 | 0.034865959 |
| Al | 0.849864919 | 0.650135081 | 0.034865959 |
| Al | 0.635758620 | 0.135758620 | 0.959469564 |
| O | 0.369027170 | 0.869027170 | 0.290601987 |
| O | 0.148750159 | 0.351249841 | 0.703181663 |
| O | 0.851249841 | 0.648750159 | 0.703181663 |
| O | 0.630972830 | 0.130972830 | 0.290601987 |
| O | 0.335078186 | 0.414094325 | 0.196872367 |
| O | 0.164122082 | 0.914737979 | 0.822883822 |
| O | 0.414737979 | 0.664122082 | 0.822883822 |
| O | 0.085905675 | 0.164921814 | 0.196872367 |
| O | 0.664921814 | 0.585905675 | 0.196872367 |
| O | 0.585262021 | 0.335877918 | 0.822883822 |
| O | 0.914094325 | 0.835078186 | 0.196872367 |
| O | 0.835877918 | 0.085262021 | 0.822883822 |
| O | 0.000000000 | 0.500000000 | 0.152989729 |
| O | 0.500000000 | 0.000000000 | 0.804300289 |

**Table S2**. Atomic positions of relaxed isotropically compressed (0.99) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.163802386 | 0.663802386 | 0.500610926 |
| Ca | 0.344125296 | 0.155874704 | 0.481102868 |
| Ca | 0.655874704 | 0.844125296 | 0.481102868 |
| La | 0.836197614 | 0.336197614 | 0.500610926 |
| Al | 0.000000000 | 0.000000000 | 0.013945274 |
| Al | 0.500000000 | 0.500000000 | 0.013945274 |
| Al | 0.364291425 | 0.864291425 | 0.959240289 |
| Al | 0.151541926 | 0.348458074 | 0.034639616 |
| Al | 0.848458074 | 0.651541926 | 0.034639616 |
| Al | 0.635708575 | 0.135708575 | 0.959240289 |
| O | 0.369547045 | 0.869547045 | 0.291724233 |
| O | 0.150347814 | 0.349652186 | 0.701516041 |
| O | 0.849652186 | 0.650347814 | 0.701516041 |
| O | 0.630452955 | 0.130452955 | 0.291724233 |
| O | 0.335800500 | 0.413722182 | 0.200927264 |
| O | 0.164059478 | 0.916314027 | 0.820007643 |
| O | 0.416314027 | 0.664059478 | 0.820007643 |

| | | | |
|---|---|---|---|
| O | 0.086277818 | 0.164199500 | 0.200927264 |
| O | 0.664199500 | 0.586277818 | 0.200927264 |
| O | 0.583685973 | 0.335940522 | 0.820007643 |
| O | 0.913722182 | 0.835800500 | 0.200927264 |
| O | 0.835940522 | 0.083685973 | 0.820007643 |
| O | 0.000000000 | 0.500000000 | 0.148506431 |
| O | 0.500000000 | 0.000000000 | 0.802195448 |

**Table S3**. Atomic positions of relaxed isotropically compressed (0.98) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.163391423 | 0.663391423 | 0.499670850 |
| Ca | 0.344342823 | 0.155657177 | 0.482938593 |
| Ca | 0.655657177 | 0.844342823 | 0.482938593 |
| La | 0.836608577 | 0.336608577 | 0.499670850 |
| Al | 0.000000000 | 0.000000000 | 0.014879093 |
| Al | 0.500000000 | 0.500000000 | 0.014879093 |
| Al | 0.364276999 | 0.864276999 | 0.959077206 |
| Al | 0.153030308 | 0.346969692 | 0.034499155 |
| Al | 0.846969692 | 0.653030308 | 0.034499155 |
| Al | 0.635723001 | 0.135723001 | 0.959077206 |
| O | 0.370011925 | 0.870011925 | 0.292884883 |
| O | 0.151855250 | 0.348144750 | 0.699998165 |
| O | 0.848144750 | 0.651855250 | 0.699998165 |
| O | 0.629988075 | 0.129988075 | 0.292884883 |
| O | 0.336610417 | 0.413230514 | 0.204941227 |
| O | 0.164031159 | 0.918044311 | 0.817025279 |
| O | 0.418044311 | 0.664031159 | 0.817025279 |
| O | 0.086769486 | 0.163389583 | 0.204941227 |
| O | 0.663389583 | 0.586769486 | 0.204941227 |
| O | 0.581955689 | 0.335968841 | 0.817025279 |
| O | 0.913230514 | 0.836610417 | 0.204941227 |
| O | 0.835968841 | 0.081955689 | 0.817025279 |
| O | 0.000000000 | 0.500000000 | 0.143774947 |
| O | 0.500000000 | 0.000000000 | 0.800463141 |

**Table S4**. Atomic positions of relaxed isotropically compressed (0.97) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.162877897 | 0.662877897 | 0.499047028 |
| Ca | 0.344705259 | 0.155294741 | 0.484564249 |
| Ca | 0.655294741 | 0.844705259 | 0.484564249 |
| La | 0.837122103 | 0.337122103 | 0.499047028 |
| Al | 0.000000000 | 0.000000000 | 0.015695050 |
| Al | 0.500000000 | 0.500000000 | 0.015695050 |
| Al | 0.364189254 | 0.864189254 | 0.958999438 |
| Al | 0.154603062 | 0.345396938 | 0.034460395 |

| | | | |
|---|---|---|---|
| Al | 0.845396938 | 0.654603062 | 0.034460395 |
| Al | 0.635810746 | 0.135810746 | 0.958999438 |
| O | 0.370396647 | 0.870396647 | 0.294093824 |
| O | 0.153321191 | 0.346678809 | 0.698630343 |
| O | 0.846678809 | 0.653321191 | 0.698630343 |
| O | 0.629603353 | 0.129603353 | 0.294093824 |
| O | 0.337495416 | 0.412686166 | 0.208906008 |
| O | 0.164073437 | 0.919994845 | 0.813866968 |
| O | 0.419994845 | 0.664073437 | 0.813866968 |
| O | 0.087313834 | 0.162504584 | 0.208906008 |
| O | 0.662504584 | 0.587313834 | 0.208906008 |
| O | 0.580005155 | 0.335926563 | 0.813866968 |
| O | 0.912686166 | 0.837495416 | 0.208906008 |
| O | 0.835926563 | 0.080005155 | 0.813866968 |
| O | 0.000000000 | 0.500000000 | 0.138770569 |
| O | 0.500000000 | 0.000000000 | 0.799156865 |

