Magnetism in SQUIDs at Millikelvin Temperatures

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We have characterized the temperature dependence of the flux threading dc SQUIDs cooled to millikelvin temperatures. The flux increases as $1/T$ as temperature is lowered; moreover, the flux change is proportional to the density of trapped vortices. The data are compatible with the thermal polarization of surface spins in the trapped fields of the vortices. In the absence of trapped flux, we observe evidence of spin-glass freezing at low temperature. These results suggest an explanation for the universal $1/f$ flux noise in SQUIDs and superconducting qubits.

Superconducting integrated circuits are a leading candidate for scalable quantum information processing [1]. A key qubit figure of merit is the dephasing time, the time over which a superposition of the qubit $|0\rangle$ and $|1\rangle$ states maintains phase coherence. Dephasing is governed by low-frequency noise in the bias parameters that control the energy separation between the qubit states [2,3]. In the Josephson flux qubit, the Gaussian decay envelope of qubit Ramsey fringes is compatible with a magnetic flux noise with $1/f$ spectrum and a magnitude at 1 Hz of $1 \mu\Phi_0/\text{Hz}^{1/2}$, where $\Phi_0 = h/2e$ is the magnetic flux quantum [4,5]. In the Josephson phase qubit, a recent experiment used the resonant response of the qubit to measure the power spectral density of flux noise directly; again the spectrum was $1/f$, with a noise magnitude comparable to that seen in the flux qubit [6]. While these experiments identify flux noise as a dominant dephasing mechanism, they offer no clue to its microscopic origin.

The flux noise inferred from recent qubit experiments is consistent with the flux noise observed more than 20 years ago in a series of measurements performed on dc superconducting quantum interference devices (SQUIDs) cooled to millikelvin temperatures [7]. The noise was observed to be universal, that is, only weakly dependent on a wide range of parameters such as superconducting materials, SQUID loop geometry and inductance, and temperature. While these experiments ruled out many potential sources of noise, the microscopic mechanism was never identified. There has been recent theoretical interest in the possibility that the $1/f$ flux noise is due to unpaired spins on the surfaces of the superconductors [8,9]. Models for $1/f$ flux noise from surface spins are attractive, as they yield a noise power that is only weakly dependent on the overall scale of the device, compatible with the universal character of the noise [6]. To date, however, there has been no direct experimental evidence for magnetism in superconducting circuits.

Here we present clear evidence for a high density of unpaired surface spins in thin-film SQUIDs. While fluctuations of these spins produce a small but measurable effect, the coherent magnetization of the spins couples a very large flux of order $1 \Phi_0$ to the SQUID. Moreover, simple measurements of the temperature dependence of the magnetization allow determination of the surface density of unpaired spins, and indicate that interactions between spins are significant.

We first characterized the flux noise of a two-stage SQUID amplifier at millikelvin temperatures. A schematic of the experiment is shown in Fig. 1(a). Both SQUIDs are made in the conventional Ketchen-Jaycox square-washer geometry [Fig. 1(b)] [10]; fabrication details can be found in [11]. The 110 pH Al SQUID is biased with a voltage, and the current through the SQUID is monitored with the
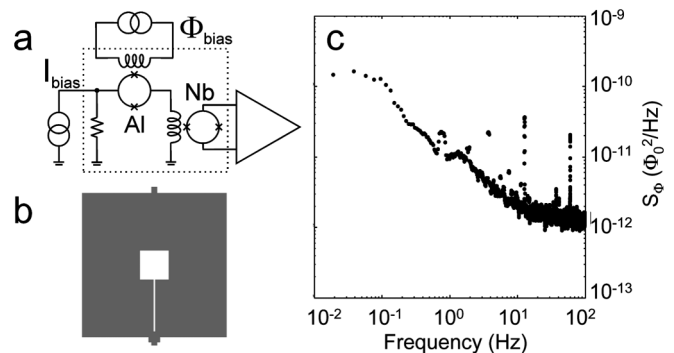

FIG. 1. (a) Schematic of two-stage SQUID amplifier. The first-stage Al SQUID is biased with a voltage, and the current through this device is read out with a Nb SQUID operated in a flux-locked loop. Both devices are operated inside a superconducting aluminum shield (dashed box). (b) Layout of the thin-film SQUIDs. The Al SQUID has inner and outer dimensions 60 and 300 μm; the Nb SQUID has inner and outer dimensions 200 μm and 1 mm. (c) Power spectral density of flux noise of the two-SQUID amplifier measured at 100 mK.

