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MRFM can be a valuable technique for condensed-matter physics. In this chapter, four future experiments are discussed in different sub-fields of condensed-matter. These fields could all benefit greatly from a wide availability of MRFM. It only becomes widely available when it becomes more user-friendly. The results in Ch. 6 invites us to think of a monolithic MRFM, which would make the technique easier to use. The combination of a valuable technique with being user-friendly paves the way of a commercial available MRFM.
8.1 Outline

The first experiment we propose is that of measuring the electronic susceptibility of the surface state of a topological insulator. We show a first attempt of such a measurement and give suggestions for improvements.

In Sec. 8.3 we propose an experiment on the two dimensional superconducting electron gas of an oxide interface (LAO/STO), of which the exact nature of the superconducting state is as yet unknown.

In Sec. 8.4, we propose an experiment in which we probe a copper layer which is part of a triplet spin valve. Measuring the Korringa relation for copper nuclei could give a direct evidence of triplet superconductivity\(^1\). Our experiment on the dangling bonds in Ch. 7 shows the sensitivity of our technique for magnetic moments.

In Sec. 8.5 we suggest that our MRFM can measure the diamagnetic response of nanometer sized domains in a doped Mott insulator, which are expected to become superconducting.

Finally, in Sec. 8.6 we propose the **Monolithic** MRFM. We propose that the pickup coil, which facilitates the read-out of the force sensor, is integrated on the cantilever chip. Since we have demonstrated in Ch. 6 that we no longer need a conventional rf current source close to the sample, all necessary MRFM components are then integrated on a single chip, which will make the technique simpler to use.

8.2 Proposal 1: Surface states of a topological insulator

**The original research proposal** by Wagenaar (2013b) included experiments of the surface state of a topological insulator. In this section we would like to discuss the proposed experiment, and the results we obtained in the first attempt hereof.

Topological insulators have some interesting properties with many foreseen applications in spintronics and quantum computers. Recent observation with angle-resolved
opportunities for mrfm 113

photoemission spectroscopy (ARPES) and scanning tunneling spectroscopy (STS) on recently cleaved crystals have shown the presence of unique surface states\(^2\). These techniques have the disadvantage that they are limited to measuring the uttermost surface. This makes it difficult to measure surface states if these are covered with an oxide layer or to obtain information whether these states extend partially to the bulk.

Conventional nuclear magnetic resonance (NMR) techniques have the disadvantage that they are equally sensitive to the bulk as to the surface. Measurements on nanometer-powdered samples with bulk NMR of Bi\(_2\)Te\(_3\) have shown a Korringa-like relation\(^3\), but it is not possible to measure this relation on larger crystals of this material. MRFM is able to perform NMR experiments locally.

The main goal of this proposed experiment is to explore topological surface states, thereby showing that we can provide a depth controlled local probe with our MRFM technique in the field of topological insulators.

The lab of Mark S. Golden in Amsterdam has been making major progress in the production of truly bulk-insulating three dimensional topological insulators in the Bi,Sb,Te,Se-family of materials. A lively collaboration has been set up with the group of Hans Hilgenkamp and Alexander Brinkman of the university of Twente on building devices for nanotransport experiments.

Researchers Chuan Li and Joris Voerman have placed

\(\text{Figure } 8.1: \text{ a) Optical image of the Bi}_2\text{Te}_3 \text{ flake, positioned inside the pickup coil. The square indicate the area we imaged at room temperature with atomic force microscopy (AFM). b) AFM measurement of the flake. The false colors indicate the height of the flake. The dashed rectangle indicates the little flake (Fig. 8.2). The black dot indicates the center of the flake (see Fig. 8.3). The circle at the bottom right indicates the diameter of the magnetic particle we use in our experiments.}\)
To compare, proton $^1\text{H}$ has a nuclear magnetic moment of 2.79 $\mu_N$. The gyromagnetic ratio of a nucleus is given by $\gamma = \mu / S$.

$^4$To compare, hydrogen $^1\text{H}$ has a nuclear magnetic moment of 2.79 $\mu_N$. The gyromagnetic ratio of a nucleus is given by $\gamma = \mu / S$.

two different types of topological insulators on two of our detection chips. One sample consists of a 300 nm thick flake of bismuth telluride (Bi$_2$Te$_3$), visible in Fig. 8.1. A second sample consists of a flake of stoichiometric BiSbTeSe$_2$ (BSTS).

