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## Miniature dc SQUID devices for the detection of single atomic spin-flips

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### Abstract

We report progress towards a superconducting quantum interference device (SQUID) based system capable of detecting single atomic spin-flips. The scaling of the flux sensitivity with SQUID loop dimension of miniature Nb dc SQUIDs is examined and shown experimentally to vary as predicted. Our smallest device, with loop size  $3 \times 3 \mu\text{m}^2$ , is capable of detecting a few spins in a 1 Hz bandwidth. We address the task of depositing a sample, of nanoscale dimension, within the SQUID loop. © 2001 Published by Elsevier Science B.V.

*Keywords:* SQUID; Electronic spin; Scanned probe microscopy

### 1. Introduction

It is well known that thin film dc SQUIDs are capable of measuring, with extreme sensitivity, the magnetic properties of micron size samples at low temperature [1–3]. To develop further such devices and extend their applicability to the study of samples of nanoscale dimension, two main issues must be addressed. First, achieving the required sensitivity for the detection of a low number of spins and secondly, facilitating the deposition of such a small sample within the loop of the SQUID. A major issue affecting the ultimate performance is the way in which the sensitivity to spin detection scales with internal dimension of the SQUID loop.

To address this issue, several niobium dc SQUID devices with varying loop dimension have been designed and commercially fabricated by HY-PRES. Elsewhere, [4] we outlined the measurement of the flux sensitivity for a device with smallest loop size ( $3 \times 3 \mu\text{m}^2$ ). Here we shall examine the variation of flux sensitivity with a range of loop dimensions. 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.

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## 2. Sensitivity measurements

### 2.1. Introduction

Reducing the loop area of a planar 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

$$\varepsilon = 16k_B T(LC)^{1/2}. \quad (1)$$

The electronic spin sensitivity is given by

$$S_n = a \frac{\phi_{ns}}{2\pi\mu_B\mu_0}, \quad (2)$$

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

### 2.2. Results and discussion

We have measured and made a comparison of the sensitivity of three devices. All incorporate resistively shunted ( $10 \Omega$ ) SQUIDs with junction size  $3 \times 3 \mu\text{m}^2$ , dielectric thickness  $0.2 \mu\text{m}$  and capacitance of  $10 \text{ fF}$ . The loop size, critical current density  $J_c$  and inductance  $L$  for our devices are given in Table 1. The devices were mounted in a helium cryostat, which was magnetically shielded using a double walled  $\mu$ -metal shield to a level of  $10 \text{ nT}$ , while external field fluctuations were less than the expected flux sensitivity of the SQUIDs. A coil inside the shield was employed to permit the

Table 1  
Summary of SQUID characteristics;  $J_c$  measured,  $L = 1.25\mu_0 a$

Loop size ( $\mu\text{m}^2$ )	$J_c$ ( $\text{A cm}^{-2}$ )	$L$ (pH)
$3 \times 3$	1050	5
$10 \times 10$	1104	16
$30 \times 30$	1251	47

application of both a constant and time-dependent magnetic field.

Measurement of the  $I$ - $V$  characteristics of each device has been carried out as a function of the applied magnetic field, at a temperature of  $8 \text{ K}$  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,  $\phi_e$ , 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  $I_c(\phi_e)$  adopts the form

$$I_c(\phi_e) = \frac{2I_{c0} \left| \cos\left(\pi \frac{\phi_e}{\phi_0}\right) \right| \left| \sin\left(\pi \frac{A_1}{A_L} \frac{\phi_e}{\phi_0}\right) \right|}{\pi \frac{A_1}{A_L} \frac{\phi_e}{\phi_0}}, \quad (3)$$

where  $I_{c0}$  the zero-field critical current, and  $A_1/A_L$  the ratio of the SQUID junction effective area and loop area, are used as fitting parameters. We deduce from the fit a value  $2I_{c0} = 180 \pm 10 \mu\text{A}$ . The ratio  $A_1/A_L$  is found to be of order 0.1, in agreement with expectations.

