Combining Electron Spin Resonance Spectroscopy with Scanning Tunneling Microscopy at High Magnetic Fields

Robert Drost,¹ Maximilian Uhl,¹ Piotr Kot,¹ Janis Siebrecht,¹ Alexander Schmid,² Jonas Merkt,² Stefan Wünsch,³ Michael Siegel,² Oliver Kieler,² Reinhold Kleiner,¹ and Christian R. Ast¹,*

¹Max-Planck-Institute for Solid State Research, Heisenbergstr. 1, 70569 Stuttgart, Germany
²Institut für Mikro- und Nanoelktronische Systeme, Karlsruhe Institute of Technology, Hertzstr. 16, 76187 Karlsruhe, Germany
³Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany

Combining electron spin resonance with STM (ESR-STM) has introduced new possibilities to the local studies of individual spins and has expanded the available parameters space substantially, but it imposes a series of strict experimental requirements, most notably on the base temperature of the cryostat. The operational frequency range of ESR-STM has thus far been prevented by challenges in signal delivery. We present a general method for augmenting existing instruments into ESR-STMs to investigate spin dynamics in the high-field limit. We demonstrate the performance of the instrument by analysing inelastic tunnelling in a junction driven by a microwave signal and provide proof of principle measurements for ESR-STM.

INTRODUCTION

The direct manipulation and detection of individual spins (see Fig. 1(a)) is one of the major goals in contemporary nanoscience [1–9]. Meeting these challenges requires a local measurement of electronic and magnetic properties with atomic precision. The scanning tunnelling microscope (STM) routinely achieves this limit of resolution and is thus an ideal tool to study the dynamics of magnetic nano-objects [10] on their own length and time scales [11–14].

Combining electron spin resonance with STM (ESR-STM) has introduced new possibilities to the local studies of individual spins and has expanded the available parameters space substantially, but it imposes a series of strict experimental requirements, most notably on the base temperature of the cryostat. The operational frequency range of the instrument determines the maximum magnetic field for ESR-STM experiments and sets the relevant energy scale in the experiment. ESR-STM relies on the thermal initialisation of the target systems into their ground state. However, in many contemporary implementations of ESR-STM, the Zeeman energy is on the order of $k_B T$ and a non-negligible excited state population remains [4, 6]. This is a significant impediment to resolving intrinsic spin dynamics at the nanoscale. The goal of coherent manipulation from a known ground state may be reached via two approaches: Reducing the base temperature of the experiment to suppress thermal excitations from the ground state, or increase the microwave frequency to operate at higher magnetic fields.

Current implementations of ESR-STM typically operate at frequencies up to 40 GHz [15–17]. To achieve thermal initialisation of the target systems at these frequencies mK temperatures are required, which are only achievable in dilution refrigerators [18]. This approach requires dedicated machines that are costly to produce and present significant challenges in everyday operation. High frequency signals in the upper GHz range, on the other hand, can be generated in an independent setup outside the ultra-high vacuum (UHV) system and routed to the tunnel junction through a set of suitable cables. This approach is, therefore, more flexible, allowing the retrofitting or modification of existing machines by the addition of dedicated high GHz cabling [19].

Extending the operational frequency range of ESR-STM has thus far been prevented by challenges in signal delivery. We have augmented a commercially available STM (Unisoku model USM1300) featuring 310 mK base temperature and a 6 T single axis magnet with an antenna assembly which permits us to deliver microwave signals of up to 105 GHz directly to the tunnel junction. This work can be used as a guideline to design new instruments or retrofit existing ones for high GHz microwave capabilities.

INSTRUMENT DESIGN

The Unisoku model USM1300 STM is a commercially available experimental platform combining ultrahigh vacuum (UHV) sample preparation and ultra-low temperature STM with high field capabilities. The STM unit, developed and manufactured by the Unisoku corporation, is installed on the insert of a superinsulated $^4$He bath cryostat produced by Cryogenic Ltd. The insert includes a 1 K pot, which is supplied with liquid helium from the bath through an adjustable needle valve, and a single-shot $^3$He cooling cycle. With a total volume of 301 $^3$He gas, the STM is capable of operating at a base temperature of 310 mK for up to 72 hours.

