INSTITUTE OF PHYSICS PUBLISHING SUPERCONDUCTOR SCIENCE AND TECHNOLOGY
Supercond. Sci. Technol. **16** (2003) 1570–1574 PII: S0953-2048(03)66450-X

Measurement of the spatial sensitivity of miniature SQUIDs using magnetic-tipped STM

P Josephs-Franks[1], L Hao[1], A Tzalenchuk[1], J Davies[1], O Kazakova[1], J C Gallop[1], L Brown[1] and J C Macfarlane[1,2]

[1] National Physical Laboratory, Teddington, TW11 0LW, UK
[2] Department of Physics, University of Strathclyde, Glasgow, G4 0NG, UK

E-mail: Patrick.josephs-franks@npl.co.uk

Received 21 July 2003
Published 13 November 2003
Online at stacks.iop.org/SUST/16/1570

Abstract
Large area superconducting quantum interference devices (SQUIDs) are known to be ultimate sensors of magnetic flux. By diminishing the size of a SQUID loop, the energy sensitivity of the device can be increased to near quantum-limited operation whilst also making it less sensitive to external magnetic field noise and improving its coupling to any nano-scale magnetic particle within the loop. This makes it an ideal nano-sensor for magnetic particles including single molecules. It is, however, important to optimize the coupling between any magnetic nano-particle placed within the SQUID and the SQUID itself in order to achieve maximum sensitivity for the spin detection. We have made some preliminary calculations of the expected SQUID response to a magnetic particle placed at different locations within the SQUID device, and using a magnetic tip in a scanning tunnelling microscope (STM) we are able to produce a spatial map of the SQUID sensitivity. Initial results on a 3 μm diameter loop SQUID are presented to demonstrate this method.

1. Introduction

Whilst the behaviour of Josephson interferometers in a magnetic field, homogeneous on the scale comparable with the size of the device, is well understood, little is known in the case of a well-localized magnetic moment, when the mean-field approximation is invalid. The use of state-of-the-art SQUIDs for nanomagnetometry has been reported recently by Wernsdorfer *et al* [1]. In particular, they observed that sensitivity of a micro-SQUID to the state of a magnetic particle depends on the position of the latter within the SQUID loop [2]. Groups developing scanning SQUID microscopes have also reported that spatial resolution can be reduced to less than the SQUID loop size [3]. In principle, thin film dc SQUIDs are capable of measuring the magnetic properties of micron-sized samples at low temperature with extreme sensitivity, even to the level of a single electronic spin [4]. By reducing the loop area of the SQUID, which reduces its inductance, the energy sensitivity of the device can be increased to near quantum-limited operation. Furthermore, a device incorporating a SQUID of small loop area has reduced sensitivity to external magnetic fields but increased coupling to any nano-scale magnetic particle within the loop, making it an ideal magnetic nano-sensor. Previously, we have shown that loops of sub-micron sizes are required for single spin, quantum-limited sensitivity [5, 6]. To achieve this in practice, however, it is important to maximize the coupling between the magnetic nano-particle and the SQUID. However, as far as we are aware, no systematic study of the spatial sensitivity of a SQUID to a localized magnetic source has been carried out. In this work we suggest a new technique, which we call *SQUID self-portraiture*, to achieve this. In this technique a magnetic STM tip is scanned over the SQUID loop region whilst simultaneously recording the SQUID response. In this way we are able to produce a map of the SQUID response and so determine the optimum placing of any nano-particle within the device. We intend to extend this method to sub-micron sized SQUIDs so that simultaneous STM topography and SQUID response can be obtained, allowing a one-to-one mapping of sensitivity to SQUID structure.