**Table S5**. Atomic positions of relaxed isotropically compressed (0.96) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.162266248 | 0.662266248 | 0.498716236 |
| Ca | 0.345178711 | 0.154821289 | 0.486081572 |
| Ca | 0.654821289 | 0.845178711 | 0.486081572 |
| La | 0.837733752 | 0.337733752 | 0.498716236 |
| Al | 0.000000000 | 0.000000000 | 0.016331326 |
| Al | 0.500000000 | 0.500000000 | 0.016331326 |
| Al | 0.364003398 | 0.864003398 | 0.958992700 |
| Al | 0.156258898 | 0.343741102 | 0.034571715 |
| Al | 0.843741102 | 0.656258898 | 0.034571715 |
| Al | 0.635996602 | 0.135996602 | 0.958992700 |
| O | 0.370675866 | 0.870675866 | 0.295335251 |
| O | 0.154777706 | 0.345222294 | 0.697456230 |
| O | 0.845222294 | 0.654777706 | 0.697456230 |
| O | 0.629324134 | 0.129324134 | 0.295335251 |
| O | 0.338472992 | 0.412094538 | 0.212810146 |
| O | 0.164186308 | 0.922171610 | 0.810479687 |
| O | 0.422171610 | 0.664186308 | 0.810479687 |
| O | 0.087905462 | 0.161527008 | 0.212810146 |
| O | 0.661527008 | 0.587905462 | 0.212810146 |
| O | 0.577828390 | 0.335813692 | 0.810479687 |
| O | 0.912094538 | 0.838472992 | 0.212810146 |
| O | 0.835813692 | 0.077828390 | 0.810479687 |
| O | 0.000000000 | 0.500000000 | 0.133524867 |
| O | 0.500000000 | 0.000000000 | 0.798345736 |

**Table S6**. Atomic positions of relaxed isotropically compressed (0.95) $CaLaAl_3O_7$

crystal cell.

| | | | |
|---|---|---|---|
| La | 0.161549975 | 0.661549975 | 0.498705291 |
| Ca | 0.345780501 | 0.154219499 | 0.487473688 |
| Ca | 0.654219499 | 0.845780501 | 0.487473688 |
| La | 0.838450025 | 0.338450025 | 0.498705291 |
| Al | 0.000000000 | 0.000000000 | 0.016739510 |
| Al | 0.500000000 | 0.500000000 | 0.016739510 |
| Al | 0.363703704 | 0.863703704 | 0.959037353 |
| Al | 0.157998257 | 0.342001743 | 0.034877724 |
| Al | 0.842001743 | 0.657998257 | 0.034877724 |
| Al | 0.636296296 | 0.136296296 | 0.959037353 |
| O | 0.370824411 | 0.870824411 | 0.296592189 |
| O | 0.156269833 | 0.343730167 | 0.696514814 |
| O | 0.843730167 | 0.656269833 | 0.696514814 |
| O | 0.629175589 | 0.129175589 | 0.296592189 |
| O | 0.339550168 | 0.411487258 | 0.216665264 |
| O | 0.164381153 | 0.924591550 | 0.806833894 |
| O | 0.424591550 | 0.664381153 | 0.806833894 |
| O | 0.088512742 | 0.160449832 | 0.216665264 |
| O | 0.660449832 | 0.588512742 | 0.216665264 |
| O | 0.575408450 | 0.335618847 | 0.806833894 |
| O | 0.911487258 | 0.839550168 | 0.216665264 |
| O | 0.835618847 | 0.075408450 | 0.806833894 |
| O | 0.000000000 | 0.500000000 | 0.128064750 |
| O | 0.500000000 | 0.000000000 | 0.798057475 |

**Table S7**. Atomic positions of relaxed isotropically compressed (0.94) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.160706393 | 0.660706393 | 0.498996184 |
| Ca | 0.346533690 | 0.153466310 | 0.488691801 |
| Ca | 0.653466310 | 0.846533690 | 0.488691801 |
| La | 0.839293607 | 0.339293607 | 0.498996184 |
| Al | 0.000000000 | 0.000000000 | 0.016974329 |
| Al | 0.500000000 | 0.500000000 | 0.016974329 |
| Al | 0.363253096 | 0.863253096 | 0.959106697 |
| Al | 0.159839332 | 0.340160668 | 0.035345874 |
| Al | 0.840160668 | 0.659839332 | 0.035345874 |
| Al | 0.636746904 | 0.136746904 | 0.959106697 |
| O | 0.370820328 | 0.870820328 | 0.297847480 |
| O | 0.157884734 | 0.342115266 | 0.695765237 |
| O | 0.842115266 | 0.657884734 | 0.695765237 |
| O | 0.629179672 | 0.129179672 | 0.297847480 |
| O | 0.340717543 | 0.410873135 | 0.220511645 |
| O | 0.164619419 | 0.927200126 | 0.802969722 |
| O | 0.427200126 | 0.664619419 | 0.802969722 |

| | | | |
|---|---|---|---|
| O | 0.089126865 | 0.159282457 | 0.220511645 |
| O | 0.659282457 | 0.589126865 | 0.220511645 |
| O | 0.572799874 | 0.335380581 | 0.802969722 |
| O | 0.910873135 | 0.840717543 | 0.220511645 |
| O | 0.835380581 | 0.072799874 | 0.802969722 |
| O | 0.000000000 | 0.500000000 | 0.122245978 |
| O | 0.500000000 | 0.000000000 | 0.798373347 |

**Table S8**. Atomic positions of relaxed isotropically compressed (0.93) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.159724899 | 0.659724899 | 0.499648560 |
| Ca | 0.347423642 | 0.152576358 | 0.489748950 |
| Ca | 0.652576358 | 0.847423642 | 0.489748950 |
| La | 0.840275101 | 0.340275101 | 0.499648560 |
| Al | 0.000000000 | 0.000000000 | 0.016941012 |
| Al | 0.500000000 | 0.500000000 | 0.016941012 |
| Al | 0.362625850 | 0.862625850 | 0.959147095 |
| Al | 0.161759811 | 0.338240189 | 0.036073591 |
| Al | 0.838240189 | 0.661759811 | 0.036073591 |
| Al | 0.637374150 | 0.137374150 | 0.959147095 |
| O | 0.370618571 | 0.870618571 | 0.299049811 |
| O | 0.159639574 | 0.340360426 | 0.695305981 |
| O | 0.840360426 | 0.659639574 | 0.695305981 |
| O | 0.629381429 | 0.129381429 | 0.299049811 |
| O | 0.342000256 | 0.410267346 | 0.224328943 |
| O | 0.164906406 | 0.930002568 | 0.798820911 |
| O | 0.430002568 | 0.664906406 | 0.798820911 |
| O | 0.089732654 | 0.157999744 | 0.224328943 |
| O | 0.657999744 | 0.589732654 | 0.224328943 |
| O | 0.569997432 | 0.335093594 | 0.798820911 |
| O | 0.910267346 | 0.842000256 | 0.224328943 |
| O | 0.835093594 | 0.069997432 | 0.798820911 |
| O | 0.000000000 | 0.500000000 | 0.116231469 |
| O | 0.500000000 | 0.000000000 | 0.799339115 |

