## **Complex Inductance, Excess Noise, and Surface Magnetism in dc SQUIDs**

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We have characterized the complex inductance of dc SQUIDs cooled to millikelvin temperatures. The SQUID inductance displays a rich, history-dependent structure as a function of temperature, with fluctuations of order 1 fH. At a fixed temperature, the SQUID inductance fluctuates with a 1/f power spectrum; the inductance noise is highly correlated with the conventional 1/f flux noise. The data are interpreted in terms of the reconfiguration of clusters of surface spins, with correlated fluctuations of effective magnetic moments and relaxation times.

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The origin of low-frequency 1/f flux noise in superconducting quantum interference devices (SOUIDs) is a long standing open question in condensed matter physics [1,2]. This question has attracted particular interest recently in the context of efforts to realize quantum bits ("qubits") based on superconducting integrated circuits [3]. It has been shown that low-frequency flux noise is a dominant source of dephasing in superconducting flux [4,5] and phase [6] qubits. Recent experiments indicate that there is a high density of unpaired surface spins in normal metal [7] and superconducting [8] thin films; it is suspected that fluctuations of these spins give rise to the 1/f flux noise. There is some experimental evidence that interactions between the surface spins are significant [8], and a recent theoretical model suggests that RKKY interactions between spins are a critical component of the flux noise mechanism [9]. However, a detailed microscopic understanding of 1/f flux noise is lacking.

Here we examine the complex frequency- and temperature-dependent inductance L = L' - iL'' of dc SQUIDs cooled to millikelvin temperatures. The SQUID inductance contains a small contribution from unpaired spins on the surface of the superconductor; our measurements thus provide access to the complex susceptibility  $\chi = \chi' - i\chi''$  of the surface spin system, and shed light on the underlying physics that drives spin fluctuations. We observe rich structure in the temperature-dependent SQUID inductance, suggesting the reconfiguration of spin domains or clusters, with correlated fluctuations of spin relaxation times. Noise in the SQUID inductance is highly correlated with but distinct from the usual 1/f flux noise. These results indicate cooperative behavior and complex dynamics in the surface spin system, and offer important clues that will help in the formulation of a microscopic model of flux noise.

As our goal is to probe spins that reside on the surface of the SQUID itself, we have fabricated asymmetric dc SQUIDs that allow injection of an ac excitation current directly into the SQUID loop. The device geometry is shown in Fig. 1. Excitation currents  $I_{ex}$  of order 100  $\mu$ A generate surface magnetic fields of order 100  $\mu$ T that couple strongly to any surface spins, inducing a small spin magnetization that is oriented radially with respect to the SQUID loop; the radial spin magnetization ensures optimal coupling of the spins to the SQUID. The flux induced by the spin magnetization constitutes only a small fraction of order  $10^{-5}$  of the flux coupled to the SQUID by the excitation current. For this reason, the SQUID is operated in a bridge configuration: a second external coil couples a compensating flux that nulls the flux induced by the excitation current, allowing sensitive measurement of inductance changes induced by polarization or fluctuations of the surface spin system. The SOUID is operated in a flux-locked loop, and the response of the SQUID at the excitation frequency  $f_0$  is measured with a lock-in amplifier. Changes in the in-phase and out-of-phase response of the SQUID to the excitation current are directly related to



FIG. 1. (a) Schematic of asymmetric dc SQUID circuit for investigation of surface magnetism. The ac excitation current  $I_{ex}$ is injected directly into the SQUID loop. The current  $I_{null}$ provides a compensating flux, and the SQUID response to the excitation current is measured with a lock-in amplifier. (b) Layout of the Nb-AlO<sub>x</sub>-Nb SQUIDs. The Nb SQUID loops have thickness 80 nm, trace width 2  $\mu$ m, and loop diameter of 40 and 80  $\mu$ m, corresponding to measured inductances of 110 and 250 pH, respectively.

changes  $\Delta L'(f_0, T)$  and  $\Delta L''(f_0, T)$  in the real and imaginary parts of the SQUID inductance.

The temperature dependence of the SQUID inductance is shown in Fig. 2(a). In the high temperature region  $\gtrsim 2$  K, we observe a strong temperature dependence to L' due to the kinetic inductance of the superconductor, while we see very little change in L'', compatible with vanishingly small quasiparticle dissipation at the low excitation frequencies. However, as we cool below 2 K we observe unexpected sharp structure in the SQUID inductance, with jumps of order 1 fH in the real part of the inductance. When the device is repeatedly cycled up and down in temperature, some of the main features of the  $\Delta L(T)$  scans are qualitatively reproduced, albeit with significant hysteresis in temperature; however, the detailed structure of the scans is different from run to run. We see that the fluctuations in L' and L'' are anticorrelated: a fluctuation that decreases L' is accompanied by an increase in L'' (figure inset). We remark that the fine structure in the



