

Towards in-situ nanoscale spatially resolved electron spin resonance in scanning electron microscopes

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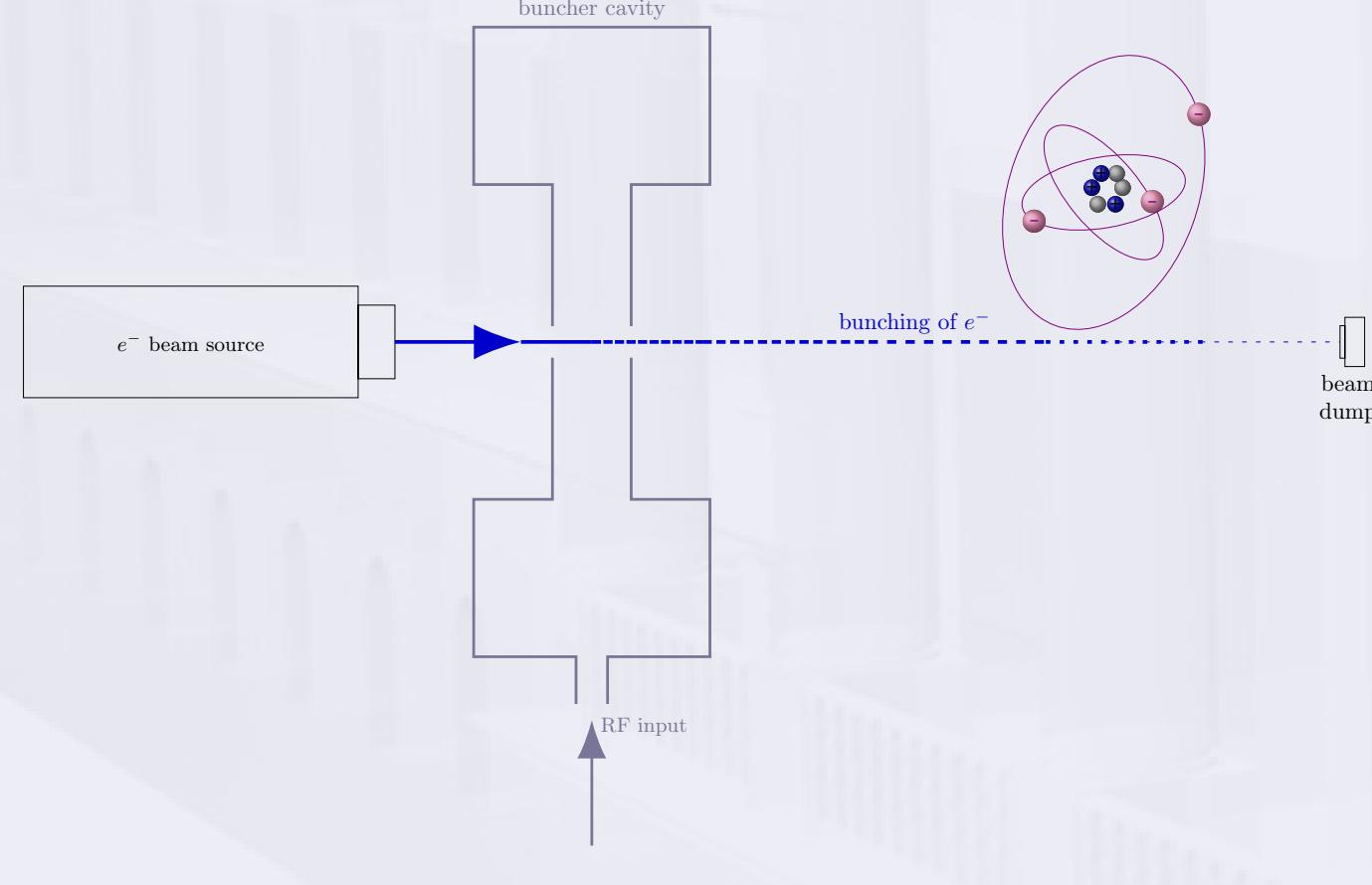
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Abstract

Electron spin resonance is a widely used analytical tool in medicine, biology and material sciences. Traditionally, electron spin systems are driven by microwaves, which offers only limited spatial resolution due to the long wavelength of microwaves, which can be optimized by sophisticated techniques such as the use of magnetic field gradients. We propose a different way of driving spin systems using the non-radiative near-field of a modulated electron beam in a scanning electron microscope by tuning the microscopes to higher beam currents and using additional beam modulation mechanisms. Driving systems with higher harmonics of the modulated beam opens up future possibilities to perform in-situ electron spin resonance analysis with high spatial resolution down to the nanoscale. To perform our experiments we modified an Philips XL30 ESEM to allow for high frequency beam modulation, cryogenic cooling of our samples and microwave frequency readout of our spin systems.