**Table S9**. Atomic positions of relaxed isotropically compressed (0.92) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.158542600 | 0.658542600 | 0.500658656 |
| Ca | 0.348531630 | 0.151468370 | 0.490514210 |
| Ca | 0.651468370 | 0.848531630 | 0.490514210 |
| La | 0.841457400 | 0.341457400 | 0.500658656 |
| Al | 0.000000000 | 0.000000000 | 0.016728452 |
| Al | 0.500000000 | 0.500000000 | 0.016728452 |
| Al | 0.361767354 | 0.861767354 | 0.959142609 |
| Al | 0.163781340 | 0.336218660 | 0.036992832 |

| | | | |
|---|---|---|---|
| Al | 0.836218660 | 0.663781340 | 0.036992832 |
| Al | 0.638232646 | 0.138232646 | 0.959142609 |
| O | 0.370161267 | 0.870161267 | 0.300206976 |
| O | 0.161663219 | 0.338336781 | 0.695065876 |
| O | 0.838336781 | 0.661663219 | 0.695065876 |
| O | 0.629838733 | 0.129838733 | 0.300206976 |
| O | 0.343381587 | 0.409667278 | 0.228156339 |
| O | 0.165177658 | 0.932876322 | 0.794464550 |
| O | 0.432876322 | 0.665177658 | 0.794464550 |
| O | 0.090332722 | 0.156618413 | 0.228156339 |
| O | 0.656618413 | 0.590332722 | 0.228156339 |
| O | 0.567123678 | 0.334822342 | 0.794464550 |
| O | 0.909667278 | 0.843381587 | 0.228156339 |
| O | 0.834822342 | 0.067123678 | 0.794464550 |
| O | 0.000000000 | 0.500000000 | 0.109767699 |
| O | 0.500000000 | 0.000000000 | 0.801129520 |

**Table S10**. Atomic positions of relaxed isotropically compressed (0.91) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.157100479 | 0.657100479 | 0.502102771 |
| Ca | 0.349908444 | 0.150091556 | 0.490966785 |
| Ca | 0.650091556 | 0.849908444 | 0.490966785 |
| La | 0.842899521 | 0.342899521 | 0.502102771 |
| Al | 0.000000000 | 0.000000000 | 0.016247564 |
| Al | 0.500000000 | 0.500000000 | 0.016247564 |
| Al | 0.360614878 | 0.860614878 | 0.959028650 |
| Al | 0.165906333 | 0.334093667 | 0.038203554 |
| Al | 0.834093667 | 0.665906333 | 0.038203554 |
| Al | 0.639385122 | 0.139385122 | 0.959028650 |
| O | 0.369358266 | 0.869358266 | 0.301262785 |
| O | 0.164040217 | 0.335959783 | 0.695132620 |
| O | 0.835959783 | 0.664040217 | 0.695132620 |
| O | 0.630641734 | 0.130641734 | 0.301262785 |
| O | 0.344885259 | 0.409096299 | 0.232000737 |
| O | 0.165419603 | 0.935785616 | 0.789822036 |
| O | 0.435785616 | 0.665419603 | 0.789822036 |
| O | 0.090903701 | 0.155114741 | 0.232000737 |
| O | 0.655114741 | 0.590903701 | 0.232000737 |
| O | 0.564214384 | 0.334580397 | 0.789822036 |
| O | 0.909096299 | 0.844885259 | 0.232000737 |
| O | 0.834580397 | 0.064214384 | 0.789822036 |
| O | 0.000000000 | 0.500000000 | 0.102874456 |
| O | 0.500000000 | 0.000000000 | 0.803944996 |

**Table S11**. Atomic positions of relaxed isotropically compressed (0.90) $CaLaAl_3O_7$

crystal cell.

| | | | |
|---|---|---|---|
| La | 0.155255504 | 0.655255504 | 0.504087782 |
| Ca | 0.351680951 | 0.148319049 | 0.490946889 |
| Ca | 0.648319049 | 0.851680951 | 0.490946889 |
| La | 0.844744496 | 0.344744496 | 0.504087782 |
| Al | 0.000000000 | 0.000000000 | 0.015579333 |
| Al | 0.500000000 | 0.500000000 | 0.015579333 |
| Al | 0.359080349 | 0.859080349 | 0.958750605 |
| Al | 0.168154625 | 0.331845375 | 0.039664312 |
| Al | 0.831845375 | 0.668154625 | 0.039664312 |
| Al | 0.640919651 | 0.140919651 | 0.958750605 |
| O | 0.368027867 | 0.868027867 | 0.302204037 |
| O | 0.166980851 | 0.333019149 | 0.695477254 |
| O | 0.833019149 | 0.666980851 | 0.695477254 |
| O | 0.631972133 | 0.131972133 | 0.302204037 |
| O | 0.346490137 | 0.408547684 | 0.235899174 |
| O | 0.165557952 | 0.938589342 | 0.784907166 |
| O | 0.438589342 | 0.665557952 | 0.784907166 |
| O | 0.091452316 | 0.153509863 | 0.235899174 |
| O | 0.653509863 | 0.591452316 | 0.235899174 |
| O | 0.561410658 | 0.334442048 | 0.784907166 |
| O | 0.908547684 | 0.846490137 | 0.235899174 |
| O | 0.834442048 | 0.061410658 | 0.784907166 |
| O | 0.000000000 | 0.500000000 | 0.095213199 |
| O | 0.500000000 | 0.000000000 | 0.808141010 |

**Table S12**. Atomic positions of relaxed isotropically compressed (0.89) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.152809509 | 0.652809509 | 0.506826043 |
| Ca | 0.354019447 | 0.145980553 | 0.490247487 |
| Ca | 0.645980553 | 0.854019447 | 0.490247487 |
| La | 0.847190491 | 0.347190491 | 0.506826043 |
| Al | 0.000000000 | 0.000000000 | 0.014725626 |
| Al | 0.500000000 | 0.500000000 | 0.014725626 |
| Al | 0.357054670 | 0.857054670 | 0.958151511 |
| Al | 0.170538915 | 0.329461085 | 0.041472147 |
| Al | 0.829461085 | 0.670538915 | 0.041472147 |
| Al | 0.642945330 | 0.142945330 | 0.958151511 |
| O | 0.365891767 | 0.865891767 | 0.302912792 |
| O | 0.170751775 | 0.329248225 | 0.696232827 |
| O | 0.829248225 | 0.670751775 | 0.696232827 |
| O | 0.634108233 | 0.134108233 | 0.302912792 |
| O | 0.348206026 | 0.408037759 | 0.239932736 |
| O | 0.165521338 | 0.941166451 | 0.779595152 |
| O | 0.441166451 | 0.665521338 | 0.779595152 |

| | | | |
|---|---|---|---|
| O | 0.091962241 | 0.151793974 | 0.239932736 |
| O | 0.651793974 | 0.591962241 | 0.239932736 |
| O | 0.558833549 | 0.334478662 | 0.779595152 |
| O | 0.908037759 | 0.848206026 | 0.239932736 |
| O | 0.834478662 | 0.058833549 | 0.779595152 |
| O | 0.000000000 | 0.500000000 | 0.086579196 |
| O | 0.500000000 | 0.000000000 | 0.814172386 |