2. Experiment

2.1. Miniature SQUIDs

Miniature SQUIDs were designed by us and produced using the HYPRES foundry service in the USA. This service uses optical lithography to produce a niobium-based multilayer system. The resolution is around 1 μm and this limits the smallest feature size to around 3 μm [5]. The SQUIDs were designed to have a 3 μm $\times$ 3 μm loop size, defined by a hole in a niobium layer, and Josephson junctions also of 3 μm $\times$ 3 μm. The junctions were shunted with resistors designed to have a resistance of 10 Ω each at 4.2 K in order to make the operation of the SQUIDs non-hysteretic. The miniature SQUIDs transition temperature, current–voltage and voltage–magnetic field relationship have all been measured. The noise power spectral density was also measured, and the flux white noise floor was found to be 4.3 $\times$ 10^{-7} Φ_0 $\mathrm{Hz}^{-1/2}$ [7]. An AFM topographic scan of one of the SQUIDs is shown in figure 1(*a*) along with a schematic of the SQUID in figure 1(*b*). The SQUID loop is shown as the dark area to the top of the figure. The device has a hole in the niobium film, extending down to the silicon substrate. The reason for this is that it gives us the possibility to image the area within the SQUID loop using our STM system and to also provide the possibility to manipulate a magnetic nano-particle within the SQUID loop using the STM. We will report this elsewhere. The two square-shaped features below the loop are the Josephson junctions. The whole device is around 50 μm $\times$ 50 μm in area and is deposited on a 5 mm $\times$ 5 mm silicon substrate along with four gold contact pads.

2.2. Modelling of the SQUID response

We have developed a simplified model to calculate the magnetic flux intersecting a circular area of radius a separated by an off-axis distance R from a single dipole magnetic moment m at the origin O and orientated in the z direction. For the z-component of the magnetic field at a distance x from the dipole, we obtained

$$B_z(x,\theta) = (3\cos^2\theta - 1)\frac{\mu_0 m}{4\pi\,|x|^3}$$

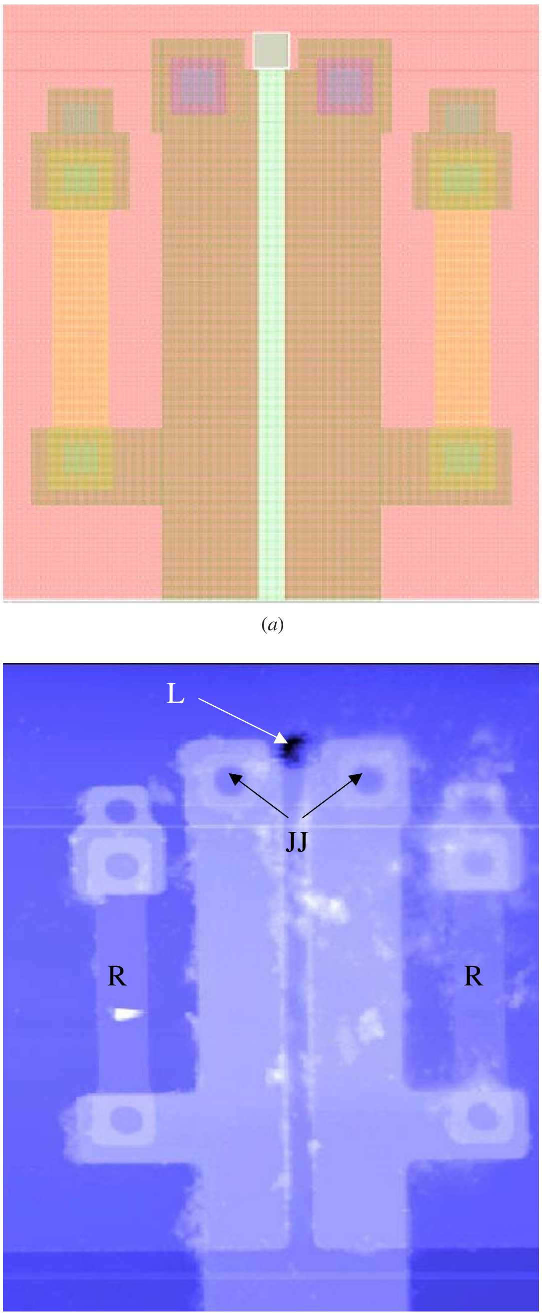
or expressing x and θ in polar coordinates

$$B_z(\varphi,\rho,r,R) = \left[\frac{3\frac{(R\cos(\rho))^2}{(R^2+r^2-2rR\sin(\rho)\cos(\varphi))} - 1}{(R^2+r^2-2rR\sin(\rho)\cos(\varphi))^{3/2}}\right]\frac{\mu_0 m}{4\pi},$$

where ϕ and ρ are defined in the inset of figure 2.