Electron Spin Resonance with Modulated Electron Beams

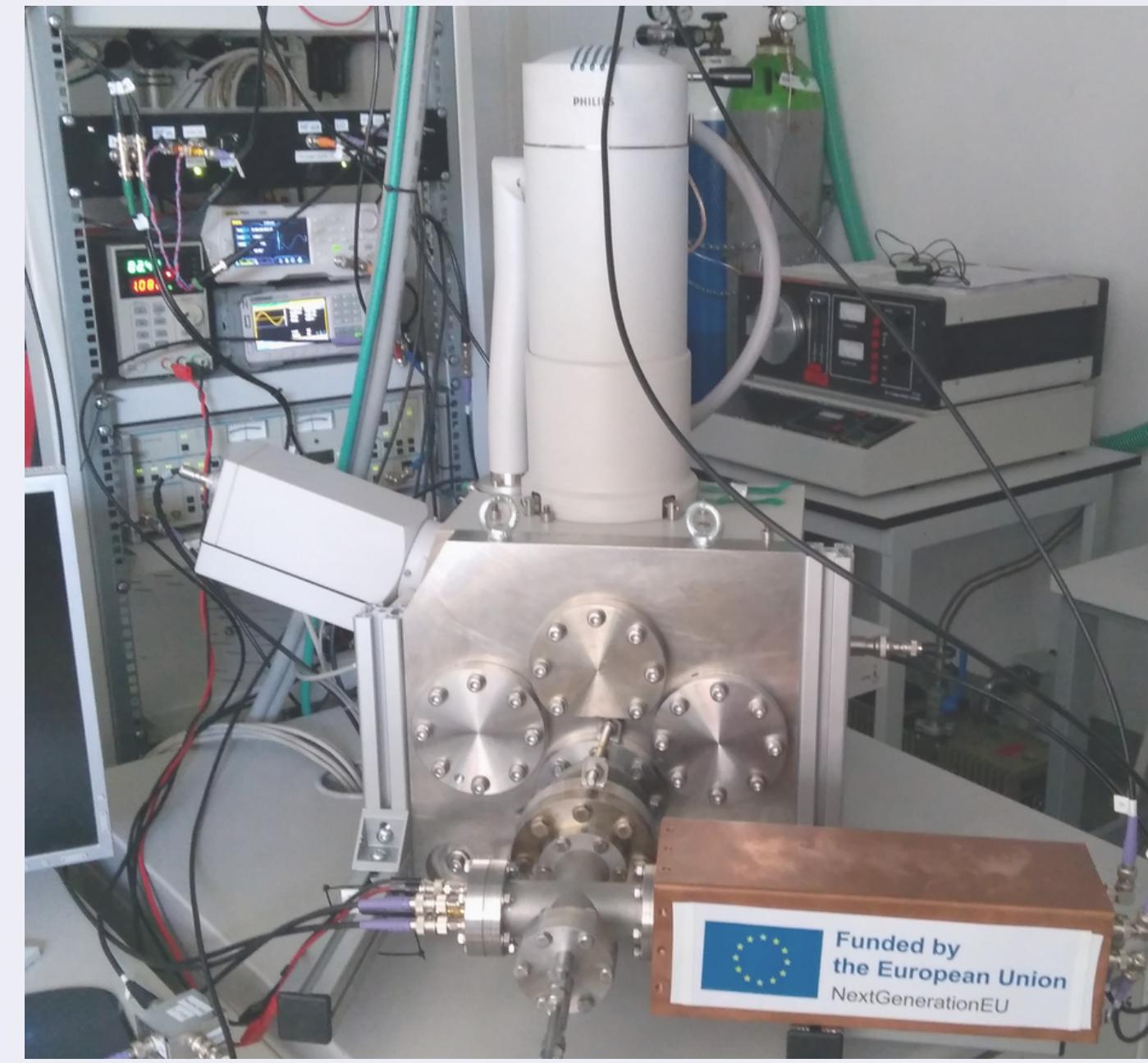
Our experiment resembles an electron spin resonance experiment. In contrast to other electron spin resonance setups where systems are excited using microwaves we use the *non-radiative electro-magnetic near-field* of an modulated electron beam



