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

T. Spielauer[1], M. Kolb[1], T. Weigner[1], A. Prüller[1], J. Toyfl[1], G. Boero[2], P. Haslinger[1]

[1]VCQ, Technische Universität Wien, Atominstitut, Stadionallee 2, 1020 Vienna, Austria; [2]EPFL, BM 3110 Station 17, CH-1015 Lausanne, Switzerland

TECHNISCHE UNIVERSITÄT WIEN Vienna | Austria

ATOMINSTITUT

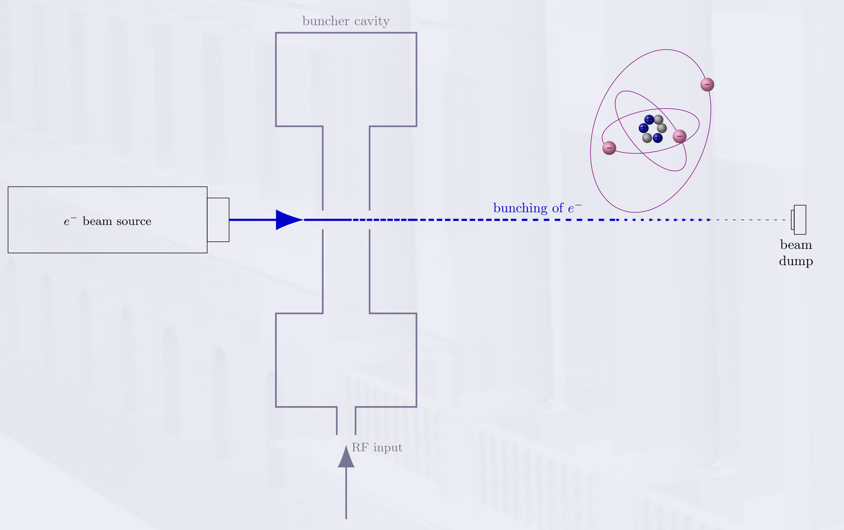
VCQ Vienna Center for Quantum Science and Technology

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 $10\mu m$ are possible in classical systems

Proposed experiment [1]:

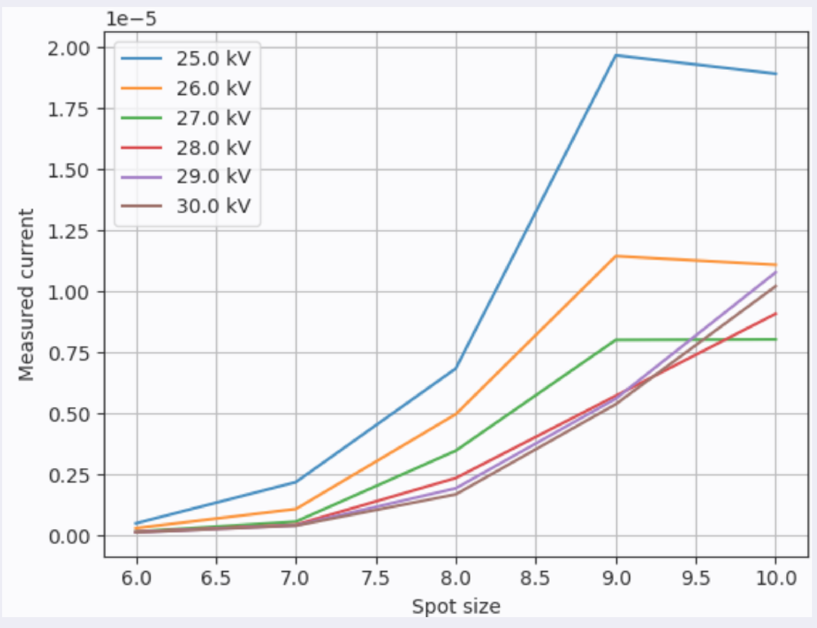
- Coherent control of spin systems
- Fundamental near-field scales $\frac{1}{r}$
- Path to nanoscale: 1^{st} harmonic scales $\frac{1}{r^2}$, 2^{nd} harmonic scales $\frac{1}{r^3}$
- Proposed addressing systems at 30nm spacing

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.

