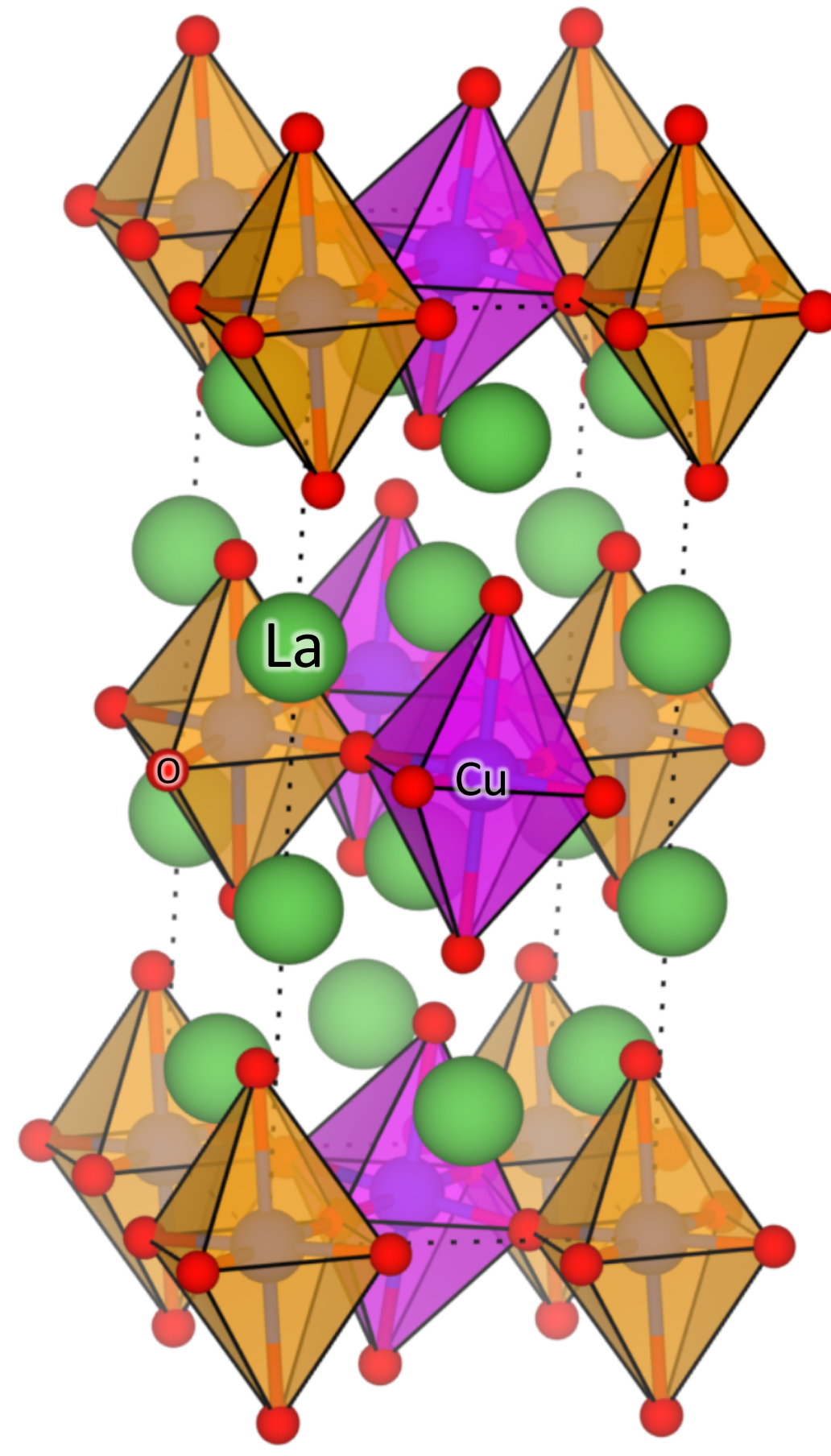


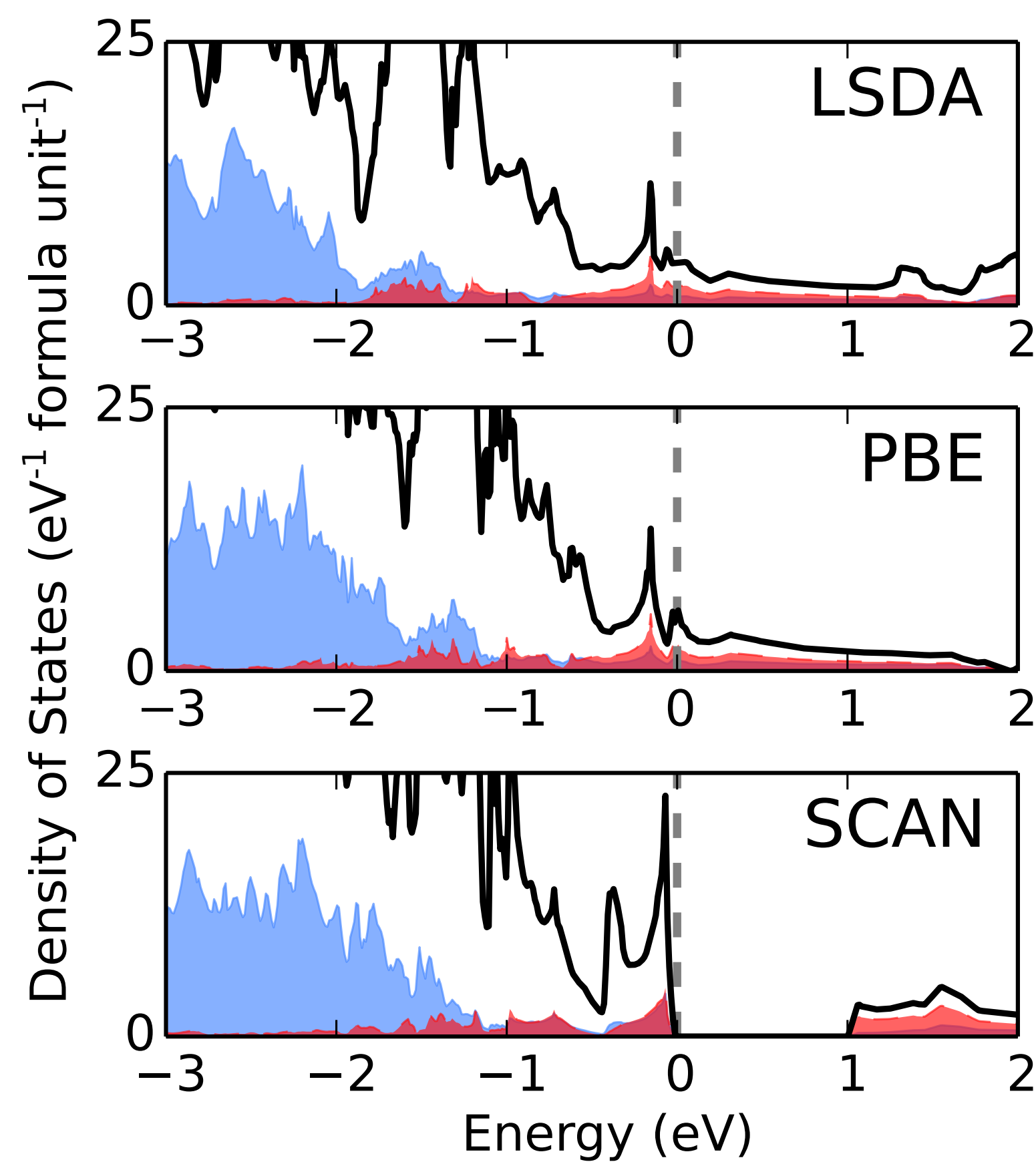
Pristine  $\text{La}_2\text{CuO}_4$ 

## Pristine Crystal Structure

- Insulator – optical leading edge band gap of 1.0 eV.
- Antiferromagnetic magnetic moments on Cu of  $0.48 \pm 0.15 \mu_B$ .