Integrating over the entire area of the loop we can obtain the dependence of the number of flux quanta in the z direction, which intersects the area of a given radius, on the distance to the dipole (figure 2). The graph shows that the flux penetrating the area scales approximately as R^3 at distances R are large compared to the loop size a, but saturates when $R \ll a$. These calculations suggest that for best sensitivity the magnetic tip should be located at a vertical distance comparable with the SQUID size and approaching the SQUID plane closer does not give a big advantage in sensitivity.

When the loop moves at a fixed vertical distance d to the magnetic dipole, which corresponds to a magnetic tip scanned across a SQUID, we should treat the angle ρ as a variable.



(*a*)

(*b*)

Figure 1. (*a*) Schematic of the SQUID layout. (*b*) AFM scan of a miniature SQUID. The scan area is approximately 66 $\times$ 66 $\mu\mathrm{m}^2$. The SQUID loop L is defined by the hole in the niobium film, which shows up as the dark feature at the top centre of the figure. The square-shaped features below and either side of the hole are the Josephson junctions (JJ). The symbol R denotes the gold shunting resistors.

(This figure is in colour only in the electronic version)

Figure 3 illustrates the result of calculation. Remarkably, the signal peaks, when the dipole is situated right under the loop perimeter and decays towards the centre of the loop. This simple model may probably explain the origin of the previously reported higher than expected spatial resolution of scanning SQUID microscopes—the resolution is mainly

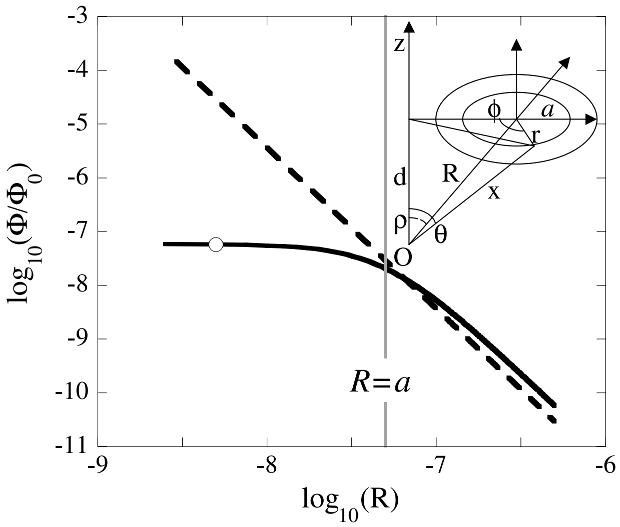


Figure 2. Calculated number of flux quanta in the z direction which intersects a circular area of radius $a = 50$ nm from a single dipole with magnetic moment $m = \mu_B = 9.3 \times 10^{-24}$ J T^{-1} situated below its centre ($\rho = 0$) as a function of vertical distance $R = d$. The dashed line corresponds to the far-field dipole approximation $\mu_0 m_B a^2/4R^3\Phi_0$. The circle marks the flux at the tip distance d of $a/10 = 5$ nm and the grey vertical line is the radius of the loop. The inset shows the geometry of the model used in the calculations.

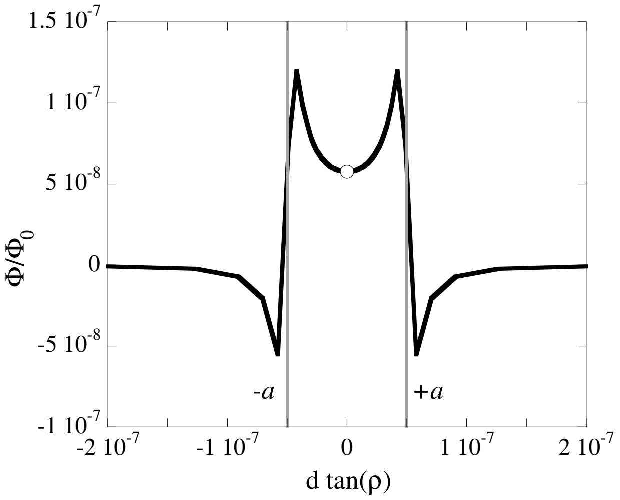


Figure 3. Number of flux quanta in the z direction which intersects a circular area of radius $a = 50$ nm from a single dipole with magnetic moment $m = \mu_B = 9.3 \times 10^{-24}$ J T^{-1} situated at a fixed vertical distance $d = a/10 = 5$ nm and variable horizontal distance $d \cdot \tan(\rho)$. The circle corresponds to the case of figure 2. The grey vertical lines mark the size of the loop.