Bi$_2$Te$_3$ consists of $^{209}\text{Bi}$ with a natural abundance of 100%, a nuclear spin $I = 9/2$ and a magnetic moment of $\mu = 4.11 \mu_N$, with $\mu_N$ the nuclear magneton$^4$. Tellurium consists mainly of isotopes with zero nuclear spin, except $^{125}\text{Te}$ with a natural abundance of 7% and a magnetic moment of $\mu = -0.89 \mu_N$.

The BSTS sample also contains selenium which consists of two different isotopes which are magnetic. In order to have as simple an experiment as possible, we have chosen the Bi$_2$Te$_3$ as choice for the first experiments. The stoichiometric BSTS sample is a sample that has not been measured extensively, since the group of Mark Golden are (one of) the first in the world able to make this sample.

The first goal is to obtain an NMR signal of the bismuth nuclei in a similar measurement as on the copper in Ch. 5. Unfortunately, the first attempts to obtain an NMR signal on the flake shown in Fig. 8.1 suffered from an experimental problem.

The interaction between the flake and the cantilever turns out to be very large, resulting in large frequency shifts. In Fig. 8.2a a room temperature conventional AFM measurement of the small flake of Fig. 8.1 is shown and in Fig. 8.2b a measurement with our MRFM setup. We measured the resonance frequency of the cantilever, while the magnet was positioned several $\mu$m above the surface. The measurement shows large shifts in the resonance frequency, even when the magnet has not fully approached the surface.

Above the big flake, the resonance frequency of the cantilever shifts hundreds of Hz. This kind of shifts causes hysteresis in images and frequency sweeps. The frequency shifts prevents us to drive the cantilever with a large amplitude, since already at amplitudes of 1 nm, a large non-linearity arises. Having to drive at such modest amplitudes results in large frequency noise. This makes it al-
most impossible to obtain a good enough signal to noise ratio to measure the small frequency shifts due to magnetic resonance, which we expect to be in the order of a millihertz.

The large frequency shifts we observe are possibly caused by electrostatic interactions. The first indication that this is the case is the large distance dependence between the surface of the magnetic particle and the surface of the flake. Magnetic interactions would cause frequency shifts with a distance dependence \( \propto (z + R_0)^n \) with \( z \) the distance between surface of the magnetic particle and \( R_0 \) the radius of the magnetic particle. The power \( n \) would depend on the details of the origin of the magnetic interaction. Instead, we observe close to the surface (Fig. 8.3) a distance dependence that goes like \( \propto z^n \), which is more in agreement with an electrostatic interaction than that of a magnetic one.

A second indication that the electrostatic interactions cause the problems in our measurements is that after touching the flake, the resonance frequency of the cantilever as function of distance changes dramatically. This agrees with changes in charges on the magnetic particle or flake, which may be expected to happen after a touch.

The frequency shifts as function of position and distance do not show a temperature dependence. Also this is in agreement with an electrostatic interaction.

For future experiments, the electrostatic interactions should be minimized in order to drive the cantilever with a larger amplitude, resulting in a lower force noise (Eq. 5.2). In order to achieve this, we recommend the following improvements for the experiment:

- The flake should be capped with a thin metallic layer in order to reduce the electrostatic interaction\(^5\).

- The use of smaller magnetic particles on the cantilever. The depth at which the measurement is sensitive is typically the same order of magnitude as the diameter of the magnetic particle. Since the thickness of the flake is 300 nm, a smaller magnetic particle can be used. Smaller magnetic particles will result in much larger signals\(^6\), since the field gradients are larger, and we ex-
pect that due to the coupling with less volume and surface of the sample, noise and electrostatic interactions are reduced.

- The possibility to apply a bias voltage on the cantilever. As shown by Moresi (2005), frequency shifts due to electrostatic interaction can be minimized by applying a bias voltage\(^7\).

8.3 **Proposal 2: The nature of the superconducting state at an oxide interface**

In the last couple of years, oxide interfaces have attracted a great deal of attention. The strong electron correlations in oxide interfaces leads to novel physical phenomena, promising novel technical applications\(^8\). Special attention in this field is for the interface between lanthanum aluminate (LaAlO\(_3\)) and strontium titanate (SrTiO\(_3\)). This interface, abbreviated as LAO/STO, has shown the existence of a superconducting state below a temperature of \(\approx 200\) mK\(^9\).

A superconducting two dimensional electron gas opens up the possibility of creating a topological superconductor\(^10\). Despite the experimental efforts in measuring the electron-electron interactions in order to reveal information about the superconducting state\(^11\), and the realization of a superconducting quantum interference device at the LAO/STO interface for phase sensitive measurements\(^12\), a microscopic understanding of the superconducting state is lacking\(^13\).

