Spin-related Electron Transfer and Orbital Interactions in Oxygen Electrocatalysis

Yuanmiao Sun, Shengnan Sun, Shibo Xi, Jose Gracia*, and Zhichuan J. Xu**

Dr. Y. Sun, Dr. S. Sun, Prof. Z. J. Xu
School of Materials Science and Engineering, Nanyang Technological University, 50
Nanyang Avenue, Singapore 639798, Singapore
E-mail: xuzc@ntu.edu.sg

Dr. S. Sun
Beijing Key Laboratory for magnetoelectric Materials and Devices (BKLMMD), Beijing
Innovation Center of Materials Science and Engineering, College of Engineering, Peking
University, Beijing 100871, China

Dr. S. Xi
Institute of Chemical and Engineering Science A*Star, 1 Pesek Road, Singapore 627833,
Singapore
E-mail: xi_shibo@nus.edu.sg

Dr. J. Gracia
MagnetoCat SL, General Polavieja 9 3I, 03012 Alicante, Spain
E-mail: magnetocat@outlook.com

Prof. Z. J. Xu
Energy Research Institute @ Nanyang Technological University, 50 Nanyang Avenue,
Singapore 639798, Singapore

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Abstract:

Oxygen evolution and reduction reactions play a critical role in determining the efficiency of the water cycling ($H_2O \Leftrightarrow H_2 + \frac{1}{2}O_2$), in which the hydrogen serves as the energy carrier. That calls a comprehensive understanding of oxygen electrocatalysis for efficient catalyst design. Current opinions on oxygen electrocatalysis have been focused on the thermodynamics of the reactant/intermediate adsorption on the catalysts. Because the oxygen molecule is paramagnetic, its production from or its reduction to diamagnetic hydroxide/water involves spin-related electron transfer. Both electron transfer and orbital interactions between the catalyst and the reactant/intermediate show spin-dependent character, making the reaction kinetics and thermodynamics sensitive to the spin configurations. This article provides a brief introduction on the spintronic explanation of the catalytic phenomena on oxygen evolution reaction (OER) and oxygen reduction reaction (ORR). The local spin configurations and orbital interactions in the benchmark transition-metal-based catalysts for OER and ORR are analysed as examples. To further understand the spintronic oxygen electrocatalysis and to develop more efficient spintronic catalysts, the challenges and future opportunities have been summarized and proposed. The spin electrocatalysis may merge as an important topic in the near future and help integrate a comprehensive understanding of oxygen electrocatalysis.

1. Introduction

The increasing desire to establish a clean and sustainable energy infrastructure requires a comprehensive understanding of oxygen electrocatalysis, which limits the efficiency of many renewable and eco-friendly energy technologies, such as fuel cells and water splitting. Currently, theoretical insights of oxygen electrocatalysis mainly focus on the thermodynamic features of the adsorption/desorption of reactants and intermediates. Besides the thermodynamic features, the electron transfer between the adsorbed reactants/intermediates and the active sites, and the charge transport within the catalysts during the electrocatalysis, are also a crucial factor influencing the reaction kinetics. That is, a comprehensible understanding of oxygen electrocatalysis should consider both the orbital interactions and the electron transfer behaviour.

In domain of oxygen electrocatalysis (at room temperature in water), the electron transfer exhibits highly spin-related character because the ground state of O_2 molecule is a triplet state (↑ O=O ↑) (Figure 1). That is why O_2 is paramagnetic. Therefore, either from H_2O/OH^- to O_2 (oxygen evolution reaction, OER) or from O_2 to H_2O/OH^- (oxygen reduction reaction, ORR), the involvement of the triplet O_2 requires spin-related electron transfer along these oxygen reactions, which plays a considerable role in the reaction kinetics.

The aspect of spin-related electron transfer has been unintentionally neglected and only a few pioneer works have noticed and emphasized its role. An early attempt can be traced back to

1990, where the correlation between oxygen reduction activity and the magnetic susceptibility was investigated on perovskites.[1a] Thereafter, R. Nazmutdinov tentatively discussed the possibility of taking advantage of the spin effects to optimise oxygen electrocatalysis.[1b] More recently, J. Gracia et al. found that, during OER and ORR, since the number of unpaired electrons is not conserved, spin-potentials in optimum magnetic catalysts act as selective gates to influence the transport of local spin currents.[1c] A persuasive phenomenon is that magnetic oxides (MMOs) with anti-ferromagnetic (AFM) spin coupling exhibits much poorer conductivity than those with ferromagnetic (FM) couplings. The OER activity increases as inter-atomic FM couplings increase, indicating direct correlation between spin-selected charge transport mobility and reaction activity.[1c] Moreover, cooperative (extended in space) FM orbital orderings in MMOs helps to moderate the binding energies of the reactants through quantum spin(-orbital) exchange interactions (QSEI), and correspondingly, from a thermodynamic point of view, their catalytic efficiencies improve in accordance with Sabatier's principle.[1d]