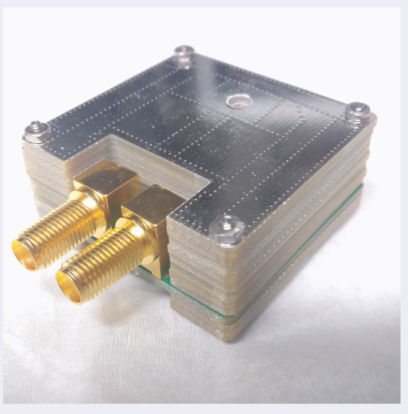
- Philips XL30 ESEM from 1993
- Up to 30 kV
- Magnification up to 500.00x, due to column tuning for high current now ≈ 100-500x
- Tungsten filament → high current capability
- Using $800\mu m$ final aperture
- Beam current: $2 - 20\mu A$ stable
- We are able to modify the machine

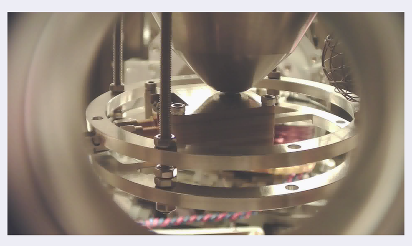
- Custom flange adapters to ConFlat system
- Front door with CF64 flanges to gain more space
- Custom column attachment system
- Python based remote control via `pyxl30`

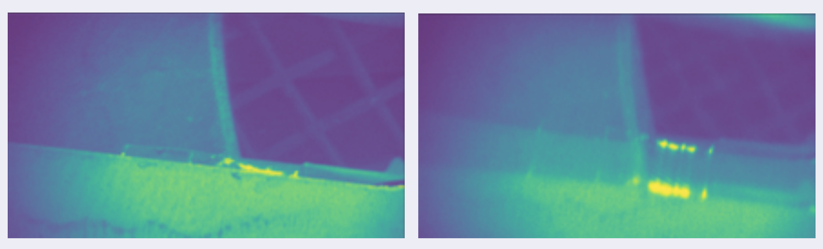


Custom PCB based beam modulator

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



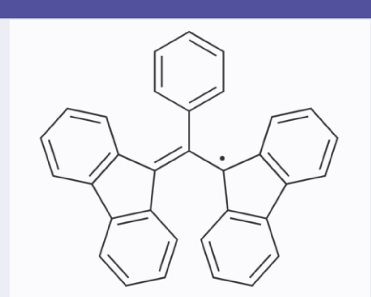




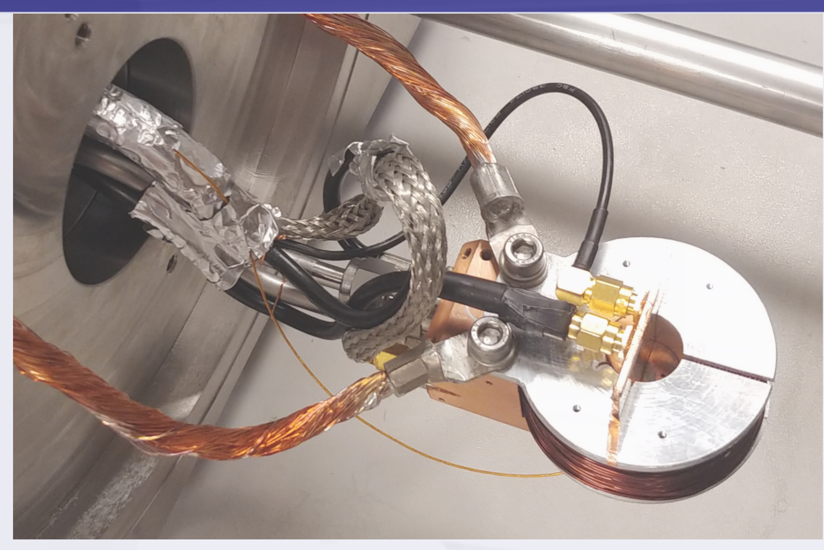
- Modulating with up to 32 dBm (2W)
- Modulator performs as modeled
- Measured by spread of visible features via imaging