As we have discussed in Ch. 1, measuring the nuclear spin-lattice relaxation time can reveal information about the pairing symmetry of a superconducting state. However, as is the case of the topological insulator, conventional NMR is not sensitive enough for measurements when only the surface exhibits the interesting feature. A second difficulty in oxide interfaces is that probing the state with STM or other surface-sensitives techniques is near impossible because the electron gas in LAO/STO only exists when the STO is covered with at least four atomic layers of LAO.
We propose to use MRFM to probe the nuclear spin-lattice relaxation time of the interface of LAO/STO. Our technique operates in the ideal temperature regime for such a measurement, and we have shown that our technique is sensitive enough in principle. By measuring $T_1$ of the nuclei in the vicinity of the interface it may be possible to obtain information about the pairing symmetry of the superconducting state at the LAO/STO interface.

Of the two oxides, lanthanum aluminate is the most suitable candidate for magnetic resonance experiments, since both lanthanum and aluminium have a nuclear magnetic moment. If we compare the density of the material and the magnitudes of the magnetic moments with the experiment on copper, we calculate that the volume sensitivity$^{14}$ will be roughly 60% of that of copper, resulting in an expected volume sensitivity of $(35 \text{ nm})^3$. If we assume that the surface state extends 2 nm in the LAO, we find that we have a typical area sensitivity of $(150 \text{ nm})^2$.

We expect that the experiment we propose can reveal crucial information for resolving the microscopic details of the superconducting state of the interface between LAO and STO. Furthermore, the rich phase diagram and many physical phenomenon in oxide interfaces pose many more questions which MRFM might contribute to.

8.4 Proposal 3: Triplet superconductivity in triplet spin valves

Spin polarized supercurrents promise important applications in superconducting spintronics devices$^{15}$. One can engineer the interface of a superconductor (S) and a ferromagnet (F) such that singlet Cooper pairs are converted into triplet pairs.

As shown by Ishida et al. (1998), one can give evidence of triplet superconductivity by measuring the Knight shift as a function of temperature. We have shown that we can perform a similar measurement by measuring $T_1 T$ as function of temperature. Our MRFM can show that triplet superconductivity is present in nanoscale devices where the existence of triplet pairs is inferred from transport ex-
We propose to show the existence of triplet superconductivity in the triplet spin valves (TSV) fabricated by Singh et al. (2015). Their spin valve consists of a S/F/N/F junction, with N indicating a normal metal which is required to magnetically decouple the two ferromagnetic layers. Currently, copper is used for this layer. The thickness of the copper layer of the TSV is originally 5 nm, but there are no reasons why this layer could not be thicker, thereby increasing the signal strength of MRFM.

The superconducting top layer is MoGe, 25 – 50 nm thick. Since the out-of-plane penetration depth of thin film MoGe is more than several times the film thickness, this will not shield the magnetic field gradient of our force sensor and MRFM can still be detected from the Cu nuclei.

In Fig. 8.4 we sketch the idea of the measurement. The measurement on the TSV is expected to show a different behavior for \((T_1 T)^{-1}\) compared to an S/N/S device. This would further substantiate the evidence for a triplet Cooper pair generation in the S/F/Cu/F device.

8.5 Proposal 4: Finding evidence for superconductivity in an iridate

The superconductors with the highest critical temperature at ambient pressures are all within the copper oxide family. Despite many experimental efforts, physicists still did not unravel the microscopic mechanism behind the glue that pairs the electrons. However, it is
widely believed that the high-temperature superconductivity emerges from the doped Mott insulator state. \cite{Keimer2015}

Recent work of Battisti et al. (2017) shows that the doped Mott insulator (Sr$_{1-x}$La$_x$)$_2$IrO$_4$, an iridate, shares a similar phase diagram as the lightly doped cuprates. Together with other work\cite{delatorre2015;Kim2016}, this implies that at a slightly higher doping concentration, the material might become a high-temperature superconductor.

Despite that the measurements with spectroscopic imaging scanning tunneling microscopy (SI-STM) can give a lot of information about the electric structure, direct evidence for superconductivity is still lacking. Conventionally this can easily be done in a transport measurement with a Hall bar to measure the resistance of a piece of material as function of temperature. Unfortunately, the electronic state in (Sr$_{1-x}$La$_x$)$_2$IrO$_4$ is highly inhomogeneous, resulting in an isolating bulk behavior\cite{Battisti2017}. The interesting melting of the Mott insulating state happens only around the dopant atoms, giving small puddles with a typical size of 2 nm in diameter. SQUID magnetometer measurements give so far an inconclusive result as well\cite{personalcommunicationMilanAllan}.