Inspired by these pioneering works, some more spin-related observations have been reported recently. For example, we have found the high spin state Co^{2+} cations ($t_{2g}^5e_g^2$) in spinel $LiCoVO_4$ act as spin channels to enhance the selective removal of spin-oriented electrons from the reactants during OER and facilitate the triplet oxygen molecule evolution.[2a] J. Galán-Mascarós observed a direct magnetic enhancement of electrocatalytic water oxidation under a moderate magnetic field and the enhancement is found to be positively correlated with the magnetization.[2b] We also found that spin-related electron transfer can switch the rate-determining step (RDS) of OER on spinel oxides, in which the RDS on the low-spin $ZnCo_2O_4$ is *O to *OOH, while that on the high-spin $ZnMn_2O_4$ is *OH to *O.[2c] Although several spin-related OER enhancements have been observed, and the quantum nature of the spin potentials that moderates the catalytic have been recognized, continuous efforts are required to address its role in more state-of-the-art oxygen electrocatalysts.

2. Examples of Spin-electrocatalysis in OER and ORR

To date, layered Ni/Fe oxyhydroxide ($Ni_xFe_{1-x}OOH$) exhibits the best alkaline OER activity. However, neither pure FeOOH nor pure NiOOH can be as active as $Ni_xFe_{1-x}OOH$ for catalysing OER and the presence of both Ni and Fe is necessary to achieve such a high activity. For example, a surface of $Ni_xFe_{1-x}OOH$ ($x > 0.7$) gives the intrinsic activity as high as 1000-fold over the pure NiOOH.[3] Nevertheless, although the remarkable activity of $Ni_xFe_{1-x}OOH$ has been widely recognised, it yet remains disputed whether nickel, iron, or the combination of nickel and iron sites are the catalytic centres. Using the X-ray adsorption spectroscopy (XAS) and electron energy loss spectroscopy (EELS) techniques, D. G. Nocera et al. proposed that Fe(III) serves as a superior Lewis acid and facilitates the formation of Ni(IV), which is responsible for the enhanced catalytic activity for OER.[4a] Employing in situ Raman spectroscopy and XAS approaches, W. Smith's group observed $NiOO^-$ species (described as adsorbed "active oxygen") in $Ni_xFe_{1-x}OOH$ and highlighted its nonnegligible role for alkaline OER.[4b] In total contrary, A. J. Bard et al. applied surface interrogation scanning electrochemical microscopy (SI-SECM) technique to observe the fast OER kinetics on iron sites and attributed the activity to the irons in the matrix.[5a] More recent studies by S. Boettcher and X. Hu also emphasized the crucial role of Fe sites, using electrochemical and operando Raman spectroscopy techniques, respectively.[3,5b] Besides, utilizing the density functional theory (DFT) calculations, Goddard's group argued that it is the synergy between Ni and Fe that delivers the optimal performance of OER.[6a] An experimental work using Mössbauer Spectroscopy also implied the insight of the synergy between Ni and Fe.[6b] The above examples have employed various approaches and tools to identify the catalytic sites in $Ni_xFe_{1-x}OOH$, providing valuable clues for establishing the current understandings of OER on $Ni_xFe_{1-x}OOH$. However, to date no attention has been paid to the local spin configurations of Ni and Fe, in relation with the overall intra- and inter- atomic QSEI, which we believe could be another angle to pinpoint the active sites in $Ni_xFe_{1-x}OOH$.

