Development of Miniature DC SQUID Devices for the Detection of Single Atomic Spin-Flips


Abstract—We report progress toward a superconducting quantum interference device (SQUID)-based system capable of detecting a few atomic spin-flips. The scaling of the flux sensitivity with SQUID loop dimension of miniature niobium dc SQUID devices is examined and shown experimentally to vary as predicted. Our smallest device, with loop size \(3 \mu m \times 3 \mu m\), is capable of detecting 40 spins in a 1-Hz bandwidth. We address the task of depositing a sample, of nanoscale dimension, within the SQUID loop.

Index Terms—Detectors, magnetic resonance, nanotechnology, SQUIDs.

I. INTRODUCTION

MINIATURE, thin-film dc SQUID devices are capable of measuring, with extreme sensitivity, the magnetic properties of micron-size samples at low temperature [1]–[3]. There are two major issues which have to be considered in order to develop further such devices and extend their applicability to the study of samples of nanoscale dimension: 1) achieving the required sensitivity for the detection of a low number of spins and 2) facilitating the deposition of such a small sample within the loop of the SQUID.

A major consideration in achieving the required sensitivity in such a device is the scaling of the internal dimension of the SQUID loop. Several niobium dc SQUID devices, with varying loop dimension, have been designed and commercially fabricated by HYPRES.\(^1\) In an earlier publication [4], we outlined the measurement of the flux sensitivity for our device with smallest loop size \(3 \mu m \times 3 \mu m\). Here, we shall examine the variation of flux sensitivity with the scaling of the loop dimension. Our smallest SQUID should be capable of detecting a few spins in a 1-Hz bandwidth. We then discuss how the design of our devices has been adapted so that a scanning probe may be employed to deposit and image a sample population within the SQUID loop. Our interest in such devices is the detection of magnetic transitions in materials with small spin populations, particularly for the growing number of potential applications in, for example, metrology and quantum computation, which may benefit from such a probe of single, isolated surface-trapped atoms.

II. SENSITIVITY MEASUREMENTS: INTRODUCTION

Reducing the loop area of the SQUID, which reduces its inductance, can increase the energy sensitivity of the device to near-quantum-limit operation. Furthermore, a device incorporating a SQUID of small loop area has reduced sensitivity to external magnetic fields, making it an ideal probe of samples situated within the SQUID loop. In the thermal limit, the energy sensitivity of a SQUID, of capacitance \(C\) and inductance \(L\), operating at a temperature \(T\), is given by

\[ \varepsilon = 16kT(LC)^{1/2} \quad (1) \]

where \(k\) is the Boltzmann constant. The electronic spin sensitivity is given by

\[ S_e = \frac{\phi_{ns}}{2\pi \mu_B \mu_0} \quad (2) \]

in units of spins (of moment \(\mu_B\)) per \(\sqrt{Hz}\), where \(\phi_{ns}\) is the flux noise density, related to the energy sensitivity by \(\phi_{ns} = (2\pi \varepsilon L)^{1/2}\), and \(\alpha\) is the internal dimension of the hole in the SQUID washer.

III. SENSITIVITY MEASUREMENTS: RESULTS AND DISCUSSION

We have measured and made a comparison of the sensitivity of three devices. All incorporate dc SQUIDs with junction size \(3 \mu m \times 0.2 \mu m\), dielectric thickness 3 \(\mu m\), and junction capacitance of 10 fF. Each junction is resistively shunted (10 \(\Omega\)). The loop size, measured critical current density \(J_c\), and calculated loop inductance \(L\) of our devices are given in Table I. The devices were mounted in a helium cryostat, which was magnetically shielded using a double-walled \(\mu\)-metal shield to a level of 10 nT, while external field fluctuations were less than the expected flux sensitivity of the SQUIDs. A coil inside the shield was employed to permit the application of both a constant and time-dependent magnetic field.