**Table 13**. Atomic positions of relaxed isotropically compressed (0.88) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.149377119 | 0.649377119 | 0.510734008 |
| Ca | 0.357260612 | 0.142739388 | 0.488495071 |
| Ca | 0.642739388 | 0.857260612 | 0.488495071 |
| La | 0.850622881 | 0.350622881 | 0.510734008 |
| Al | 0.000000000 | 0.000000000 | 0.013730012 |
| Al | 0.500000000 | 0.500000000 | 0.013730012 |
| Al | 0.354369565 | 0.854369565 | 0.957033115 |
| Al | 0.173086836 | 0.326913164 | 0.043698635 |
| Al | 0.826913164 | 0.673086836 | 0.043698635 |
| Al | 0.645630435 | 0.145630435 | 0.957033115 |
| O | 0.362407387 | 0.862407387 | 0.303272343 |
| O | 0.175858423 | 0.324141577 | 0.697513358 |
| O | 0.824141577 | 0.675858423 | 0.697513358 |
| O | 0.637592613 | 0.137592613 | 0.303272343 |
| O | 0.349981842 | 0.407594719 | 0.244195622 |
| O | 0.165217587 | 0.943315922 | 0.773737021 |
| O | 0.443315922 | 0.665217587 | 0.773737021 |
| O | 0.092405281 | 0.150018158 | 0.244195622 |
| O | 0.650018158 | 0.592405281 | 0.244195622 |
| O | 0.556684078 | 0.334782413 | 0.773737021 |
| O | 0.907594719 | 0.849981842 | 0.244195622 |
| O | 0.834782413 | 0.056684078 | 0.773737021 |
| O | 0.000000000 | 0.500000000 | 0.076387545 |
| O | 0.500000000 | 0.000000000 | 0.822928796 |

**Table S14**. Atomic positions of relaxed isotropically compressed (0.87) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.144530683 | 0.644530683 | 0.516185259 |
| Ca | 0.361727932 | 0.138272068 | 0.485290803 |
| Ca | 0.638272068 | 0.861727932 | 0.485290803 |
| La | 0.855469317 | 0.355469317 | 0.516185259 |
| Al | 0.000000000 | 0.000000000 | 0.012529231 |
| Al | 0.500000000 | 0.500000000 | 0.012529231 |
| Al | 0.350907412 | 0.850907412 | 0.955060063 |
| Al | 0.175765756 | 0.324234244 | 0.046550004 |

| | | | |
|---|---|---|---|
| Al | 0.824234244 | 0.675765756 | 0.046550004 |
| Al | 0.649092588 | 0.149092588 | 0.955060063 |
| O | 0.356946911 | 0.856946911 | 0.303028087 |
| O | 0.182768835 | 0.317231165 | 0.699670887 |
| O | 0.817231165 | 0.682768835 | 0.699670887 |
| O | 0.643053089 | 0.143053089 | 0.303028087 |
| O | 0.351838880 | 0.407207822 | 0.248836876 |
| O | 0.164550243 | 0.944797920 | 0.766964585 |
| O | 0.444797920 | 0.664550243 | 0.766964585 |
| O | 0.092792178 | 0.148161120 | 0.248836876 |
| O | 0.648161120 | 0.592792178 | 0.248836876 |
| O | 0.555202080 | 0.335449757 | 0.766964585 |
| O | 0.907207822 | 0.851838880 | 0.248836876 |
| O | 0.835449757 | 0.055202080 | 0.766964585 |
| O | 0.000000000 | 0.500000000 | 0.064676236 |
| O | 0.500000000 | 0.000000000 | 0.835489252 |

**Table S15**. Atomic positions of relaxed isotropically compressed (0.86) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.138740046 | 0.638740046 | 0.522606181 |
| Ca | 0.366952998 | 0.133047002 | 0.480664956 |
| Ca | 0.633047002 | 0.866952998 | 0.480664956 |
| La | 0.861259954 | 0.361259954 | 0.522606181 |
| Al | 0.000000000 | 0.000000000 | 0.011074035 |
| Al | 0.500000000 | 0.500000000 | 0.011074035 |
| Al | 0.347106867 | 0.847106867 | 0.951988369 |
| Al | 0.178307541 | 0.321692459 | 0.050154842 |
| Al | 0.821692459 | 0.678307541 | 0.050154842 |
| Al | 0.652893133 | 0.152893133 | 0.951988369 |
| O | 0.349998654 | 0.849998654 | 0.301839093 |
| O | 0.190727617 | 0.309272383 | 0.702900079 |
| O | 0.809272383 | 0.690727617 | 0.702900079 |
| O | 0.650001346 | 0.150001346 | 0.301839093 |
| O | 0.353813541 | 0.406830368 | 0.253799266 |
| O | 0.163666251 | 0.945758048 | 0.759346788 |
| O | 0.445758048 | 0.663666251 | 0.759346788 |
| O | 0.093169632 | 0.146186459 | 0.253799266 |
| O | 0.646186459 | 0.593169632 | 0.253799266 |
| O | 0.554241952 | 0.336333749 | 0.759346788 |
| O | 0.906830368 | 0.853813541 | 0.253799266 |
| O | 0.836333749 | 0.054241952 | 0.759346788 |
| O | 0.000000000 | 0.500000000 | 0.054129508 |
| O | 0.500000000 | 0.000000000 | 0.850831164 |

**Table S16**. Atomic positions of relaxed isotropically compressed (0.85) $CaLaAl_3O_7$

crystal cell.

| | | | |
|---|---|---|---|
| La | 0.133627136 | 0.633627136 | 0.528453362 |
| Ca | 0.371544956 | 0.128455044 | 0.475836040 |
| Ca | 0.628455044 | 0.871544956 | 0.475836040 |
| La | 0.866372864 | 0.366372864 | 0.528453362 |
| Al | 0.000000000 | 0.000000000 | 0.009398066 |
| Al | 0.500000000 | 0.500000000 | 0.009398066 |
| Al | 0.343782144 | 0.843782144 | 0.948155056 |
| Al | 0.180383967 | 0.319616033 | 0.054216444 |
| Al | 0.819616033 | 0.680383967 | 0.054216444 |
| Al | 0.656217856 | 0.156217856 | 0.948155056 |
| O | 0.343591446 | 0.843591446 | 0.299751851 |
| O | 0.197688883 | 0.302311117 | 0.706613745 |
| O | 0.802311117 | 0.697688883 | 0.706613745 |
| O | 0.656408554 | 0.156408554 | 0.299751851 |
| O | 0.355915350 | 0.406440872 | 0.258632378 |
| O | 0.162983060 | 0.946962351 | 0.751661911 |
| O | 0.446962351 | 0.662983060 | 0.751661911 |
| O | 0.093559128 | 0.144084650 | 0.258632378 |
| O | 0.644084650 | 0.593559128 | 0.258632378 |
| O | 0.553037649 | 0.337016940 | 0.751661911 |
| O | 0.906440872 | 0.855915350 | 0.258632378 |
| O | 0.837016940 | 0.053037649 | 0.751661911 |
| O | 0.000000000 | 0.500000000 | 0.048267203 |
| O | 0.500000000 | 0.000000000 | 0.865706512 |