Here we propose a new point of view to identify and explain the catalytically active sites in $Ni_xFe_{1-x}OOH$ through analysing the local spin configurations of iron and nickel. Figure 2a shows the *d*-electron configurations of iron and nickel cations at the edge sites, where reaction likely takes place.[7] The 5-coordinated cations are considered as possible reaction sites because the unsaturated coordination allows the formation of adsorbed intermediates (the site becomes 6-coordinated after the adsorption).[8] The iron and nickel cations with +3 valency are analysed based on our previous investigations.[9] The stable spin configurations of iron and nickel cations are in high spin state and low spin state, respectively. The high spin state of Fe^{3+} enables each the t_{2g} and e_g* orbitals to be occupied by one unpaired electron with single spin, making the whole *d*-shell acting as selective gates to promote the transfer of local spin currents. On the other hand, the low spin configuration of Ni^{3+} initially only has unpaired spin at e_g* orbitals. The spin distribution in Figure 2b evidently suggests a spin channel around the Fe sites, while no such channel can be propagated at Ni sites. During the OER process, oxygen molecule evolves via the intermediates of *OH, *O, and *OOH (* denotes the active site). The possible orbital interactions between iron and nickel cations at the surface of $Ni_xFe_{1-x}OOH$ and the intermediates species are shown in Figure 2c. Because of symmetry conservation, the interactions between the d_{x2-y2} and d_{xy} orbitals of cations and the orbitals of the adsorbed intermediates are negligible.[10] Therefore, neither d_{x2-y2} nor d_{xy} is shown in the diagram. Thermodynamically, to guarantee an easy start of reactant capture, a good OER catalyst should adsorb *OH strongly. To quantify the adsorption strength, we have analysed the bond order of each intermediate bound to Fe^{3+} and Ni^{3+} (Figure 2c). The bond order is defined as half the difference between the number of bonding electrons and the number of antibonding electrons (bond order = (number of bonding electrons – number of antibonding electrons)/2). Due to the spin orbital interactions, the bond orders of *OH on Ni^{3+} and Fe^{3+} are 1 and 2, respectively.

Since a higher value of bond order indicates a stronger orbital interaction between the cation and the reaction intermediate, compared to the nickel site, the ion site is more favourable for starting an OER cycle. The oxygen gas releases after the formation of *OOH (the final OER intermediate), whilst another *OH (the first OER intermediate) occupies the catalytic site and a new OER cycle begins. To facilitate an effortless and continuous product release, a good OER site should adsorb *OOH less strongly than *OH. On Ni^{3+} site, *OH and *OOH show equal bond order; while on Fe^{3+} site, the thermodynamic advantage of product release emerges, as *OH exhibits higher bond order than *OOH. That is, in $Ni_xFe_{1-x}OOH$, iron sites promote OER in both the reactant capture and product release. It's worth noting here that the different reaction thermodynamics on the two sites originate from the distinct *d*-orbital occupations of Ni^{3+} and Fe^{3+}, which are intrinsically determined by the spin state of the cations. Therefore, the local spin configurations on iron and nickel determine their mutual inter-atomic QSEI couplings and thus the features of electron transfer mobility and thermodynamic oxygen evolution pathway, making a valid possibility that Fe^{3+} ions are more active than Ni^{3+} in the presence of extended FM couplings.

However, in the case of FeOOH, where all the possible sites are Fe^{3+}, the catalytic activity for OER is instead inferior, which could likely be attributed to the poor electrical conductivity, as its activity is observed to increase at high overpotentials, where conductivity increases.[11] The intrinsic conductivity limitation is caused by the AFM QSEI in FeOOH, where the next-to-nearest neighbour iron cations respectively exhibit spin-up and spin-down character (Figure 2d up).[12] This brings about the opening of a band gap (Figure 2e up) associated with pronounced Jahn-Teller distortions. Therefore, in FeOOH, the oxygen ligand connecting to two adjacent iron cations shows quite low spin density (0.080 e^-) and the long-range spin-related charge transport inside the FeOOH catalyst is poor (Figure 2f up). In contrast, in the case of $Ni_xFe_{1-x}OOH$ ($x > 0.7$), the coupling between high-spin Fe^{3+} and low-spin Ni^{3+} leads to a FM QSEI, where the spin density in ion is parallel to that in nickel (Figure 2d down). This enables a spinhole[13] (0.207 e^- spin density) in the oxygen atoms bound between iron and nickel cations to facilitate charge transport inside the catalyst, which leads to a half-metal density of states (DOS) pattern (Figure 2e down) and creates unblocked spin channel along the lattice (Figure 2f down). Thus, the presence of both Ni and Fe in the oxyhydroxide with Ni as dominant component is necessary for a high OER activity. This is exactly what have been found experimentally to date as mentioned in the above references.

