Detection of single magnetic nanobead with a nano-superconducting quantum interference device

L. Hao,[1,a)] C. Aßmann,[2] J. C. Gallop,[1] D. Cox,[1] F. Ruede,[2] O. Kazakova,[1] P. Josephs-Franks,[1] D. Drung,[2] and Th. Schurig[2]

[1]National Physical Laboratory, Hampton Road, Teddington TW11 0LW, United Kingdom

[2]Physikalisch-Technische Bundesanstalt, Abbestrasse 2-12, D-10587 Berlin, Germany

We report the use of an ultralow noise nano-superconducting quantum interference device nanoSQUID_ to measure the hysteretic magnetization behavior of a single FePt nanobead at a temperature of around 7 K in a magnetic field of only 10 mT. We also show that the nanobead can be accurately positioned with respect to the SQUID loop and then removed without affecting SQUID performance. This system is capable of further development with wide applications in nanomagnetism.

Detection of ever-smaller magnetic-particles is of crucial technological and scientific importance, driven both by the needs of the information technology IT and telecom communities but also by medical and biological requirements, in addition to improved understanding of the physics of small numbers of coupled spins. Superconducting quantum interference devices SQUIDs, the most sensitive detectors for a wide range of physical parameters, are also exquisitely suitable for the measurements on magnetic nanoparticles.[1-4] We have proposed that the sensitivity of a sufficiently small SQUID should be adequate to detect the reversal of a single Bohr magneton moment.[5] Here, we describe recent progress toward this grand challenge.

Our nano-SQUIDs use microbridge junctions formed from a bilayer of Nb and amorphous tungsten. Using a focused ion beam the bilayer is narrowed into a micro/nanobridge 65 nm wide and 60–80 nm long and the thickness reduced to 70 nm. Nano-SQUID loop sizes range from 250 to 800 nm. For more fabrication details see Refs. 6 and 7. These SQUIDs operate over a limited temperature range from 4.2 up to _9 K. The mean operating temperature may be tuned by adjusting the Nb film thickness.

Using a SQUID series array SSA preamplifier in two-stage configuration,[8] we have shown these devices exhibit a white flux noise spectral density as low as 2_10^{-7}_$_0$ /Hz$^{1/2}$ above 1 kHz, one of the lowest noise figures achieved for SQUIDs operating above 1 K.[9] An idea key to this work is that SQUID sensitivity to the magnetization of a smaller nanoscale particle increases as the loop size decreases toward the dimensions of the particle itself. This introduces a complication. Although SQUID microsusceptometers, which have a separate pick-up coil sensitively measure the field from a micron-scale magnetic sample[10,11] it is not currently feasible to reduce the coil size below a few microns, leading to suboptimal coupling between them. To circumvent this, we couple a particle directly to the SQUID loop, eliminating the need for an intermediate coil system, allowing optimal coupling to submicron particles to be approached. However, in this case, we must both magnetize the particle with an external field and also tune the operating flux applied to the SQUID loop. Solutions to this problem are outlined elsewhere,[12] and here, we deal only with direct measurement of a submicron particle coupled to a microbridge SQUID loop.

For a uniformly magnetized spherical particle the external magnetic field is that of its total dipole moment concentrated at the sphere's center. Integration of this dipole field over the area of the SQUID loop yields the flux coupled into the SQUID. Simulations show that coupling strongly depends on the height of the sphere above the SQUID and that the flux signal falls rapidly as the dipole is moved outside the loop.

A SQUID can only sense a magnetic particle if the direction or magnitude of the particle's magnetization changes the flux coupled to it. The simplest detection method involves changing magnitude or direction of an applied polarizing field. Any variation in the magnetic moment component perpendicular to the SQUID loop will change the SQUID readout voltage. Note that the polarizing field also contributes to the flux coupled to the SQUID. This contribution may be calibrated out, as shown below, if the system is measured before and after particle attachment. FePt beads with 40:60 Fe:Pt composition diameter d70 to 200 nm_ were fabricated from a water-based solution of FePt nanoparticles with average diameter 3.8 nm. Transmission electron microscopy TEM analysis shows the beads consist of a large number of close-packed FePt nanoparticles in an fcc chemically disordered state_, with separation of 0.5 nm. Magnetization measurements on a large ensemble of FePt beads demonstrated their superparamagnetic properties at room temperature. At low temperatures, T10 K FePt beads are characterized by an open hysteresis loop and well pronounced remanent magnetization. The magnetic moment of the 150 nm sphere is _1_10$_7$_B in a field of B=3 T, estimated by SQUID magnetometry and x-ray magnetic circular dichroism measurements.[13]

We describe next, a reversible and accurate particle positioning method. First a single FePt nanobead is identified in a scanning electron microscope SEM and a sharpened probe carbon fiber_ is brought close to it using a micromanipulator. A brief exposure of the tip-nanobead contact region deposits sufficient amorphous carbon to "weld" the tip to the nanobead which may then be lifted and positioned close to the SQUID loop see Fig. 1. Once in position another burst from the electron beam, with a slightly longer duration, is used to attach the nanobead to the loop edge. The tip can now be withdrawn, leaving the bead fixed close to the SQUID. Once measurement is complete, the nanobead can be removed, using the same methodology in reverse, leaving the SQUID loop in its original condition.