<table>
<thead>
<tr>
<th>Loop size ((\mu m))</th>
<th>(J_c) (A/cm(^2))</th>
<th>(L) ((\mu H))</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 (\mu m \times 3 \mu m)</td>
<td>1050</td>
<td>5</td>
</tr>
<tr>
<td>10 (\mu m \times 10 \mu m)</td>
<td>1104</td>
<td>16</td>
</tr>
<tr>
<td>30 (\mu m \times 30 \mu m)</td>
<td>1251</td>
<td>47</td>
</tr>
</tbody>
</table>

\(^1\)HYPRES, Inc., 175 Clearbrook Rd., Elmsford, NY 10523 USA.
Measurement of the current–voltage ($I$–$V$) characteristics of each device has been carried out as a function of temperature and of the applied magnetic field. The zero-field data demonstrate hysteresis for $T < 6$ K. This suggests a value of the McCumber parameter $\beta_L$ of 3 at 4.2 K, compared to a design value of unity, presumably due to an excess capacitance over that due purely to the junctions. Measurement of the magnetic field dependence of the $I$–$V$ characteristics have been carried out at a chosen operating temperature of 8 K, for which no hysteresis was observed in the transport measurements of any of our SQUIDs. Typical results, showing the observed variation of critical current $I_c$ with applied magnetic flux $\phi_F$ is shown in Fig. 1. The data are for the SQUID of smallest loop size; the inset shows the extent of modulation of $I_c$ for the larger devices. The solid lines are a theoretical fit to the experimental points, assuming $I_c(\phi_F)$ adopts the form

\[ I_c(\phi_F) = \frac{2I_{c0}}{\alpha} \left[ \cos\left( \frac{\pi \phi_F}{\phi_0} \right) \right] \left[ \sin\left( \frac{\pi \phi_F}{\phi_0} \right) \right] \]  

where $I_{c0}$, the zero-field critical current, and $A_I/A_L$, the ratio of the SQUID junction effective area and loop area, are used as fitting parameters. We deduce from the fit a value $2I_{c0} = (180 \pm 10) \mu A$, in agreement with that predicted by the device manufacturer. The ratio $A_I/A_L$ is found to be of order 0.1, which also agrees with that expected.

An elementary measurement of the sensitivity of our smallest device has been made by modulating the applied field at low amplitude, such that the flux induced in the SQUID is much less than a flux quantum, and tuning the SQUID bias current to give a maximum in the voltage response at the modulation frequency. The measured peak voltage response of $2.0 \text{ mV/}\phi_0$ (at a bias of $100 \mu A$) is in accord with the calculated value of $R\delta I_c/\delta \phi_F$, obtained from Fig. 1. Noise measurements for this SQUID show a white noise floor level of $5 \times 10^{-7} \phi_0/\sqrt{Hz}$, which is limited by the head amplifier noise of $1 \text{ nV/}\sqrt{Hz}$ of our commercial SQUID electronics (Oxford Instruments dc SQUID Controller). This corresponds to a measured spin sensitivity [2] for our smallest device of 38.7 spins (of moment $\mu_B$)/$\sqrt{Hz}$. In the thermal noise limit [(1)], a spin sensitivity of $2.5 \text{ spins/}\sqrt{Hz}$ is anticipated at our chosen operating temperature of 8 K.

Evaluating $R\delta I_c/\delta \phi_F$ for the other devices yields a scaling of the flux noise density with SQUID loop dimension, as shown in Fig. 2, where we have taken the head amplifier noise as limiting the performance of each device. $\phi_{th}$ appears to adopt the expected linear variation with $\alpha$, except in the limit $\alpha \rightarrow 0$ where $R\delta I_c/\delta \phi_F$ approaches the calculated maximum device response, $R\delta I_c/\phi_0$. The scaling, with SQUID loop dimension, of the spin sensitivity of our devices is represented in Fig. 3. In addition to the experimental data, limited by amplifier noise, we illustrate the expected thermal-limited sensitivity (1) at 8 K and the quantum limit ($\varepsilon = \hbar/2$).

IV. PROBING THE SQUID SAMPLE SPACE

In order to utilize the potential spin sensitivity of our SQUID device, a sample must be located within the loop of the SQUID. Scanning probe techniques have evolved into a reliable method of imaging and manipulating surfaces with atomic resolution. Making use of a cryogenic UHV scanning tunnelling microscope (STM), we aim to study a number of potential techniques for the deposition of a sample within the loop of our devices.

