Low-Temperature Scanning Probe Techniques for Studying Spins on Surfaces

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There is increasing interest in quantum science, which promises advances in sensing and computation technology. At the core of quantum science are quantum two-level systems that can act as quantum bits (qubits). For quantum sensors, "sharp" energy transitions that are sensitive to electrostatic and magnetic fields from the environment are desirable. To execute quantum algorithms, long phase coherence times are key, as is the ability to couple multiple qubits. Electron and nuclear spins are ideally suited for this purpose due to their tunable transitions via magnetic fields and well-developed all-electrical control and readout techniques.

A major issue with established nanoscale qubits is their lack of reproducibility and the challenges in coupling them. In this respect, spins in atoms and molecules on a surface are well-defined by their electronic structure. When combined with low-temperature scanning probe microscopy (SPM), it becomes possible to manipulate atomic positions—bringing them close together—and control or read their spin states. Unfortunately, most quantum science efforts have so far been restricted to thin insulating films on metal surfaces, and—until recently—there have been few strategies for studying spins on metals and thick insulators.

In this talk, I will introduce atomic-scale quantum sensing (ASQS) and single-atom magnetic exchange force (MxF) microscopy. In ASQS, a single molecule with an unpaired electron, attached to the metallic tip of an SPM, acts as a sensor. We follow the electron spin resonance of the molecule while approaching single atoms on a metal surface. From changes in the resonance frequency, we can deduce dipolar magnetic and electrostatic coupling between the adatom and molecule with ~100 neV resolution. To detect spins on insulators, a different approach is required, making atomic force microscopy the method of choice. I will show how the MxF between a tip and a single atom on the surface can be determined, as well as how the sign and strength of the interactions can be manipulated. Interestingly, we can track the MxF into a chemical bond formation at the condensed-phase interface and gain insights into the fundamental nature of spin-spin interactions in adsorption.

So far, advanced low-temperature SPM methods have only occasionally been applied to physical chemistry at surfaces and heterogeneous catalysis. I look forward to discussing possible applications and the combination of methods needed to make surface chemistry a more exact science.