Similarly, we also notice the logics of using local spin configurations to explain the catalytic phenomena in ORR. As alternatives to Pt-based materials, MN_4 moieties (M = Mn, Fe, and Co) embedded in partially graphitized carbon have emerged as remarkable catalysts to achieve both high activity and low cost.[14] Among the various MN_4 coordination catalysts, FeN_4 is reported to exhibit the highest ORR activity, whose intrinsic activity is reported as high as 6.89 mA m^{-2}.[15a] The current explanation of the promising ORR activity on FeN_4 mainly focus on the viewpoint of electronic structures or free energy diagram computed through DFT calculations. For example, G. Wu et al. have computed the density of states (DOS) of FeN_4 with and without a 2% Fe–N bond contraction (Figure 3a) and attributed the better activity of FeN_4 with Fe–N bond contraction to the positive shifting of the 3d orbitals of central Fe, which facilitates oxygen adsorption on FeN_4 sites (Figure 3b).[15b] Y. Xie et al. calculated the free energy diagrams of ORR on pyrrole-type and pyridine-type FeN_4. They found that pyrrole-type FeN_4 exhibits lower thermodynamic overpotential (0.35 eV) from the initial state (O_2) to the final state (H_2O) than that of pyridine-type (0.67 eV) in a typical $4e^-$ pathway.[15a] Combining computational calculations and electrochemical experiments, J. Li et al. revealed the formation of an axial hydroxyl ligand on the Fe center under certain electrode potentials, which weakens the following adsorption of ORR intermediates and results in improved ORR activities.[15c] However, the viewpoint from the local iron spin configurations has rarely been proposed and it can be a new, but important angel to understand the ORR on such single atom catalysts. Taking the work by G. Wu et al.[15b] as an example, we describe here how to take advantage of the local spin configurations to analyse the ORR performance on FeN_4. According to the DOS pattern (Figure 3a), an obvious increase in the spin density (the intensity of spin-up minus spin-down) is observed on FeN_4 with Fe–N bond contraction, indicating a high spin state. The possible spin configurations of Fe are shown in Figure 3c. The higher spin state of Fe in FeN_4 with bond contraction can create a wider spin-related channel in FeN_4 (Figure 3d), promoting the charge transport during ORR. The enhanced charge transport mobility is also evidently demonstrated by the decreased band gap in FeN_4 with Fe–N bond contraction (Figure 3a). Moreover, the oxygen molecule can be more easily captured by FeN_4 with Fe–N bond contraction because of higher bond order resulted from the spin-orbital interactions between Fe and O_2 (Figure 3e), which should be the intrinsic factor dominating the DFT calculated trend of O_2 adsorption.

Another explanation given by F. Kang et al. indicates that the on-site magnetic moment of the Fe center in FeN_4 sites can be linearly correlated with the key thermodynamic descriptor of *OH adsorption free energy (ΔG_{*OH}).[16] By conducting DFT calculations on the models of a series of FeN_4 sites with different amount of neighbouring graphitic nitrogen (Figure 4a), they constructed a volcano plot of the theoretical ORR overpotential (V *vs.* RHE) as a function of ΔG_{*OH} (Figure 4b). The relatively poorer activity of pristine FeN_4 site is due to the excessively strong adsorption energy of *OH, which leads to a low ΔG_{*OH} value (low value of ΔG_{*OH} indicates strong chemisorption). Of the screened FeN_4 sites with different amount of neighbouring nitrogen, FeN_4-3N-1 is calculated to have the optimal ΔG_{*OH} and locates at the summit of the volcano, estimating the lowest overpotential of 0.28 V. Further computational analysis found a negatively-linear correlation between ΔG_{*OH} and the on-site magnetic moment of the Fe center (Figure 4c), clearly indicating the spin configurations on Fe center is the intrinsic factor dominating the thermodynamics of ORR. Compared to pristine FeN_4 where the on-site magnetic moment of Fe center is close to 2.0 μ_B, the on-site magnetic moment on FeN_4-3N-1 is significantly low (~0.25 μ_B), suggesting ORR benefits more from low magnetic moment Fe center. The correlation between ΔG_{*OH} and the magnetic moment on Fe center can also be qualitatively analysed from the perspective of spin-orbital interactions between Fe center and *OH. As shown in Figure 4d, the decreased magnetic moment intrinsically indicates a lower spin state. For high magnetic moment Fe center, the orbital interaction with *OH results in a bond order of 1.5; while for the low magnetic moment one, a bond order of 1 is resulted (Figure 4e). As discussed earlier, a lower value of bond order represents a weaker chemical bond; thus, the negative correlation between $\Delta G_{*\mathrm{OH}}$ and on-site magnetic moment can be spintronically explained. It's worth noting that although enhanced reaction thermodynamics are gained, the decreased magnetic moment may impact, to some extent, the local electron transfer at the interface between the Fe site and the adsorbed reactants because of the narrower spin-related channel around Fe site (Figure 3d). Ideally, spin configurations of cations can be engineered to promote both features of electron transfer and reaction thermodynamics; however, in some cases, one of the features shows significantly positive response while the other relatively negative. Therefore, the phenomena resulted from spin manipulation should be case-by-case analysed. As for the above case of FeN_4-3N-1 site, although low magnetic moment Fe center shows slightly narrower spin-related channel than high magnetic moment Fe center, the overall electric conductivity of the 2D and carbon-rich structure is still superior because of the high mobility of electrons inside the material. Thus, the spin-related thermodynamic feature of the Fe site plays much more important role than spin-related electron transfer, and the significant enhancement of the thermodynamic feature eventually makes FeN_4-3N-1 a much better catalytic site than pristine FeN_4.

