Understanding field driven electrochemistry at the example of CO_2 reduction

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At the example of CO_2 reduction, we present new experimental and theoretical data that highlights the significance of the electric double layer field in electrochemistry. For this purpose, we developed a multi-scale modeling approach combining field-dependent Density Functional Theory with continuum electrolyte simulations. We find the model to provide an unprecedentedly quantitative explanation of cation effects on CO production on Au and Ag and C_2 product formation at Cu. The cation sensitivity of these product pathways is explained by their high sensitivity to the interfacial field and the field variation with cation size. By combining the model with mass transport effects, we show that the field not only controls the kinetics, but also the migration of charged species and local pH. From these insights, we develop new promising handles to boost field-sensitive reactions, as the potential of zero charge (PZC) and the interfacial capacitance. Finally, we present the impact of field effects on other energy conversion processes and discuss possible refinements of the theory via explicit electrolyte simulation approaches.