## Accurate Electronic and Magnetic Structures



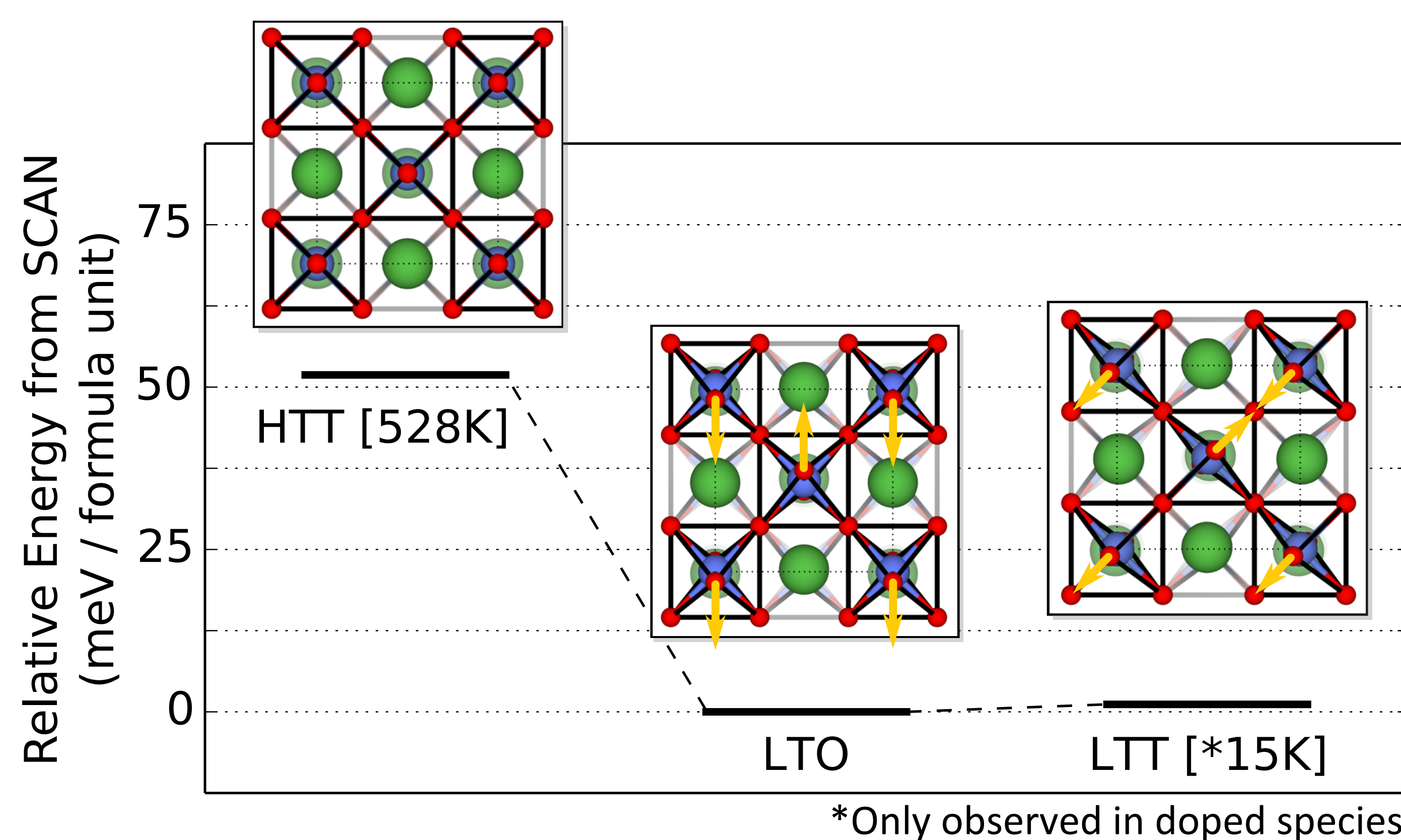
- LSDA and PBE incorrectly predict metallic gap.
- SCAN predicts correct insulator gap of 1.0 eV.

- LSDA and PBE cannot stabilise copper magnetic moments.
- SCAN correctly stabilises copper magnetic moments.

LSDA	$0.11\mu_B$
PBE	$0.27\mu_B$
SCAN	$0.49\mu_B$
Exp.	$0.48\mu_B$

## Accurate Ionic Structure

- $\text{La}_2\text{CuO}_4$  exists in 3 isomorphs defined by local octahedral tilt mode.
- SCAN reproduces relative energetic ordering.



\*Only observed in doped species.