3. The Future of Spintronic Oxygen Electrocatalysis: Challenges and Opportunities

We have described here how spin-related electron transfer and orbital interactions affect the oxygen electrocatalysis with two examples of benchmark catalysts. With the growing global energy consumption, the demand for a sustainable energy infrastructure will continuously increase, making the usage of efficient oxygen electrocatalysts based on earth-abundant elements a critical need. Thus, the study of oxygen electrocatalysis on the cost-effective 3d transition metal-based catalysts, whose properties are precisely spin-dependent, should be a worthwhile topic in the near future. Fortunately, ever since some pioneering works to address the significant role of spin in oxygen electrocatalysis[1], growing efforts are being spent on the spintronic aspects. We thereby believe a substantial consideration of the spin-related electron transfer and orbital interactions will help establish a more intelligible understanding of oxygen electrocatalysis, and spin electrocatalysis may emerge as one of important future topics. Here, we would like to highlight the challenges and opportunities of spintronic oxygen electrocatalysis, which might provide some inspirations for the development of this field.

I. Spin-polarized DFT calculations with oriented spin ordering. Spin-polarized DFT calculations are able to precisely reflect most of the thermodynamic feature of reactions on magnetic materials, and J. Gracia et al.[17] have demonstrated that catalyst with different spin ordering exhibits different binding affinity to the reactants and intermediates of ORR. Thus, to accurately describe the spin-related catalysis phenomena and provide more reliable predictions for catalyst design, spin-polarized DFT calculations with oriented spin ordering should be employed. Besides, spin-polarized DFT calculations can also simulate and illustrate the spin channel by optimising the spin structure of the electrocatalysts, which can be used to qualitatively evaluate the electron transfer ability. Therefore, a proper utilization of the DFT calculation could not only obtain the thermodynamic feature of the reaction, but also forecast the trend of spin-related electron transfer on different catalysts.

II. The synthesis of magnetic materials with targeted spin ordering. It has been demonstrated theoretically by J. Gracia et al. that materials with dominating FM spin orderings generally exhibit better spin-selected charge transport mobility.[17] The oxygen electrocatalysis could benefit significantly if materials dominated by FM spin orderings can be synthesized through simple and practicable processes. However, to date, one-step synthesis of the commonly employed transition metal oxides (TMOs), such as perovskite and spinel oxides, with dominant FM ordering is technically difficult. Also, to improve the catalytic activity of catalysts, TMOs are intensively synthesized with metal dopants and substitutes; the doping and substitution can create active atomic sites, but may also interrupt the local spin ordering and create localized AFM spin orderings, impacting the charge transport mobility.

III. The usage of applied magnetic field. Many magnetic materials exhibit paramagnetic feature under ambient conditions, where the unpaired spins are randomly ordered in the material. Applying a magnetic field can effectively reorient the spins and create long-range ordered FM spin structure. However, it's worth noting that different materials possess different sensibility to the applied magnetic field and some can only respond to extremely high external field. Therefore, for practical applications, the balance between the applied high magnetic field and the resulted catalytic enhancement should be carefully considered. Besides, in some cases, the spin ordering in materials will revert back to paramagnetic after the removal of outer magnetic field. Thus, to ensure an aligned spin ordering under electrocatalysis conditions, strategies should be designed to maintain the spin ordering in catalysts. A simple approach is to constantly apply the magnetic field along the electrocatalysis process. The challenge of this approach is that the magnetic field will not only reorient the spin ordering of catalysts, but also influence, to some extent, the diffusion of ions in the solutions by Lorenz force[18]. Therefore, the overall catalysis performance with applied magnetic field under the reaction shall be a complicated phenomenon and requires more detailed investigations.

