

Near-Room-Temperature Easy-Plane p-Orbital Ferromagnetism in Half-Metallic Monolayer La₂CO₂

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S1. Robustness of Electronic Structure Calculation Methods

To verify the reliability of the half-metallic ferromagnetism predicted by the PBE functional[1] in the main text, we performed comparative calculations with higher precision methods. First, considering the heavy atomic mass of La, the influence of Spin-Orbit Coupling (SOC) on the band structure was evaluated. As shown in Figure S1(a), upon introducing SOC[2], the overall topological structure of the bands remains highly congruent with the non-SOC results. The energy deviation near the Fermi level is negligible (< 3 meV), indicating that SOC primarily affects the magnetocrystalline anisotropy direction rather than altering the electronic dispersion characteristics.

Secondly, to correct for the deficiency of the PBE functional in describing strong electronic correlations, calculations were performed using the HSE06 hybrid functional[3,4]. As depicted in Figure S1(b), the HSE06 results show that the band gap of the spin-up channel is corrected and increased to 3.33 eV, and the exchange splitting near the Fermi level is significantly enhanced. Crucially, the spin-down channel retains its metallic characteristic crossing the Fermi level. This provides

compelling evidence that the half-metallic nature of La_2CO_2 is an intrinsic and robust property of the material, rather than an artifact of the PBE functional.

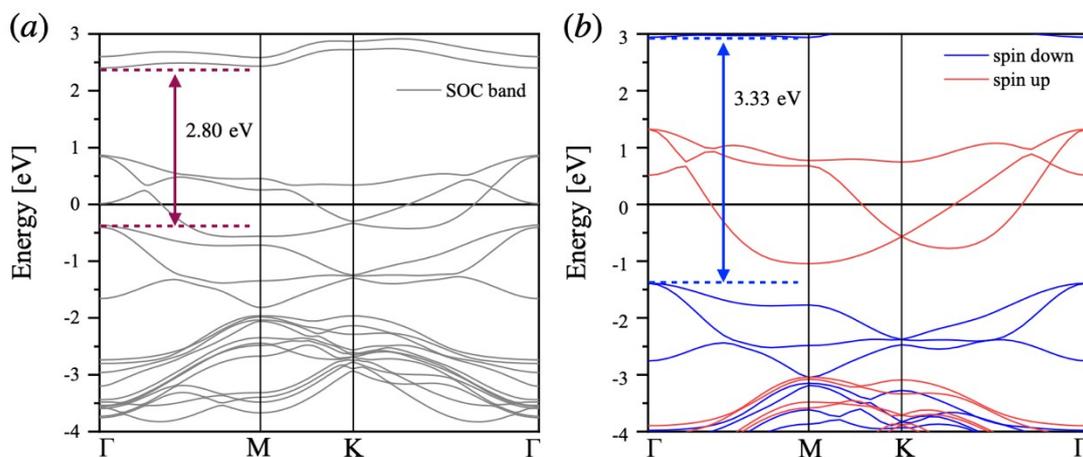


Figure 1 **Validation of the electronic band structure of monolayer La_2CO_2 .** (a) Band structure calculated with Spin-Orbit Coupling (SOC) included. The band gap is 2.80 eV, and the dispersion remains largely consistent with non-SOC results. (b) Band structure obtained using the HSE06 hybrid functional. The band gap of the spin-up channel (red) is corrected to 3.33 eV, while the spin-down channel (blue) remains metallic. This confirms the robustness of the half-metallic ferromagnetism against electronic correlation effects.

S2. Projected Band Structure and Hybridization Mechanism

To micro-scopically elucidate the "band inversion" and the origin of anisotropy discussed in the main text, a projected band (Fat-band) analysis was conducted. As shown in Figure S2(a), the p-orbitals of the C atom exhibit strong spin-dependent occupation: in the spin-up channel, the valence band maximum is dominated by in-plane p_x, p_y orbitals, while the vertical p_z orbital is nearly empty and located in the conduction band; conversely, in the spin-down channel, the metallic states crossing the Fermi level are primarily contributed by the p_z orbital. This selective occupation of p-orbitals directly leads to the anisotropic distribution of the electron cloud.

Furthermore, Figure S2(b) displays the orbital contributions from La and O atoms. La atoms exhibit significant weight near the Fermi level and at the conduction band minimum, with a dispersion shape highly matching that of the C- p_z orbital. This directly confirms that the La d-f orbitals are not fully localized but undergo strong p-d/f hybridization with the C p-orbitals. It is the additional Coulomb repulsion introduced by this hybridization that drives the shift of the C- p_z band relative to the p_x, p_y bands, thereby establishing the unique electronic structure.