We suggest to perform a very sensitive magnetic force experiments with our setup, without the need of resonance techniques. Chapter 7 has shown that our setup is very sensitive to paramagnetic effects of the sample under study. We expect that the diamagnetic effect of superconducting colloids can be measured and may give the final verdict for the existence of superconductivity in the doped Mott insulator (Sr$_{1-x}$La$_x$)$_2$IrO$_4$.

Below we first derive the expression for the frequency shift of our cantilever due to the diamagnetic response of the superconducting puddles/colloids. Secondly we calculate the expected frequency shift for typical expected parameters and flake size.

### 8.5.1 Diamagnetism measured with MFM

When a superconductor is placed within an external magnetic field $H$, a magnetization $M$ is induced according to:

$$\mathbf{M} = \chi V \mathbf{H}$$

\cite{DiamagnetismmeasuredwithMFM}
Here $\chi_V = -1$ for a bulk superconductor of type I, which means a perfect diamagnetic response. $H$ is the external magnetic field as if there was no magnetic material.

We expect the coherence length $\xi$ similar as in the cuprates\textsuperscript{23}, which means several nanometers. For the London (bulk) penetration depth we assume a typical value of $\lambda_L = 200$ nm.

When the average size of the superconducting domains $D$ becomes smaller than $\lambda_L$, the effective penetration depth becomes effectively larger\textsuperscript{24}. Prozorov and Giannetta (2006) give the effective susceptibility $\chi_V$ in the case of small spherical superconducting domains ($D < \lambda_L$) as:

$$\chi_V = -\frac{3}{2} \left( 1 - \frac{3\lambda_L}{D} \coth \left( \frac{D}{\lambda_L} \right) + \frac{3\lambda_L^2}{D^2} \right) \quad (8.2)$$

With $D$ the average radius of the spherical domains. If we evaluate this expression in the case $D \ll \lambda_L$, we obtain:

$$\chi_V = -\frac{1}{10} \frac{D^2}{\lambda_L^2} \quad (8.3)$$

The above equation shows that the shielding is poor compare with a bulk superconductor ($|\chi_V| \ll 1$). We can approximate the magnetic field for all colloids to be unaffected by the magnetization, since even inside the colloid the total magnetic field $B$ is given by:

$$B = \mu_0 (H + M) \approx \mu_0 H \quad (8.4)$$

If we assume that the dynamics of the diamagnetic response is much faster than the cantilever dynamics, which has a resonance frequency $f_0 = 3$ kHz, we can assume that the magnetization of the domain is always anti-parallel with the applied magnetic field, and also proportional to it. In that case we derive for the stiffness associated with the interaction:

$$E_V = -M \cdot B = \frac{1}{10\mu_0} \frac{D^2}{\lambda_L^2} B^2 \quad [\text{J/m}^3] \quad (8.5)$$

$$\Delta k_{x,V} = \frac{d^2 E_V}{dx^2} = \frac{1}{5\mu_0} \frac{D^2}{\lambda_L^2} \left( B \frac{d^2 B}{dx^2} + \left( \frac{dB}{dx} \right)^2 \right) \quad [\text{N/m}^4] \quad (8.6)$$
This result is almost in agreement with Eqs. 3.10 for a semi-classical spin in the limit \( T_1 = T_2 = 0 \) and \( T = 0 \). The difference is that the result has an additional factor \( B \), because in the case for a spin the magnetization is pointed towards \( B \), but the size of the magnetization is fixed. Furthermore there is an additional factor \(-1\) to account for the diamagnetism instead of paramagnetism. Also, there is no temperature dependence in our equation, but note that \( \lambda_L \) depends on temperature\(^{25}\).

With the stiffness of the interaction we can find the shift in resonance frequency per unit volume:

\[
\Delta f_V = \frac{f_0 \Delta k_x}{2 k_0} = \frac{f_0 D^2}{10 \mu_0 k_0 \lambda_L^2} \left( B \frac{d^2 B}{dx^2} + \left( \frac{dB}{dx} \right)^2 \right) \text{ [Hz/m}^3\text{]} \tag{8.7}
\]

### 8.5.2 Simulation

Using Eq. 8.7, we calculate in Fig. 8.5 the expected frequency shift when the cantilever approaches a sample consisting of the spherical superconducting domains as described above. The sample is 2 \( \mu m \) wide and deep and 1 \( \mu m \) thick. We have used the parameters described above and similar as in the experiments described throughout this thesis, i.e. \( D = 2 \text{ nm}, \lambda_L = 200 \text{ nm}, \mu_0 = 4\pi \cdot 10^{-7} \text{Tm/A}, f_0 = 3 \text{ kHz}, k_0 = 7 \cdot 10^{-5} \text{ N/m} \).