IV. Manipulation of the spin state of metal cations. Spin state refers to the spin configuration of the metal's *d*-orbital electrons, which is associated with the *d*-orbital occupation and therefore determines the orbital interactions between metal cation and reaction intermediates. Depending on the arrangement of *d*-orbital electrons and the ligands coordinated with, the spin state of metal cations may vary from low-spin to intermediate-spin and high-spin state. As demonstrated in above discussions, cations with different spin state show different reaction thermodynamics towards oxygen electrocatalysis. Thus, engineering the spin state of cations is an effective and feasible approach to control the reaction activity. In transition metal oxides, the spin state of a manganese cation in octahedral coordination is usually in high spin state independent of its valence state[19a] due to the smaller crystal field splitting energy than spin-pairing energy in oxides[19b], while most other 3d metal cations in oxides may vary their spin state depending on the valence state, particle size, and even the synthesis conditions. For example, the spin state of cobalt cations on the surface of perovskite $LaCoO_3$ can be varied along with the particle size.[19c] That leads to a change of the e_g electron filling number on Co surface site and thus a change on OER activity.

V. In-situ characterization of the spintronic structure of catalysts. The spin-related catalysis phenomena are sensitive to the spin structure of the catalysts, which makes the in-situ spintronic structure characterization a significantly valuable technology to understand the theory and mechanism of spin-catalysed oxygen electrocatalysis. Unfortunately, this technology is currently inaccessible and future efforts are desired.

VI. Electrocatalysts made by 2D materials. 2D materials exhibit unique magnetic and electronic properties. One interesting assumption that is the electrons inside the layer are more flexible due to the weak van der Wall restriction and thus the rigid crystal limit can be broken to allow the spin manipulation to influence the reaction.

Conflict of Interest

The authors declare no conflict of interest.

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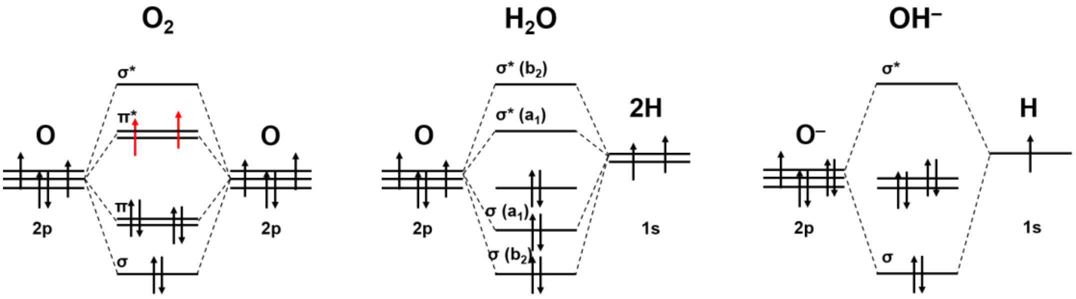

Figure 1. Molecular orbital diagram of O_2, H_2O, and OH^-.

