Application of Surface Coil for Nuclear Magnetic Resonance Studies of Semi-conducting Thin Films

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(Dated: October 19, 2017)

We conduct a comprehensive set of tests of performance of surface coils used for nuclear magnetic resonance (NMR) study of quasi 2-dimensional samples. We report $^{115}$In and $^{31}$P NMR measurements on InP, semi-conducting thin substrate samples. Surface coils of both zig-zag meander-line and concentric spiral geometries were used. We compare reception sensitivity and signal-to-noise ratio (SNR) of NMR signal obtained by using surface-type coils to that obtained by standard solenoid-type coils. As expected, we find that surface-type coils provide better sensitivity for NMR study of thin films samples. Moreover, we compare the reception sensitivity of different types of the surface coils. We identify the optimal geometry of the surface coils for a given application and/or direction of the applied magnetic field.

I. INTRODUCTION

The nuclear magnetic resonance (NMR) technique is a very powerful scientific tool, both in medical imaging and basic science. More precisely, NMR is a bulk microscopic probe of magnetism that can be used to effectively determine spatial variations of local magnetic properties in matter. Therefore, additionally to being a valuable tool for studying ordered magnetic states, it can be used to access real space features in spatially inhomogeneous states and those characterized by short range magnetic order. Furthermore, the element site-specific nature of our probe permits the separation of itinerant from local electronic properties. The applicability of NMR over a broad range of magnetic fields up to 45 T allows an unparalleled exploration of phase diagrams. However, the problem is that detection of the NMR signal typically requires an order of $10^{20}$ spins. For this reason, the application of NMR to study properties of nano-structures, 2D materials, and devices is precluded.

Inherently low sensitivity of NMR can be improved by enhancing the difference of Boltzmann population of the nuclear Zeeman energy levels by either lowering temperature and/or increasing the applied magnetic field. However, we often require knowledge of physical properties of matter as a function of temperature and/or applied field. Thus, sensitivity has to be improved by varying some other parameter. The signal-to-noise ratio (SNR) in an NMR experiment is also controlled by the filling fraction, which is the ratio of the volume of the effective radio frequency (RF) field to the sample volume. Consequently, by increasing the filling factor of the solenoid coil, the sensitivity can be improved, as illustrated in Fig. 1. Nonetheless, this strategy does not work for volume limited samples and/or 2D thin-films. In such cases the application of surface micro-coils can be beneficial. This is because the effective RF field strength rapidly decays away from the surface of the coil. Since the effective RF field is confined to a region adjacent to the coil, the filling factor, and thus SNR, is maximized for 2D-like samples or for probing sizable surface area to a limited depth. In this paper, we investigate the performance of surface coils for NMR studies of quasi 2D materials. We compare the reception sensitivity of different geometries of surface coils and identify one which is optimal for a given application. We emphasize that in addition to the gain in sensitivity for such samples the application of the surface coils offers ready access to the sample. This is important if other external parameters, such as bias gate voltages and/or applied strain are to be varied.

All surface coils produce an RF field whose magnitude decays away from the coil in the direction perpendicular to its plane. The inhomogeneity of the RF excitation field is one of the major challenges in the application of surface coils to imaging and high resolution NMR spectroscopy. However, for the following reasons, this is not an issue in the NMR study of fundamental properties of quantum materials. Often intrinsic magnetic and/or electronic inhomogeneities and textures far exceed the inhomogeneity of the excitation field generated by the surface coil. The penetration of the RF field is spatially varying due to electronic and superconducting currents shielding effect in metallic and superconducting samples, respectively. As far as applications to condensed matter physics examined here, the main advantage for using

![FIG. 1: Sketches of different type of coils typically used in an NMR experiment: a) solenoid, b) flat solenoid, and c) spiral surface coil. Flat gray boxes illustrate thin samples used in our study.](image-url)
a) b)

FIG. 2: Sketches of two different geometries of surface coils investigated here: a) meander-line and b) spiral. Mutual separation between the parallel conductors of the meander-line coil is denoted by \( a \), while \( r_0 \) denotes the opening radius of the spiral.

Surface coils is increased sensitivity for resonance studies of devices and quasi 2D materials, tuning of the probing depth of sizable surface area of the sample, and ready access to the sample.

The paper is organized as follows. We discuss basic principles and assumptions necessary for the intuitive understanding of the NMR performance of different surface coils in section II. In section III, we describe experimental details about our NMR set-up, coil fabrication, and the samples. Measurements of the NMR spectra acquired by separate surface coils and for the different orientations of the magnetic field are presented in section IV.

