Supplementary Information for "Optical spectroscopy and band gap analysis of hybrid improper ferroelectric $Ca_3Ti_2O_7$"

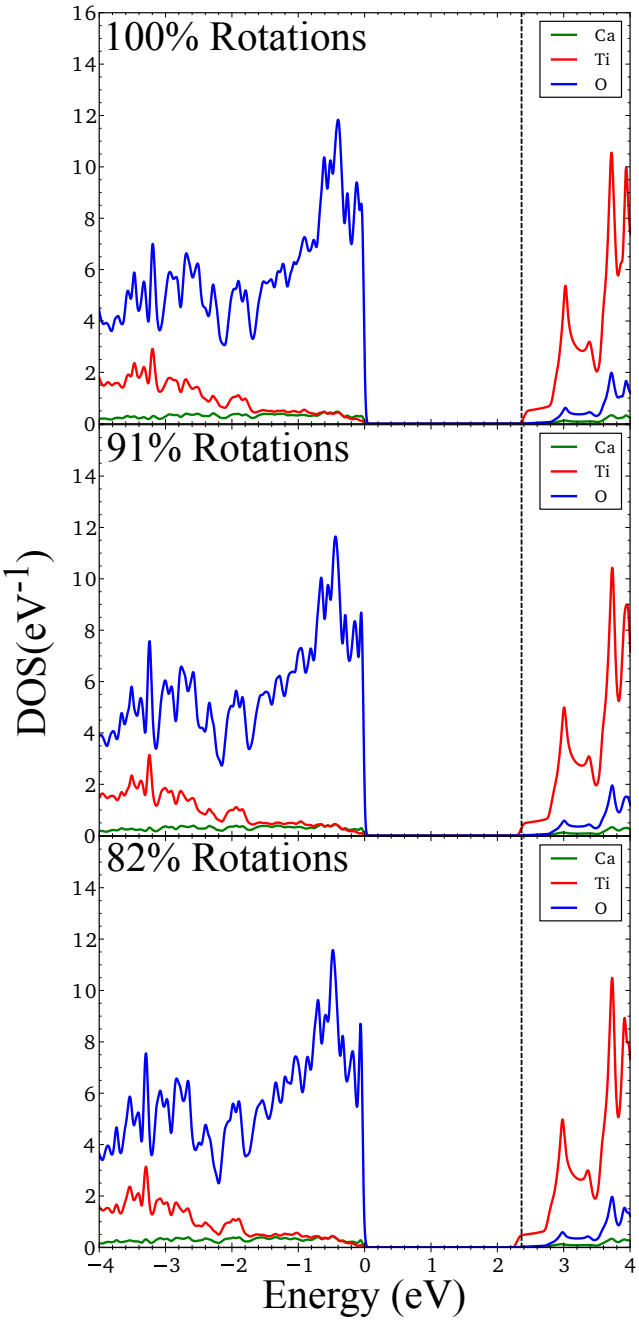

Figure 1: Densities of states for $Ca_3Ti_2O_7$ for the experimentally reported crystal structure, and hypothetical structures with smaller octahedral rotation angles and polarizations. The top panel corresponds to the experimentally reported crystal structure, whereas the lower ones are obtained by interpolating between the experimental structure and a hypothetical non-polar high-symmetry structure with the point group I4/*mmm*. Group theoretical analysis of the lattice distortions are made using the Isotropy software package.[1] Even decreasing the octahedral rotations and the accompanying polarization by 18% has a minor effect on the bandgap. Note that Sr doping also increases the lattice constant, which is expected to further increase the bandgap. Our DFT calculations here does not take into account this effect of the Sr doping, and so the change in the gap seen on the figure is probably larger than the experimental one.

References

[1] B. J. Campbell, H.T. Stokes, D.E. Tanner, and D.M. Hatch, J. Appl. Cryst. **39**, 607 (2006)