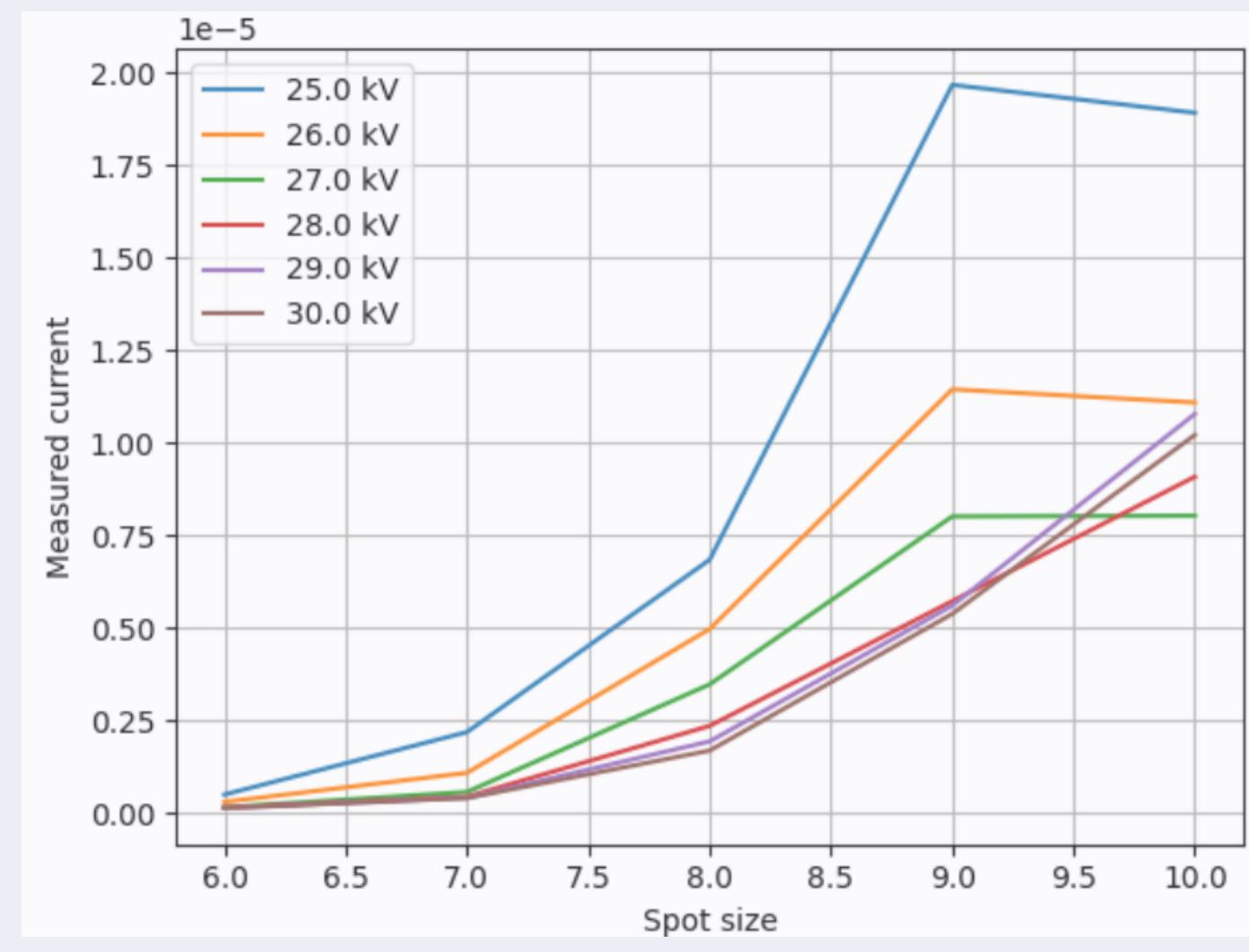
- Classical EPR requires large field gradients for spatial resolution
- Currently resolutions down to $10\mu\text{m}$ are possible in classical systems
- Proposed experiment [1]:
 - Coherent control of spin systems
 - Fundamental near-field scales $\frac{1}{r}$
 - Path to nanoscale: 1st harmonic scales $\frac{1}{r^2}$, 2nd harmonic scales $\frac{1}{r^3}$
 - Proposed addressing systems at 30nm spacing

Customized Philips XL30 ESEM

We customized an scanning electron microscope from the Institute of Material Science and Technology at TU Vienna (disassembled, transported and reassembled) as our experimental platform to fit our special requirements to perform EPR experiments.

