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Single-electron-spin resonance by microwave photon counting

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Electron spin resonance (ESR) spectroscopy is the method of choice for characterizing paramagnetic impurities in a sample, with applications ranging from chemistry to quantum computing but it gives access only to ensemble-averaged quantities due to its limited signal-to-noise ratio. Here, we report a new method for the detection of single electron spins at millikelvin temperatures. Analogous to the optical fluorescence detection of atoms or molecules, it consists in detecting the microwave photons spontaneously emitted by an electron spin when it relaxes radiatively to its equilibrium ground state after being excited by a pulse [1]. To enhance the radiative relaxation rate [2], the spins are inductively coupled to a small-mode-volume, high-quality-factor, superconducting resonator patterned on top of the sample. The microwave fluorescence photons are then routed towards a single-microwave-photon counter based on a superconducting qubit [3].

The method applies to all paramagnetic species with sufficiently low non-radiative decay rate; here, we demonstrate it on rare-earth-ion spins (Er3+) doped in a scheelite CaWO4 host matrix. We resolve individual narrow peaks in the fluorescence signal, on which we observe microwave photon anti-bunching, proving that they originate from single spins [4]. We reach a signal-to-noise ratio of 1 in 1 second integration time for each Er3+ ion located in a detection volume of ~20µm3. We observe spin coherence times above a millisecond, limited by the radiative lifetime. I will discuss the new perspectives opened by these results for practical single electron spin resonance spectroscopy and spin-based quantum computing.

[1] E. Albertinale et al., Nature 600, 434 (2021)

[2] A. Bienfait et al., Nature 531, 74 (2016)

[3] R. Lescanne et al., Phys. Rev. X 10, 021038 (2020)

[4] Z. Wang et al., arXiv:2301.02653

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