S1: **Emmanuel Flurin** (CEA Saclay)

"Single Atom Magnetic resonance by Microwave Photon Counting"

Electron spin resonance (ESR) spectroscopy is a widely used technique for characterizing paramagnetic impurities, with applications ranging from chemistry to quantum computing. However, it is typically limited to ensemble-averaged measurements due to its restricted signal-to-noise ratio. Sensitivity sufficient to detect single electron spins has been achieved through methods such as spin-dependent photoluminescence, transport measurements, and scanning probes. Unfortunately, these techniques are often system-specific or sensitive to a small detection volume, leaving practical single-spin detection an ongoing challenge. Here, we demonstrate single-electron magnetic resonance via spin fluorescence detection [1,2], utilizing a microwave photon counter based on a superconducting transmon qubit operating at millikelvin temperatures [4]. In our experiment, individual paramagnetic erbium ions in a scheelite $CaWO_4$ crystal are manipulated and read out, enabled by magnetic coupling with a small-mode-volume, high-quality-factor superconducting microwave resonator. Leveraging this capability, we perform nuclear magnetic resonance of the nearby ^{183}W nucleus [3], achieving single-shot nuclear spin readout and demonstrating second-scale coherence times for individual atoms. This quantum control over individual high-coherence nuclei opens new avenues for quantum computing. Our method, applicable to arbitrary paramagnetic species with sufficiently long non-radiative relaxation times, enables large detection volumes ($\sim 10\ \mu m^3$), paving the way for ESR at the single-molecule level with unprecedented sensitivity and spectral resolution.

[1] Albertinale, E. et al. Nature 600, 434– 438 (2021).

[2] Z. Wang, et al. Nature 619, 276–281 (2023).

[3] J. Travesedo et al. arXiv:2408.14282.

[4] L. Balembois, et al. PRApplied (2024).