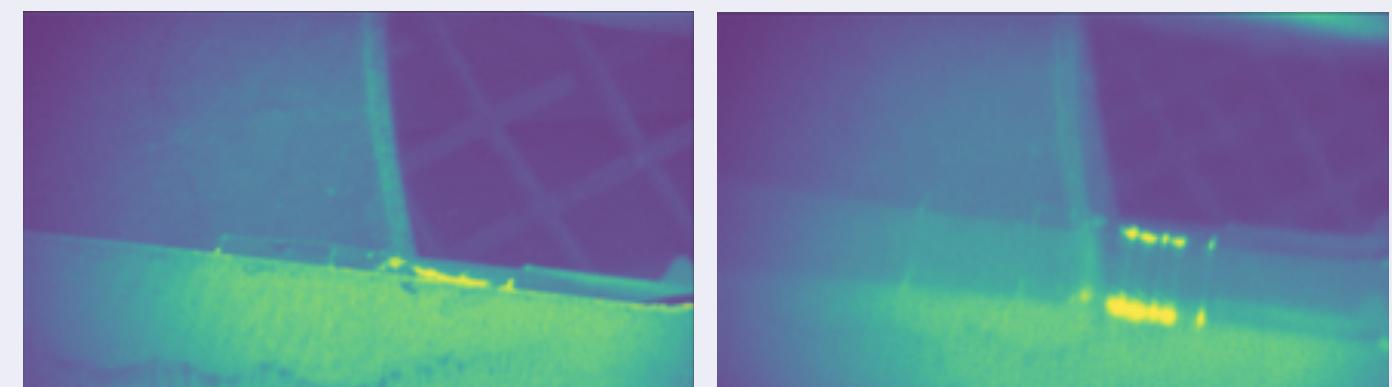


- Philips XL30 ESEM from 1993
- Up to 30 kV
- Magnification up to 500.00x, due to column tuning for high current now \approx 100-500x
- Tungsten filament \rightarrow high current capability
- Using $800\mu\text{m}$ final aperture
- Beam current: $2 - 20\mu\text{A}$ stable
- We are able to modify the machine



Custom PCB based beam modulator

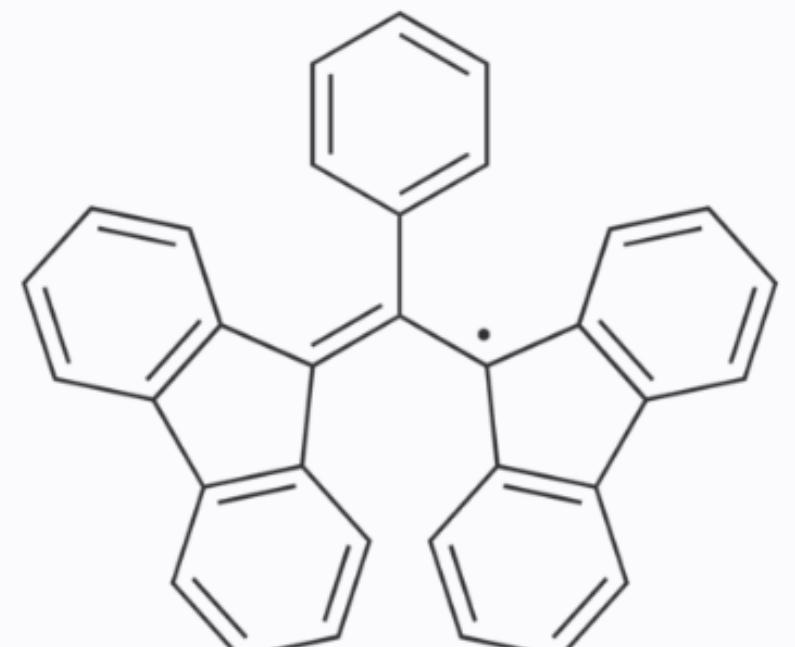
- Cannot use column deflection coils (kHz range)
- Require up to 500 MHz modulation frequency
- Using deflection as in our other experiments
- Other option would be bunching
- Deflector based on modular PCB stack
- Attaches to column attachment system



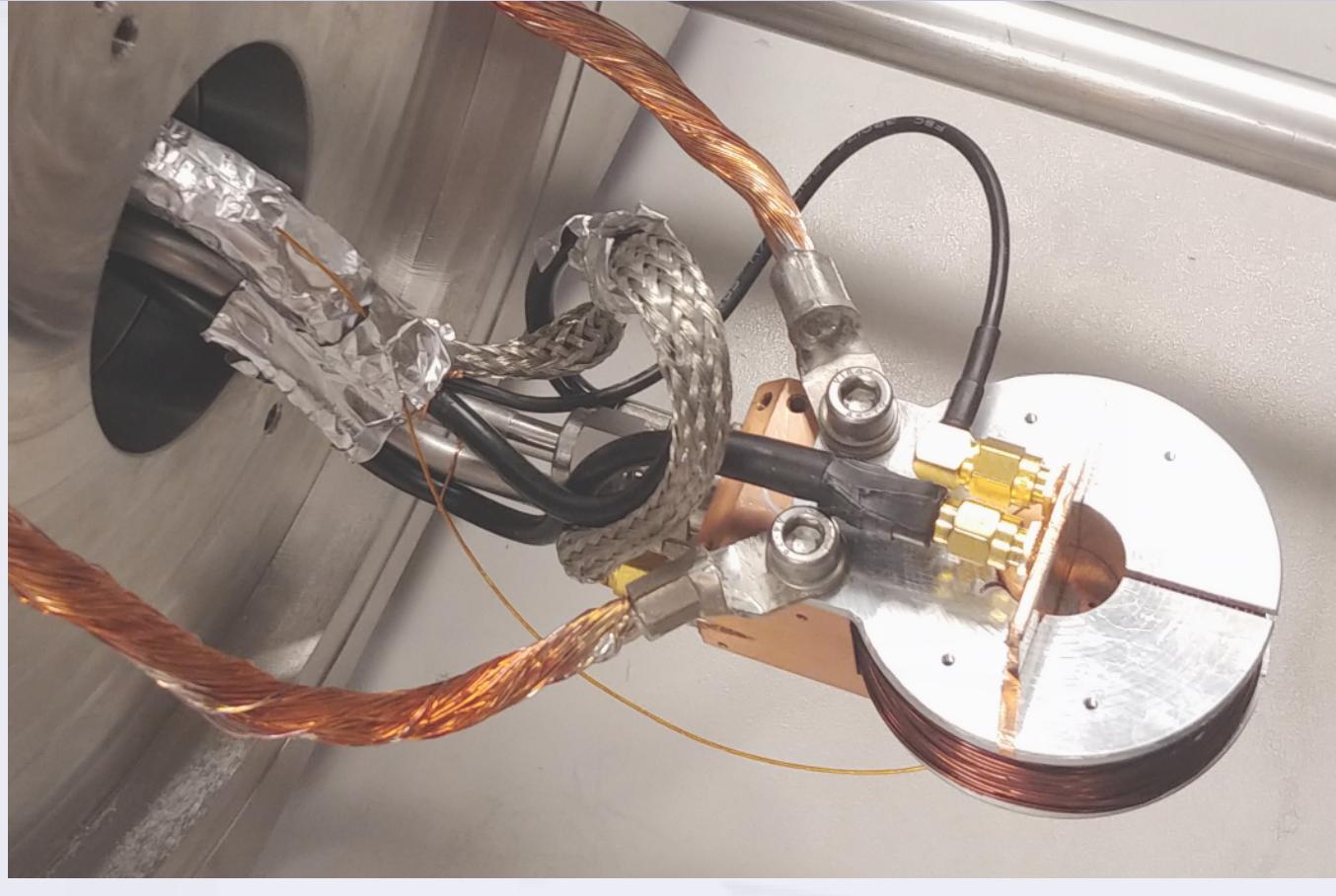
- Modulating with up to 32 dBm
- Modulator performs as modeled
- Measured by spread of visible features via imaging