We assume that the flake fully consists of the superconducting colloids, but in reality the amount of superconducting will vary around 50 \%, based on the measurements of Battisti et al. (2017). Still, the signal is significant. We can determine the resonance frequency with < 1 mHz accuracy when sweeping the driving frequency (Fig. 2.6) or with a PLL (Fig. 5.2). We expect signals from the iridate that are several orders of magnitude larger. By scanning the sample in \( x,y \) and \( z \) one should be able to reconstruct an image and resolve the domains.

### 8.6 The monolithic MRFM

All proposed experiments above show that MRFM could be a promising tool for condensed-matter physics. Phys-
Figure 8.6: We have designed a new cantilever holder where the pickup coil (red) is placed close to the tip of the cantilever. In this way, we can detect the motion of the cantilever without the sample-positioning and lithography steps discussed in Ch. 2.

However, the details of the experimental setup in Ch. 2 show that MRFM is a difficult technique. One needs to combine sample and cantilever fabrication with a millikelvin experimental setup. This together with the possibility to move mechanically in three dimensions, while performing magnetic resonance experiments in conditions that are quite different from conventional NMR, due to the high field gradients (see Ch. 4). The experimental results need a thorough analysis as we have shown in Chs. 5 and 7. All together, our technique is not very attractive as a technique that can be widely used by research groups who are not specialized in the development of MRFM.

A good approach to lower the threshold for groups to start using MRFM, is to think about how we can integrate all components into a single device: The Monolithic MRFM. A device where the force detector, rf source, detection method and the three dimensional positioning system are all integrated, such that a sample can be easily loaded and unloaded in a setup without the need for extra fabrication and modification steps on top of the sample. This Monolithic MRFM could be designed and delivered by a commercial company, which can specialize and further improve the MRFM. A more friendly MRFM will allow research groups to focus on experimenting, for example one of the proposals above.

The results in Ch. 6 are an important step towards The Monolithic MRFM. One of the experimental challenges in conventional MRFM is to apply rf fields to the sample under study. This forces one to combine sample fabrication with that of an rf line. We show that for saturation magnetic resonance experiments, no additional rf source is needed, since the force sensor itself can be used as rf source.

At the side of the sample, we still have our detection method, the pickup coil. It is possible to replace our detection method with a laser interferometer based readout scheme, but as discussed before (Sec. 2.2), this would
heat up our cantilever and possibly excite our sample under study. Therefore, we have designed a new cantilever holder where the pickup coil is integrated\(^\text{26}\). The design, which is currently being developed, is sketched in Fig. 8.6.

One possible extension to the above design would be an additional superconducting wire to excite the higher modes of the cantilever, as discussed in Ch. 6. This wire could than also be used to generate rf fields at GHz frequencies for ESR experiments.

When we complete the proposed steps above, we have created a device with all ingredients are combined, such that a sample can be placed and approached from a single side. The Monolithic MRFM could be combined with a three dimensional nanopositioning system\(^\text{27}\), for an even easier implementation. The measures we have taken to reduce the vibrations caused by the pulse tube and dilution refrigerator (Sec. 2.1) are currently further developed and made available commercially by Leiden Spin Imaging, a spin-off company of our research group.

### 8.7 Conclusions

The experiments described here predicts MRFM to be promising for future experiments in condensed-matter physics. The measurements of the nuclear spin-lattice relaxation time as a function of temperature, down to 42 mK in Ch. 5, opens up the possibility for experiments in the field of superconductivity (Secs. 8.3 and 8.4) together with systems with an inhomogeneous electron state such as topological insulators (Sec. 8.2). The sensitivity of the developed magnetic probes can also be used to measure quantitatively the density of dangling electron bonds and their relaxation times (Ch. 7), and we argue that it could be used to measure the diamagnetic response of a possible new family of superconductors (Sec. 8.5).

The technical achievements described in Ch. 6 and Sec. 8.6 show us a route to develop a Monolithic MRFM, a device that can be more widely used in condensed-matter physics by research groups who do not have a focus on
the development of MRFM.

Altogether, we expect MRFM to become a powerful technique for experiments in the field of condensed-matter physics.