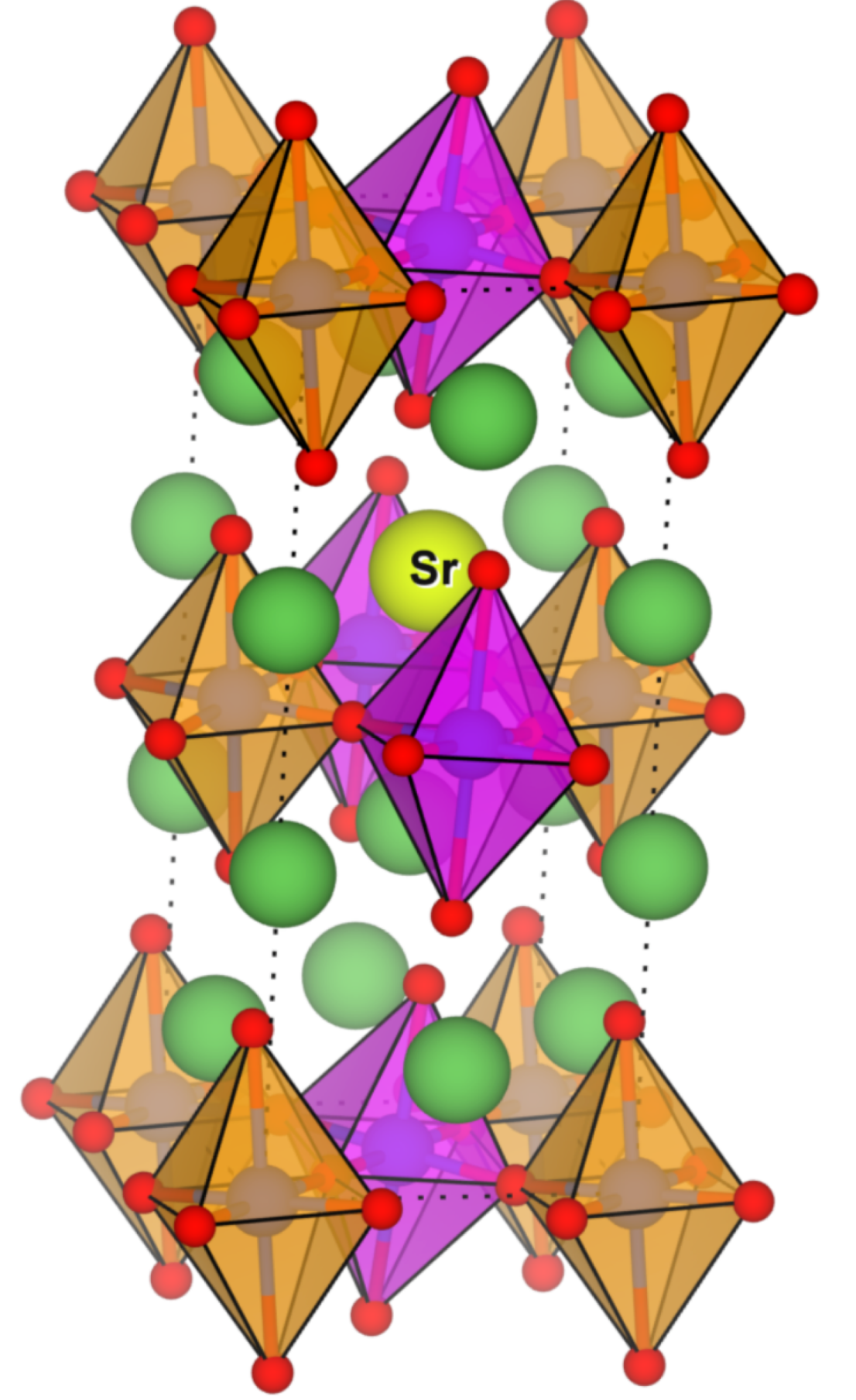
## Abstract

A first-principles density-functional description of the electronic structures of the high- $T_c$  cuprates has remained a long-standing problem since their discovery in 1986, with calculations failing to capture either the insulating (magnetic) state of the pristine compound or the transition from the insulating to metallic state with doping.

Here, by taking lanthanum cuprate as an exemplar high- $T_c$  cuprate, we show that the recently developed non-empirical, strongly-constrained and appropriately-normed (SCAN) density functional accurately describes both the antiferromagnetic insulating ground state of the pristine compound and the metallic state of the doped system. Our study yields new insight into the low-energy spectra of cuprates and opens up a pathway toward wide-ranging first-principles investigations of electronic structures of cuprates and other correlated materials.