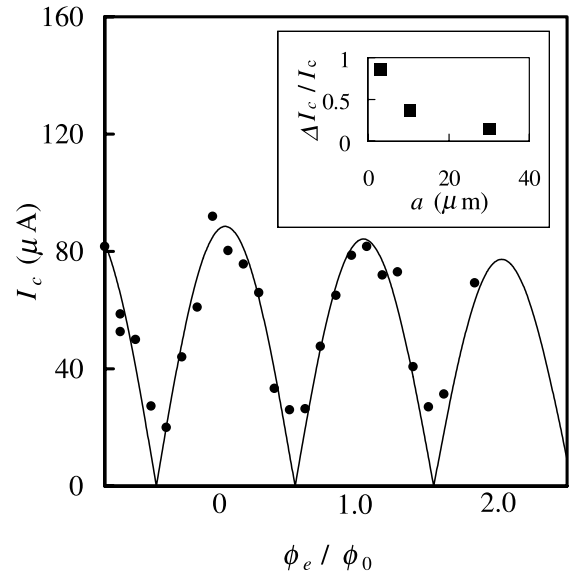


Fig. 1. Variation of  $I_c$  with applied magnetic flux,  $\phi_e$ , for the  $3 \times 3 \mu\text{m}^2$  SQUID. Inset: variation of critical current modulation,  $\Delta I_c/I_c$ , with loop dimension.

A simple 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  $100 \text{ }\mu\text{A}$ ) is in accord with the calculated value of  $R\partial I_c/\partial\phi_e$ , obtained from Fig. 1. Noise measurements for this SQUID show a white floor level of  $5 \times 10^{-7} \phi_0/\sqrt{\text{Hz}}$ , which is limited by the head amplifier noise of  $1 \text{ nV}/\sqrt{\text{Hz}}$  of our SQUID electronics. This corresponds to a measured spin sensitivity for our smallest device of  $38.7/\sqrt{\text{Hz}}$ . In the thermal noise limit (Eq. (1)), a spin sensitivity of  $2.5/\sqrt{\text{Hz}}$  is anticipated at  $8 \text{ K}$ .

Evaluating  $R\partial I_c/\partial\phi_e$  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.  $\phi_{\text{ns}}$  appears to adopt the expected linear variation with  $a$ , except in the limit  $a \rightarrow 0$  where  $R\partial I_c/\partial\phi_e$  approaches the calculated maximum device response,  $RI_c/\phi_0$ .

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 instrumental noise, we illustrate the ex-

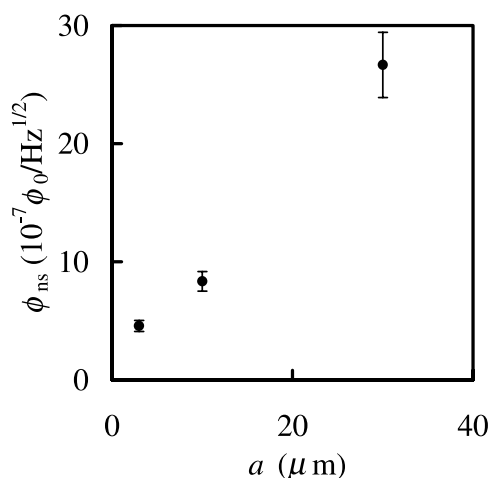


Fig. 2. Variation of measured flux noise density with SQUID loop dimension.

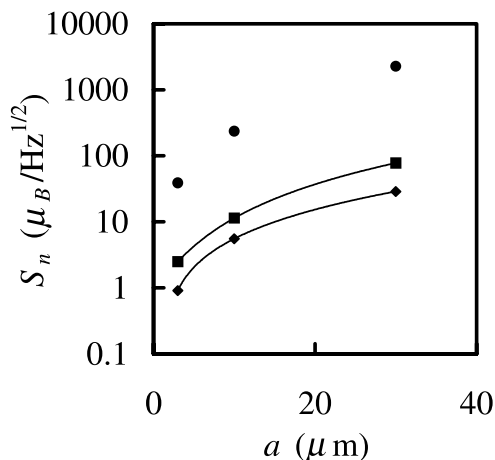


Fig. 3. Variation of spin sensitivity with SQUID loop dimension: (●) experimental data (instrumental noise limited), (■) expected spin sensitivity (thermal noise limit), (◆) quantum limit. The lines are a guide to the eye.

pected thermal-limited sensitivity (Eq. (1)) and the quantum limit ( $\varepsilon = \hbar/2$ ).