Material for proof of principle experiment

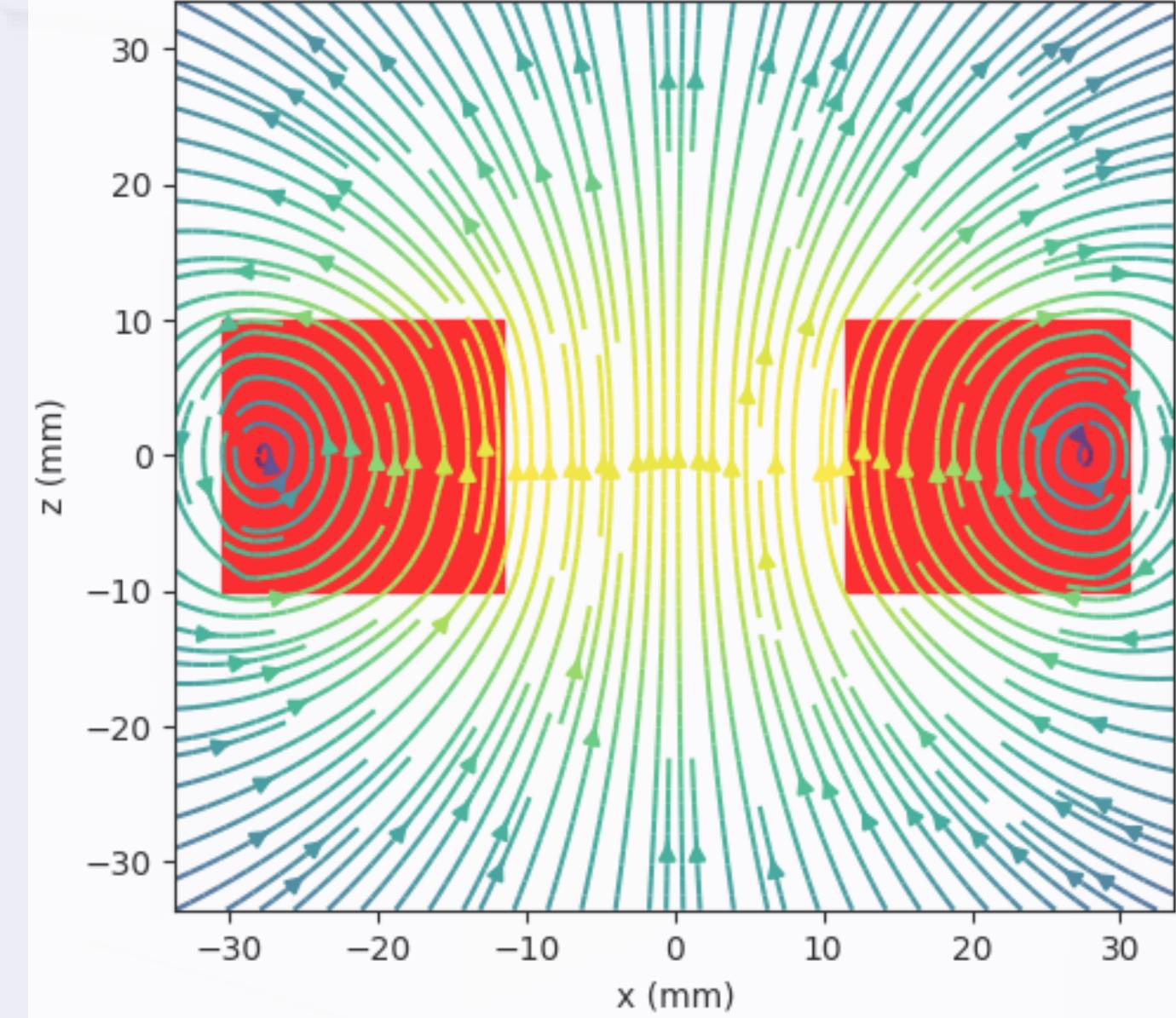
- Koelsch radical (α, γ -Bisdiphenylene- β -phenylallyl - BDPA; $C_{33}H_{21}$)
- high spin density (1 free spin per 51 atoms)



