

Research Highlight

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# Spin polarization effect enhances photocatalytic CO<sub>2</sub> reduction

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Efficient CO<sub>2</sub> conversion to high-value hydrocarbons such as CH<sub>4</sub> is crucial for carbon neutrality but is hindered by sluggish proton-coupled electron transfer and charge recombination. Enhancing electron density at active sites has been a key strategy, with approaches such as defect engineering, noble metal loading, and heterojunction construction yielding significant progress. However, inherent charge carrier limitations necessitate alternative strategies to further improve photocatalytic efficiency<sup>[1]</sup>.

Spin polarization, defined as the unequal distribution of spin-up and spin-down electrons within a system, has recently garnered significant attention as an effective strategy to modulate charge carrier dynamics in photocatalytic processes. This spin asymmetry can be induced via the incorporation of magnetic metal dopants (e.g., Mn, Fe), the introduction of cationic vacancies, or through structural asymmetry, and can be further amplified under the application of an external magnetic field. During the transport of



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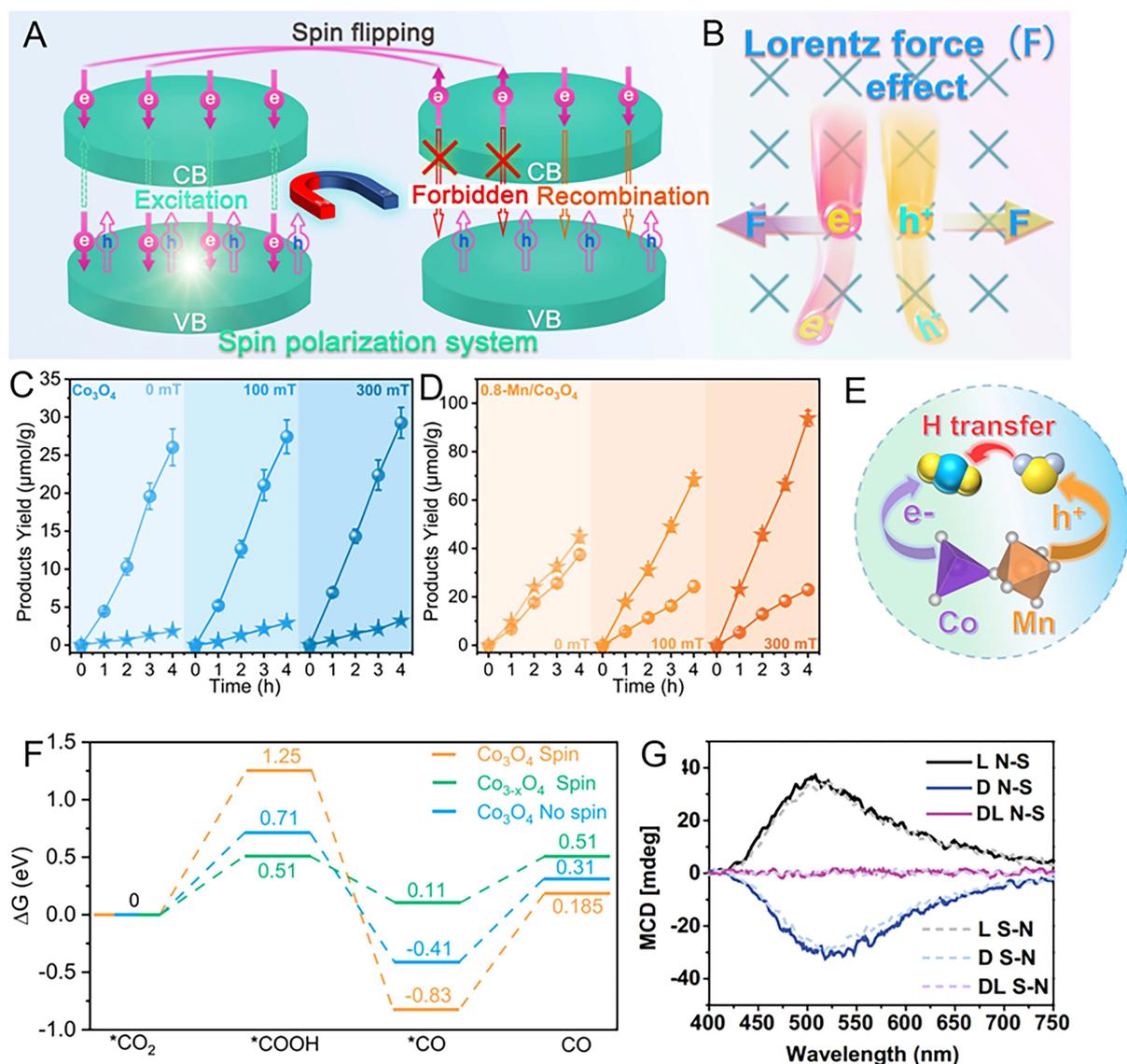
photogenerated electrons, spin flipping may occur as a result of spin-orbit coupling and hyperfine interactions. In conventional, non-spin-polarized systems, such spin-flipped electrons can still recombine readily with holes of opposite spin. By contrast, in spin-polarized systems, flipped electrons are less likely to encounter spin-compatible holes [Figure 1A], thereby effectively suppressing charge recombination. This mechanism prolongs the lifetime of photogenerated carriers and facilitates enhanced spatial separation of charges. In addition to reinforcing spin polarization, an external magnetic field also induces Lorentz forces that drive photogenerated electrons and holes in opposite directions [Figure 1B], thereby accelerating carrier migration and further diminishing recombination probability. The synergistic effects of spin-selective recombination suppression and magnetically induced charge separation ultimately lead to significantly enhanced photocatalytic efficiency<sup>[2]</sup>.