**Table S17**. Atomic positions of relaxed anisotropically compressed (0.95, a-axis) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.167022837 | 0.661179997 | 0.499622881 |
| Ca | 0.340637104 | 0.153179740 | 0.482263641 |
| Ca | 0.659362896 | 0.846820260 | 0.482263641 |
| La | 0.832977163 | 0.338820003 | 0.499622881 |
| Al | 0.000000000 | 0.000000000 | 0.014606463 |
| Al | 0.500000000 | 0.500000000 | 0.015105023 |
| Al | 0.366239465 | 0.862176322 | 0.959288981 |
| Al | 0.152038440 | 0.346561219 | 0.033842269 |
| Al | 0.847961560 | 0.653438781 | 0.033842269 |
| Al | 0.633760535 | 0.137823678 | 0.959288981 |
| O | 0.369794018 | 0.869779241 | 0.287820520 |
| O | 0.152836000 | 0.349637162 | 0.704537116 |
| O | 0.847164000 | 0.650362838 | 0.704537116 |
| O | 0.630205982 | 0.130220759 | 0.287820520 |
| O | 0.338165347 | 0.412084986 | 0.208409810 |
| O | 0.161508133 | 0.912626106 | 0.814324926 |
| O | 0.421341966 | 0.664472504 | 0.822046550 |

| | | | |
|---|---|---|---|
| O | 0.086825215 | 0.163903389 | 0.199424229 |
| O | 0.661834653 | 0.587915014 | 0.208409810 |
| O | 0.578658034 | 0.335527496 | 0.822046550 |
| O | 0.913174785 | 0.836096611 | 0.199424229 |
| O | 0.838491867 | 0.087373894 | 0.814324926 |
| O | 0.000000000 | 0.500000000 | 0.147534107 |
| O | 0.500000000 | 0.000000000 | 0.799592565 |

**Table S18**. Atomic positions of relaxed anisotropically compressed (0.95, b-axis) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.161148870 | 0.667067926 | 0.499609953 |
| Ca | 0.347065395 | 0.159642206 | 0.482285271 |
| Ca | 0.652934605 | 0.840357794 | 0.482285271 |
| La | 0.838851130 | 0.332932074 | 0.499609953 |
| Al | 0.000000000 | 0.000000000 | 0.015357401 |
| Al | 0.500000000 | 0.500000000 | 0.014389415 |
| Al | 0.362202814 | 0.866213833 | 0.959286717 |
| Al | 0.153449007 | 0.347968804 | 0.033825908 |
| Al | 0.846550993 | 0.652031196 | 0.033825908 |
| Al | 0.637797186 | 0.133786167 | 0.959286717 |
| O | 0.369841132 | 0.869755326 | 0.287826454 |
| O | 0.150428455 | 0.347215341 | 0.704515809 |
| O | 0.849571545 | 0.652784659 | 0.704515809 |
| O | 0.630158868 | 0.130244674 | 0.287826454 |
| O | 0.336094080 | 0.413311103 | 0.199333070 |
| O | 0.164393834 | 0.921135810 | 0.822328742 |
| O | 0.412818167 | 0.661588630 | 0.814036305 |
| O | 0.088041062 | 0.161838351 | 0.208527786 |
| O | 0.663905920 | 0.586688897 | 0.199333070 |
| O | 0.587181833 | 0.338411370 | 0.814036305 |
| O | 0.911958938 | 0.838161649 | 0.208527786 |
| O | 0.835606166 | 0.078864190 | 0.822328742 |
| O | 0.000000000 | 0.500000000 | 0.147517469 |
| O | 0.500000000 | 0.000000000 | 0.799583684 |

**Table S19**. Atomic positions of relaxed anisotropically compressed (0.95, c-axis) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.162501620 | 0.662501620 | 0.499896353 |
| Ca | 0.345406535 | 0.154593465 | 0.482593843 |
| Ca | 0.654593465 | 0.845406535 | 0.482593843 |
| La | 0.837498380 | 0.337498380 | 0.499896353 |
| Al | 0.000000000 | 0.000000000 | 0.014049000 |
| Al | 0.500000000 | 0.500000000 | 0.014049000 |
| Al | 0.364227894 | 0.864227894 | 0.958773097 |
| Al | 0.152332298 | 0.347667702 | 0.035786294 |

| | | | |
|---|---|---|---|
| Al | 0.847667702 | 0.652332298 | 0.035786294 |
| Al | 0.635772106 | 0.135772106 | 0.958773097 |
| O | 0.369825618 | 0.869825618 | 0.302403755 |
| O | 0.151155246 | 0.348844754 | 0.691627491 |
| O | 0.848844754 | 0.651155246 | 0.691627491 |
| O | 0.630174382 | 0.130174382 | 0.302403755 |
| O | 0.335299831 | 0.414188742 | 0.202671533 |
| O | 0.165304980 | 0.917383153 | 0.818548979 |
| O | 0.417383153 | 0.665304980 | 0.818548979 |
| O | 0.085811258 | 0.164700169 | 0.202671533 |
| O | 0.664700169 | 0.585811258 | 0.202671533 |
| O | 0.582616847 | 0.334695020 | 0.818548979 |
| O | 0.914188742 | 0.835299831 | 0.202671533 |
| O | 0.834695020 | 0.082616847 | 0.818548979 |
| O | -0.000000000 | 0.500000000 | 0.139033956 |
| O | 0.500000000 | -0.000000000 | 0.805824328 |

**Table S20**. Atomic positions of relaxed anisotropically compressed (0.90, a-axis) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.171860848 | 0.656620213 | 0.496711262 |
| Ca | 0.334729905 | 0.147163563 | 0.485624900 |
| Ca | 0.665270095 | 0.852836437 | 0.485624900 |
| La | 0.828139152 | 0.343379787 | 0.496711262 |
| Al | 0.000000000 | 0.000000000 | 0.014031741 |
| Al | 0.500000000 | 0.500000000 | 0.019484256 |
| Al | 0.368069633 | 0.859663531 | 0.959645938 |
| Al | 0.153764115 | 0.342282437 | 0.032306362 |
| Al | 0.846235885 | 0.657717563 | 0.032306362 |
| Al | 0.631930367 | 0.140336469 | 0.959645938 |
| O | 0.368448752 | 0.872708612 | 0.285203446 |
| O | 0.156866324 | 0.348448568 | 0.705633159 |
| O | 0.843133676 | 0.651551432 | 0.705633159 |
| O | 0.631551248 | 0.127291388 | 0.285203446 |
| O | 0.342775097 | 0.405940066 | 0.220695949 |
| O | 0.158041741 | 0.908756465 | 0.804284057 |
| O | 0.428629651 | 0.664291218 | 0.823958228 |
| O | 0.084905501 | 0.163139250 | 0.200454434 |
| O | 0.657224903 | 0.594059934 | 0.220695949 |
| O | 0.571370349 | 0.335708782 | 0.823958228 |
| O | 0.915094499 | 0.836860750 | 0.200454434 |
| O | 0.841958259 | 0.091243535 | 0.804284057 |
| O | 0.000000000 | 0.500000000 | 0.140336016 |
| O | 0.500000000 | 0.000000000 | 0.797112518 |