350 pH Nb SQUID, which is operated in a flux-locked loop [12]. The current through the first-stage Al SQUID oscillates with applied flux, with periodicity Φ_0. When the first-stage SQUID is biased at a flux $(n \pm 1/4)\Phi_0$, where n is an integer, sensitivity to applied flux is maximum.

The power spectral density of flux noise in the Al first-stage SQUID is shown in Fig. 1(c). The noise scales as $1/f$ at low frequencies, with a noise magnitude of $3 \ \mu\Phi_0/\text{Hz}^{1/2}$ at 1 Hz. The flux noise is roughly independent of temperature over the range from 30 to 500 mK (not shown). These results are compatible with previous measurements of SQUIDs and qubits [4–7].

At the lowest temperatures, however, we observe a surprising dependence of SQUID flux on bath temperature. We characterize the temperature dependence of the flux in the SQUIDs by tracing out the full current-flux (I-Φ) characteristic of the first-stage SQUID as bath temperature is varied [Fig. 2(a)]. By following shifts in the position of the extrema of the I-Φ curve along both the Φ and I axes, we monitor changes in the quasistatic fluxes threading the first- and second-stage SQUIDs, respectively. The results of this experiment are shown in Fig. 2(b).

The fluxes threading the SQUIDs change dramatically as bath temperature is lowered. At the lowest accessible temperatures $\sim$90 mK, the temperature-to-flux transfer coefficients are in excess of 10 Φ_0/K. This behavior has been observed in both Al and Nb devices, made both with and without a wiring dielectric, and prepared in different facilities according to different fabrication recipes.

Temperature-induced flux drift in SQUIDs has been studied in the past. There have been investigations of flux shifts due to the thermal motion of magnetic flux [13,14], and due to temperature-dependent critical currents [13]. Howeve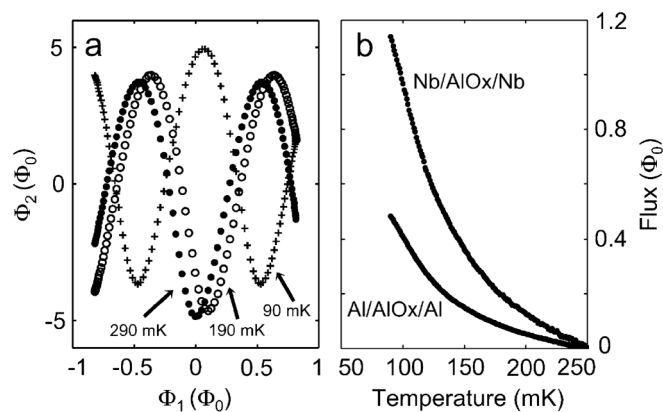r, we do not expect these mechanisms to play a role here, since our experiments are conducted far below the superconducting transition temperature T_c. Indeed, the large magnitude and low energy scale of the effect observed here suggest a new mechanism. Over a broad temperature range below 500 mK, the flux threading the SQUIDs scales inversely with temperature. Paramagnetic impurities in the materials of the SQUID would naturally give rise to such a signature, and we interpret the $1/T$ dependence of the flux as strong evidence for unpaired spins, most likely in the native oxides of the superconductors. However, as these experiments are performed in a nominal zero field, the source of the orienting magnetic field is not immediately clear.

In order to clarify the source of the temperature-dependent flux, we have performed a series of field-cool experiments in which a magnetic field B_{fc} is applied to a 350 pH square-washer Nb SQUID as it is cooled through T_c; the field freezes magnetic flux vortices into the Nb film, with density $\sigma_v \approx B_{\text{fc}}/\Phi_0$ [15]. When the device is well below T_c, the field is removed, and the SQUID is maintained in a flux-locked loop as it is cooled to millikelvin temperatures. In Fig. 3(a) we plot the flux through the SQUID as a function of temperature for eight different values of B_{fc}. The cooling field strongly affects the temperature-dependent flux, enhancing or even reversing the polarity of the observed signal. In Fig. 3(b) we plot the flux change on cooling from 500 to 100 mK as a function of B_{fc}; a linear fit to the data yields a slope of 1.3 Φ_0/mT. Clearly, vortices contribute significantly to the measured flux shift.