FIG. 2. Temperature dependence of complex inductance L = L' - iL'' of a 110 pH SQUID. (a)  $\Delta L'$  (upper panel) and  $\Delta L''$  (lower panel) vs temperature, measured at an excitation frequency  $f_0 = 100$  Hz. The direction and ordering of the temperature sweeps are indicated by the arrows. The increase in  $\Delta L'$  at higher temperatures is due to the temperature-dependent kinetic inductance of the superconductor. The inset shows a blowup of the fine structure in  $\Delta L(T)$ , demonstrating strong correlation between fluctuations in L' and L''. (b)  $\Delta L'(T)$  probed at  $f_0 = 10$  Hz and 100 Hz. The sharp structure in  $\Delta L'$  below 2 K is independent of probe frequency. (c) As in (b), but for  $\Delta L''(T)$ ; the 10 Hz data have been scaled by a factor 10 to facilitate comparison with the data at 100 Hz. The fluctuations in L'' scale linearly with probe frequency.

 $\Delta L(T)$  scans is not due to the noise of the measurement system, or to any conventional SQUID noise mechanism, but rather represents actual fluctuations of the inductance of the SQUID.

By injecting a two-tone excitation and performing simultaneous lock-in detection at the two frequencies, we can examine correlations between inductance fluctuations at different frequencies [Figs. 2(b) and 2(c)]. We observe that the inductance fluctuations at different frequencies are highly correlated. While the real part of the inductance fluctuations  $\Delta L'$  is independent of probe frequency, the imaginary part of the fluctuations scales linearly with frequency:  $\Delta L'' \propto f_0$ . At an excitation frequency around 50 Hz, the magnitudes of the real and imaginary parts of the inductance fluctuations are comparable.

To investigate the nature of the inductance fluctuations in detail, we have stabilized the device at fixed temperature, and examined the power spectra of fluctuations of the SQUID response to the excitation current for excitation frequencies  $f_0$  in the range from 100 to 300 Hz (Fig. 3). Here the white noise level is set by the noise of the SQUID system at the excitation frequency, and is typically in the range from 1 to 2  $\mu \Phi_0/\text{Hz}^{1/2}$ . At low frequencies, how-



FIG. 3. 1/f inductance noise in a 250 pH SQUID. (a)  $S_L(f; f_0 = 100 \text{ Hz})$  for excitation amplitudes  $I_{\text{ex}} = 30$  and 100  $\mu$ A, measured at 300 mK. (b) Power spectra  $S_{L'}(f; f_0)$  and  $S_{L''}(f; f_0)$  of fluctuations in the real and imaginary parts of the SQUID inductance, for probe frequencies  $f_0 = 150$  and 277 Hz. The data were obtained at 300 mK. (c)  $S_L(f; f_0 = 100 \text{ Hz})$  measured from 100 mK to 2 K.

ever, we observe excess 1/f noise in the SQUID response that far exceeds the system white noise level. The excess noise in the SQUID response scales as the square of the excitation current for excitation amplitudes in the range from 10 to 100  $\mu$ A [Fig. 3(a)]; thus, the fluctuating response is due to an apparent noise  $S_L(f; f_0)$  in the SQUID inductance.

In Fig. 3(b) we show the power spectra of fluctuations of the real and imaginary parts of the SQUID inductance  $S_{L'}(f; f_0)$  and  $S_{L''}(f; f_0)$  for probe frequencies  $f_0$  of 150 and 277 Hz. At these relatively high probe frequencies, noise in the imaginary part of the inductance exceeds noise in the real part of the inductance,  $S_{L''}(f; f_0) > S_{L'}(f; f_0)$ . Moreover, the noise in the real part of the inductance  $S_{L'}(f; f_0)$  is independent of probe frequency, while noise in the imaginary part of the inductance  $S_{L''}(f; f_0)$  scales with frequency as  $f_0^2$ . These observations are compatible with the scaling of the individual inductance jumps described in Fig. 2. Thus, we conclude that the structure in the  $\Delta L(T)$  scans and the excess 1/f inductance noise at fixed temperature are manifestations of the same underlying mechanism.

In Fig. 3(c) we show the temperature dependence of the inductance noise over the range from 100 mK to 2 K. The noise increases as temperature is lowered, and saturates at a temperature around 200 mK.

We now turn to possible physical explanations for our observations. We have performed numerical simulations based on the method of [10] which indicate that critical current fluctuations will not lead to an apparent inductance noise. We note that fluctuations in the density of nonequilibrium quasiparticles would give a similar signature, namely, in-phase fluctuations in the kinetic inductance that are independent of the excitation frequency  $f_0$ , and out-of-phase fluctuations due to quasiparticle dissipation that scale linearly with  $f_0$ . While there is evidence for long-lived nonequilibrium quasiparticles from other measurements of superconducting devices, one expects to see negligible quasiparticle resistance at the low excitation frequencies employed in these experiments.