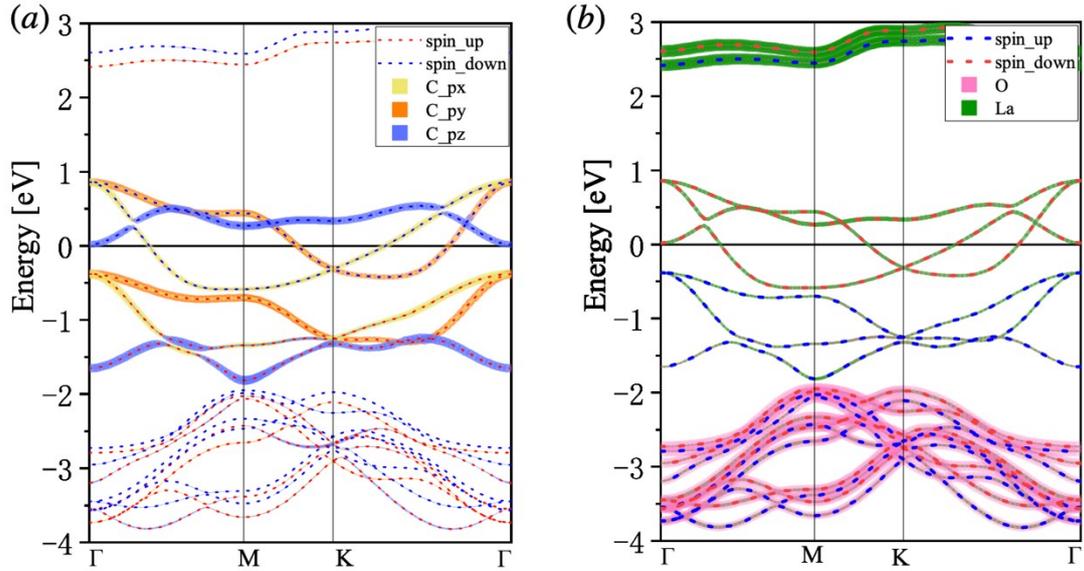


Figure S2 **Projected band structures (fat-band plots) of monolayer La₂CO₂**. (a) Band projections onto C p orbitals. Yellow, orange, and blue dots represent the weights of px, py, and pz orbitals, respectively. The plot reveals that in the spin-up channel, the valence band maximum is dominated by occupied px,y orbitals while the pz orbitals are empty (in the conduction band); conversely, the metallic states crossing the Fermi level in the spin-down channel originate primarily from the pz orbital. (b) Band projections onto La and O atoms. Green and pink dots denote contributions from La and O atoms, respectively, highlighting the significant orbital hybridization of La near the Fermi level and in the conduction bands.

S3. Physical Origin of Ferromagnetism: The Stoner Mechanism

The main text identifies La₂CO₂ as an itinerant ferromagnet. To quantitatively validate this conclusion, we calculated the electronic Density of States (DOS) in the non-spin-polarized (NM) state. As shown in Figure S3, a remarkably sharp DOS peak appears at the Fermi level (E_F) in the non-magnetic state, with a magnitude of $N(E_F) \approx 8.34$ states/eV.

Such a massive $N(E_F)$ implies a high degree of electronic instability in the paramagnetic state. According to the Stoner criterion for ferromagnetism, $I \cdot N(E_F) > 1$ (where I is the Stoner exchange parameter), this instability forces a spontaneous splitting of the spin-up and spin-down bands to lower the total system energy. This result provides thermodynamic proof that the magnetism in La₂CO₂ originates from the itinerant ferromagnetism of C-p electrons, rather than traditional local moment mechanisms.