### 3. Probing the SQUID sample space

In order to utilise 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) and an ambient environment atomic force microscope (AFM) 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 utilised as a surface for sample deposition. Fig. 4 shows an AFM topographic image of a SQUID with loop size  $3.5 \times 3.5 \text{ }\mu\text{m}^2$ . Within the loop, a hole of depth  $1 \text{ }\mu\text{m}$  is fabricated; the sides of the hole slope in such a way as to expose a region of substrate of dimension  $1 \text{ }\mu\text{m}$ . Measurements of the sensitivity

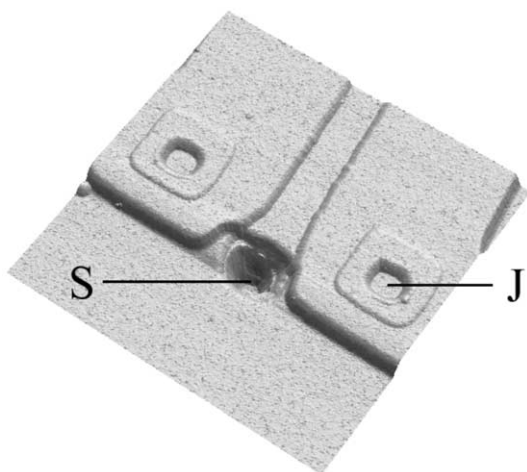


Fig. 4. AFM image ( $50 \times 50 \mu\text{m}^2$ ) of a SQUID with loop dimension  $3.5 \mu\text{m}$ , showing exposed substrate (S) and Josephson junctions (J).

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  $100 \mu\text{m}$  SQUID show the underlying substrate to have a roughness of a few nanometers over a region of dimension  $20 \text{ nm}$ . 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. To exploit the use of the SQUID devices for the detection of magnetic transitions in a small number of atoms, we must be capable of depositing materials of such a small dimension within the reasonably large area of the SQUID loop. The AFM has a demonstrated capability for translating small particles (e.g. carbon nanotubes) across a substrate. The weak van der Waals forces between tip and particle are sufficient to allow

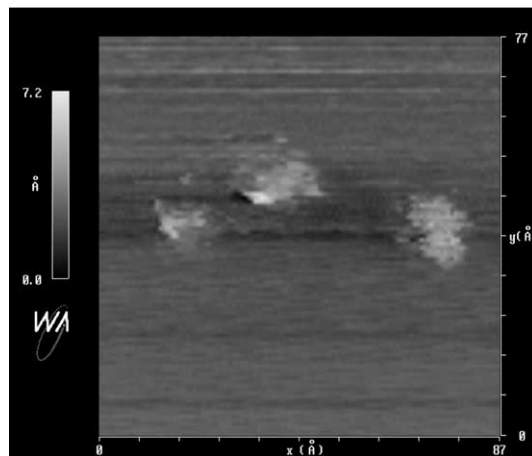


Fig. 5. STM image ( $8 \times 8 \text{ nm}^2$ ) showing clusters of Fe deposited on a surface of HOPG. Each cluster is formed by bias-pulsing a Fe STM tip.

movement across a flat smooth surface. However, release of the particle from the tip is problematic though possible. Consequently, we are also exploring a second deposition technique by field emission from a bias-pulsed STM tip [5,6]. Fig. 5 shows an example in which Fe clusters of  $5$ – $10 \text{ nm}$  dimension are deposited on a highly orientated pyrolytic graphite (HOPG) surface via field emission from a Fe STM tip. Refinement of this technique, by control of the bias pulse and tip-sample distance, may allow us to deposit sub-nanometre 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 a STM tip both in macroscopic quantities using an electron beam evaporator and at the level of a few atoms by field-induced desorption from a surface.

#### 4. Spin-flip detection

When we have successfully placed one or more magnetic nanoparticles within a micro-SQUID loop, it will be necessary to investigate methods for inducing spin flips within the nanoparticle while measuring the SQUID output. At present we envisage applying a static magnetic field within the plane of the SQUID, to minimise direct coupling

between static field and SQUID. RF pulses will then be applied at the Larmor frequency of the spin system to induce free induction decay. Given the broad band nature of SQUIDs (as high as GHz frequencies when used in the open-loop mode) real time observation of the decay should prove possible with sub-micrometre SQUID loops. Alternatively, if the relaxation time of the spin system proves too short compared with the SQUID recovery time we will investigate alternative detection methods such as adiabatic fast passage.

## 5. Conclusions

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 the 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 nanometre-sized sample using a STM or

AFM. The spin-detection system will be employed in exploring a number of applications in the areas of precision metrology [3], quantum information processing [7] and biomolecule investigations [8].

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