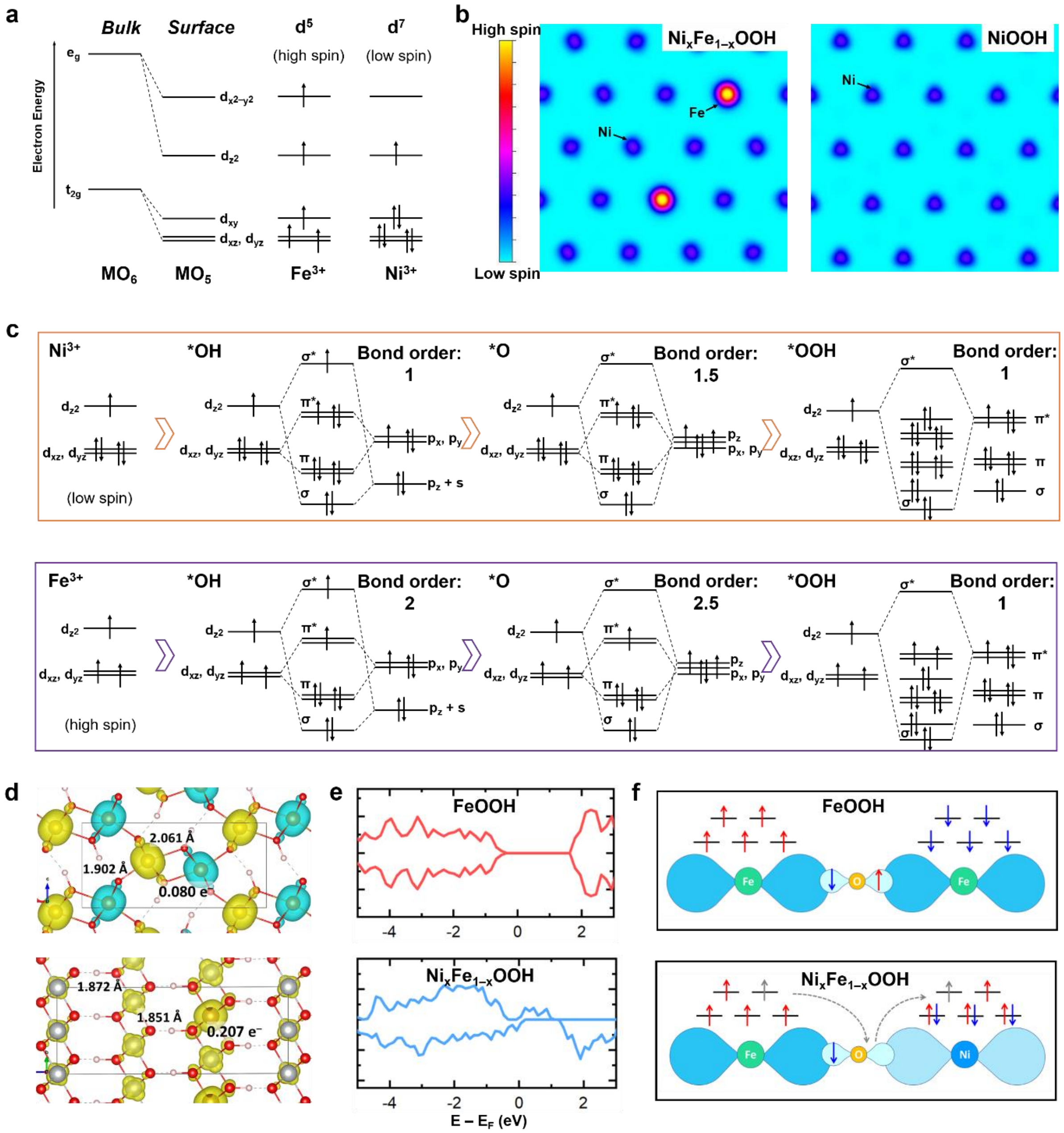

Figure 2. The explanation of OER on $Ni_xFe_{1-x}OOH$ from a spintronic point of view. a) *d*-electron configurations of iron and nickel cations at the surface. b) The spin channel of Fe sites and Ni sites in $Ni_xFe_{1-x}OOH$ and NiOOH. c) The orbital interactions between cations and the OER intermediates. d) The spin density pattern of FeOOH (up) and $Ni_xFe_{1-x}OOH$ (down). The $Ni_xFe_{1-x}OOH$ model is constructed with x equalling to 0.83. The isosurfaces in yellow and blue represent spin-up and spin-down densities, respectively. e) The density of states (DOS) pattern of FeOOH and $Ni_xFe_{1-x}OOH$. f) The spin transfer route in FeOOH (showing the Fe–O–Fe bonds) and $Ni_xFe_{1-x}OOH$ (showing the Fe–O–Ni bondsolecular orbital diagram of O_2, H_2O, and OH^-.