II. THEORY

The optimal choice of surface coil geometry relies on the specifics of a particular application. We will discuss the two geometries illustrated in Fig. 2. They differ in uniformity of the in-plane RF fields. These are meander-line and Archimedean spiral coils. As shown in Fig. 2, the meander-line coil is a serpentine array of parallel conductors of mutual separation \( a \). The Archimedean spiral is defined as \( r = s \theta \) in polar coordinates, where \( s \) is the spiral constant. Coils of spiral geometry induce a more uniform in-plane RF fields, and thus represent a more appropriate geometry for applications requiring uniform excitations.

The excitation field pattern of the surface coil is fully described by the spatial distribution of the magnetic field \( B_1 \) produced by unit current through the coil, and can be calculated from the Biot-Savart law [4]. For a given surface coil, excitation and reception patterns are identical [5]. Therefore by calculating the spatial distribution of the \( B_1 \) magnetic field, one can also obtain information about the reception sensitivity of the coil. In materials such as metals, where RF field penetration is spatially non-uniform, calculation of the spatial distribution of \( B_1 \) does not necessarily give precise reception sensitivity. Thus, to compare true reception sensitivity of different coils in our study we have independently optimized excitation pulses for each coil and applied field orientation.

The spatial distribution of the \( B_1 \) magnetic field has been calculated for both spiral [3] and meander-line [2, 9] coils. In what follows, we give a brief overview of the main results of these calculations. The distribution of the magnitude of \( B_1 \) calculated near a conductor of a meander-line coil, defining the \( x - y \) plane, in the near field region, for \( z/a \ll 1 \), is given by

\[
B_1(x, y, z) = \mu_0 I \frac{e^{-\pi z/a}}{2\pi z(1 + x^2/z^2)^{1/2}},
\]

for \(-a/2 \leq x \leq a/2\). Here \( z \) is the distance away from the coil plane, and the coil strips are considered as infinitely thin but of finite width. The magnitude of \( B_1 \) is the quantity of interest in NQR detection, while the individual components of \( B_1 \) perpendicular to the applied field define excitation/detection pattern in NMR. The important finding is that the RF magnetic field has the periodicity of the meander-line itself, in planes parallel to the surface of a meander-line coil. However, its strength at a distance \( z \) away from the coil is given by \( \exp(-\pi z/a) \). Therefore, the effective RF field is confined to a region adjacent to the coil and its penetration depth is determined only by the spacing \( a \) and not by the overall size of the coil. Since the signal-to-noise ratio in an NMR experiment is proportional to the filling fraction, the meander-line coils are ideally suited for probing thin 2D-like samples or sizable surface area to a limited depth [2, 5]. Consequently, by adjusting only the spacing \( a \), one can control SNR and sensitivity depth.

For spiral coils, the on-axis field decays more slowly at a distance \( z \) away from the coil plane than that for the meander-line coil. In the limit of spacing between conducting traces (\( \Delta \)) going to zero, on-axis field reduces to the expression for a single loop RF coil carrying a current \( I \) in the AC conductor limit [3, 9],

\[
B_1(0, z) = \frac{\mu_0 I}{2} \frac{r_0^2}{(r_0^2 + z^2)^{3/2}}.
\]

Evidently, the RF field and consequently SNR decreases with increasing axial distance from the coil’s center. Total \( B_1 \) field can be increased by adding turns to the spiral. Each additional turn strengthens total \( B_1 \) field by superposition. However, as additional turns add to the total resistance to the coil, there is an optimal number of turns for each coil configuration that maximizes SNR. For our designs we did not find decreasing reception sensitivity for the coils with up to 7 turns.

III. EXPERIMENTAL

A. NMR set-up

The measurements were done using a high homogeneity superconducting magnet with field strength of 7 T. Data was taken at room temperature. The NMR data was recorded using a state-of-the-art laboratory-made NMR spectrometer. The coils were mounted on a homemade broadband NMR probe, constructed based on the design described in [10]. Variable capacitors were used
to tune each coil to the desired resonance frequency and assure that circuit is matched to 50Ω. NMR absorption spectra were obtained from the Fourier transform of the spin-echo. We used a standard spin echo sequence \((\pi/2 - \tau - \pi)\), with pulses independently optimized for each coil and applied field orientation. This was done to assure that only reception sensitivity is compared in each case. Because the samples are highly inhomogeneous, only a single line was observed for \(^{115}\text{In}\) NMR. That is, we did not observe nine distinct quadrupolar satellite lines expected for \(^{115}\text{In}\) with nuclear spin \(I = 9/2\) in non-cubic local environment.