Two devices, further to those discussed in the previous section, have been designed and commercially fabricated. Both have their underlying silicon substrate exposed within the SQUID loop, so that the substrate may be probed and utilized as a surface for sample deposition. Fig. 4 shows an optical photograph of a SQUID with a $100 \mu m \times 100 \mu m$ loop, through which a hole of depth $1 \mu m$ is fabricated, exposing the underlying substrate. Fig. 5 shows an atomic force microscope (AFM) topographic image of a SQUID with loop size $3.5 \mu m \times 3.5 \mu m$. Within the loop, a hole of depth $1 \mu m$
is fabricated; the sides of the hole slope in such a way as to expose a region of substrate of diameter 1 μm. Measurements of the sensitivity of this device demonstrate a spin sensitivity comparable with our smallest device studied in the previous section.

The devices are designed so that they can be incorporated into the sample stage of our STM, which consists of several electrical contacts, permitting in-situ transport measurements. Measurements of the I-V characteristics of the devices demonstrate that the SQUIDs can operate within the STM vacuum system. STM topographic images of the exposed region of substrate within the loop of our 100-μm SQUID show the underlying substrate to have a roughness of a few nanometers over a region of size 20 nm × 20 nm. We are currently in the process of cleaning the substrate by exposure to an Ar-Ion beam through the SQUID loop.

We are pursuing two approaches to using the STM in the deposition of sample material within the SQUID loop. The first is to use an STM to assess the sample population of a sub-mono-layer of adsorbed atoms deposited either by gas deposition or by electron beam evaporation. Such a sample may of course contain a very large number of spin centres. However, the SQUID devices should find several useful applications in the detection of magnetic transitions of macroscopic samples. We have used our device incorporating the SQUID of loop size 3.5 μm × 3.5 μm to detect the superconducting transition in a sample of lead, of thickness a few microns and diameter approximately 50 μm, deposited directly onto the SQUID through a pinhole of diameter 50 μm. While the spin sensitivity of our SQUID is far beyond that required for such a measurement, our devices may be useful as an accurate fixed-point thermometer, as we shall describe in a later publication.

To exploit the use of the SQUID devices for the detection of magnetic transitions in a low number of atoms, we must be capable of depositing materials of such a small dimension within the reasonably large area of the SQUID loop. With this aim we are exploring a second STM deposition technique by field emission from a bias-pulsed STM tip [5], [6]. Fig. 6 shows an example in which Au clusters of 5–10-nm dimension are deposited on a highly orientated pyrolytic graphite surface via field emission from a Au STM tip. Refinement of this technique, by control of the bias pulse and tip-sample distance, may allow us to deposit sub-nanometer clusters and single atoms of material. In addition, we are addressing the possibility of depositing sample material of many different species of atom, the material being pre-evaporated onto an STM tip both in macroscopic quantities

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Fig. 4. Photograph of a SQUID with loop dimension 100 μm, showing exposed underlying substrate (S), Josephson junctions (J), and shunt resistors (R).

Fig. 5. AFM image (20 μm × 20 μm) of a SQUID with loop dimension 3.5 μm, showing exposed substrate (S) and Josephson junctions (J).

Fig. 6. STM image (100 nm × 100 nm) showing clusters of Au on a surface of HOPG. Tunnelling current 1 nA; STM tip bias 20 mV. Each cluster is formed by bias-pulsing a Au STM tip at −2 V for 10 ms.
using an electron beam evaporator and at the level of a few atoms by field-induced desorption from a surface.

V. CONCLUSION

We have shown that the flux noise sensitivity of a miniature dc SQUID exhibits the expected linear scaling with the dimension of the SQUID loop. Our smallest device is found to have a spin sensitivity of about 40 spins in a 1-Hz bandwidth; we are currently improving upon the head amplifier of our commercial SQUID instrumentation to exploit the full sensitivity capability of this device of a few spins in a 1-Hz bandwidth in the thermal noise limit. Our devices have been designed so that the underlying Si substrate within the SQUID loop is exposed; we are addressing the task of depositing a sub-nanometer sample using an STM.

REFERENCES