Controllable Ordering of the Fractional Double Perovskite, $EuTa_2O_6$

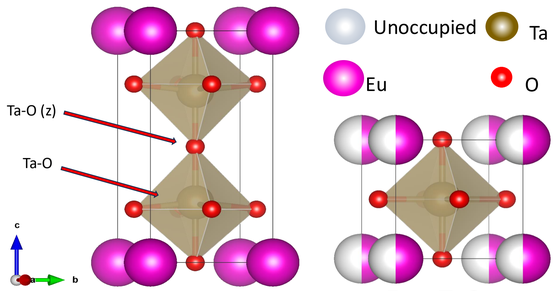
Brenan Palazzolo[1], Tobias Schwaigert[2,3], Darrell Schlom[2,3]

[1]Department of Physics and Astronomy, Clemson University, Clemson, SC 29641, USA, [2]Department of Materials Science and Engineering, Cornell University, Ithaca, NY 14853, USA, [3]PARADIM, Cornell University, Ithaca, NY 14853, USA

Motivation: *A*-site ordering of the fractional double perovskite, $EuTa_2O_6$, has shown to form a quasi-2D Electron Gas (quasi-2DEG) that is controllable by an external magnetic field.

Goal: Control over the amount of *A*-site ordering in the crystal structure during growth by Molecular Beam Epitaxy.

Structural features of fractional double perovskites



$EuTa_2O_6$ X = 1 $Eu_{0.5}TaO_3$ X = 0.5

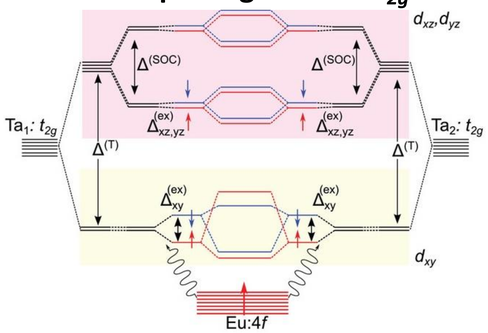
Lattice Constants of $EuTa_2O_6$	
a = b (Å)	3.926
c (Å)	7.929
Ta-O (z)	1.906
Ta-O	1.977

The growth of $Eu_xTa_{2x}O_{6x}$ can result in a range of ordering:

X = 1 being the **most ordered structure** with alternating layers of fully occupied and fully unoccupied Eu-sites along the c-axis

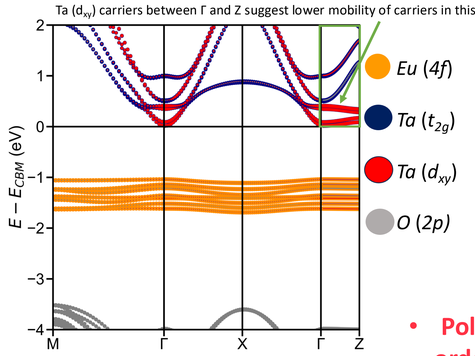
X = 0.5 to be **the most disordered structure** with only half occupied Eu-sites.

Splitting of the *d*-t_{2g} Ta orbitals by ordering



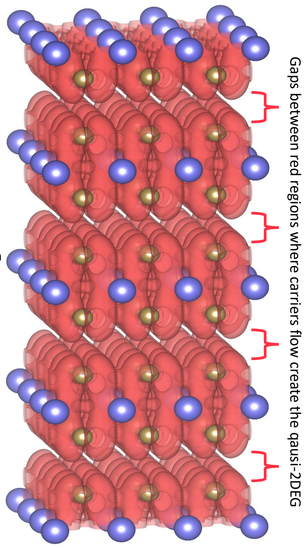
Splitting of *d*-t_{2g} orbitals where spins are differentiable (red and blue lines)[1].

Ta (d_{xy}) carriers between Γ and Z suggest lower mobility of carriers in this region



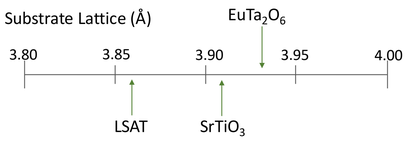
Energy band diagram of ordered $EuTa_2O_6$

At low temperatures application of an external magnetic field can control the spin polarized bands orbital Ta-manifolds can be controlled by the unpaired electrons of Eu^{2+}.



- **Polarization of Ta atoms when ordered causes splitting of d-t_{2g} orbitals**

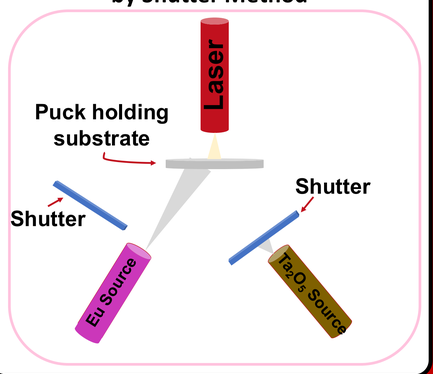
Application of compressive strain to control orientation and vacancy order in ETO