### B. Coil fabrication

We used CST Microwave Studio, a popular commercial tool for 3D EM simulation of microwave and RF frequency, to build 3D models of the various surface coils. This software generates the 3D field distribution using a discretized solution of the integral formulation of Maxwell’s equation. The calculated field distribution depends on the exact boundary conditions used. We used open boundary conditions for the spatial part and in time domain by a so-called “transient finite difference time domain” approach. These conditions produce results that are sufficiently accurate for MRI imaging applications. However, we used the software just as a rough guide for the field distribution and used our measurements to identify an ideal coil geometry for a particular application. More importantly, the CST Microwave Studio output file is used by CircuitCAM software to build the design for machining the coil. CircuitCAM outputs the file in .lmd format (common PCB format) which is then loaded to BoardMaster software to control CNC machine for physical coil fabrication. We used 300 \(\mu\text{m}\) thick PC board with Cu conducting layer of thickness of 25 \(\mu\text{m}\) to machine the coils of desired geometry. Coils were fabricated at the NMR facility, Division of Structural and Synthetic Biology Centre for Life Science Technologies, RIKEN Yokohama Campus, Japan, led by Prof. H. Maeda.

We also fabricated meander-line micro-coils with 100 nm spacing between the conductors using lithographic techniques. The gold conducting leads were wire bound and used to connect to the NMR tank circuit. We were able to send sufficient power to detect an NMR echo signal without burning the coil with the RF power. These type of coils can be very beneficial for the NMR studies of nano-devices.

### C. Test sample

For this study, we used 400 \(\mu\text{m}\) thick InP substrate as our test samples made at the IBM research center in the group of Dr. D. Sadana. The InP substrates are semi-insulating films doped with Fe, with room temperature resistance exceeding 1 MΩ. Such samples were chosen for the following reasons. Both \(^{115}\text{In}\) and \(^{31}\text{P}\) nuclei provide good NMR sensitivity. These substrates can be easily cut to the desired shape, allowing us to perform a comprehensive set of desired performance tests. The thickness of the substrates was selected to be comparable to the spacing between neighboring Cu conductors in the surface coil. For meander-line coils this assured that the effective RF field would penetrate the entire thickness of the sample. To eliminate artifacts associated with the skin-depth and induced surface currents, Fe doped insulating InP samples were taken.

### IV. RESULTS AND DISCUSSION

In a typical NMR experiment a sample is placed in a solenoid coil, as described above. Such coil provides very low filling fraction, and thus low SNR, for flat 2D-like samples. This can be easily resolved in part by using a flat solenoid, as depicted in Fig. 1b. As a matter of fact, we found that SNR can be improved by a a factor of two, as shown in Fig. 2. However, significant improvement of reception sensitivity by a factor of six can be achieved when a surface coil is employed as compared to the solenoid coil. In Fig. 3, we plot magnitude of the \(^{115}\text{In}\) NMR spectra for the same square sample acquired using three different coils, as denoted. The results clearly demonstrate the advantage of the surface coil. Here, we used the spiral surface coil, with the coil plane oriented parallel to the applied field and the sample covering significant area of the coil.

Our next step is to determine the optimal geometry of the surface coil for a given sample size and the appropriate variation of the external parameters, such as the applied magnetic field. This is particularly important when specific anisotropic and/or inhomogeneous quantities are to be investigated.

### A. Coil plane parallel to the applied magnetic field

In this section, we will examine the performance of different surface coils with the plane oriented parallel to the applied field. We first consider spiral coils. As discussed in Sec. II, the strongest effective RF field is induced at the center of the spiral and quickly decays as one moves away from the plane of the coil. When the applied field is oriented parallel to the surface of the coil, this component of the effective RF field is responsible for the spin flip since it is perpendicular to the applied field. This also implies that the most sensitive reception region is in the center cavity of the spiral coil. Therefore, for this applied field orientation, we expect that placing the sample in the center cavity of the spiral should provide the best reception sensitivity, and consequently SNR.

To test this hypothesis, we used the same spiral coil to obtain NMR signal from two samples of different sizes, as illustrated in Figs. 4a & b. One sample is such that it
FIG. 3: Comparison of the magnitude of the In signal, i.e. $^{115}$In spectra, recorded using three different coils (solenoid, spiral, and flat solenoid) illustrated in Fig. 1. Square sample of 3 mm $\times$ 3 mm was used. The applied field was oriented parallel to the plane of the spiral coil. Optimal excitation pulse conditions were determined separately for each coil. Relative ratios of the magnitudes are approximately 6:2:1.

FIG. 4: Sketch of the two samples of different sizes and their placement in the plane of the spiral surface coil: a) 3.5 $\times$ 3.5 mm in the cavity of the spiral coil and b) 13 $\times$ 13 mm covering most of the area of the surface coil. c) Comparison of the magnitude of the $^{115}$In signal per unit area from two samples, placed in the plane of the spiral surface coil as depicted in part a) and b). The normalized signal per unit area from the sample placed in the cavity of the spiral surface coil is approximately 3.7 times that of the sample covering the entire area of the coil.