determined by the sharp peak in Φ when the dipole crosses the loop. Conversely, in our experiments one would expect to see a very sharp contrast from the inner edge of the SQUID loop, a somewhat smaller and negative signal at the outer edge (from the return flux), and by far less details elsewhere. The model is also instructive in devising optimal conditions for the spin detection experiments. Perhaps counterintuitively, the magnetic particles or molecules should be positioned as close to the SQUID perimeter as possible, and not in the centre of the loop.

In the future, we shall expand the model to include finite penetration depth and the finite thickness of the superconductor, location and critical current of the Josephson junctions.

2.3. Mapping SQUIDs using a magnetic STM tip

The STM sample holder has four gold–platinum pads, which contact gold electrical spring-pins in the STM system, thus enabling four-point *I–V* measurements whilst the system is in the STM chamber. The SQUID device was wire bonded to our STM sample holder using gold wire. The bonded device was then placed into the variable temperature UHV STM system. This system typically operates at a pressure of 1×10^{-10} mbar. We are able to cool the STM to around 4.5 K using continuously flowing liquid helium supplied from an external dewar via a transfer tube. The consumption of liquid helium is around 2.5 l h^{-1} which means we can run for several hours at base temperature using a standard 60l dewar. Spring and eddy-current damping provide the vibration isolation, and the whole STM system floats on a piezo-electric driven active suspension system. We are able to routinely observe atomic resolution using this system at all temperatures from 4.5 K to 300 K. The STM was equipped, for these experiments, with an iron tip, made by simply cutting a 0.25 mm diameter iron wire. Naturally, such wire produces a large stray field originating from its whole body, rather than the end, which only weakly decays with the distance. We plan in the future to use coated tips, made from thin film coatings of nickel and cobalt onto etched tungsten tips, or, ideally, anti-ferromagnetic tips, where the stray field is compensated on the atomic level, while the very end of the tip remains magnetic. Although cut iron wire is definitely not the ideal material to use as a magnetic tip, for the purpose of testing the idea it was straightforward to install into our system.

The STM system also incorporates a piezo-electric driven *x–y* stage, so we are able to position the tip over a particular region of the sample with a precision of the order 50 μm. This resolution is limited by the optical resolution of the CCD device used to view the tip on approach to the sample. Thus we are able to bring the tip down very close to the SQUID device; but are unable to guarantee that the tip will be directly over the SQUID hole. Unfortunately, the HYPRES devices have an insulating layer of SiO over the SQUID area, which prevents us from obtaining STM topography on these devices. Using single- or bi-layer devices will remedy this. Since the scan range of the STM at low temperatures is around 1.5 μm, this means that, at best, we are able to scan over half of the SQUID loop. It is much more likely, however, that the tip will come down in an unknown position on the device. The tunnelling mode of the STM can help in accurate positioning of the tip. Using the iron tip we are able to obtain topographic resolution of the order 100 nm. Figure 4 shows an STM topographic image of one of the gold electrical contact pads of the SQUID device. Such lithographically defined gold features can be used as alignment marks for precise navigation to the SQUID loop.

Using the software that controls the STM we are able to record data from an analog-to-digital (ADC) channel whilst scanning the tip over the SQUID loop region. In this mode

