$YBa_2Cu_3O_7$ and Nb NanoSQUIDs for the Investigation of Magnetization Reversal of Individual Magnetic Nanoparticles

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***Abstract*—We report on the fabrication, performance and application of sensitive $YBa_2Cu_3O_7$ (YBCO) and Nb nanoSQUIDs to magnetization reversal measurements of individual magnetic nanoparticles. The YBCO SQUIDs are based on grain boundary Josephson junctions and are patterned in a single layer of epitaxially grown YBCO films by Ga focused ion beam milling. The Nb SQUIDs contain sandwich-type Josephson junctions with normal conducting HfTi barriers; they are fabricated with a multilayer technology that includes patterning by e-beam lithography and a combination of milling techniques and chemical-mechanical polishing. Due to the small inductance of the SQUID loops, ultralow white flux noise at 4.2 K can be achieved, which yields spin sensitivities of down to a few Bohr magnetons per unit bandwidth for a magnetic nanoparticle placed at 10 nm distance to the SQUID loop.**

***Keywords*—nanoSQUID; YBCO; Nb; focused ion beam milling; flux noise; magnetic nanoparticle**

I. Introduction

Magnetic properties of micro- and nanoscale objects, are currently a topic of intensive research. Their investigation requires the development of appropriate tools, e.g. for detection of the magnetization reversal of individual magnetic nanoparticles (MNPs) [1, 2]. Promising candidates for this task are strongly miniaturized superconducting quantum interference devices (SQUIDs) – so-called nanoSQUIDs [3, 4]. The magnetization hysteresis loop of a ferromagnetic nanoparticle can be detected via the induced change of stray magnetic field coupled to a nanoSQUID by a MNP that is placed in close vicinity to the SQUID loop. The figure of merit for this kind of SQUID application is the spin sensitivity $S_\mu^{1/2}$, defined as the rms flux noise $S_\Phi^{1/2}$ of the nanoSQUID divided by the coupling factor $\phi_\mu=\Phi/\mu$ (flux Φ coupled to the SQUID per magnetic moment μ of the MNP).

II. Device Fabrication

A. YBCO nanoSQUIDs

We fabricate devices from thin films of $YBa_2Cu_3O_7$ (YBCO), epitaxially grown by pulsed laser deposition on bicrystal $SrTiO_3$ (STO) or MgO substrates, with 24° misorientation angle of the grain boundary. The grain boundary formed in the YBCO film (typically 120 nm thick) acts as a Josephson junction [5]. Subsequently, we deposit in-situ by sputtering or electron beam evaporation 50-70 nm thick Au on top of YBCO. The Au film serves as a protection layer and for providing electrical contacts. Next, we pattern 4-8 µm wide bridges straddling the grain boundary by photolithography and Ar ion milling. As a final patterning step, we use Ga focused ion beam (FIB) milling to pattern the SQUID loop, to define the width of the grain boundary junctions and to pattern a narrow constriction (100-300 nm wide) into the SQUID loop [6-9]. The constriction in the SQUID loop provides the location of highest sensitivity to the stray field produced by a MNP placed on top of the constriction [9]. Moreover, by sending a current I_{mod} through the constriction, the flux coupled to the SQUID can be controlled and modulated. This feature can be conveniently used for on-chip flux modulation of the nanoSQUIDs and for their operation in flux-locked loop readout [8].

B. Nb nanoSQUIDs

The Nb SQUIDs are fabricated by a multilayer process, involving in-situ sputtering of a Nb/HfTi/Nb trilayer plus ex-situ sputtered SiO_2 insulating and Nb wiring layer and a combination of electron beam lithography, various etching steps and chemical-mechanical polishing. The Nb films are typically 160-200 nm thick, and the normal conducting HfTi barriers have typical thicknesses ranging from 17 to 25 nm. For

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details of the fabrication process and superconductor-normal metal-superconductor (SNS) Josephson junction characteristics see [10-13]. The typical nanoSQUID geometry of our devices is of a microstrip-type: two Nb lines (typically 150-250 nm wide) are patterned on top of each other (separated by SiO_2) and are vertically connected by two trilayer Nb/HfTi/Nb Josephson junctions. The lateral spacing of the two junctions is a few µm down to ~100 nm, and the vertical spacing of the two Nb lines is ~200 nm; both quantities together define the size of the SQUID loop with a loop plane that is perpendicular to the substrate surface. This microstrip geometry allows us to conveniently control and modulate magnetic flux coupled to the SQUID loop via a modulation current I_{mod}, which is flowing along one of the two Nb lines [13].

III. NanoSQUID Performance

The YBCO and Nb nanoSQUIDs have nonhysteretic current-voltage characteristics (IVCs) at temperature T=4.2 K that are reasonably well described by the resistively and capacitively shunted junction model. Due to the small inductance L of the SQUID loop (in the pH range), the rms flux noise in the thermal white noise region $S_{\Phi,w}^{1/2}$ is very low – typically a few 100 $n\Phi_0/Hz^{1/2}$ (Φ_0 is the magnetic flux quantum). The lowest values obtained so far are $S_{\Phi,w}^{1/2}$=45 $n\Phi_0/Hz^{1/2}$ [8] and 110 $n\Phi_0/Hz^{1/2}$ [4] for our best YBCO and Nb nanoSQUIDs, respectively. For a MNP placed at 10 nm distance from the SQUID loop, this corresponds to spin sensitivities $S_\mu^{1/2}$~4 $\mu_B/Hz^{1/2}$ and ~10 $\mu_B/Hz^{1/2}$, respectively (μ_B is the Bohr magneton). For the determination of $S_\mu^{1/2}$, we calculate the coupling factor by numerical simulation of the supercurrents flowing in the SQUID loop, for any given planar SQUID geometry. For these simulations we use 3D-MLSI [14], a finite-element-based software, solving the London equations in 2D sheets; this takes into account the thickness of the superconducting films forming the SQUID loop and the value of the London penetration depth [4, 6, 9, 15].

The YBCO nanoSQUIDs offer the advantage of operation over a very wide temperature range, so far from 300 mK up to 80 K [9]. Moreover, due to the huge upper critical field of YBCO, these devices offer the potential for operation up to very strong magnetic fields. So far, we operated devices at 4.2 K up to 3 T, and performed flux noise measurements up to 1 T [7]. A major drawback of the YBCO nanoSQUIDs is their strong low-frequency excess noise, scaling approximately as S_Φ~1/f (f is the frequency) [8]. This is due to strong critical current fluctuations, as typically observed for SQUIDs based on cuprate superconductors [16]. Moreover, 1/f noise may further increase upon applying strong magnetic fields, unless the entry of Abrikosov vortices can be avoided [17].

The temperature range of operation of the Nb nanoSQUIDs is much more restricted, as compared to the YBCO nanoSQUIDs. Typically our devices operate below ~6 K. Upon cooling to below 4.2 K, the devices start to develop hysteresis in their IVCs which can be attributed to Joule heating in the junctions. The temperature below which hysteresis appears increases with increasing critical current density and area of the junctions [18]. Operation in strong magnetic fields, although restricted to below ~1 T is also possible, in particular for devices with strongly reduced linewidths, for which operation up to ~0.5 T has been demonstrated [12].

The mature multilayer technology for Nb nanoSQUID fabrication offers the possibility to develop quite complex device layouts which can be used e.g. to develop gradiometric designs for operation in strong homogeneous magnetic fields. One promising approach is the development of 3D vector nanoSQUIDs that have been realized recently [19]. This device combines two orthogonal microstrip-type Nb nanoSQUIDs (loop normal along the x- and y-axis) with a gradiometric nanoSQUID with loop normal along the z-axis. For a MNP placed in the center of one of the gradiometer loops, the switching of its magnetic moment upon applying a magnetic field in z-direction can then be traced by all three orthogonal nanoSQUIDs, to record simultaneously all three vector components of the magnetic moment of the MNP. This approach shall be particularly useful for studies of the magnetic anisotropy of individual MNPs.

IV. Applications of nanoSQUIDs to Magnetization Reversal Measurements of Magnetic Nanoparticles

We have used YBCO and Nb nanoSQUIDs to perform magnetization reversal measurements of individual MNPs of different geometries: nanopillars, -disks, -wires and -tubes.

A. MNP measurements with YBCO nanoSQUIDs

A Fe nanowire, embedded in a carbon nanotube, has been placed on top of a YBCO nanoSQUID with ~300 nm distance from the SQUID loop (on the side opposite to the constriction in the loop). A magnetic field was applied along the wire axis (the easy axis of magnetization). While sweeping the magnetic field, the flux coupled to the SQUID was recorded, yielding an almost ideal rectangular-shaped magnetization hysteresis curve at 4.2 K, as expected for a single-domain state [8]. The detected flux change of ±82.5 $m\Phi_0$ in the saturated states was in very good agreement with the calculated signal (from the simulated coupling factor integrated over the volume of the Fe wire), assuming the literature value for the saturation magnetization of Fe. The observed switching field ~100 mT was ten times smaller than predicted from a simple Stoner-Wohlfarth reversal mechanism. This low switching field, however, was in very good agreement with estimates based on magnetization reversal via curling mode [8].

We note that further measurements with similar YBCO nanoSQUIDs have been performed on Co nanowires grown by focused-electron-beam-induced deposition (FEBID), as reported in [20]. In this case, the nanowires were suspended above the nanoSQUIDs. These measurements clearly showed improved performance of the nanowires that had been annealed after FEBID growth [21].

In a further series of measurements, we have demonstrated the benefit of using YBCO nanoSQUIDs for MNP measurements over a wide temperature range [9]. In this case, Co nanopillars and nanodisks were grown by FEBID directly on top of the constriction in the SQUID loop. For the Co MNPs that revealed single-domain states, we recorded hysteresis loops from 300 mK up to 80 K. The observed T-dependence of the switching fields was shown to be in very good agreement with predictions from a model for thermally induced magnetization reversal [9].

B. MNP measurements with Nb nanoSQUIDs

Nb nanoSQUIDs have been integrated into the torque magnetometer setup of the Poggio group at Univ. Basel to investigate individual Ni, permalloy and CoFeB nanotubes. The combined system enables simultaneous measurements of the integral magnetization by dynamic cantilever torque magnetometry and local magnetization by nanoSQUID magnetometry [13]. Combined torque and SQUID measurements on individual Ni nanotubes, supported by micromagnetic simulations of magnetization configurations, suggest reversal via the formation of vortexlike states within the nanotube [22]. Such stray-field free states can have applications for memory and noninvasive sensing.

Conclusions and Outlook

YBCO and Nb nanoSQUIDs have been developed for the investigation of magnetization reversal of individual magnetic nanoparticles. Very small SQUID inductances enable the realization of ultralow flux noise of the nanoSQUIDs in the thermal white noise limit. For MNPs placed in 10 nm distance to the SQUID loop, this translates into spin sensitivities down to only a few Bohr magnetons per unit bandwidth, which is appropriate for many studies on individual MNPs. Apart from further suppression of $1/f$ noise, a key challenge is the development of reliable routines for placing MNPs in a controlled way in close vicinity to the nanoSQUIDs, ideally at variable position and temperature. For YBCO nanoSQUIDs, the recently developed creation of Josephson junctions and SQUIDs by focused He ion irradiation [23, 24] can provide new perspectives for creating advanced nanoscale devices

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