Pulsed electron spin resonance on picoliter radicals samples

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Abstract

Electron paramagnetic resonance (EPR) is a powerful technique for studying unpaired electrons in materials. It is widely used in chemistry, biology, and physics, and its importance continues to grow as researchers develop more advanced EPR methods. Implementing EPR in superconducting quantum circuits has dramatically improved sensitivity, reaching the level where a single electron spin can be detected. This exceptional sensitivity is achieved thanks to resonators with very small mode volumes and narrow linewidths, operation at millikelvin temperatures, and the use of quantum-limited amplifiers to boost the microwave signals emitted by spins.
Although high-quality superconducting resonators enable extremely sensitive spin detection, their long ring-down times make them unsuitable for studying spins that dephase quickly. To address this limitation, we developed a resonator that can be tuned dynamically using the kinetic inductance of niobium–titanium–nitride (NbTiN), a superconducting alloy that remains stable in magnetic fields. By applying fast DC current pulses, we were able to adjust the resonator linewidth on microsecond timescales while maintaining state-of-the-art performance for electron spin resonance (ESR) detection.

The resonator is compatible with a wide range of paramagnetic samples. We tested it by performing time-resolved spectroscopy on a 20-picoliter sample of α,γ-bisdiphenylene-β-phenylallyl at millikelvin temperatures. We measured both the longitudinal and transverse relaxation times of this molecule, achieving a sensitivity of about six hundred thousand spins per square-root hertz. This is only two orders of magnitude below the best experiments performed on highly coherent spin ensembles with similar coupling strengths.
These results demonstrate a flexible ESR platform operating at millikelvin temperatures. It offers promising opportunities for fast control of quantum spins and hybrid systems, for materials characterization, and for detecting unpaired electrons in individual biological cells

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