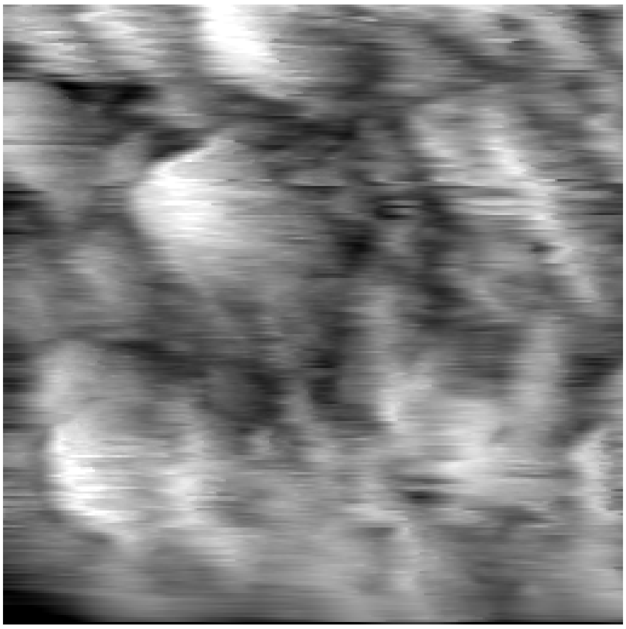
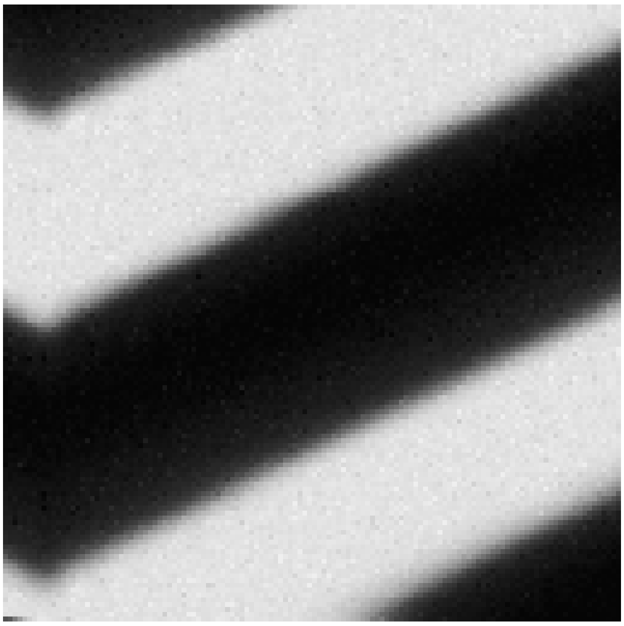
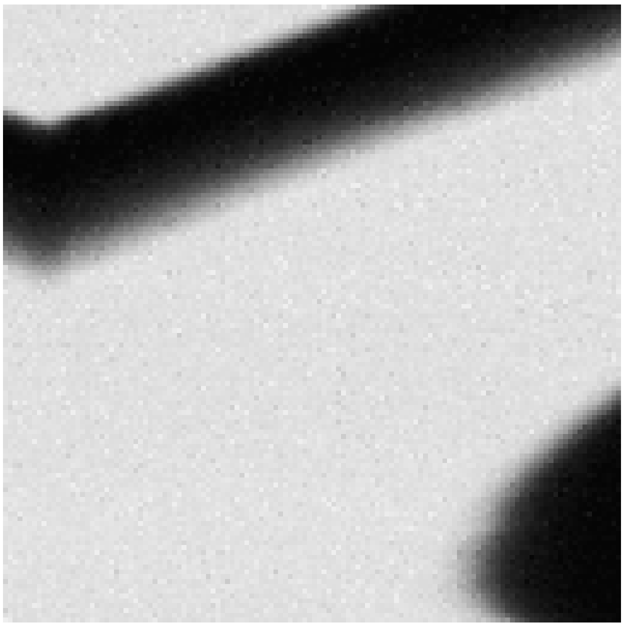


Figure 4. STM scan using an iron tip of one of the gold contact pads on the SQUID device. The STM conditions were, bias of 0.1 V, relative to the sample and tunnelling current of 0.1 nA. The image is approximately 1000 nm × 1000 nm with a height variation of approximately 50 nm.