The spin polarization effect has achieved considerable advancements in photocatalytic hydrogen production and has recently emerged as a promising strategy for CO<sub>2</sub> photoreduction. For instance, Lin *et al.* investigated the role of spin polarization in manganese-ion (Mn<sup>2+</sup>)-doped halide perovskite nanoplates (CsPbBr<sub>3</sub> NPLs)<sup>[3]</sup>. Their study demonstrated that Mn doping, in conjunction with an external magnetic field, enabled precise modulation of spin-polarized electrons, significantly enhancing the efficiency of photocatalytic CO<sub>2</sub> reduction. However, while this work highlighted the role of spin polarization in promoting charge carrier separation, it had a negligible impact on product selectivity in CO<sub>2</sub> reduction.

Building upon this strategy, we incorporated Mn dopants into Co<sub>3</sub>O<sub>4</sub> to induce spin polarization. The Mn sites functioned as hole-enriching centers adjacent to Co, forming spatially proximate redox-active sites<sup>[4]</sup>. The Mn-induced spin polarization effect, further amplified by an external magnetic field, effectively suppressed charge recombination between these neighboring redox sites. Therefore, the spatial confinement of redox centers facilitated efficient proton transfer, which not only led to a remarkable 28.8-fold increase in CO<sub>2</sub> reduction efficiency but also promoted multi-electron-proton coupling [Figure 1C-E], thereby enhancing methane selectivity from 30.7% to 94.2%<sup>[4]</sup>.

To further elucidate the role of spin polarization in photocatalytic CO<sub>2</sub> reduction, we designed cobalt oxide with engineered Co vacancies. Through comprehensive experimental and theoretical investigations, we demonstrated that spin polarization not only enhances charge separation kinetics but also optimizes the energy landscape of CO<sub>2</sub> reduction [Figure 1F]<sup>[2]</sup>. Notably, for the first time, we provided a thermodynamic framework to explain how spin polarization facilitates photocatalytic CO<sub>2</sub> reduction, offering profound mechanistic insights into its fundamental role in guiding charge dynamics and reaction thermodynamics. Although the application of spin polarization in CO<sub>2</sub> photoreduction remains limited, existing studies collectively suggest that its promotional effects can be primarily attributed to two aspects: first, the suppression of charge carrier recombination and the enhancement of carrier migration, which improve the reaction kinetics; second, the modulation of energy barriers in key reaction steps, thereby favorably influencing reaction thermodynamics. These combined effects not only enhance the overall catalytic activity but may also steer the reaction pathway, potentially altering product selectivity.

Despite the significant potential of spin polarization in CO<sub>2</sub> photoreduction, research in this field remains in its early stages. Notably, in electrocatalysis and other photocatalytic domains, the modification of materials with chiral molecules or the fabrication of spin-polarized materials has emerged as a growing research focus. For example, Ai *et al.* utilized chiral methionine molecules as symmetry-breaking agents to synthesize chiral ZnO [Figure 1G]<sup>[5]</sup>. Their study revealed that the chiral structure functions as a spin filter, inducing spin polarization, which not only prolongs carrier lifetimes but also increases the population of triplet species, ultimately enhancing photocatalytic oxygen evolution efficiency by more than twofold.



Looking ahead, the incorporation of chiral molecular modification or the development of intrinsically spin-polarized materials is expected to advance the design of highly efficient spin-polarized materials for CO<sub>2</sub> photoreduction. Moreover, such approaches will provide deeper mechanistic insights into the intrinsic role of spin polarization in governing charge dynamics and reaction pathways in CO<sub>2</sub> photoreduction.

Beyond CO<sub>2</sub> photoreduction, spin polarization has demonstrated significant potential in various catalytic applications, including electrocatalysis and other photocatalytic reactions, suggesting that spin polarization

may serve as a universal strategy for improving charge dynamics across diverse catalytic systems. However, challenges remain in precisely controlling spin polarization effects in non-magnetic materials, as well as in designing stable spin-polarized interfaces under reaction conditions. Developing advanced characterization techniques and theoretical models will be critical for further elucidating these mechanisms and guiding material design.

In summary, spin polarization represents a groundbreaking advancement in photocatalytic CO<sub>2</sub> reduction, addressing critical challenges in charge carrier dynamics and product selectivity. The rational design of spin-polarized materials, particularly through chiral modifications and defect engineering, will be key to unlocking new levels of efficiency in CO<sub>2</sub> photoreduction. Future research should focus on deepening the mechanistic understanding of spin polarization at the atomic level and exploring synergistic strategies that combine spin-selective effects with advanced photocatalytic architectures.

## DECLARATIONS

### Authors' contributions

Wrote the draft manuscript: Wang, L.

Revised and rewrote some parts of the manuscript: Wu, S.; Zhang, J.

### Availability of data and materials

Not applicable.

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### Conflicts of interest

All authors declared that there are no conflicts of interest.

### Ethical approval and consent to participate

Not applicable.

### Consent for publication

Not applicable.

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Shiqun Wu received his Ph.D. in 2021 from East China University of Science and Technology under the supervision of Prof. Jinlong Zhang, and subsequently continued his postdoctoral research at the same institution. In 2024, he was appointed as an Associate Researcher at East China University of Science and Technology. His research interests focus on the design and modulation of atomically dispersed active sites in photocatalysts, with applications in the photocatalytic conversion of methane and carbon dioxide. He is currently a Youth Committee Member of the Photochemistry Division of the Chinese Renewable Energy Society, a Youth Editorial Board Member of *Carbon Neutralization*, and a Member of the Working Group of Youth Editor of *Chemical Synthesis*.

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