First the nano-SQUID without nanobead was connected to the input of the SSA and both installed in a cryogenic probe maintained at 4.2 K while the nano-SQUID's temperature may be adjusted anywhere between 5 and 12 K using an auxiliary heater see Fig. 2. The nano-SQUIDs characteristics were extensively tested using this SSA setup, measuring noise, critical current versus temperature, voltage versus flux, etc. Essential for this work was the measurement of the small signal nano-SQUID response to an external magnetic field, applied perpendicular to the SQUID loop see Fig. 3a. Note the response to an increasing magnetic field is periodic with constant period corresponding to addition of one flux quantum to the loop. On decreasing the field the nano-SQUID voltage closely follows the up-sweep trace, with no detectable hysteresis _such as would arise from flux trapping or any irreversible movement of flux_. Due to its small size the nano-SQUID cannot at present be operated in flux-locked mode so we use it in periodic response mode. Next, the nano-SQUID with single nanobead _d_150 nm_ attached was measured. Again an external magnetic field, applied by the coil shown in Fig. 2_b_, is ramped up from zero to some maximum value then back to zero, reversed to the same negative maximum and returned to zero. This process was repeated for successively larger field sweep amplitudes, up to a maximum of 10 mT. The presence of the nanobead magnetization contributes to the flux at the SQUID. If the magnetization exhibits hysteretic behavior then there will be a shift between the up-sweep and down-sweep periodic responses and this is just what is shown in the Fig. 3_b_.

The flux signal at the SQUID arising from the nanobead may now be measured by subtracting the empty SQUID response _see Fig. 3_a__ from that with the nanobead present _see Fig. 3_b__. Although in principle it is possible to calculate the total magnetic flux at the SQUID for any small signal output of the SSA _within only an uncertainty modulo the integer flux quantum number_, in practice this is difficult as the SQUID current-voltage characteristic does not exactly follow the analytical response predicted by the resistively shunted junction model. However, turning points in the periodic SSA response to external applied flux accurately correspond to successive half-integer flux quanta at the SQUID. We plot the external applied flux value for each turning point of the SSA response for both up and down-sweep directions to allow the magnetization loop of the particle to be calculated. We have done this for a range of different applied field sweeps to derive a set of single nanobead magnetization loops _shown in Fig. 4_a__ with increasing magnetic field sweep values up to 10 mT. The SQUID integer flux quantum scale origin is arbitrary so each magnetization loop has been centered on the same origin. Note that even for the lowest field sweep, to 3.2 mT, the nanobead shows a remanent moment, suggesting that at 7.8 K it is already below its blocking temperature T_B. Following these measurements the nanobead was removed from the SQUID, which was then remeasured. The flux shift had now disappeared, proving it arose from the presence of a net magnetic moment of the nanobead. Using the known geometry of the nano-SQUID and nanobead in Fig. 1 and the estimated Pearl penetration depth for Nb at this temperature,[14] one can estimate the nanobead dipole moment of _10^6_$_B$ which produces the observed flux signal at the nano-SQUID. To compare this estimate with different experimental results for FePt nanobeads we have measured the low temperature magnetization loops for a large ensemble of these FePt beads at temperatures from 2 to 30 K and in fields up to 4.5 T by SQUID magnetometry _see Fig. 4_b__. The sample consists of a variety of FePt beads ranging from 70 to 200 nm. The inset indicates the coercive field as a function of temperature. It is interesting to note that the coercive field of the sample characterized by the inhomogeneous distribution of bead sizes is around _120 mT at T=8 K, much higher than the value of less than 1 mT _although this is a minor hysteresis loop_ observed for a single 150 nm sized bead in our nano-SQUIDs setup. This indicates the effect of the bead size as well as importance of interbead coupling in large ensembles of magnetic beads.