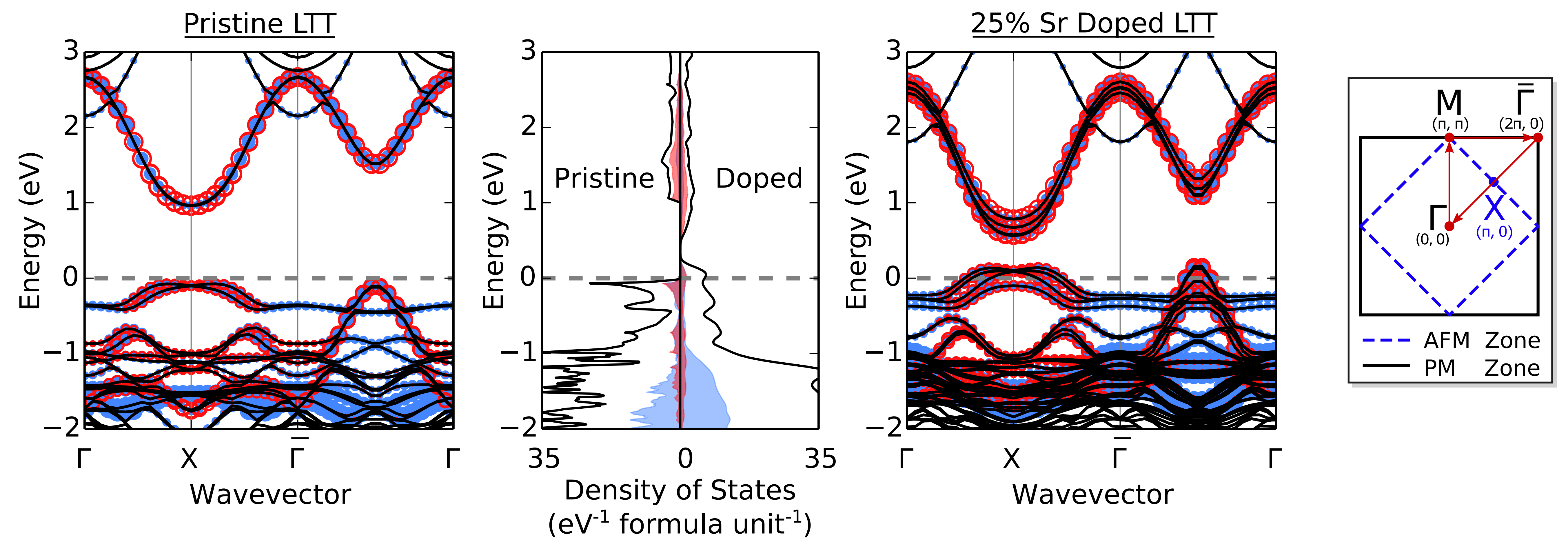
Doped  $\text{La}_{1.75}\text{Sr}_{0.25}\text{CuO}_4$  $\delta$ -Doped Crystal Structure

- Single La atom replaced by Sr in alternating layers.<sup>[1]</sup>
- Metallic Conductor.
- More complicated ferrimagnetic moments on copper atoms.



<sup>[1]</sup>Suter, A. et al.; Superconductivity drives magnetism in delta-doped  $\text{La}_2\text{CuO}_4$  (2017); arXiv:1706.07599

## Doping Dependent Transition



- The SCAN functional correctly captures the doping dependent insulator-metal transition
- Previous functionals fail to reproduce this transition, predicting metallic (LSDA, GGA) or insulator (Hybrid GGA) behaviour for both species.

## Functional Comparison

		LTO	Exp. LTT	HTT	LTO	SCAN LTT	HTT	LTO	PBE LTT	HTT	LTO	LSDA LTT	HTT
$\Delta E$	(meV FU <sup>-1</sup> )	10K	15K	528K	0.0	1.2	51.9	0.0	6.9	96.3	0.0	-0.1	22.1
Cu Magnetic Moment	( $\mu_B$ )	0.495	-	-	0.491	0.492	0.479	0.273	0.107	0.262	0.109	0.073	0.100
Indirect Band Gap	(eV)	1.0 <sup>†</sup>	-	-	0.979	1.006	0.918	0.026	0.000	0.000	0.000	0.000	0.000
Lattice Constants	a (Å)	5.335	5.360	5.391	5.323	5.391	5.348	5.352	5.471	5.401	5.220	5.285	5.258
	b (Å)	5.421	5.360	5.391	5.459	5.391	5.348	5.576	5.471	5.401	5.353	5.285	5.256
	c (Å)	13.107	13.236	13.219	13.088	13.071	13.125	13.101	13.075	13.163	12.956	12.956	12.989
	V (Å <sup>3</sup> )	379.1	380.3	384.2	380.3	379.8	375.4	391.0	391.4	384.0	362.0	361.8	358.9
Octahedral Tilt	axial (degrees)	5.5	3.8	0.0	7.2	6.9	0.0	8.7	8.5	0.0	5.8	5.5	0.0
	$\Delta E_d$ (meV FU <sup>-1</sup> )	-	15K	-	0.0	-0.5	18.3	0.0	-4.4	46.0	0.0	-0.4	2.7

<sup>†</sup>Leading edge in optical spectrum.  $\Delta E$  and  $\Delta E_d$  are energies relative to the LTO phase in the pristine and doped cases, respectively. FU denotes formula unit.

- The SCAN functional provides an accurate model for the prototypical high- $T_c$  cuprate, allowing further study of detailed magnetic behaviour.
- This opens the way for further investigation of strongly correlated materials using the SCAN functional.

- Nature Communications: Physics, Accepted.
- Physical Review Letters, Submitted.