**Table S21**. Atomic positions of relaxed anisotropically compressed (0.90, b-axis)

$CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.157780414 | 0.670808956 | 0.496438283 |
| Ca | 0.349945419 | 0.162735879 | 0.486250494 |
| Ca | 0.650054581 | 0.837264121 | 0.486250494 |
| La | 0.842219586 | 0.329191044 | 0.496438283 |
| Al | 0.000000000 | 0.000000000 | 0.017881535 |
| Al | 0.500000000 | 0.500000000 | 0.015720531 |
| Al | 0.360251231 | 0.867778904 | 0.959400348 |
| Al | 0.157323059 | 0.345610128 | 0.032478183 |
| Al | 0.842676941 | 0.654389872 | 0.032478183 |
| Al | 0.639748769 | 0.132221096 | 0.959400348 |
| O | 0.373104956 | 0.868655639 | 0.284802322 |
| O | 0.150026442 | 0.341964228 | 0.705812129 |
| O | 0.849973558 | 0.658035772 | 0.705812129 |
| O | 0.626895044 | 0.131344361 | 0.284802322 |
| O | 0.336812602 | 0.413917214 | 0.201278035 |
| O | 0.164906356 | 0.928939407 | 0.822558900 |
| O | 0.408748687 | 0.657543957 | 0.805993959 |
| O | 0.093095133 | 0.157084893 | 0.220034263 |
| O | 0.663187398 | 0.586082786 | 0.201278035 |
| O | 0.591251313 | 0.342456043 | 0.805993959 |
| O | 0.906904867 | 0.842915107 | 0.220034263 |
| O | 0.835093644 | 0.071060593 | 0.822558900 |
| O | 0.000000000 | 0.500000000 | 0.139789310 |
| O | 0.500000000 | 0.000000000 | 0.796514719 |

**Table S22**. Atomic positions of relaxed anisotropically compressed (0.90, c-axis) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.160424199 | 0.660424199 | 0.499873296 |
| Ca | 0.347185152 | 0.152814848 | 0.484220444 |
| Ca | 0.652814848 | 0.847185152 | 0.484220444 |
| La | 0.839575801 | 0.339575801 | 0.499873296 |
| Al | 0.000000000 | 0.000000000 | 0.014972864 |
| Al | 0.500000000 | 0.500000000 | 0.014972864 |
| Al | 0.363626278 | 0.863626278 | 0.957400039 |
| Al | 0.154606344 | 0.345393656 | 0.038046734 |
| Al | 0.845393656 | 0.654606344 | 0.038046734 |
| Al | 0.636373722 | 0.136373722 | 0.957400039 |
| O | 0.369709652 | 0.869709652 | 0.315579323 |
| O | 0.153768418 | 0.346231582 | 0.679442610 |
| O | 0.846231582 | 0.653768418 | 0.679442610 |
| O | 0.630290348 | 0.130290348 | 0.315579323 |
| O | 0.335937372 | 0.414112243 | 0.208209458 |
| O | 0.166224074 | 0.920049559 | 0.813748649 |
| O | 0.420049559 | 0.666224074 | 0.813748649 |

| | | | |
|---|---|---|---|
| O | 0.085887757 | 0.164062628 | 0.208209458 |
| O | 0.664062628 | 0.585887757 | 0.208209458 |
| O | 0.579950441 | 0.333775926 | 0.813748649 |
| O | 0.914112243 | 0.835937372 | 0.208209458 |
| O | 0.833775926 | 0.079950441 | 0.813748649 |
| O | 0.000000000 | 0.500000000 | 0.121917932 |
| O | 0.500000000 | 0.000000000 | 0.811179021 |

**Table S23**. Atomic positions of relaxed anisotropically compressed (0.85, a-axis) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.174213049 | 0.655612653 | 0.494130342 |
| Ca | 0.333509237 | 0.146596718 | 0.487278259 |
| Ca | 0.666490763 | 0.853403282 | 0.487278259 |
| La | 0.825786951 | 0.344387347 | 0.494130342 |
| Al | 0.000000000 | 0.000000000 | 0.017912613 |
| Al | 0.500000000 | 0.500000000 | 0.017147466 |
| Al | 0.371082340 | 0.857216323 | 0.962220336 |
| Al | 0.152961190 | 0.339170427 | 0.029274305 |
| Al | 0.847038810 | 0.660829573 | 0.029274305 |
| Al | 0.628917660 | 0.142783677 | 0.962220336 |
| O | 0.368259918 | 0.874415976 | 0.284119077 |
| O | 0.160190722 | 0.348820945 | 0.706857842 |
| O | 0.839809278 | 0.651179055 | 0.706857842 |
| O | 0.631740082 | 0.125584024 | 0.284119077 |
| O | 0.346055320 | 0.404052897 | 0.227223022 |
| O | 0.154624313 | 0.907223535 | 0.797983289 |
| O | 0.431652778 | 0.664527653 | 0.825799011 |
| O | 0.084764863 | 0.163187177 | 0.200659309 |
| O | 0.653944680 | 0.595947103 | 0.227223022 |
| O | 0.568347222 | 0.335472347 | 0.825799011 |
| O | 0.915235137 | 0.836812823 | 0.200659309 |
| O | 0.845375687 | 0.092776465 | 0.797983289 |
| O | -0.000000000 | 0.500000000 | 0.138298186 |
| O | 0.500000000 | -0.000000000 | 0.795552152 |

**Table S24**. Atomic positions of relaxed anisotropically compressed (0.85, b-axis) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.157946011 | 0.673306235 | 0.492023857 |
| Ca | 0.348878282 | 0.164481202 | 0.490048885 |
| Ca | 0.651121718 | 0.835518798 | 0.490048885 |
| La | 0.842053989 | 0.326693765 | 0.492023857 |
| Al | 0.000000000 | 0.000000000 | 0.013978682 |
| Al | 0.500000000 | 0.500000000 | 0.018555870 |
| Al | 0.359342528 | 0.869517455 | 0.962824123 |

| | | | |
|---|---|---|---|
| Al | 0.158899613 | 0.346692024 | 0.029178366 |
| Al | 0.841100387 | 0.653307976 | 0.029178366 |
| Al | 0.640657472 | 0.130482545 | 0.962824123 |
| O | 0.377030059 | 0.869390036 | 0.285462648 |
| O | 0.147036129 | 0.337534111 | 0.705839179 |
| O | 0.852963871 | 0.662465889 | 0.705839179 |
| O | 0.622969941 | 0.130609964 | 0.285462648 |
| O | 0.336604156 | 0.413831793 | 0.202526602 |
| O | 0.165048808 | 0.928653220 | 0.823707728 |
| O | 0.408291033 | 0.655540802 | 0.802507542 |
| O | 0.094877221 | 0.154891813 | 0.221581922 |
| O | 0.663395844 | 0.586168207 | 0.202526602 |
| O | 0.591708967 | 0.344459198 | 0.802507542 |
| O | 0.905122779 | 0.845108187 | 0.221581922 |
| O | 0.834951192 | 0.071346780 | 0.823707728 |
| O | -0.000000000 | 0.500000000 | 0.140401556 |
| O | 0.500000000 | 0.000000000 | 0.795662190 |

**Table S25**. Atomic positions of relaxed anisotropically compressed (0.85, c-axis) $CaLaAl_3O_7$ crystal cell.

| | | | |
|---|---|---|---|
| La | 0.164098530 | 0.664098530 | 0.501909253 |
| Ca | 0.344079287 | 0.155920713 | 0.478851824 |
| Ca | 0.655920713 | 0.844079287 | 0.478851824 |
| La | 0.835901470 | 0.335901470 | 0.501909253 |
| Al | 0.000000000 | 0.000000000 | 0.012962363 |
| Al | 0.500000000 | 0.500000000 | 0.012962363 |
| Al | 0.364241380 | 0.864241380 | 0.959469564 |
| Al | 0.150135081 | 0.349864919 | 0.034865959 |
| Al | 0.849864919 | 0.650135081 | 0.034865959 |
| Al | 0.635758620 | 0.135758620 | 0.959469564 |
| O | 0.369027170 | 0.869027170 | 0.290601987 |
| O | 0.148750159 | 0.351249841 | 0.703181663 |
| O | 0.851249841 | 0.648750159 | 0.703181663 |
| O | 0.630972830 | 0.130972830 | 0.290601987 |
| O | 0.335078186 | 0.414094325 | 0.196872367 |
| O | 0.164122082 | 0.914737979 | 0.822883822 |
| O | 0.414737979 | 0.664122082 | 0.822883822 |
| O | 0.085905675 | 0.164921814 | 0.196872367 |
| O | 0.664921814 | 0.585905675 | 0.196872367 |
| O | 0.585262021 | 0.335877918 | 0.822883822 |
| O | 0.914094325 | 0.835078186 | 0.196872367 |
| O | 0.835877918 | 0.085262021 | 0.822883822 |
| O | 0.000000000 | 0.500000000 | 0.152989729 |
| O | 0.500000000 | 0.000000000 | 0.804300289 |

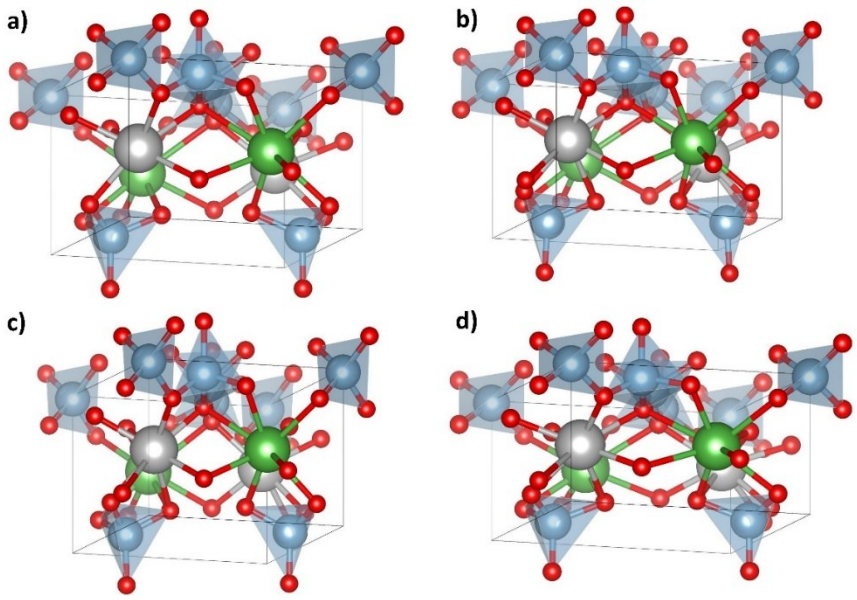


**Figure S6**. Comparison between fully relaxed $CaLaAl_3O_7$ cell (a) and anistropically compressed relaxed cells (0.85 multiplier): (b) alongside a-axis, (b) alongside b-axis, (c) alongside c-axis. Red color stands for O atoms, blue – Al, silver – Ca, green – La.