In summary, we have developed an ultralow noise nano- SQUID detector capable of measuring the hysteretic magnetization behavior of a single FePt nanobead at T_8 K in a magnetic field of only _10 mT. Our manipulation method shows that any nanoscale particle can be accurately positioned with respect to the SQUID loop and then removed without affecting the SQUID properties and performance. We next plan to implement movable nanobead positioning, with the nanobead mounted on a thin cantilever which may be moved relative to the SQUID. An alternative technique has already been demonstrated, where a micrometre scale pick-up loop coupled to a small SQUID, is scanned across a cryogenic sample.[15] This has a resolution of _10 _m. Combining a direct-coupled nano-SQUIDs with a scanning stage should provide even smaller spatial resolution. These future developments should have wide applications in nanomagnetism.

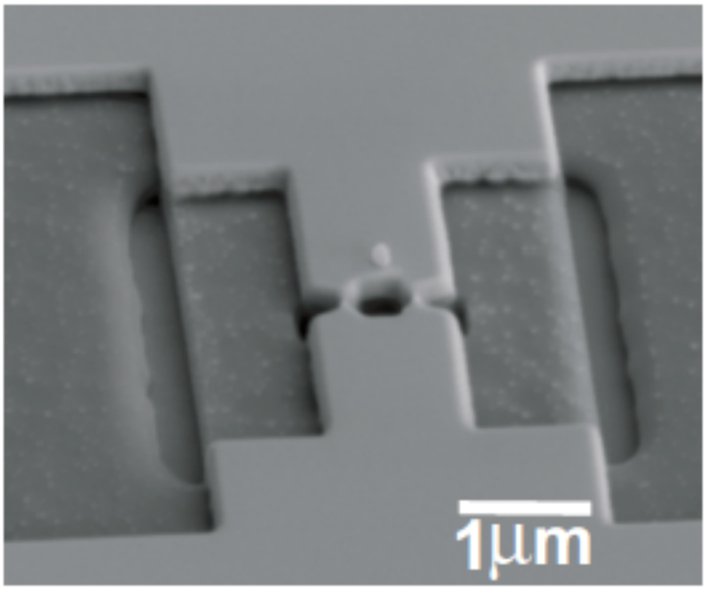

FIG. 1. SEM image of a single FePt particle bead _d_150 nm_ positioned at the Nb nano-SQUID perimeter. In the experimental setup the applied dc magnetic field B perpendicular to the SQUID loop _geometric area is _400 nm2_, whereas the effective area from measured flux period is _700 nm2. Thus, the penetration depth appears _150 nm but this ignores flux focusing/defocusing. SSA chip Coil

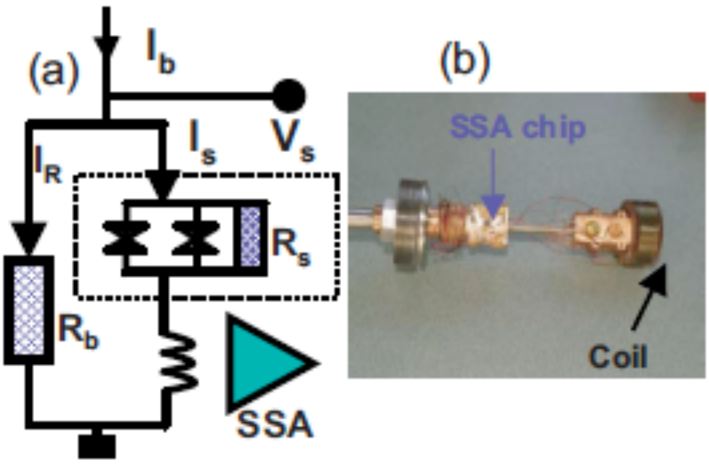

FIG. 2. Schematic circuit diagram showing nano-SQUIDs _in the dotted line box_ and SSA as preamplifier in two-stage configuration, where R_b__0.5 __ is the bias resistance. _b_ Cold stage of the measurement probe with a SSA chip, superconducting coil and nano-SQUID chip _located inside the coil_.