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)

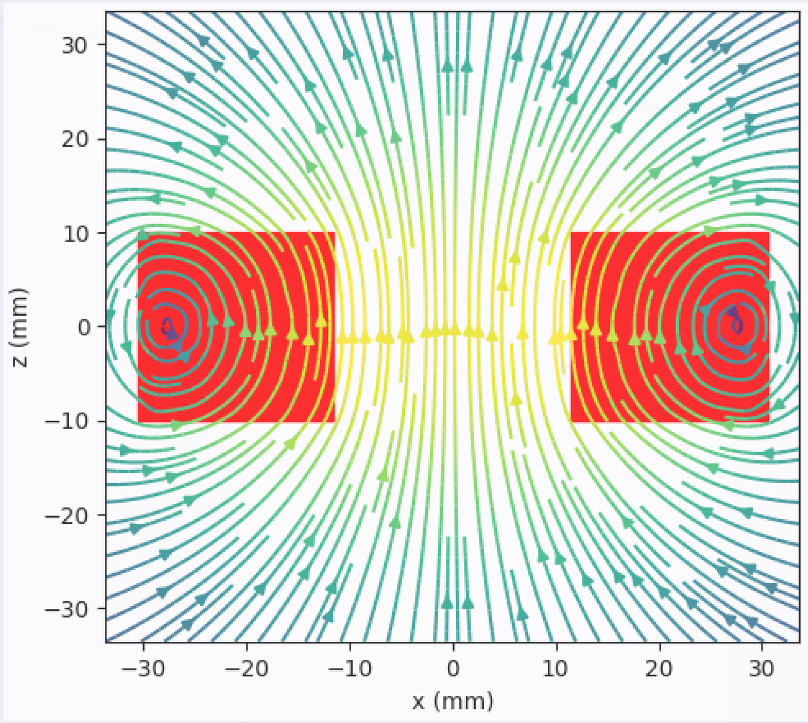


Our Cryostage

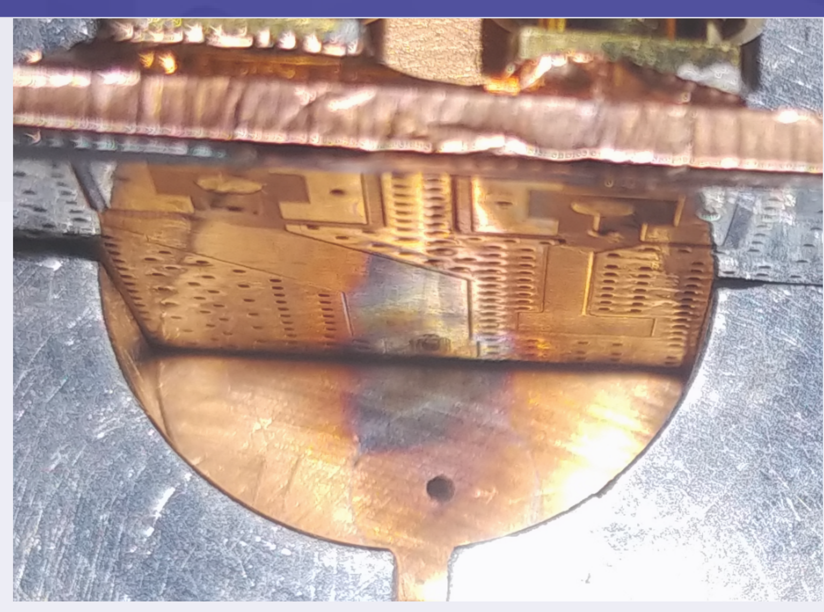


- Lower than eucentric → larger beam deflection with lower parasitic radiated field
- Allows for cooling with water or liquid nitrogen (signal gain ≈ 4, SNR gain ≈ 7.7)
- Sample thermally coupled to stage
- Stage thermally anchored to front door
- Alignment manually via port aligner

- Two concentric coils
- Main B_0 coil (up to $15mT$)
- Modulation coil ($\approx 200\mu T$)
- Largest possible field with working imaging $\approx 30mT$
- Doing *frequency* scans instead of B_0 scans to keep beam position stable
- Coils cooled via thermal coupling to stage

