Supplementary Information (SI) for Physical Chemistry Chemical Physics.

Tuning the Electronic and Magnetic Ground States of Layered Honeycomb Material Na_2IrO_3 via Strain Engineering

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Supporting Information

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1 Computational details

The electronic structure and magnetic ground state of Na_2IrO_3 have been investigated using density functional theory (DFT) as implemented in the Vienna ab initio Simulation Package (VASP) [1, 2]. The exchange-correlation interactions are treated within the generalized gradient approximation (GGA), using the projector augmented-wave (PAW) method [3]. Consistent with previous studies [4–6], we employ the GGA+SO+U formalism to account for spin-orbit coupling and on-site electron correlations, using $U = 1.7$ eV and Hunds exchange $J = 0.6$ eV. A plane-wave energy cutoff of 600 eV is used for describing valence electrons. A Γ-centered k-point mesh of $6\times4\times6$ is adopted for Brillouin zone sampling during self-consistent field (SCF) calculations. Electronic convergence is achieved with an energy tolerance of 10^{-8} eV between successive SCF steps. Ionic relaxation is performed until the Hellmann-Feynman forces on all atoms are below 5 meV/Å, corresponding to an energy convergence threshold of 10^{-4} eV between successive ionic steps. Phonon dispersion relations are calculated using density functional perturbation theory (DFPT) as implemented in the Phonopy package [7, 8]. Phonon calculations are performed on a $2 \times 1 \times 2$ supercell at the Γ point, with structural relaxation carried out until residual forces fall below 5×10^{-3} meV/Å. All calculations under strain are performed for both C2/c and C2/m crystal symmetries. Isotropic strain is applied within a range of -4% (compressive) to +4% (tensile), where the negative and positive signs indicate lattice compression and elongation, respectively.

2 Supporting Figures

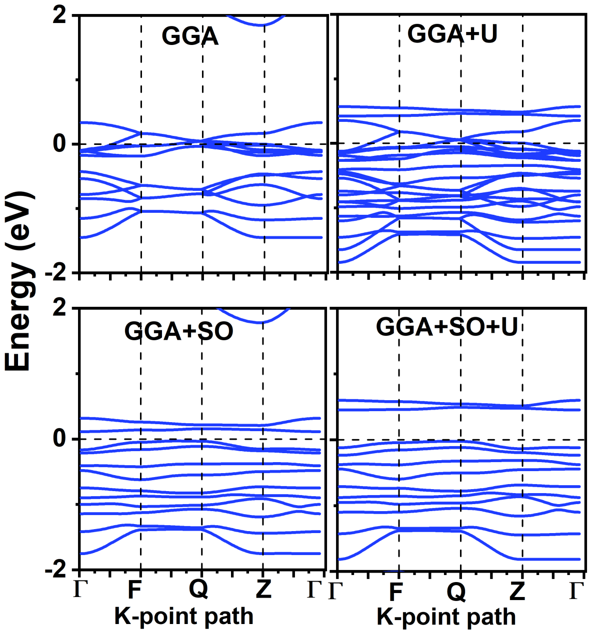

Fig. S1 Band dispersion plots using GGA, GGA+U, GGA+SO and GGA+SO+U methodologies for unstrained Na_2IrO_3.

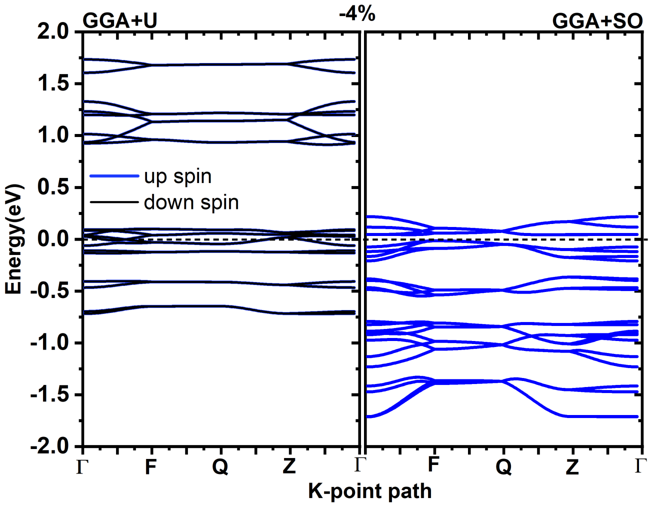

Fig. S2 Band dispersion plots under GGA+U and GGA+SO formalisms for −4% strained Na_2IrO_3.

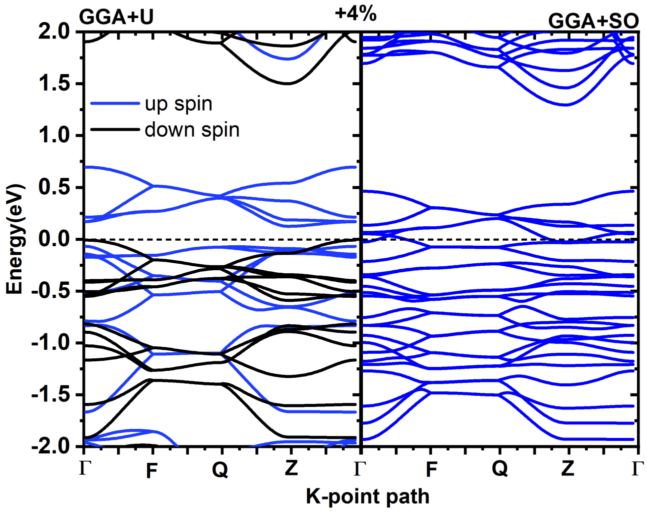

Fig. S3 Band dispersion plots under GGA+U and GGA+SO formalisms for +4% strained Na_2IrO_3.

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