of operation the role of the tip is merely to scan over the device at a close, but unknown distance, from the surface of the device. However, we are able to record the SQUID response as the magnetic tip is traversed over the SQUID. We call this method *SQUID self-portraiture*. The SQUID was driven from a constant current source, which supplied a dc current just sufficient to put the SQUID into the linear mode of operation. This was checked by moving the STM tip in the vertical direction and monitoring the voltage response of the device. The SQUID is operated in an open-loop small-signal configuration. The *I–V* characteristics of the SQUIDs showed no evidence of additional noise rounding due to operation in the STM chamber compared to preliminary measurements in a well-shielded cryostat. At a fixed tip position, we were able to vary the flux through the SQUID by several flux quanta by changing the height of the tip above the device. We observed the standard sinusoidal response of the SQUID. During the scanning of the tip over the SQUID we made sure that the SQUID response did not vary by more than a single flux quanta over the scan image, by adjusting the tip height, as this would cause a sudden change in the recorded voltage, which would produce a false edge on the sensitivity plot. Figures 5(*a*) and (*b*) show a typical response of the SQUID as the tip is scanned over the device. The brightness in the images is proportional to the flux in the SQUID and the full range corresponds to approximately one-half flux quantum. By relating these images to the AFM topographic images, we are able to tentatively assign figure 5(*a*) to a region close to one of the Josephson junctions and figure 5(*b*) to an area showing the edge of the SQUID loop, based on the relative sizes and angles of the observed features. To verify this technique we replaced the iron tip with a standard electrochemically etched tungsten tip. The SQUID then showed no change of voltage output as the tip was scanned. Also the images were reproducible when the tip was scanned over the same region but using different scan directions. Some additional features in the images were noted that we could not immediately associate with structural features of the device and may be due to magnetic imaging effects. This is under investigation.



(*a*)



(*b*)

Figure 5. Response of the SQUID as an iron tip is scanned over the SQUID region. The scan range is approximately 1×1 μm, limited by the STM piezo. We believe this to show the region near one of the Josephson junctions (*a*) and near to the SQUID loop (*b*). The SQUID is operated in a constant current mode, at slightly above the critical current during the scan. The intensity is proportional to the SQUID output voltage and corresponds to a flux change of the order of one-half flux quanta.

3. Conclusion and future work

Using this new 'self-portraiture' technique we will extend our studies to even smaller SQUIDs, of the order 100 nm loop sizes. This will enable us to scan the whole of the device in one image, thus mapping out the SQUID response over the entire device. Also by examining devices produced using a single layer of niobium will enable us to obtain simultaneous STM topographic images along with the SQUID self-portraiture. This will enable a one-to-one correlation to be made between device structure and sensitivity and comparisons made with the predicted behaviour. We shall also study the effects of using both ferromagnetic and anti-ferromagnetic tips. In particular, anti-ferromagnetic tips should have a more localized effect and give a higher resolution. In conclusion, we have demonstrated a new technique using a magnetically tipped STM system combined with open-loop operation of a SQUID to enable a map of the SQUID response to be produced. This is important in order to be able to optimize the coupling between any magnetic nano-particle and the SQUID so as to achieve maximum sensitivity of spin detection. The SQUID system may then be used in a number of applications requiring operation towards the single spin limit such as precision metrology [5], quantum information processing [8] and biomolecule investigations [9].

Acknowledgment

This work is supported by the UK Department of Trade and Industry under the NMS Quantum Metrology Programme, project FQM132.

References

[1] Wernsdorfer W, Hasselbach K, Mailly D, Barbara B, Benoit A, Thomas L and Suran G 1995 *J. Magn. Magn. Mater.* **145** 33
[2] Wernsdorfer W 1996 *Ph.D. Thesis* Joseph Fourier University Grenoble
[3] Kirtley J R, Ketchen M B, Stawiasz K G, Sun J Z, Gallagher W J, Blanton S H and Wind S J 1995 *Appl. Phys. Lett.* **66** 1138
[4] Gallop J C and Radcliffe W J 1985 *IEEE Trans. Magn.* **21** 602–5
[5] Josephs-Franks P, Reed R R and Pakes C I 2000 *Physica* B **280** 540–1
[6] Gallop J C, Josephs-Franks P, Davies J, Hao L and Macfarlane J 2002 *Physica* C **368** 109–13
[7] Hao L, Macfarlane J C, Josephs-Franks P and Gallop J C 2003 Inductive superconducting transition-edge photon and particle detector *IEEE Trans. Appl. Supercond.* **16**
[8] Tejada J *et al* 2001 *Nanotechnology* **12** 1–6
[9] Tejada J *et al* 1997 *Phys. Rev. Lett.* **79** 1754–7