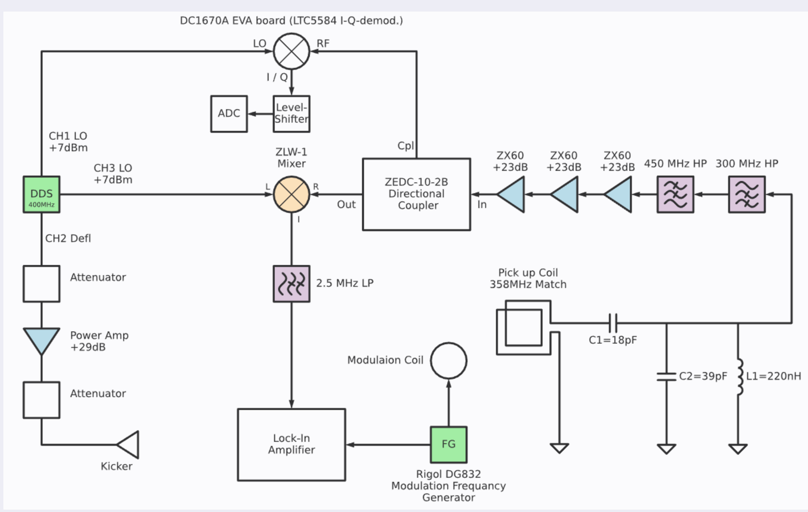


Microcoil and RF setup



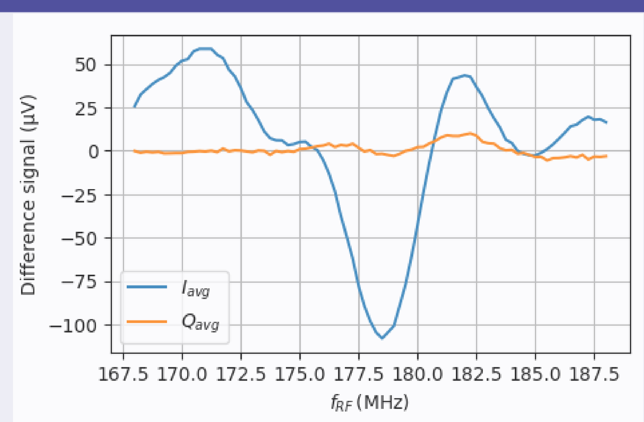
- PCB microcoil ($1.7 * 0.5mm$)
- Resides in center of B_0 coil (minimal field inhomogenity)
- Impedance match near coil
- Beam passes aloof
- Faraday cup below the microcoil to measure beam current

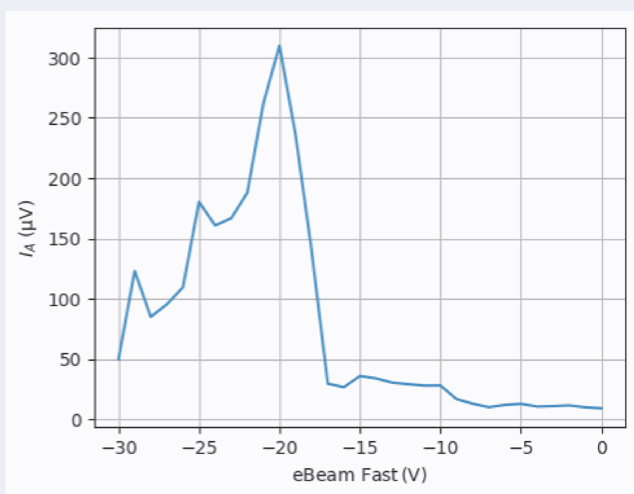
- AD9959 based 4 channel DDS
- Beam modulation via 2W RF amplifier
- Measuring via LTC5584 I/Q demodulator and Lock-In amplifier
- Fast system control via RP2040 controller
- Python based experimental control system



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)*

FWF Österreichischer Wissenschaftsfonds
FFG Austrian Research Promotion Agency
ÖAW ÖSTERREICHISCHE AKADEMIE DER WISSENSCHAFTEN