Our Cryostage



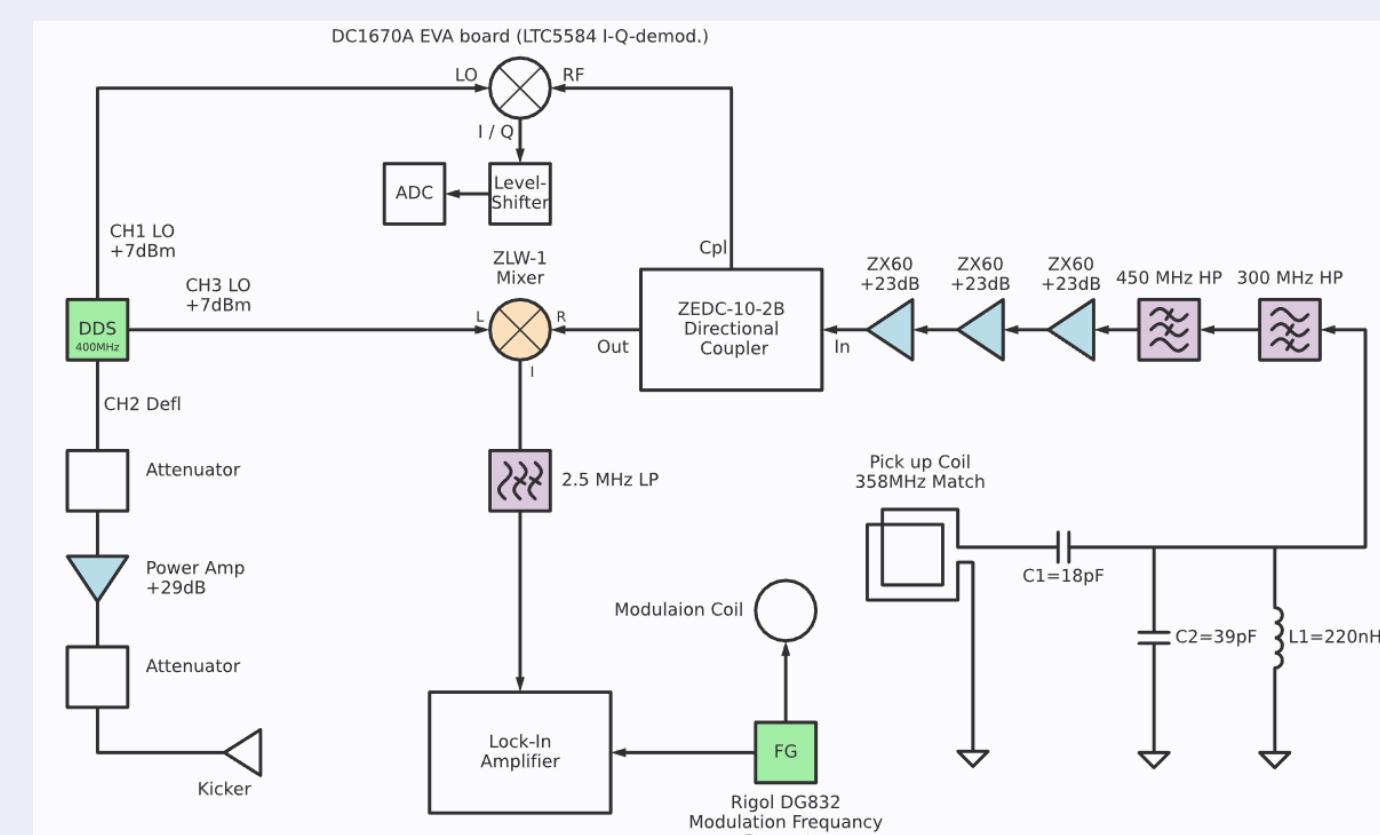
- Lower than eucentric \rightarrow larger beam deflection with lower parasitic radiated field
- Allows for cooling with water or liquid nitrogen (signal gain ≈ 4 , SNR gain ≈ 7.7)
- Sample thermally coupled to stage
- Stage thermally anchored to front door
- Alignment manually via port aligner