Figure 1(b) shows a sketch of the cryostat insert including the modifications we implemented as part of the ESR-
STM augmentation. The main addition to the base setup is the installation of a series of 0.047 inch semirigid coaxial cables, rated to 110 GHz, and a radio frequency antenna into the system. We solve the challenges of finding leak-tight vacuum feedthroughs and thermalisation of the RF assembly to produce a high-performance machine capable of delivering high-frequency signals at large amplitudes onto the tunnel junction. Our approach extends the operational frequency range of ESR-STM by a factor of more than two while maintaining signal amplitudes comparable with previous efforts.

Below is a step-by-step discussion of the design philosophy and implementation of our custom modifications to the base system.

**High-frequency wiring**

The geometry of STMs presents a severe challenge to the integration of high-frequency signals. The confined space of the scanner housing and complex shape of the tip-sample system thwart any attempt to realise an impedance-matched connection to the tip. The resulting scattering of electromagnetic waves in the instrument will inevitably lead to high losses. This makes it all the more difficult to bring a high amplitude signal close to the tunnel contact. The requirement for high signal amplitudes leads to a conflict with a key design principle for cryostats to use high-resistance cabling in order to limit the thermal load on the experiment.

We overcome these issues by using a combination of high conductance coaxial cables from different materials to achieve maximum power transmission to the antenna. We use semirigid cables with a copper jacket and silver-plated copper weld (SPCW) conductor on the air side and in the upper sections of the cryostat to the 1K-pot (see red wire section in Fig. 1(b)). Their high conductance ensures small signal losses even at room temperature. We installed a coaxial cable with NbTi shield and conductor running from the 1K-pot to the \(^3\)He stage (see blue wire section in Fig. 1(b)). NbTi is a superconductor with a transition temperature of 10 K and a high critical field of 15 T. As superconductors are excellent conductors of electricity, but very poor conductors of heat [20], the NbTi cable provides excellent signal transmission at low temperatures while essentially eliminating thermal loads on the low-temperature parts of the experiment. Finally, a flexible coaxial cable with silver plated copper shield and conductor connects the RF antenna in order to preserve STM motion during sample transfer and spring damping during regular operation.

We use semirigid coaxial cables with 0.047 in outer diam-
eter for all applications. This cabling standard is rated to 110 GHz with an impedance of 50 Ω. We installed 1 mm connectors (Anritsu W1 series), also rated to 110 GHz, on all cable segments. Before installation in the machine, all cables were repeatedly immersed in liquid helium and rigorously tested for any temperature related damages.

Our combination of copper/SPCW and superconducting cables requires excellent heat management in order to work effectively. To ensure proper thermalisation, all cables are anchored at several points inside the cryostat. The thermal anchors consist of a copper braid, glued to the outer conductor of the cable over a large surface area using thermally conductive silver epoxy, and fastened to the anchor points with a screw and lug. The copper/SPCW cable is anchored at the baffles of the cryostat, the sorption pump, and the 1K-pot. The NbTi cable is thermally anchored at the 1K-pot and 3He-pot as described above and wound in a wide loop around the UHV column to accommodate the thermal expansion and contraction of the wiring during cool-down or warm-up. The installation of our custom radio frequency cabling did not affect the base temperature of the instrument.

High-frequency UHV feedthrough

To our knowledge, there are no commercially available hermetically sealed double ended coaxial vacuum feedthroughs rated to 90 GHz or above. In practice, however, vacuum feedthroughs with 1.85 mm connectors, rated to 65 GHz, show very low losses up to at least 90 GHz and can act as a substitute. Fig. 2 shows transmission measurements through the hermetically sealed feedthrough KPC185FFHA by Kawashima Manufacturing Corporation from 60 GHz to 90 GHz. The performance of KPC185FFHA is comparable to 1 mm female-female adapters rated to 110 GHz. A pair of 1.85 mm to 1 mm adapters (e.g. CentricRF C8186) is needed to mate the feedthrough to the high-frequency cabling.

High-frequency antenna

The implementation of the dedicated high-frequency line requires a solution for coupling the high-frequency signal to the tunnel junction. We designed an antenna to transform the electric signal in the high-frequency line into electromagnetic radiation illuminating the tunnel junction, with the STM tip effectively acting as a receiver. A dedicated antenna ensures efficient coupling into the vacuum, eliminating losses. Mounting it in close proximity to the tunnel contact further improves the signal strength.

We chose an on-chip antenna design for a compact and integrated solution [21]. The silicon chip is mounted in a phosphorous bronze carrier attached directly to the side of the STM scanner housing, shown in Fig. 3(a). A flange mount connector (Anritsu W1-103F) provides the electrical contact to the high-frequency wiring. To increase the microwave power incident on the tunnel junction, we fitted the carrier with a hyper-hemispherical silicon lens flush with the underside of the antenna chip to partially collimate the radiation.

A broadband bowtie antenna affords the most flexibility, covering the entire intended frequency range of 60 to 90 GHz. Fig. 3(b) shows a dimensional drawing of the optimised antenna structure installed in the microscope. The antenna is assembled in a thin AuPd film on a high resistivity silicon substrate of 380 μm thickness. Design and parameter optimisation for the antenna was performed using the CST Microwave Studio software with performance tests in 8:1 scale models. Fig. 3(c) shows the simulated reflectance of the antenna from DC up to 120 GHz [21]. The design achieves excellent power dissipation across a wide band beginning at about 60 GHz.

RADIO FREQUENCY GENERATION

We use a multi-stage generation scheme to reach our intended operational frequency window (see Fig. 1(b)). The first stage is a baseband generator (Keysight 8257D) capable of producing signals up to 20 GHz. This generator feeds a frequency extension module (VDI WR12SGX), which multiplies the input frequency by a factor of six. The frequency extension module is intended to operate with input frequencies between 10 and 15 GHz, producing a constant amplitude signal between 60 and 90 GHz. We found that upper limit can be extended by feeding the module with higher input frequencies,
but at a significant cost in amplitude. Still, overdriving allows us to extend the operational frequency range of our ESR-STM to 105 GHz. Expanding the frequency range at the lower end in the same fashion is not possible due to the sharp cut-off of the WR12 waveguide output on the extension module. We regulate the source amplitude through a computer-controlled rotary vane attenuator (Mi-Wave 511E/387ND). This device allows us to regulate the power entering the high-frequency wiring in steps of 0.1 dB from the source power of the frequency extension module.

**INSTRUMENT PERFORMANCE**

Key operational parameters of the instrument, such as base temperature, \( z \)-stability and \(^3\)He hold time, were unaffected by our modifications. We demonstrate the excellent STM performance in a series of test measurements on V(100). Fig. 4(a) shows the oxygen reconstruction of the V(100) surface [22, 23] in atomic resolution at the base temperature of 310 mK after the system upgrade. Tunnelling spectroscopy between a superconducting vanadium tip and the V(100) sample shows well-developed coherence peaks and a clear energy gap typical of superconductor-insulator-superconductor junctions (see Fig. 4(b)) [20, 24, 25].

**Microwave signal delivery**

We evaluate the performance of the microwave assembly by observing microwave-assisted tunnelling processes occurring in a superconductor-insulator-superconductor (SIS) junction between a V(100) sample and a vanadium tip driven by a high-frequency signal from the antenna. SIS junctions are well studied and a robust framework for data analysis is readily available [19, 26–28]. The current in presence of a radio frequency signal is [26, 29]:

\[
I_{QP}(V_0, V_\omega) = \sum_{n=-\infty}^{\infty} J_n^2 \left( \frac{eV_\omega}{\hbar\omega} \right) I_{QP} \left( V_0 - \frac{n\hbar\omega}{e}, 0 \right), \tag{1}
\]

where \( I_{QP}(V, 0) \) is the quasiparticle current in absence of any RF radiation, \( V_0 \) the DC bias voltage applied to the tunnel junction, \( V_\omega \) the AC voltage dropping across the junction as a result of the RF signal, \( J_n \) are the Bessel functions of the first kind of order \( n \), and \( e \) is the elementary charge. Further, \( \hbar \) is the reduced Planck constant and \( \omega = 2\pi\nu \), where \( \nu \) is the microwave frequency. The sharp coherence peaks in the SIS tunnel data allow us to observe directly the effects of changing frequency. To analyse our data, we use reference spectra acquired using the same parameters as the corresponding microwave-assisted tunnelling spectrum, but with the RF signal switched off. This reference measurement is used as input for \( I_{QP} \) in Eq. (1). Fitting our model to the experimental data allows us to extract the frequency and amplitude of the RF signal arriving at the junction. All data shown is acquired at a base temperature of 310 mK.

---

**Figure 4. Performance test using a SIS tunnel junction.**

(a) Atomic resolution image of the oxygen reconstruction on a V(100) surface. (b) A typical reference spectrum of the SIS contact in absence of an RF signal (blue) is shown alongside conductance spectra under irradiation by a RF signal of 75 GHz (orange) and 60 GHz (yellow). Replica of the superconducting coherence peaks are clearly resolved. Their separation from the original position depends on the frequency of the RF signal. (c) Experimental data for a junction under irradiation by 60 GHz radiation of varying power (blue and yellow) with fits from the model in Eq. (1) superimposed (orange and purple).
Figure 5. Individual atoms on a thin insulating layer for ESR-STM experiments. (a) A double layer MgO island on an Ag(100) surface serves as a decoupling layer for individual Fe atoms and TiH molecules. (b) IETS spectrum on a TiH molecule in 6 T out of plane external field. (c) IETS spectrum on an individual Fe atom in 2.3 T out of plane external field. The width of the excitation gap around zero bias is a fingerprint for the unambiguous identification of Fe and TiH.

Only a single frequency is needed in the model to reproduce the experimental results [19].

ESR-STM

The prototypical spin systems for ESR-STM are individual metal atoms on an MgO decoupling layer on Ag(100) [3]. We grow double-layer MgO islands by sublimation of Mg in a 1×10^{-7} mbar oxygen atmosphere onto the sample held at 700 K. Sublimation of Ti and Fe from an electron beam evaporator onto the cold sample (T≤20 K) yields individual Fe atoms and TiH molecules. We use individual metal atoms on MgO/Ag(100) as a model system to provide proof of principle ESR-STM measurements. Fig. 5(a) shows an STM topograph of a typical sample, once again demonstrating that the machine can resolve single atoms with ease. The tip is a sharpened Pt/Ir wire, further prepared by field emission in the microscope and repeated indentation into the Ag(100).

Figure 6. Generation of constant amplitude radio frequency sweeps at the tunnel junction. (a) Step-like feature measured on a TiH molecule (blue), spectrum under radio frequency irradiation (orange), and fit using Eq. (1) (yellow) with ν=64.9 GHz and \( V_0 = 7.5 \text{ mV} \). The radio frequency amplitude determined from the fit is used to compute the transfer function. (b) Calibration curve mapping the measured lock-in signal to RF amplitude. The orange line is a smoothed spline interpolation, which we use to interpolate the calibration curve. (c) Transfer function measured from 60 GHz to 105 GHz. The operational range of the instrument extends far past the intended 60 to 90 GHz. (d) Constant amplitude radio frequency sweep at 8 mV setpoint value.

We identify individual Fe atoms and TiH molecules through their fingerprints in inelastic excitation tunnel spectroscopy (IETS), see Fig. 5(b) and (c).

The essential ingredient of any ESR measurement is the tuning of the spin system across its resonance with the microwave signal. This may be achieved through either changing the excitation frequency at fixed field (ν sweep), or the external field at fixed excitation frequency (B sweep). In either case, it is necessary to calibrate the microwave amplitude in order to perform consistent and comparable measurements.

Frequency sweeps further require the compensation of the antenna-junction transfer function to ensure a constant amplitude signal at throughout the measurement to suppress spurious signals.

We mainly follow Ref. [15] to calibrate the microwave signal generate constant amplitude radio frequency sweeps at the junction. A similar procedure is given in Ref. [6]. Indi-
Figure 7. High-frequency ESR STM. ESR-STM signals from frequency sweeps at 175 pA (a) and 50 pA (b) setpoint current. The legend gives the centre frequency of the signal. The probe current is a source of decoherence that contributes to the broadening of the signal. (c) ESR-STM signals from field sweeps at 175 pA setpoint current. The frequency of the excitations signal for each sweep is given in the annotations. (d) All ESR-STM measurements yield a $g$-factor close to 2.

individual TiH molecules on MgO show a prominent step-like feature at a bias voltage of about $-80 \text{ mV}$ (see blue spectrum in Fig. 6(a)). We first determine the transfer function of the antenna assembly at one fixed frequency using Eq. (1). We acquire a reference spectrum in absence of any RF signal and a tunnel spectrum under RF irradiation, as for the SIS case above. Then, we extract the signal amplitude $V_\omega$ through a fit (see yellow spectrum in Fig. 6(a)) of the data for the irradiated junction (see red spectrum in Fig. 6(a)). The transfer function $T$ is then easily found from the known source amplitude $V_S$ according to

$$T = 20 \log(V_\omega/V_S).$$

We then acquire a calibration curve by measuring the conductance near the centre of the slope of the TiH step as a function of source amplitude, see Fig. 6(b). We use a smoothing cubic spline interpolation to fit the calibration function more accurately for arbitrary shapes. The smoothing parameter must be calculated such that the resulting smoothed spline can be inverted. This produces better fits than directly fitting to the inverted data. With the known transfer function value, these values can be converted into signal amplitudes at the junction. Since the TiH step is much broader than $\hbar \nu$ in the frequency range of interest, the frequency itself has no measurable effect on the RF spectrum and the calibration curve becomes a universal injective mapping of the conductance on $V_\omega$.

By measuring the conductance of our tunnel junction with fixed signal attenuation while varying the source frequency and mapping the acquired values onto amplitudes using the inverted calibration curve, we measure the transfer function of the antenna assembly over a large frequency range, see Fig. 6(c). The instrument performs well over the complete intended operational range of 60 to 90 GHz and beyond. Losses do not become insurmountable until about 105 GHz. Using the experimentally determined transfer function, we can compensate the losses in the antenna assembly through the adjustable attenuator to generate constant amplitude radio frequency sweeps at the tunnel junction, see Fig. 6(d). We achieve signal amplitudes of 10 mV throughout most of the frequency range, and up to about 90 mV at select frequencies.

ESR Signal

We demonstrate the ESR capabilities of our setup using TiH molecules on double layer MgO/Ag(100) as a model system (see Fig. 5). TiH adsorbed on the bridge site (between two oxygen atoms) of MgO is a spin-$1/2$ system with a $g$-factor close to 2 in an out-of-plane field $[6, 30–33]$. We attach one or more Fe atoms to the STM tip to generate a spin-polarised probe by picking them up from the surface $[3]$. An out-of-plane magnetic field lifts the degeneracy between the spin states of the TiH molecule. In our frequency range, resonance is achieved in fields between about 2.15 T and 3.75 T. The Zeeman energy in these fields is about an order of magnitude larger than the thermal energy at the base temperature of 310 mK and the spin systems are thermally initialised to their ground state. All of the data presented here are measured at a bias voltage of 100 mV and the setpoint currents are referenced to this voltage.

The RF signal drives the resonant transition between the ground and excited states, resulting in a reduction of the spin-polarised signal on the atom as the time-averaged spin population is no longer thermal. By chopping the RF driving signal and locking in to the chopping frequency with a lock-in amplifier, we single out and record those parts of the signal that are affected by the driving, i.e. the ESR-STM signal $[3, 15]$. Our results are summarised in Fig. 7. All data shown was acquired with the same spin-polarized microtip on the same TiH molecule adsorbed on a bridge site. Figure 7(a) and (b) show frequency sweep ESR-STM measurements at 175 pA and 50 pA setpoint current, respectively. The RF amplitudes are 10 mV and 15 mV for the data at 175 pA and 50 pA, respectively. We find reasonably spaced intervals throughout the frequency band between 60 GHz and 95 GHz, where a $\nu$
sweep is possible (as indicated in the insets). As the probe current is itself a source of decoherence, the line width is substantially reduced at lower current. This reproduces the general trend that has been observed previously [4]. Also, the line widths of the measured ESR peaks are comparable to the published literature at lower frequencies (≤ 40 GHz).

Fig. 7(c) shows ESR data from B sweep measurements at sixteen different excitation frequencies spanning the available frequency range between 60 GHz and 100 GHz of the instrument. The microwave amplitude was set to 10 mV. The baseline has been offset to zero for all sweeps. We find a fairly even distribution of suitable frequencies across a magnetic field range of almost 1.5 T. Such a spread allows for a more accurate determination of the $g$-factor, which contains valuable information on the molecule and its environment [32].

We calculate the corresponding Zeeman energies $E_Z = h \nu_{\text{res}}$ for all data sets, where $\nu_{\text{res}}$ is the resonance frequency. The $g$-factor of our spin system can then be extracted through a simple linear fit according to

$$E_Z = 2 \mu_B s g B, \quad (3)$$

where $\mu_B$ is the Bohr magneton and $s = 1/2$ is the spin of the TiH molecule. The results are shown in Fig. 7(d). We consistently obtain $g$-factors close to 2 for all our measurements, in agreement with previous results for TiH on the bridge site and in an out-of-plane field [6]. The data points lie very well on the line given by Eq. (3) for both the $\nu$ sweep as well as the $B$ sweep, which were extracted from independent measurements at 175 pA setpoint current shown in Fig. 7(a) and (c). The fitted $g$-factors are $g = 2.005$ ($\nu$ sweep) and $g = 2.008$ ($B$ sweep), which lie within 0.15%. For the $\nu$ sweep at 50 pA setpoint current, we find a $g$-factor of $g = 1.988$, which is slightly smaller than for the higher setpoint current. The offset between the $\nu$ sweeps at 50 pA and 175 pA setpoint current can be attributed to the different tip fields $B_{\text{tip}}$ felt by the TiH molecule. The tip fields are 95.7 mT (B sweep) and 95.1 mT ($\nu$ sweep) at 175 pA current setpoint and 46.1 mT for the 50 pA current setpoint. The small changes in the $g$-factor can be attributed to tip-induced changes in the TiH bond length [32, 33].

**CONCLUSION**

We have augmented a commercially available low-temperature STM system into a high-performance ESR-STM by the addition of a dedicated high-frequency line with antenna assembly to deliver radio frequency signals between 60 and 105 GHz to the tunnel junction. Using commercially available components rated to high GHz frequencies, we achieve a high signal amplitude across a wide frequency range. The compensation of the transfer function allows us to keep the signal amplitude constant throughout our frequency range. In a series of proof-of-principle measurements, we measure an ESR signal on individual TiH molecules on a MgO decoupling layer in both frequency sweep and field sweep modes. In an operational field of several Tesla and at a base temperature of 310 mK, excited state populations of typical spins-$\frac{1}{2}$ systems at a resonance frequency above 60 GHz are lower than 0.01%. With these parameters, it becomes possible to study the intrinsic dynamics of individual spins with atomic resolution. Our approach can serve as a template to convert typical STMs into ESR-STMs capable of resolving the coherent dynamics of individual spins and magnetic nanostructures.

**ACKNOWLEDGEMENTS**

We gratefully thank Andreas Heinrich, Klaus Kern, Aparajita Singha, Markus Ternes, and Philip Willke for fruitful discussions. This work was funded in part by the ERC Consolidator Grant AbsoluteSpin (Grant No. 681164).
[12] T. L. Cocker, V. Jelic, M. Gupta, S. J. Molesky, J. A. J. Burgess, G. D. L. Reyes, L. V. Titova, Y. Y. Tsui, M. R. Freeman, and F. A. Hegmann, An ultrafast terahertz scanning tunnelling microscope, Nat Photon 7, 620 (2013).

[13] S. Yoshida, Y. Aizawa, Z.-h. Wang, R. Oshima, Y. Mera, E. Matsuyama, H. Oigawa, O. Takeuchi, and H. Shigekawa, Probing ultrafast spin dynamics with optical pump–probe scanning tunnelling microscopy, Nature Nanotechnology 9, 588 (2014).

[14] R. Gützler, M. Garg, C. R. Ast, K. Kuhnke, and K. Kern, Light–matter interaction at atomic scales, Nature Reviews Physics, 1 (2021).

[15] W. Paul, S. Baumann, C. P. Lutz, and A. J. Heinrich, Generation of constant-amplitude radio-frequency sweeps at a tunnel junction for spin resonance STM, Review of Scientific Instruments 87, 074703 (2016).

[16] F. D. Natterer, F. Patthey, T. Bilgeri, P. R. Forrester, N. Weiss, and H. Brune, Upgrade of a low-temperature scanning tunneling microscope for electron-spin resonance, Review of Scientific Instruments 90, 013706 (2019), publisher: American Institute of Physics.

[17] J. Friedlein, J. Harm, P. Lindner, L. Bargsten, M. Bazarnik, S. Krause, and R. Wiesendanger, A radio-frequency spin-polarized scanning tunneling microscope, Review of Scientific Instruments 90, 123705 (2019), publisher: American Institute of Physics.

[18] W. M. J. van Weerdenburg, M. Steinbrecher, N. P. E. van Mullekom, J. W. Gerritsen, H. von Allwörden, F. D. Natterer, and A. A. Khajetoorians, A scanning tunneling microscope capable of electron spin resonance and pump–probe spectroscopy at mK temperature and in vector magnetic field, Review of Scientific Instruments 92, 033906 (2021).

[19] P. Kot, R. Drost, M. Uhl, J. Ankerhold, J. C. Cuevas, and C. R. Ast, Microwave-assisted tunneling and interference effects in superconducting junctions under fast driving signals, Physical Review B 101, 134507 (2020).

[20] M. Tinkham, Introduction to Superconductivity (Dover, 1996).

[21] J. Merlet, Entwurf und Untersuchung einer Antenne für 84GHz zur Strahlungseinkopplung in ein STM, Bachelor’s Thesis, Karlsruher Institut für Technologie, (2017).

[22] P. W. Davies and R. M. Lambert, Surface chemistry of the metal-halogen interface: Bromine chemisorption and related studies on vanadium (100), Surface Science 95, 571 (1980).

[23] J. S. Foord, A. P. C. Reed, and R. M. Lambert, The (100) surfaces of chromium and vanadium: Reconsiderations of their structure and reactivity, Surface Science 129, 79 (1983).

[24] C. R. Ast, B. Jäck, J. Senkpiel, M. Eltschka, M. Etzkorn, J. Ankerhold, and K. Kern, Sensing the quantum limit in scanning tunnelling spectroscopy, Nature Communications 7, 13009 (2016).

[25] B. Jäck, M. Eltschka, M. Assig, M. Etzkorn, C. R. Ast, and K. Kern, Critical Josephson current in the dynamical Coulomb blockade regime, Phys. Rev. B 93, 020504 (2016).

[26] G. Falci, V. Bubanja, and G. Schön, Quasiparticle and Cooper pair tunneling in small capacitance Josephson junctions, Zeitschrift für Physik B Condensed Matter 85, 451 (1991).

[27] A. Roychowdhury, M. Dreyer, J. Anderson, C. Lobb, and F. Wellstood, Microwave Photon-Assisted Incoherent Cooper–Pair Tunneling in a Josephson STM, Phys. Rev. Applied 4, 034011 (2015).

[28] O. Peters, N. Bogdanoff, S. Acero González, L. Melischek, J. R. Simon, G. Reecht, C. B. Winkelmann, F. von Oppen, and K. J. Franke, Resonant Andreev reflections probed by photon-assisted tunneling at the atomic scale, Nature Physics 16, 1222 (2020).

[29] P. K. Tien and J. P. Gordon, Multiphoton Process Observed in the Interaction of Microwave Fields with the Tunneling between Superconductor Films, Physical Review 129, 647 (1963).

[30] Y. Bae, K. Yang, P. Willke, T. Choi, A. J. Heinrich, and C. P. Lutz, Enhanced quantum coherence in exchange coupled spins via singlet-triplet transitions, Science Advances 4, eaau4159 (2018).

[31] K. Yang, Y. Bae, W. Paul, F. D. Natterer, P. Willke, J. L. Lado, A. Ferrón, T. Choi, J. Fernández-Rossier, A. J. Heinrich, and C. P. Lutz, Engineering the Eigenstates of Coupled Spin-1/2 Atoms on a Surface, Physical Review Letters 119, 227206 (2017).

[32] M. Steinbrecher, W. M. J. van Weerdenburg, E. F. Wolvaren, N. P. E. van Mullekom, J. W. Gerritsen, F. D. Natterer, D. I. Badridinov, A. N. Rudenko, V. V. Mazurenko, M. I. Katsnelson, A. van der Avoird, G. C. Groenenboom, and A. A. Khajetoorians, Quantifying the interplay between fine structure and geometry of an individual molecule on a surface, Physical Review B 103, 155405 (2021).

[33] T. S. Seifert, S. Kovarik, D. M. Juraschek, N. A. Spaldin, P. Gammel, and S. Stepnow, Longitudinal and transverse electron paramagnetic resonance in a scanning tunneling microscope, Science Advances 6, eabc5511 (2020).