# Supporting Information

## Curvature Geometry - Spin Electronics - Catalytic Dynamics Coupling in Emerging Catalytic Engineering

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## S1. Mathematical Description of Curvature–Spin Magnetic Effects

Curvature introduces a geometric perturbation that modifies the local electronic environment, altering orbital arrangements and lifting degeneracies in a manner highly sensitive to spatial asymmetry. This geometric reshaping directly affects spin–orbit interactions, strengthening the coupling between orbital and spin degrees of freedom and enabling spin-selective modulation of surface states and reaction intermediates.

When curvature breaks inversion symmetry or generates interfacial electric-field gradients, it induces momentum–spin locking analogous to Rashba-type behavior, creating spin-polarized surface channels that bias electron transfer and intermediate stabilization. Curvature and strain further modify magnetic exchange interactions, allowing transitions between different spin configurations of catalytic metal centers—an effect that can drastically alter the accessibility of spin-sensitive elementary steps such as oxygen activation, O–O coupling, or radical-mediated pathways.

By integrating geometric perturbation, spin–orbit effects, magnetic exchange, and the reaction coordinate into a unified theoretical picture, the mathematical framework clarifies how curvature-driven spin rearrangements influence activation barriers, rate-determining steps, and pathway selectivity. In essence, these formulations establish a rigorous physical foundation for the curvature–spin–catalytic dynamics triple-coupling mechanism: curvature reshapes electronic structure, electronic structure reorganizes spin manifolds, and the resulting spin landscape modulates the kinetics and selectivity of catalytic reactions. This provides not only conceptual clarity but also actionable guidance for designing next-generation catalysts that leverage curvature and spin as coupled reactivity modulators.

### S1.1 Curvature-induced geometric potential

For a curved two-dimensional surface with mean curvature $H$ and Gaussian curvature $K$, the effective geometric potential that modifies the local electronic states can be described as:

$$V_{geo} = -\frac{\hbar^2}{2m}(H^2 - K)$$

This term breaks local symmetry, perturbs orbital degeneracy, and shifts $d$-orbital energy levels, thereby influencing spin-state filling and catalytic intermediate binding.

S1.2 Spin–orbit coupling Hamiltonian

The SOC effect that couples spin polarization with curvature-modified electronic structure can be represented by:

$$H_{SOC} = \lambda L \cdot S$$

Where $\lambda$ is the SOC constant; L is orbital angular momentum; S is spin angular momentum.

Curvature modifies the orbital basis, effectively changing L, enhancing SOC in regions of large curvature.

S1.3 Rashba-type spin splitting (for asymmetric curved interfaces)

When curvature introduces local inversion asymmetry and interfacial electric fields, the Rashba Hamiltonian becomes relevant:

$$H_R = \alpha_R(\sigma \times k)\cdot\hat{z}$$

Where $\alpha_{\mathrm{R}}$ is the Rashba parameter; $\sigma$ is the Pauli spin matrix; k is momentum.

This term explains curvature-induced spin splitting that selectively stabilizes spin-allowed catalytic pathways.

S1.4 Exchange coupling under curvature-induced strain

Local strain or curvature affects magnetic exchange coupling:

$$H_{ex} = -J(\varepsilon)\sum_{\langle i,j\rangle} S_i \cdot S_j$$

Where J(ε) is strain/curvature-dependent exchange integral.

Curvature modifies J, enabling control over high-spin ↔ low-spin transitions at catalytic metal sites.

S1.5 Effective kinetic barrier under spin selection rules

For spin-dependent elementary steps, the effective activation energy becomes:

$$E_{a,eff} = E_a - \Delta_{spin}$$

$$\Delta_{spin} = E_{(spin-allowed)} - E_{(spin-forbidden)}$$

Curvature–SOC coupling increases the energy penalty for spin-forbidden channels, pushing the reaction toward spin-allowed low-barrier pathways.

S1.6 Curvature–spin–kinetics triple coupling

A general form of the Hamiltonian incorporating curvature, SOC, exchange coupling, and catalytic reaction coordinate $q$ is:

$$H = H_0 + H_{geo}(K,H) + H_{SOC} + H_R + H_{ex}(\varepsilon) + H_{kin}(q)$$

with the kinetic term:

$$H_{kin}(q) = \frac{1}{2}k(q - q^{\dagger})^2$$

where $q^{\ddagger}$ is the curvature- and spin-modulated transition state.

This form highlights:

geometry modifies electronic potential ($H_{\mathrm{geo}}$)

electronic potential reshapes spin textures ($H_{\mathrm{SOC}}$, $H_{\mathrm{R}}$)

spin textures modulate exchange and reaction pathways ($H_{\mathrm{ex}}$, $H_{\mathrm{kin}}$)

Establishing the curvature–spin–kinetic coupling triad.