The linear dependence of flux drift on vortex density suggests the following interpretation. As the device is cooled through T_c in an applied field, vortices nucleate and relatively large magnetic fields of order 10 mT are frozen into the films. The observed signal is due to the
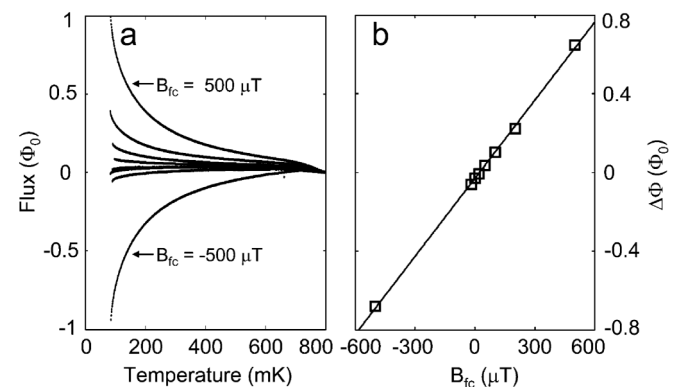

FIG. 2. Temperature dependence of the flux threading the SQUIDs of the amplifier described in Fig. 1. (a) I-Φ characteristic of the Al first-stage SQUID measured at three temperatures; flux Φ_1 through the Al first-stage SQUID is plotted on the horizontal axis, and current I through the Al first-stage SQUID is expressed as a flux Φ_2 coupled to the Nb second-stage SQUID and plotted on the vertical axis. (b) Quasistatic flux threading the SQUIDs vs temperature.

FIG. 3. (a) Temperature dependence of the flux threading a 350 pH Nb/Al oxide/Nb SQUID, for different fields B_{fc} applied as the device was cooled through the superconducting transition (from bottom to top, $B_{\text{fc}} = -500, -20, 0, 20, 50, 100, 200, 500 \ \mu\text{T}$). (b) Temperature-induced flux change $\Delta\Phi = \Phi(100 \ \text{mK})-\Phi(500 \ \text{mK})$ on cooling from 500 to 100 mK vs cooling field B_{fc}. A linear fit to the data yields a slope $\Delta\Phi/B_{\text{fc}} = 1.3 \ \Phi_0/\text{mT}$.

magnetization of unpaired electron spins on the surface of
the superconductor in the strong fields of the vortices.
Indeed, careful analysis of the field-cool data allows us
to extract the surface density σ_s of spins, a key parameter
in models of $1/f$ noise from surface magnetism. We note
that there is negligible *direct* coupling to the SQUID from
spins polarized out of the plane of the superconducting
films; this is easily understood from reciprocity, as surface
magnetic fields due to currents in the SQUID have vanish-
ing perpendicular component. However, the topology of
our device is quite different from that of a continuous
superconducting washer, due to the presence of the vorti-
ces. The thermal polarization of unpaired surface spins in
the vortex forces a redistribution of the circulating super-
currents in the vortex, due to the requirement to conserve
magnetic flux. These currents, in turn, couple strongly to
the SQUID loop. Calculation of the spin density from the
data of Fig. 3 therefore proceeds in two stages: (1) calcu-
lation of the coupling between a vortex and the SQUID
loop, and (2) calculation of the coupling between surface
spins and a vortex. (A detailed discussion of this calcula-
tion can be found in [11].)

A single vortex trapped in the SQUID washer induces an
effective flux offset in the SQUID; the magnitude of the
flux offset approaches 0 Φ_0 and 1 Φ_0 for vortices posi-
tioned at the outer and inner edges of the SQUID washer,
respectively. Numerical calculations for a circular washer
show that this effective flux offset averaged over the area of
the washer is 0.5 Φ_0 for a thin washer (with width much
less than radius), as expected; for our device dimensions
[inner radius 100 μm and outer radius 500 μm—see
Fig. 1(b)], we find an average flux offset of 0.14 Φ_0. The
physical source of the flux is vortex currents that drop off
slowly with distance from the core and circulate around the
hole in the SQUID washer. Polarization of the surface spins
couples a flux $\Phi_v(T)$ to the vortex, requiring the circulating
currents to change due to the quantization condition. The
change in flux coupled to the SQUID is thus $\Phi = -0.14\Phi_v(T)$.

To calculate $\Phi_v(T)$, we rely on reciprocity: knowledge
of the current distribution in the vortex allows us to deter-
mine the coupling to a spin at an arbitrary location. We
model the vortex as a hole in a superconducting ground
plane with radius equal to the estimated coherence length
$\xi = 30$ nm and a thin-film penetration depth $\Lambda = 100$ nm.
Numerical solution of the London equations allows us to
determine the currents in the vortex, from which we de-
termine the in-plane and out-of-plane magnetic fields $B_r(r)$
and $B_z(r)$. One can show that the flux coupled to the vortex
by the spins is given by

$$\Phi_v(T) = \mu_B \sigma_s L_v P_{\text{eff}}(T), \qquad (1)$$

where L_v is the vortex self-inductance, and where we have
defined an effective spin polarization $P_{\text{eff}}(T)$:

$$P_{\text{eff}}(T) = \frac{1}{\Phi_0} \int_0^\infty 2\pi r B(r) \tanh\left(\frac{\mu_B B(r)}{2k_B T}\right) dr, \qquad (2)$$

where $B(r) = [B_r(r)^2 + B_z(r)^2]^{1/2}$. A change in tempera-
ture yields a change in effective spin polarization $P_{\text{eff}}(T)$ in
the vortex, which in turn induces a change in flux $\Phi_v(T)$
coupled to each vortex. The total flux change $\Delta\Phi$ at the
SQUID is obtained by summing over all vortices, taking
into account the coupling factor 0.14. Thus, we can relate
the measured temperature-dependent flux to spin density as
follows:

$$\frac{\Delta\Phi}{B_{\text{fc}}} = 0.14 \frac{A_{\text{SQ}}}{\Phi_0} \mu_B \sigma_s L_v \Delta P_{\text{eff}}, \qquad (3)$$

where A_{SQ} is the area of the SQUID washer. For the
materials parameters given above, we find $\Delta P_{\text{eff}} \equiv
P_{\text{eff}}(100 \text{ mK}) - P_{\text{eff}}(500 \text{ mK}) = 0.037$ and $L_v =
0.24$ pH. Using $A_{\text{SQ}} = 0.96$ mm^2 and the slope $\Delta\Phi/B_{\text{fc}} =
1.3 \Phi_0/\text{mT}$ from Fig. 3(b), we find a spin density $\sigma_s =
5.0 \times 10^{17}$ m^{-2}. This density of surface spins is compat-
ible with densities considered in recent theoretical models
of $1/f$ flux noise from surface spins [8,9], and with the
surface spin density postulated in a recent model for spin-
flip scattering in superconducting nanowires [16,17].

Yet while spin densities of this order of magnitude are
clearly required to explain the measured $1/f$ flux noise in
terms of surface magnetism, the microscopic physics that
drives the spin fluctuations is by no means obvious. A
recent theory explains flux noise in terms of spin diffusion
and interaction between surface spins via the RKKY
mechanism [18]. A key parameter in this model is the
characteristic energy of spin interactions. In the field-
cool experiments described above, any possible spin inter-
actions are obscured by strong coupling of the spins to the
fields of the vortices. To probe interactions between spins,
it is necessary to examine devices that have no trapped flux.
To this end, we have performed additional experiments
using SQUIDs made from narrow (2 μm) superconduct-
ing lines. It is energetically unfavorable for a vortex to
nucleate in a superconducting strip of width w provided the
strip is cooled through T_c in a magnetic field that is less
than the threshold field $B_{\text{thr}} \approx \Phi_0/w^2$ [15,19,20]. As
$B_{\text{thr}} \approx 500 \mu$T for our narrow SQUIDs, we expect no
trapped vortices when these devices are cooled in a nomi-
nal zero field ($\lesssim 10 \mu$T).