**Table S26**. Changes in atomic charges due to isotropic $CaLaAl_3O_7$ cell compression.

| | Charge | ΔCharge | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 1 | 0.99 | 0.98 | 0.97 | 0.96 | 0.95 | 0.94 | 0.93 | 0.92 | 0.91 | 0.9 | 0.89 | 0.87 | 0.86 | 0.85 |
| La(1) | 56.977 | -0.016 | -0.019 | -0.019 | -0.020 | -0.024 | -0.025 | -0.029 | -0.032 | -0.033 | -0.036 | -0.036 | -0.038 | -0.039 | -0.042 |
| Ca(1) | 1.583 | -0.070 | -0.075 | -0.077 | -0.080 | -0.083 | -0.085 | -0.089 | -0.091 | -0.094 | -0.098 | -0.102 | -0.105 | -0.109 | -0.112 |
| Ca(2) | 1.583 | -0.070 | -0.075 | -0.077 | -0.080 | -0.083 | -0.085 | -0.089 | -0.091 | -0.094 | -0.098 | -0.102 | -0.105 | -0.109 | -0.112 |
| La(1) | 56.977 | -0.016 | -0.019 | -0.019 | -0.020 | -0.024 | -0.025 | -0.029 | -0.032 | -0.033 | -0.036 | -0.036 | -0.038 | -0.039 | -0.042 |
| Al(1) | 2.524 | 0.001 | 0.000 | 0.002 | 0.004 | 0.000 | 0.003 | 0.002 | -0.001 | 0.004 | 0.001 | 0.000 | 0.002 | 0.004 | 0.003 |
| Al(2) | 2.524 | 0.001 | 0.000 | 0.002 | 0.004 | 0.000 | 0.003 | 0.002 | -0.001 | 0.004 | 0.001 | 0.000 | 0.002 | 0.004 | 0.003 |
| Al(3) | 2.505 | 0.003 | 0.001 | 0.000 | -0.001 | -0.001 | 0.000 | 0.001 | 0.004 | -0.001 | 0.003 | 0.004 | 0.000 | 0.002 | 0.000 |
| Al(4) | 2.505 | 0.003 | 0.001 | 0.000 | -0.001 | -0.001 | 0.000 | 0.001 | 0.004 | -0.001 | 0.003 | 0.004 | 0.000 | 0.002 | 0.000 |
| Al(5) | 2.516 | 0.003 | -0.001 | 0.000 | 0.001 | 0.004 | -0.001 | 0.004 | 0.000 | 0.001 | 0.004 | 0.001 | 0.000 | 0.002 | 0.003 |
| Al(6) | 2.516 | 0.003 | -0.001 | 0.000 | 0.001 | 0.004 | -0.001 | 0.004 | 0.000 | 0.001 | 0.004 | 0.001 | 0.000 | 0.002 | 0.003 |
| O(1) | -1.701 | -0.001 | -0.002 | 0.006 | 0.010 | -0.001 | -0.001 | -0.002 | -0.001 | -0.002 | -0.006 | 0.002 | 0.006 | -0.006 | -0.001 |
| O(2) | -1.725 | -0.001 | -0.002 | 0.005 | -0.001 | -0.001 | -0.001 | -0.002 | -0.001 | -0.001 | 0.002 | 0.010 | 0.006 | -0.001 | -0.006 |
| O(3) | -1.725 | 0.054 | 0.057 | 0.059 | 0.064 | 0.068 | 0.072 | 0.075 | 0.080 | 0.082 | 0.085 | 0.087 | 0.089 | 0.093 | 0.095 |
| O(4) | -1.699 | 0.004 | -0.001 | -0.002 | -0.001 | -0.006 | 0.006 | 0.010 | -0.001 | 0.010 | -0.006 | -0.002 | -0.006 | -0.010 | -0.012 |
| O(5) | -1.703 | 0.002 | -0.001 | -0.002 | -0.006 | 0.005 | -0.001 | -0.002 | -0.006 | -0.001 | 0.006 | 0.010 | 0.015 | 0.029 | 0.021 |
| O(6) | -1.703 | 0.002 | -0.001 | -0.002 | -0.006 | 0.001 | -0.001 | -0.002 | -0.006 | -0.001 | 0.006 | 0.010 | 0.015 | 0.029 | 0.021 |
| O(7) | -1.748 | 0.058 | 0.061 | 0.063 | 0.065 | 0.068 | 0.070 | 0.072 | 0.076 | 0.079 | 0.081 | 0.084 | 0.085 | 0.089 | 0.090 |
| O(8) | -1.703 | 0.002 | -0.001 | -0.002 | -0.006 | 0.001 | -0.001 | -0.002 | -0.006 | -0.001 | 0.006 | 0.010 | 0.015 | 0.029 | 0.021 |

| | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| O(9) | -1.748 | -0.001 | -0.001 | -0.001 | 0.002 | 0.006 | -0.006 | 0.000 | -0.001 | -0.006 | 0.005 | -0.001 | -0.001 | 0.002 | 0.006 |
| O(10) | -1.748 | -0.001 | -0.001 | -0.001 | 0.002 | 0.006 | -0.006 | 0.000 | -0.001 | -0.006 | 0.001 | -0.001 | -0.001 | 0.002 | 0.006 |
| O(11) | -1.703 | 0.000 | 0.002 | 0.006 | 0.010 | 0.006 | -0.001 | -0.002 | -0.001 | 0.002 | -0.002 | -0.006 | 0.002 | -0.001 | 0.005 |
| O(12) | -1.703 | 0.000 | 0.002 | 0.006 | 0.010 | 0.006 | -0.001 | -0.002 | -0.001 | 0.002 | -0.002 | -0.006 | 0.002 | -0.001 | 0.005 |
| O(13) | -1.895 | 0.000 | 0.002 | 0.001 | 0.004 | 0.002 | 0.001 | 0.003 | 0.005 | 0.001 | 0.004 | 0.006 | 0.005 | 0.008 | 0.012 |
| O(14) | -1.649 | -0.001 | -0.001 | -0.001 | -0.002 | -0.006 | -0.001 | 0.000 | 0.002 | 0.006 | 0.005 | 0.002 | 0.001 | 0.006 | 0.012 |

**Table S27**. Changes in atomic charges due to anisotropic $CaLaAl_3O_7$ cell compression.

| | Charge | ΔCharge | | |
|---|---|---|---|---|
| | 1 | 0.85 (a) | 0.85 (b) | 0.85 (c) |
| La(1) | 56.977 | -0.038 | -0.028 | -0.027 |
| Ca(1) | 1.583 | -0.104 | -0.099 | -0.102 |
| Ca(2) | 1.583 | -0.104 | -0.099 | -0.102 |
| La(1) | 56.977 | -0.008 | -0.005 | -0.001 |
| Al(1) | 2.524 | 0.003 | -0.002 | -0.002 |
| Al(2) | 2.524 | 0.003 | -0.002 | 0.003 |
| Al(3) | 2.505 | 0.001 | 0.002 | 0.003 |
| Al(4) | 2.505 | 0.001 | 0.002 | 0.003 |
| Al(5) | 2.516 | 0.004 | -0.001 | 0.001 |
| Al(6) | 2.516 | 0.004 | -0.001 | 0.001 |
| O(1) | -1.701 | -0.001 | -0.005 | -0.004 |
| O(2) | -1.725 | -0.005 | -0.006 | -0.007 |
| O(3) | -1.725 | 0.084 | 0.091 | 0.095 |
| O(4) | -1.699 | -0.008 | -0.005 | -0.003 |
| O(5) | -1.703 | 0.017 | 0.014 | 0.019 |
| O(6) | -1.703 | 0.002 | 0.003 | 0.012 |
| O(7) | -1.748 | 0.094 | 0.089 | 0.090 |
| O(8) | -1.703 | 0.007 | 0.004 | 0.001 |
| O(9) | -1.748 | 0.004 | 0.003 | 0.017 |
| O(10) | -1.748 | 0.004 | 0.002 | 0.000 |
| O(11) | -1.703 | 0.005 | 0.005 | 0.000 |
| O(12) | -1.703 | 0.005 | 0.005 | 0.001 |
| O(13) | -1.895 | -0.002 | -0.001 | 0.001 |
| O(14) | -1.649 | -0.002 | 0.000 | -0.002 |

## References


1. J. P. Perdew, M. Ernzerhof and K. Burke, *The Journal of Chemical Physics*, 1996, **105**, 9982-9985.

2. J. P. Perdew, K. Burke and M. Ernzerhof, *Physical Review Letters*, 1996, **77**,

3865-3868.

3. P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. Dal Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Sbraccia, S. Scandolo, G. Sclauzero, A. P. Seitsonen, A. Smogunov, P. Umari and R. M. Wentzcovitch, *Journal of Physics: Condensed Matter*, 2009, **21**, 395502.

4. A. Otero-de-la-Roza, E. R. Johnson and V. Luaña, *Computer Physics Communications*, 2014, **185**, 1007-1018.

5. K. Momma and F. Izumi, *Journal of Applied Crystallography*, 2008, **41**, 653-658.