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Tracing coherent dynamics of electron and nuclear spins using pump-probe spectroscopy

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Abstract

The implementation of electron spin resonance (ESR) at the probe tip of a scanning tunneling microscope (STM) has dramatically enhanced our ability to investigate and control individual atomic spins on a surface. A particularly interesting avenue is the possibility to observe the free coherent dynamics of a spin that is brought into a non-eigenstate, which was demonstrated for a single spin using pulsed ESR experiments [1]. For coupled spins, however, the maximum attainable Rabi rate can typically barely compete with the coupling dynamics, complicating state initialization. As an alternative approach, we employ pump-probe spectroscopy using DC pulses to initialize and read out an electron spin located underneath the STM tip, that is coupled to one or multiple spins not directly probed by the tip. In this approach, ESR-STM is used as a method to identify avoided crossings between energy levels and tune the system to those crossings. The technique, initially demonstrated on a pair of coupled titanium atoms [2], can be extended to chains of atoms, providing access to the dynamics of magnons traversing the chain. Most recently, we further refined the technique to include dynamics between an electron spin and the nuclear spin on the same atom [3], yielding a complex evolution pattern resulting from the combination of coherent oscillations between multiple hybridized quantum states.

[1] Yang et al., Science 366, 509 (2019)
[2] Veldman et al., Science 372, 964 (2021)
[3] Veldman et al., arXiv:2309.03749 (2023)