In Fig. 4 we show typical flux vs temperature data for
two narrow linewidth Nb/Al oxide/Nb SQUIDs config-
ured as square loops with dimensions $d = 50$ and 200 μm
(see inset). These devices show a strongly temperature-
dependent flux even in the absence of trapped vortices.
Although the magnitude of the flux change varies from
cooldown to cooldown, suggesting a dependence of the
magnetization on the details of the cooldown, we consis-
tently observe a flux shift of order 10 mΦ_0 on cooling to
30 mK. These data confirm that the flux drift observed in
our experiments is not due to thermally active vortices.
Moreover, these measurements allow us to extract an en-
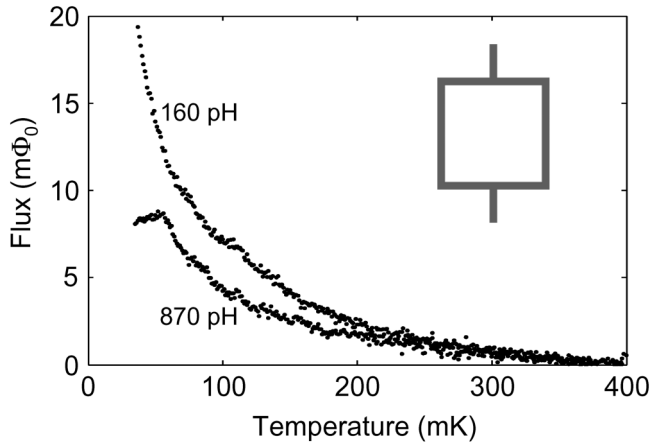ergy scale of spin interactions, as we explain below.

FIG. 4.   Temperature dependence of the flux threading narrow (2 μm) linewidth Nb/Al oxide/Nb SQUIDs with inductances 160 and 870 pH. SQUID loop geometry is shown in the inset.

First, the magnitude of the temperature-dependent flux in these devices indicates a high degree of spin order. Following [6], one can show that the maximum flux coupled to the SQUID from surface spins (achieved when the spins are fully polarized and oriented radially) is given by

$$\Phi_{\max} = 2\mu_0\mu_B\sigma_s d. \qquad (4)$$

The flux scales linearly with device dimension, and is independent of the width of the superconducting lines for $w \ll d$. For the 160 pH SQUID with $d = 50$ μm, a spin density $\sigma_s = 5 \times 10^{17}$ m^{-2} yields $\Phi_{\max} = 280$ mΦ_0; the measured flux change of 19 mΦ_0 therefore suggests a high degree of spin polarization at 30 mK. While surface spin densities in the narrow SQUIDs could be higher than that inferred from the field-cool experiments (the narrow devices are not passivated by a dielectric), the large flux shift in these devices in the absence of vortices suggests an energy scale for spin interactions of order 10 mK. The RKKY model of [18] predicts a spin interaction strength of 20 mK for the spin densities and materials parameters considered here, assuming a uniform distribution of spins through the native oxides of the superconducting wires; for spins concentrated at the metal-insulator interface, the interaction strength approaches 500 mK.

Moreover, we note the appearance of a cusp at 55 mK in the flux vs temperature data of the 870 pH SQUID. This is reminiscent of the cusp in the first-order susceptibility of a spin glass at the freezing temperature [21]. This feature again indicates the importance of spin interactions and suggests the possibility of spin-glass freezing on the time scales of our experiment.

In summary, we have demonstrated that superconducting circuits are magnetically active at millikelvin temperatures. This observation points to a microscopic explanation for the excess $1/f$ flux noise in Josephson circuits, a dominant source of dephasing in superconducting qubits

and an open question for more than 20 years. We observe evidence of interactions between surface spins and possible spin-glass freezing at millikelvin temperatures. A high density of surface spins could play a deleterious role in other schemes for quantum information processing in the solid state, notably those based on electron spin. It is possible that suitable surface treatments of the superconducting films will lower the density of magnetic states, leading to superconducting devices with lower noise and solid-state qubits with improved coherence times [22].

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