ESR-STM of single FePc molecules at strong spin-photon coupling

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The integration of electron spin resonance with scanning tunneling microscope (ESR-STM) has enabled the detection and coherent control of single surface-bound spins with high energy and spatial resolution (1, 2), establishing them as locally addressable quantum states. In this setup, the spin-polarized STM tip plays a multifaceted role: it probes the quantum state, renormalizes the Zeeman splitting, and serves as the driving source for the localized spin (3). Here, we demonstrate the realization of a strong driving regime in an ESR-STM experiment on Fe-phthalocyanine (FePc) molecules deposited on Ag(100)/MgO(100) (2, 4, 5), mediated by the electric field modulation of the tip-molecule exchange coupling. Unlike macroscopic cavities that require large spin ensembles (N) to leverage a $\sqrt{N}$ - enhanced collective magnetic dipolar coupling (6, 7), the ESR-STM platform achieves a strong coupling regime at the single-spin limit via highly localized exchange interactions. At large RF excitation, the single ESR resonance peak evolves into a series of uniformly spaced peaks as the magnetic field is swept, which we identify as multi-photon transitions. Under the same conditions, we also observe the splitting of the ESR signal into what resembles an Autler-Townes doublet during pulsed frequency sweeps. Both phenomena are a direct consequence of the strong microwave photon-spin coupling, offering a promising pathway for the formation of spin-photon dressed states (8).

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