Microcoil and RF setup

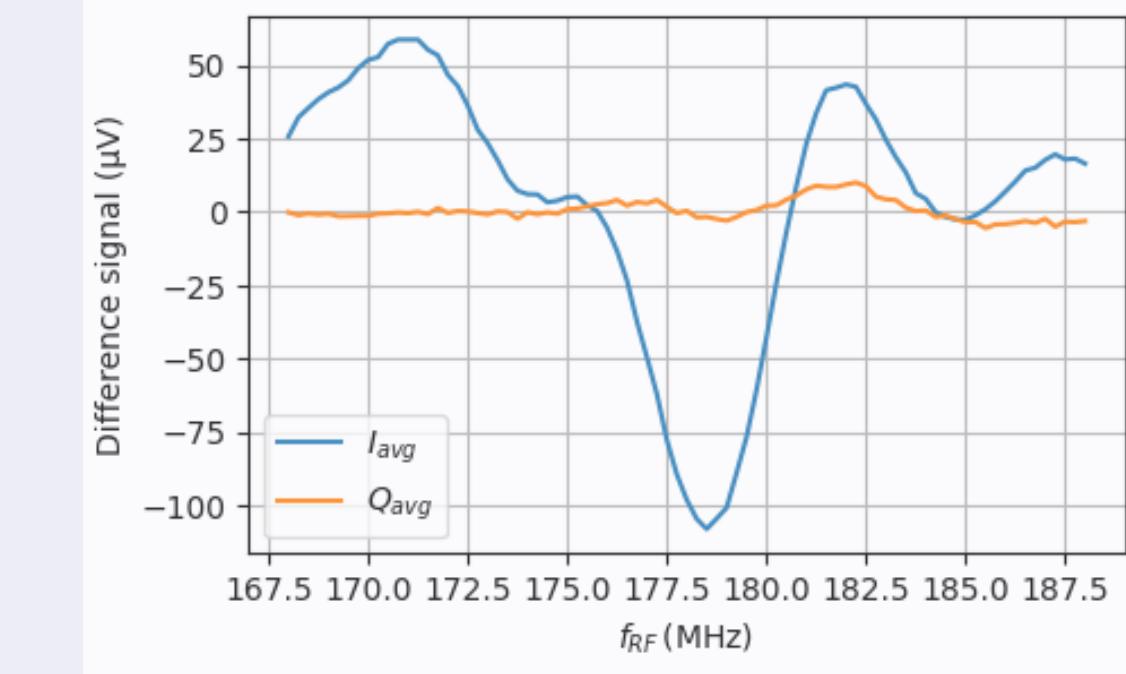
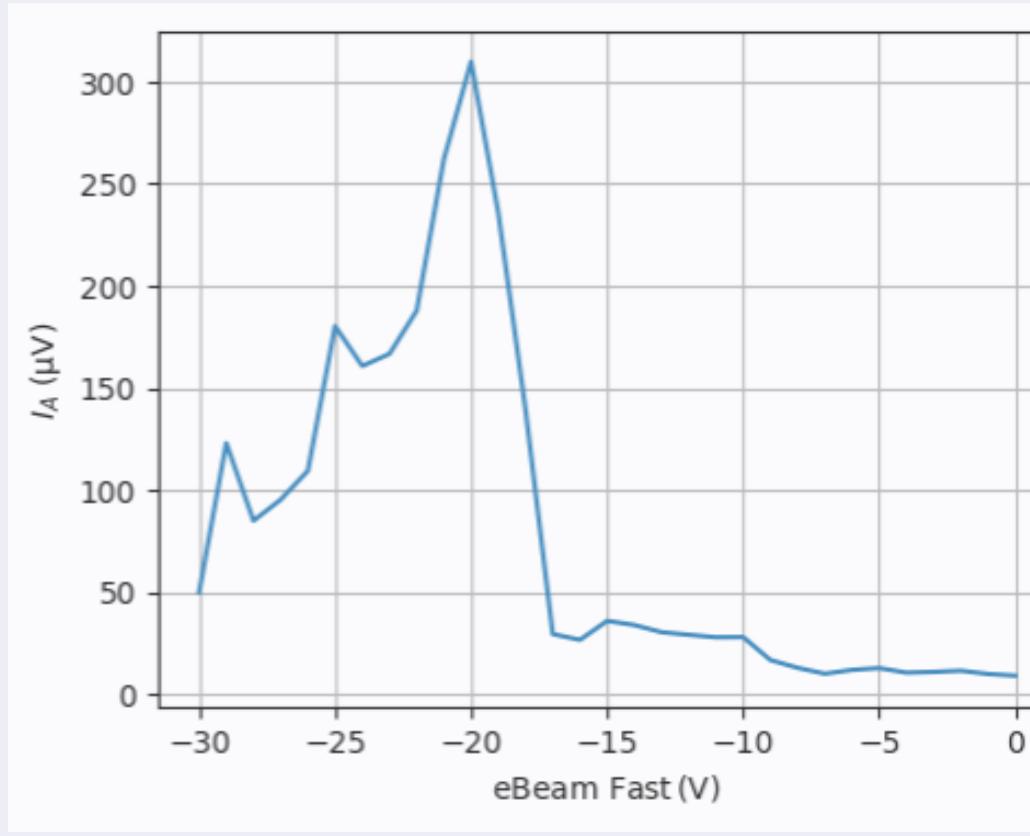


- PCB microcoil ($1.7 * 0.5\text{mm}$)
- Resides in center of B_0 coil (minimal field inhomogeneity)
- Impedance match near coil
- Beam passes aloof
- Faraday cup below the microcoil to measure beam current



First test measurements

- Typical measurement sequence is differential
- Measuring once with electron beam and once without
- Keeping magnetic fields and all other settings constant
- Measuring with beam in spot mode



- We already see position dependent signals induced by the electron beam
- We have to:
 - Verify these are EPR signals
 - Verify these are induced by near field

References & Acknowledgments

- [1] D. Rätzel, D. Hartley, O. Schwartz, P. Haslinger, A Quantum Klystron - Controlling Quantum Systems with Modulated Electron Beams. *Phys. Rev. Research* 3, 023247 (2021)