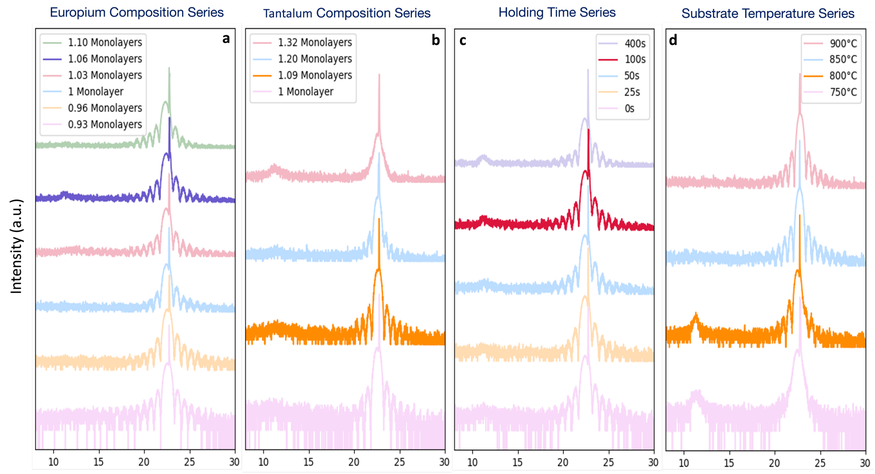
It has been shown in other materials that applying compressive strain on film by substrate can improve ordering of your crystal.

Using either LSAT or $SrTiO_3$ will apply this compressive strain as the lattice constant is smaller than ETO's a=b lattice constant.

Growth with Molecular Beam Epitaxy by Shutter Method



Finding control over higher order structures by modulation of growth recipes

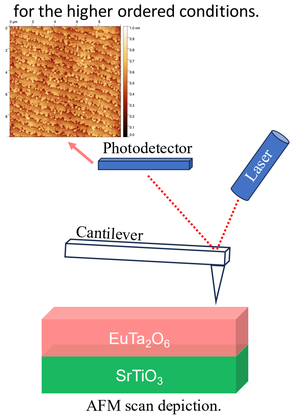


X-ray Diffraction scan series depict variation of growth parameters to determine optimal conditions for higher ordering in ETO. a) Europium composition series b) Tantalum composition series c) Holding time between unit cells series d) Substrate Temperature series

Atomic Force Microscopy rms roughness values for films in the series

Series	Conditions	RMS Roughness (pm)
Europium	0.93 Monolayer	198.2
	0.96 Monolayer	232.7
	1.00 Monolayer	240.7
	1.03 Monolayer	205.8
Highest order	1.06 Monolayer	113.7
	1.10 Monolayer	192.2
Tantalum	1.00 Monolayer	298.7
Highest order	1.09 Monolayer	173.4
	1.20 Monolayer	393.0
	1.32 Monolayer	794.4
Holding Time	0 seconds	180.1
	25 seconds	177.6
	50 seconds	190.9
Highest order	100 seconds	113.7
	400 seconds	236.4
Substrate Temperature	900 °C	307.4
	850 °C	115.7
Highest order	800 °C	285.2
	750 °C	526.0

AFM scans revealed the rms (root-mean square) roughness is lower for the higher ordered conditions.

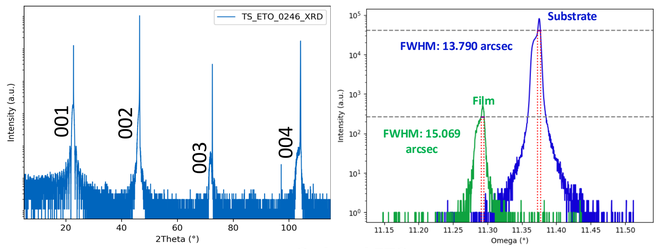


AFM scan depiction.

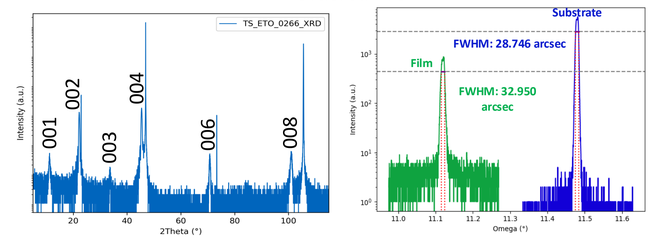
The 850°C substrate temperature reveals a high crystalline quality disordered film resulting in the low roughness.

XRD Shows High Crystalline Quality of both Disordered and Ordered ETO

Disordered ETO



Ordered ETO

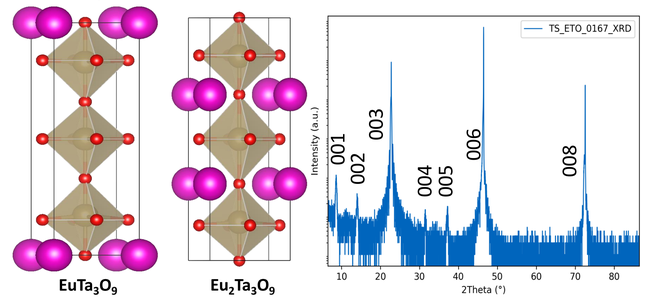


XRD scans (left) of disordered and ordered ETO with corresponding rocking curves (right) that show close full-width half max values of the substrate and film.

Conclusions and Future Plans

Successful control over which polymorph of $Eu_xTa_{2x}O_{6x}$ is being grown by Molecular Beam Epitaxy, allowing exploration of the levels of ordering and the effect on the formation quasi-2DEG.

There has been some speculation of different *A*-*B* site stacking forming by X-ray Diffraction. Exploration of this possibility is underway.



Potential structures of ETO with an additional *B*-site layer.

XRD data shows two "ordering peaks", i.e., 001 and 002, before the fundamental peak, 003.

Acknowledgements

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References

[1] O. Dowinton, D. Maryenko, R. V. Belosludov, B.-J. Yang, M. S. Bahramy, *Adv. Funct. Mater.* 2023, 33, 2300995.