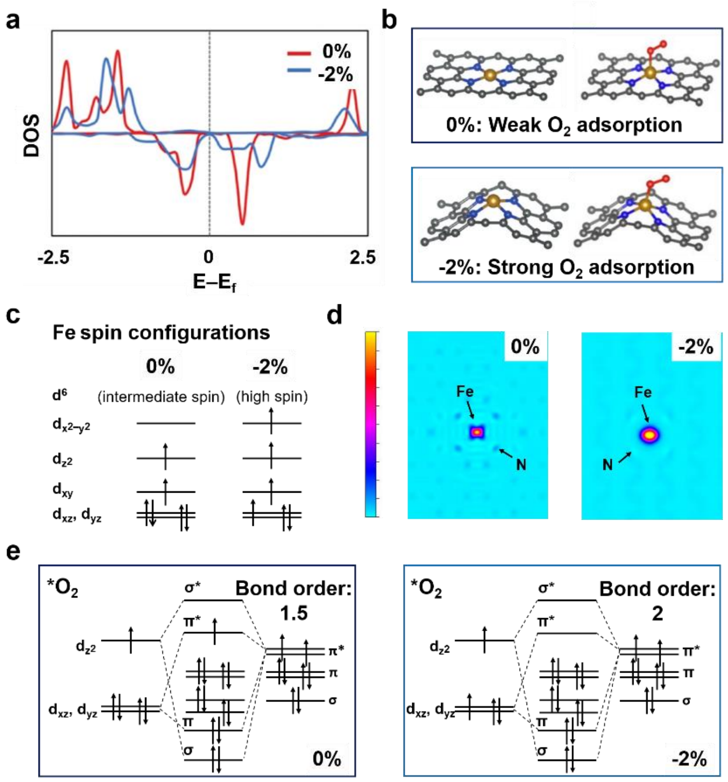

Figure 3. Spin-related charge transfer and orbital interactions of ORR on FeN_4 with and without 2% Fe–N bond contraction. a) Density of states (DOS) of the 3*d* orbitals of the central Fe cation in the two FeN_4 sites. 0% and -2% denotes pure FeN_4 site and FeN_4 site with 2% Fe–N bond contraction, respectively. b) Adsorption configuration of O_2 adsorption on two FeN_4 sites. c) Possible Fe spin configurations in FeN_4. d) The spin channel of Fe site in FeN_4. e) The orbital interactions between Fe sites and the triplet O_2 molecule. a and b are reproduced with permission from ref. 15b © 2019 Wiley‑VCH Verlag GmbH & Co. KGaA, Weinheim.

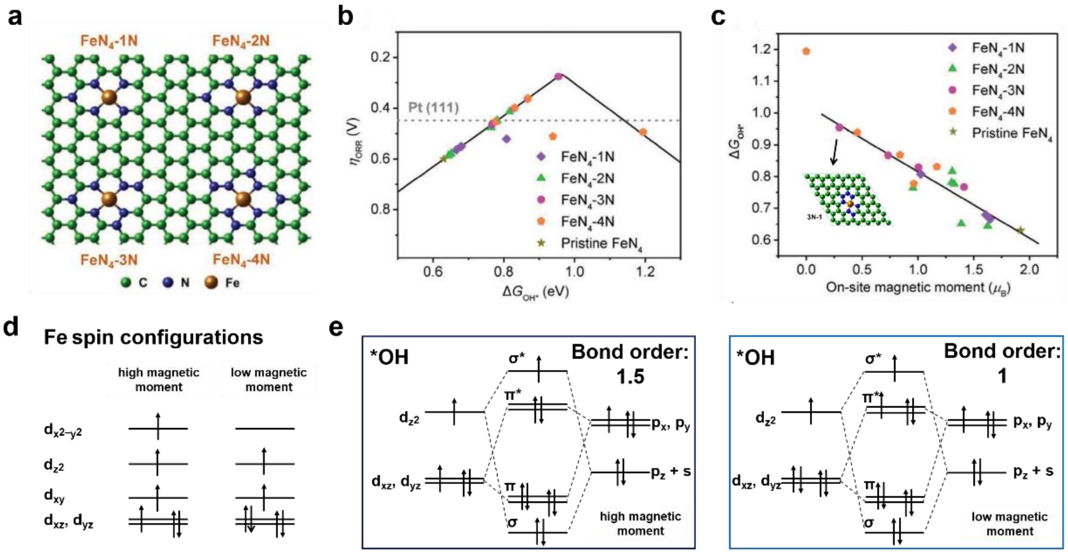

Figure 4. The spin-related orbital interactions on pristine FeN_4 and spin-engineered FeN_4. a) Representative configurations of FeN_4 with different amount of surrounding graphitic N atoms. b) Overpotential (η_{ORR}) as a function of *OH adsorption free energy (ΔG_{*OH}). c) Linear relationship between ΔG_{*OH} and on-site magnetic moments (namely, the spin) of the Fe center in FeN_4 sites. d) The possible spin state of the Fe center with high and low magnetic moment. e) The orbital interactions between Fe center and *OH intermediate. a, b and c are reproduced with permission from ref. 16 @ 2019 Wiley - VCH Verlag GmbH & Co. KGaA, Weinheim.

This article provides a brief discussion on spin-related charge transfer and orbital interactions in oxygen electrocatalysis. It is expected that a more comprehensive understanding on oxygen electrocatalysis probably can be established in the near future by including the role of electron spin.

Keyword: oxygen electrocatalysis; spin; orbital; water oxidation; oxygen reduction

Yuanmiao Sun, Shengnan Sun, Shibo Xi*, Jose Gracia*, Zhichuan J. Xu*

Spin-related Electron Transfer and Orbital Interactions in Oxygen Electrocatalysis

TOC figure: