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**Title:** Advances in SQUID-detected magnetic resonance force microscopy  
**Issue Date:** 2019-06-18
Magnetic Resonance Force Microscopy is a technique with immense potential, but in practice it is hindered by the complexity of the setup and strong restrictions on possible samples. For these reasons only approximately 10 groups work on this technique worldwide, and MRFM setups are not commercially available. In this valorisation chapter, we explain the necessity for a more simplified MRFM setup, if necessary with a slightly reduced functionality. This would make the technique more broadly applicable, in which case it could offer invaluable information to a variety of fields, some of which will be exemplified at the end of this chapter. We show current progress in the development of this new instrument, and describe the modifications required to reach the desired specifications for full operation.
9 Valorisation: the Easy-MRFM

9.1 Necessity for a New Characterization Tool

The current performance of many nanodevices is limited by the presence of fluctuating two-level systems (TLS) that couple to the device. These systems are often associated with the presence of dangling chemical bonds on the surface of the substrate or inside the device itself. Examples of the limited performance of devices include the reduced coherence times in superconducting qubits [193, 206–211], the short $T_2$ times of shallow NV-centers [135, 137–139, 212, 213], and excess flux noise in SQUIDs and Josephson junctions [214–217].

Attempts to solve these issues follow two general approaches. On the one hand people try to reduce the device’s sensitivity to the noise by tweaking the design [195, 218, 219]. On the other hand they try to remove the source of the noise by changing materials and fabrication procedures [141, 220, 221]. However, real progress is hindered by the fact that we are blind to the effects of various adjustments until the finished device is tested, as only the final performance of the device gives a decisive answer. This means improvements are costly and time-consuming, as changes to one of the first steps of the production still require the completion of all subsequent steps. Additionally, when it is suspected that the unwanted electron spins are introduced in one of the fabrication steps, it is currently very challenging to find out which particular step is responsible. What is needed is an independent characterization tool that is capable of quantifying the impact of an action at any stage in the device fabrication process on the density of two level systems.

Our magnetic resonance force microscope (MRFM) might be a viable option for this purpose. We have demonstrated that we are able to do ultra sensitive magnetic force microscopy measurements on dilute paramagnetic spin systems to extract spin densities, spin-lattice relaxation times, and even rudimentary dynamics such as spin diffusion. Over the years, we have used this technique on a variety of samples, such as the dangling bonds in the native oxide of silicon [77], the surface spins and $(N^-)$ bulk spins in diamond [34], and the Fe$^{3+}$ defects in the bulk of single-crystal strontium titanate (SrTiO$_3$) [53]. In our group, the ultra sensitive magnetic force technique has become a standard sample characterization before the start of MRFM experiments.

The method of the characterization of a sample’s spin properties is based on the observation that the resonance frequency and damping of the magnetically-tipped cantilever are altered by the vicinity of nearby spins. By measuring the temperature- and distance-dependence of both the resonance frequency and the damping of the cantilever at very low temperatures ($< 250$ mK), the density and $T_1$ times of the spins on the surface and in the bulk of a sample can be determined [52]. The lowest
surface spin density that we have found was 0.07 spins/nm$^2$. In other words, these surface spins are on average 4 nm apart. The sensitivity of our method would be sufficient to detect 100 times fewer spins than this, i.e. electron surface spins that are up to 40 nm apart on average.

However, our current MRFM setup is ill-suited as a general sample characterization tool, since measurements can only be done in the proximity of a pickup loop necessary for the detection of the motion of the magnetically-tipped cantilever. This means that either the sample has to be placed on a detection chip, or a superconducting pickup loop has to be fabricated on the sample. Using optical detection instead would not allow to reach the low temperatures that are called for to polarize the magnetic spins with the field of our magnetic particle. An additional drawback is that changing the sample requires a partial deconstruction of the setup, so it takes at least several days. This makes the current setup incompatible with, for instance, testing surface treatments concerning the removal of oxide layers, where a fast throughput time is essential.

Therefore, we have developed a new setup, called the easy-MRFM, that is still compatible with milliKelvin operating temperatures (and therefore is still based on SQUID technology), and where all vital MRFM components are separated from the sample. This opens the possibility to characterize more general samples with a higher turn-over rate.

9.2 Progress of the easy-MRFM

The idea of the easy-MRFM is to fabricate a superconducting pickup loop on a silicon arm that can be placed next to the cantilever, rather than fabricating the pickup loop on the sample itself. This would allow for detection of the motion of the cantilever with a constant coupling independent of the position of the cantilever with respect to the sample. The ability to measure the properties of the cantilever is all that is required for the magnetic force measurements to extract the spin densities of the sample under investigation. More complex MRFM protocols, requiring radio-frequency (RF) pulses, can be performed by using the higher modes of the cantilever for the mechanical generation of RF fields [50].

The first prototype of the easy-MRFM is shown in Figs. 9.1(a)-(d). It was designed to be a module for the current MRFM setup, where it can simply be exchanged with the old cantilever holder without major adjustments. As a result, the entire easy-
Figure 9.1: (a) Photograph of the easy-MRFM prototype with the most important components labeled. (b) Easy-MRFM detection chip and cantilever. (c) Alignment of the cantilever with respect to the pickup loop with area $(40 \, \mu m)^2$. (d) Easy-MRFM positioned above a conventional MRFM detection chip before cooling down. (e) Detected cantilever response to a driving force. The left image shows the polar plot (raw data), the right image shows the squared amplitude response, corrected for the direct crosstalk between the drive and detection circuits. The solid red line is a Lorentzian fit to the data.
MRFM has a diameter of only 8 mm. The prototype contains the cantilever chip, mounted on top of a piezoelectric element, the easy-MRFM detection chip made of 100 nm thick NbTiN, fabricated by Delft Circuits \cite{222}, and a gradiometric transformer to match the detection chip pickup loop to the input coil of a SQUID, which is placed in a superconducting shielding several centimeters away from the tip of the easy-MRFM. The cantilever, which is one of the double-magnet cantilevers described in Ch. 8 of this thesis, can be placed within a distance of 50 µm from the pickup loop (see the inset of Fig. 9.1(c)).

Initial results show that it is possible to detect the driven motion of the cantilever and extract its properties, as shown in Fig. 9.1(e). Despite large crosstalk, we can extract the properties of the cantilever which gives access to the two parameters, \( f_{\text{res}} \) and \( Q \), that are necessary to extract a spin density. In the right image of Fig. 9.1(e) we have manually subtracted the offset due to the crosstalk. The same can be achieved using a circle fit to the cantilever signal in the polar plot \cite{72}. Protocols for the fits to the polar plot data are available \cite{65}.

Even though this data gives the proof-of-concept for the easy-MRFM, further optimization is desirable. The coupling between the magnetically tipped cantilever and the detection pickup loop is currently too small to see the thermal motion of the resonator (required to calibrate the absolute coupling strength). Furthermore, the low coupling requires the cantilever to be driven to a large amplitude for its motion to become detectable. This large amplitude may induce non-linearities in the cantilever, which hinder the determination of the cantilever’s resonance frequency and quality factor. For these reasons, the coupling should be improved by at least one order of magnitude. This can be done by optimizing the position of the cantilever with respect to the pickup loop, increasing the radius of the magnet at the end of the cantilever, or improving the matching between the pickup loop and the input coil of the SQUID. The latter can be done by reducing the parasitic inductance and tweaking the design of the transformer.

When the easy MRFM is operational, the turn-over rate will be dictated by the cooldown time of the dilution refrigerator in which it is operated, which is several days. The turn-over rate can be improved further by mounting the instrument on a cold-insertable probe \cite{223}, in which case samples can be exchanged without the need to open the cryostat. Then it should become possible to determine spin densities on one sample per day.
9.3 Future applications

When the easy-MRFM is further developed into a mature technique, applications can be found in many fields of physics and industry. In the greater quantum computation community, it could be used to pinpoint the critical fabrication steps that should be removed, adapted, or included to remove the dangling bonds responsible for the low coherence times. As a pilot experiment we propose to measure the spin density on a thin layer of NbTiN, a common material used for superconducting qubits, before and after the in-situ application of a small amount of hydrofluoric acid to remove the surface layer on top of the NbTiN.

Apart from the prospect of using the easy-MRFM technology to specifically address the problem of unwanted electron spins in quantum computation devices, we believe that it may also be useful to study chemically amplified resists after they have been illuminated with UV light. The radicals that are produced in the process have an unpaired electron [224][226]. It is estimated that a typical density of broken bonds is about $10^{20} \text{ cm}^{-3} = 0.1 \text{ nm}^{-3}$ [227]. At this density, these radicals should be readily detectable as a frequency shift of our magnetic force sensor. Any additional information about the detailed response of the resist to various radiation doses could be invaluable for improving the performance of the new high resolution resists.