B. Coil Plane perpendicular to the applied magnetic field

In what follows, we will examine the performance of surface coils when their plane is oriented perpendicular to the applied field. This is an important case, as this geometry is required in the study of physical phenomena such as vortices in superconductors and quantization of 2D electron gas. In the previous section, we established that placing the sample in the center cavity of the spiral coil provides the best reception sensitivity because the
strongest effective RF field is induced at the center of the spiral. However, for the applied field oriented perpendicular to the plane of the coil, this RF field, being aligned with the applied field, cannot induce spin flips. Therefore, no NMR signal can be detected in this geometry.

For this applied field orientation, the NMR signal is generated by the effective in-plane RF fields. We compare the NMR signal acquired on the same sample by three different coil geometries that produce such RF fields. In Fig. 6, we plot the magnitude of $^{115}$In NMR signals acquired by meander-line, square, and spiral surface coils, as denoted. In all three cases the sample covers a significant area of the coil. For this insulating sample, the signal acquired by the meander-line coil is the weakest. However, for studies of metallic samples with finite skin depth the use of a meander-line coil with a spacing between conducting lines that matches the skin depth provides optimal filling fraction, and thus sensitivity, as described in Sec. II. Another example of potentially effective use of meander-line coils includes NMR studies of vortices in superconductors. Here, the optimal sensitivity is achieved by a spacing between conducting lines that matches the superconducting penetration depth.

C. Different orientations of the applied magnetic field

To study intrinsic anisotropies, it is crucial to investigate sample properties as a function of the orientation of the applied magnetic field with all other parameters being fixed. It important to compare relative magnitude of the signals acquired with the plane of the coil oriented parallel and perpendicular to the applied field. Next, we present such comparison for three different coil designs described above.

Relative magnitude of the $^{115}$In signals obtained using meander-line, square, and spiral surface coils with the plane of the coil oriented perpendicular to the applied field, $H_0$. Sample of $3.5 \times 3.5$ mm area is used. Relative magnitude of signals scale as 6:5:2, for spiral, square, and meander-line coils respectively. We used coils with the following specifications: square coil of 10.1 mm in length, with the opening radius/length of $r_0 = 1.05$ mm, and 1060 $\mu$m conductor width; spiral coil of 14 mm in diameter, with the opening radius of the spiral of $r_0 = 1.1$ mm, and 580 $\mu$m conductor width; and, meander-line coil of 10.2 mm in length and 3.4 mm width, with spacing between conducting lines of $\approx 1$ mm, and 530 $\mu$m conductor width.
two field orientations is observed for signal acquired by a meander-line coil. That is, when the plane of meander-line coil is perpendicular to the applied field, a signal 1.8 times stronger is detected. On the other hand, the signal for a spiral coil varies by a factor of three for two different field orientations. Therefore, meander-line surface coils are best suited for the studies of intrinsic anisotropic properties as their use minimizes artifacts associated with the effective RF field anisotropy.

V. CONCLUSIONS

We investigate the performance of surface coils for NMR studies of quasi 2D materials. That is, we compared the reception sensitivity of surface micro-coils of spiral and meander-line geometries. The optimal geometry of the surface coil for a given application and the direction of the applied field is identified. In this study we were not concerned with the homogeneity of the effective RF field induced by the coil, since the phenomena in condensed matter systems often produce intrinsic inhomogeneity exceeding by far those associated by the RF field.

For the magnetic field applied parallel to the coil plane, the best sensitivity can be achieved by employing the spiral coil designed so that the entire sample can be placed in the center cavity of the coil. However, this geometry will yield no signal for the magnetic field applied perpendicular to the coil plane. In this case, the best sensitivity can be achieved by employing the spiral coil designed so that the sample covers a significant area of the coil.

Meander-line surface coils are best suited for the studies of intrinsic anisotropic properties, since their use minimizes artifacts associated with the effective RF field anisotropy. Furthermore, meander-line geometry is best suited for the study of samples in which RF penetration is spatially inhomogeneous, such as metals and superconductors. In this case, optimal sensitivity is obtained if the spacing between conducting lines matches the RF penetration depth. Finally, in addition to the gain in sensitivity for 2D-like samples, application of the surface coils offers ready access to the sample, which can be crucial in allowing in-situ variation of parameters, such as bias gate voltage.
Acknowledgments

We would like to thank Dr. Devendra K. Sadana, IBM - T. J. Watson Research Center, for providing InP thin substrate samples for our work, and Prof. Hideaki Maeda, RIKEN Yokohama Campus, for hosting Lu Lu and making coil fabrication facility available to us. We acknowledge guidance for software use from Dr. Yoshi-nori Yanagisawa and Dr. Masato Takahashi. This research was supported in part by the National Science Foundation under Grant No. DMR-1608760.

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