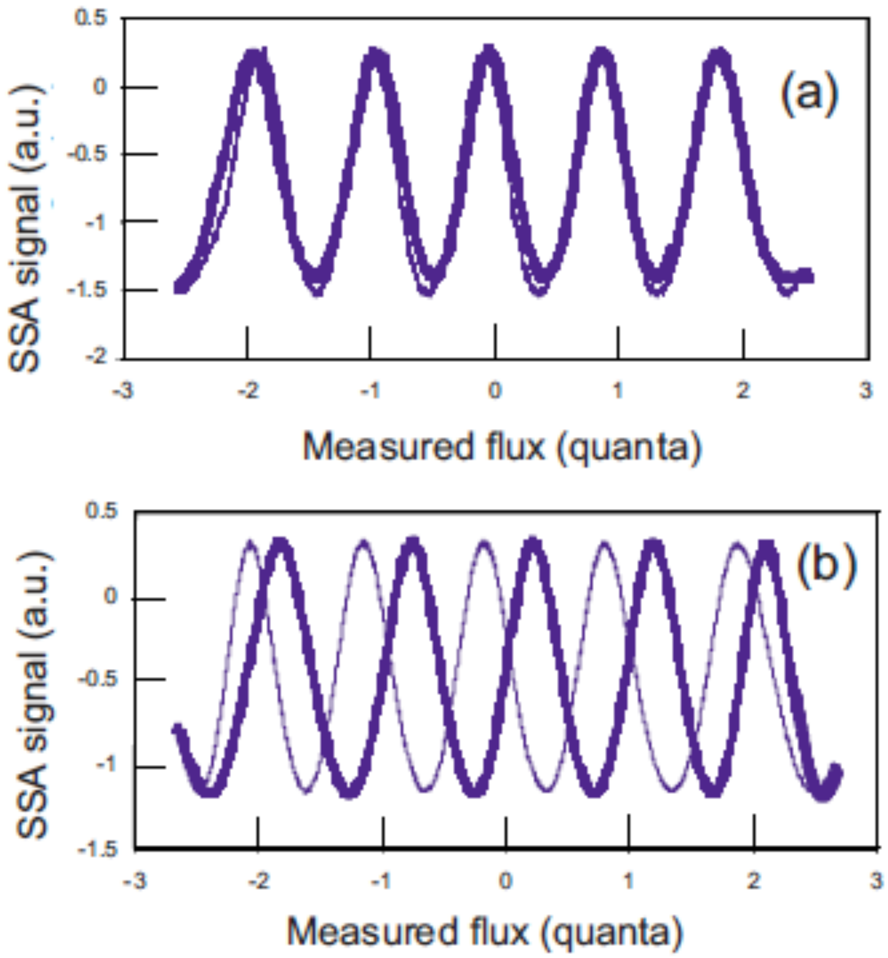

FIG. 3. _Color online_ _a_ SQUID output at 7.8 K with no particle present _the thin line is field sweep up, thick line is field sweep down_. _b_ SQUID output with a single FePt nanobead present at same temperature showing hysteresis.

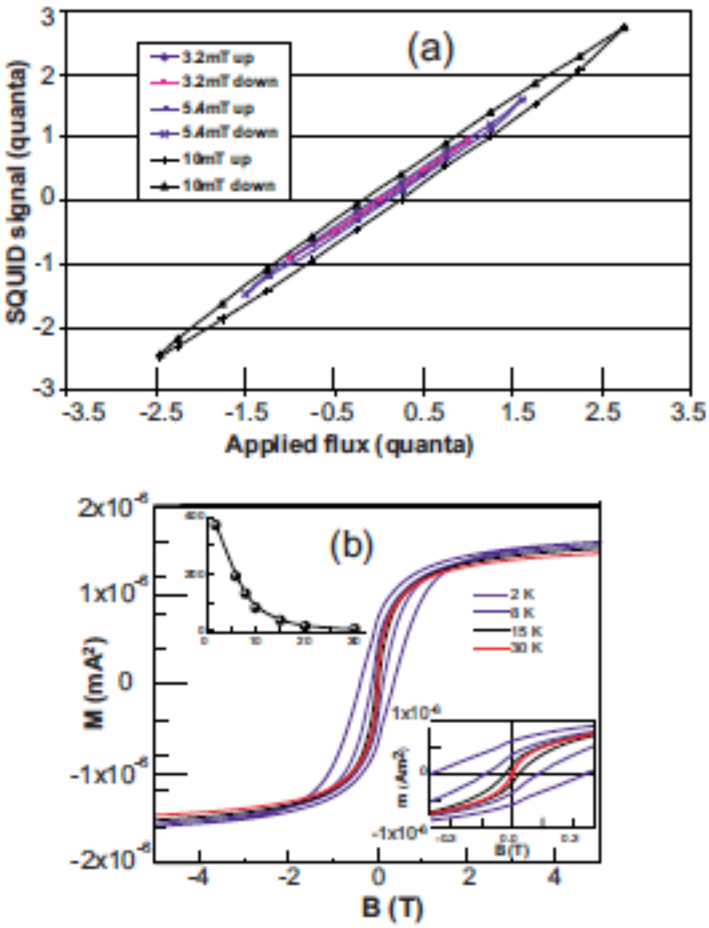

FIG. 4. _a_ Hysteresis plots for the single FePt nanobead for a range of applied magnetic fields. _b_ Magnetization measurements of a large ensemble of FePt beads of various sizes at different temperatures, indicating the blocking temperature, T_b, at about 10 K. 092504-3 Hao et al. Appl. Phys. Lett. 98, 092504 _2011_

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