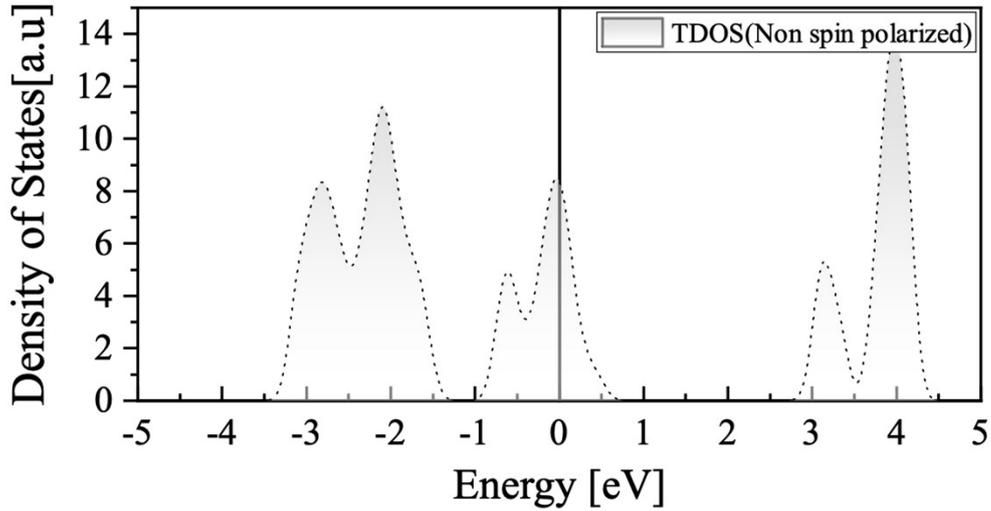


Figure S3 **Total density of states (TDOS) of monolayer La_2CO_2 in the non-spin-polarized state.** The plot reveals a prominent peak at the Fermi level ($E_F=0$ eV) with a magnitude of $N(E_F) \approx 8.34$ states/eV. This exceptionally high DOS signifies a strong electronic instability in the non-magnetic state, which is the crucial factor satisfying the Stoner criterion ($I \cdot N(E_F) > 1$) and driving the spontaneous ferromagnetic polarization of the system.

S4. Strain-Tunable Electronic and Magnetic Properties

To assess the feasibility of strain engineering as a tuning strategy for monolayer La_2CO_2 in flexible spintronic applications, we examined its electronic and magnetic responses under biaxial strain. Figure S4(a) presents the strain-dependent spin-polarized band structures over a range of -3% to $+3\%$. Remarkably, the half-metallic character remains intact across the entire strain window, with the Fermi level preserving 100% spin polarization despite the applied deformation—a key requirement for stable device operation under mechanical stress.

The strain response of the magnetic exchange coupling J is shown in Figure S4(b). The ferromagnetic interaction persists throughout the investigated strain range, confirming the robustness of magnetic order against external perturbations. Notably, J exhibits a non-monotonic behavior, reaching a maximum under mild compression ($\approx -1\%$). Given the proportionality between J and the Curie

temperature T_C , this enhancement suggests that strategic strain application could

further elevate the already near-room-temperature T_C of La_2CO_2 .

These findings highlight the dual role of strain as both a stabilizer of the half-metallic state and a modulator of magnetic interaction strength. The ability to tune magnetic

properties via mechanical deformation, combined with the material's intrinsic structural flexibility, positions monolayer La_2CO_2 as a promising candidate for next-generation flexible spintronic devices where strain engineering can be readily integrated into device design.

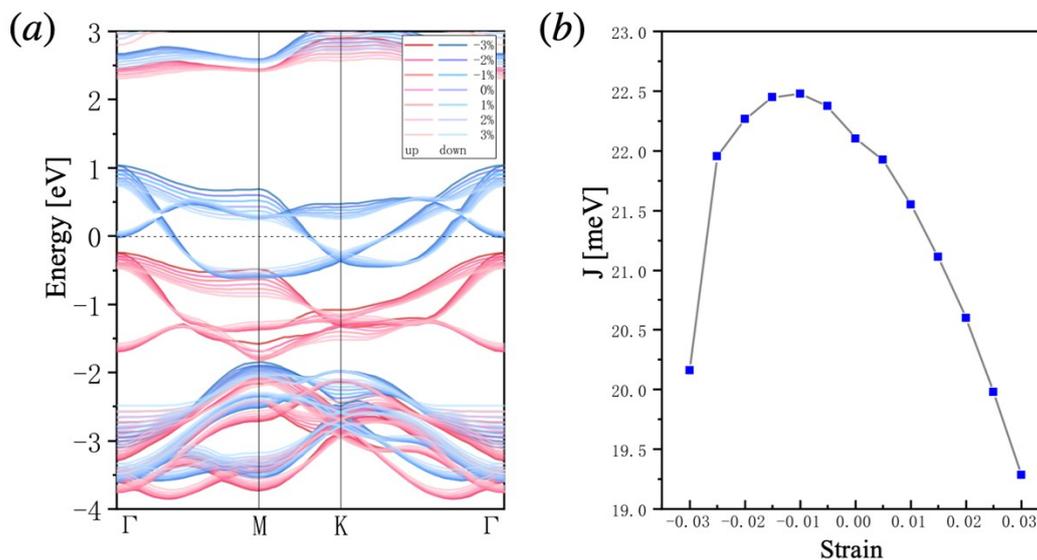


Figure S4 **Strain engineering of monolayer La_2CO_2 .** (a) Spin-polarized band structures under various biaxial strains ranging from -3% to 3%. The robust half-metallicity is preserved across the entire strain range. (b) The variation of magnetic exchange energy (J) as a function of biaxial strain.