

# Supplemental Materials - Is room temperature multiferroicity in cupric oxide under high pressure achievable? A combined experimental and computational investigation

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The Magnetocrystalline Anisotropy (MCA) describes the variation of the total energy induced by the rotation of the overall rotation of magnetic moments for a given magnetic ground state. During this study, we investigated the MCA by taking into account the Spin-Orbit Coupling as a perturbation at the second order of the total energy of such state. For a given orientation of magnetic moments along the  $[uvw]$  crystallographic direction, this quantity expresses as:

$$MCA_{[uvw]} = E_{[uvw]} - E_{min} \quad (1)$$

where  $E_{[uvw]}$  is the corresponding total DFT energy, and  $E_{min}$  is the DFT energy of the most stable orientation of magnetic moments.

Figure S 1 evidences the evolution of the MCA in the  $(\mathbf{a}, \mathbf{c})$  plane with respect to the hydrostatic pressure. By increasing the pressure from 0 GPa to 38 GPa, we showed that i) the easiest magnetization direction is still along the  $\mathbf{b}$  axis, ii) the MCA slightly rotates as evidenced by the direction associated to the maximum which moves away from the  $\mathbf{c}$  axis, and iii) the maximum is slightly decreasing. Our results are in good agreement with our previous DFT simulations [1].

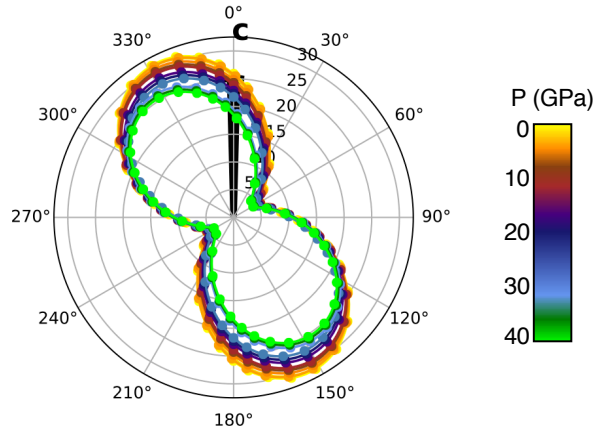


Figure S1: Dependency of the MCA (in  $\mu\text{eV}$ ) within the  $(\mathbf{a}, \mathbf{c})$  plane with the hydrostatic pressure (in GPa). The origin of the polar grid is set to the  $[001]$  direction.

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- [1] X. Rocquefelte, K. Schwarz, P. Blaha, S. Kumar, and J. van den Brink, Room-temperature spin-spiral multiferroicity in high-pressure cupric oxide, *Nat Commun* **4**, 2511 (2013).