

# Supplemental material: *Ab initio* description of the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ electronic structure

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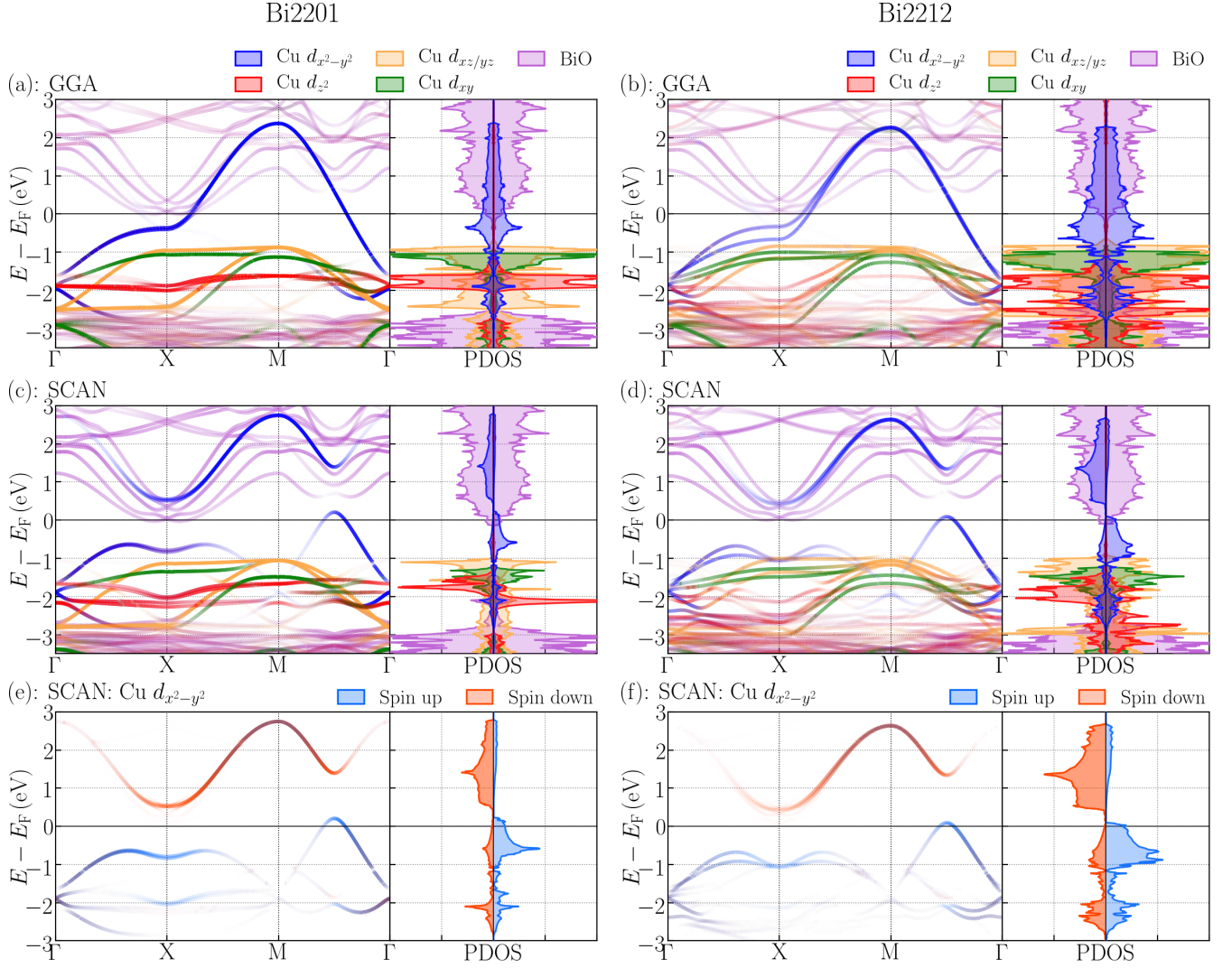
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## 1 $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ (Bi2201)

We have also performed calculations on the  $\text{CuO}_2$  monolayer compound Bi2201 which does not contain any calcium layers. Relaxing the structure gives a unit cell of  $5.294 \text{ \AA} \times 5.407 \text{ \AA} \times 24.860 \text{ \AA}$ . The corresponding experimental values are  $5.376(1) \text{ \AA} \times 5.383(1) \text{ \AA} \times 24.384(7) \text{ \AA}$  [97]. The agreement is reasonable, but not as good as in the case of Bi2212. Notably, the relaxed structure exhibits significant elongation in the *ab* plane. This feature of Bi2201 might be due to the doubling of the Bi/Cu ratio compared to Bi2212, so that the BiO zigzag chains include a greater reduction of the *a* axis (see the discussion in Sec. II A of the main text). In a physical sample this effect might be smaller due to the presence of type-B oxygen dopants, which would relieve the stress in the BiO layers. Also Bi atoms being substituted by other elements could result into this stress relief effect, as for example in Ref. [94] 20 % of the Bi atoms have been substituted with Ca.

The electronic structure of undoped Bi2201 is shown in Fig. 1 and compared with that of Bi2212. Three important differences between Bi2201 and Bi2212 can be observed as follows. (1) The self-doping is significantly stronger in Bi2201 because the bismuth pockets in Bi2201 reach further below  $E_F$  (compared to Bi2212) and the Cu  $d_{x^2-y^2}$  bands extend further above  $E_F$ . Also the energy gap in Cu  $d_{x^2-y^2}$  is decreased with about 0.1 eV. Consequently, the Cu magnetic moment is only  $0.395 \mu_B$ , which is  $0.030 \mu_B$  less than in Bi2212. These differences are due to the smaller number



of  $\text{CuO}_2$  planes in Bi2201, so that the effective Bi/Cu ratio in Bi2201 is twice as large, which makes the self-doping due to Bi more prominent. (2) There is no bilayer splitting in Bi2201. As a result, there is only one van Hove singularity at  $-0.6\text{ eV}$  instead of two singularities in Bi2212 at around  $-0.65\text{ eV}$  and  $-0.88\text{ eV}$ . Moreover, the  $d_{z^2}$  and  $t_{2g}$  related peaks in the DOS are significantly sharper in Bi2201 compared to Bi2212. (3) Curiously, our estimation for the value of  $U$  is  $4.3\text{ eV}$ , which is  $0.4\text{ eV}$  less than in Bi2212. However, the value of Hund's splitting ( $-1.34\text{ eV}$ ) in Bi2201 is almost equal to that of Bi2212.