Instead we interpret our data in terms of fluctuations in the dispersion  $\chi'$  and absorption  $\chi''$  of a surface spin system, possibly associated with the freezing or blocking of spin clusters. We first consider the magnitude of the observed structure in the  $\Delta L'(T)$  scans in light of a simple model of surface magnetic states. We consider a toroidal SQUID geometry with loop radius *R* and wire radius *r*, covered with a surface density  $\sigma$  of magnetic states with moment  $\mu$ . In the case of noninteracting spins with relaxation time  $\tau$  much shorter than the inverse excitation frequency  $\tau \ll 1/2\pi f_0$ , we expect to find a Curie-like contribution to *L'* from the spins given by

$$L'_{\rm spin} = \frac{\mu_0^2 \mu^2}{3k_B T} \sigma \frac{R}{r}.$$
 (1)

For R/r = 20,  $\mu = \mu_B$ , and for the surface spin density  $\sigma = 5 \times 10^{17} \text{ m}^{-2}$  inferred from [8], we expect to find a spin contribution to the SQUID inductance of order 30 aH upon cooling to 1 K; a more detailed calculation that takes into account the current distribution in our thin-film geometry yields an inductance contribution that is larger by a factor 1.5. In our experiments, we observe inductance structure spanning a full range of order fH. The large magnitude of the inductance fluctuations is not compatible with any existing theoretical models for 1/f flux noise from surface spins. We take the observed magnitude of the inductance fluctuations as evidence for the formation of clusters of spins with effective moment  $\mu = \alpha \mu_B$ , with  $\alpha$ in the range from 10 to 100. The emergence of spin clusters would lead to enhancement of the spin contribution to the SQUID inductance by the factor  $\alpha$ , and could account for the pronounced structure, hysteresis, and variation in the  $\Delta L(T)$  scans.

We now examine the frequency dependence of the inductance fluctuations. We consider an ensemble of spin clusters with a distribution of relaxation times  $g(\tau)$ . The susceptibility of the spin system is given by

$$\chi(f_0) = \int g(\tau) \frac{\chi_T(\tau)}{1 + i2\pi f_0 \tau} d\tau, \qquad (2)$$

where  $\chi_T$  is the static isothermal spin susceptibility, which scales linearly with cluster size  $\alpha$ . The observation that fluctuations in the real part of the spin inductance L' are independent of frequency indicates that spin relaxation is fast for all frequencies considered,  $2\pi f_0 \tau \ll 1$ , and suggests fluctuations of the sizes of spin clusters, i.e., fluctuations in  $\chi_T$ . However, the large magnitude of fluctuations in the imaginary inductance  $\Delta L''$  compared to fluctuations in the real inductance  $\Delta L'$  cannot be explained by fluctuations in  $\chi_T$  alone. Indeed, the large magnitude and frequency dependence of  $\Delta L''$  are suggestive of fluctuations of cluster relaxation times that are correlated with the fluctuations in  $\chi_T$ . While relaxation time fluctuations have minimal effect on the dispersion  $\chi'$  in the limit  $2\pi f_0 \tau \ll 1$ , they induce fluctuations in the absorbtion  $\chi''$  that scale linearly with frequency. It is possible that the fluctuation of relaxation times is associated with spin glass freezing of the surface spin system. In canonical spin glass systems, weight in the relaxation time distribution shifts dramatically toward longer times at the freezing temperature  $T_G$ , corresponding to the freezing out or blocking of clusters of spins [11]. Spin glass freezing is thus accompanied by a shift in the absorption spectrum to lower frequencies, resulting in the onset of significant absorption [12,13]. The abrupt jumps that we observe in L'' upon cooling, predominantly in the direction of increased absorption, are thus qualitatively compatible with the freezing of spin clusters. The significant departure in our data from canonical spin glass behavior-namely, a cusp in the dispersion and an inflection point in the ab-



FIG. 4. Amplitude of the cross spectrum  $S_{L''\Phi}/(S_{L''}S_{\Phi})^{1/2}$  of inductance and flux fluctuations of a 250 pH SQUID, measured from 100 mK to 3 K.

sorption at a well-defined freezing temperature—along with the substantial variation in the detailed behavior of  $\Delta L(T)$ , reflect the importance of fluctuations in our samples and suggest the participation of a relatively small number of dominant fluctuation modes. The apparent discrepancy in the freezing temperatures inferred from these measurements ( $T_G \sim 2$  K) and from earlier studies of static magnetization in similar samples ( $T_G \approx 55$  mK [8]) is not presently understood.

We now address the relation of the excess inductance noise to the 1/f flux noise that has previously been observed in SQUIDs and qubits. Is it possible that the flux noise is in fact due to low-frequency fluctuations of the SQUID loop inductance? For example, during normal SQUID operation, currents of order the junction critical current  $I_0$  flow around the SQUID loop. Fluctuations in the loop inductance will therefore couple a fluctuating flux to the SQUID. For a SQUID that is flux-biased at  $\Phi_0/4$ , we expect a flux noise due to inductance fluctuations  $S_{\Phi}/\Phi_0^2 = (\beta_L^2/16)(S_L/L^2)$ , where  $\beta_L \equiv 2LI_0/\Phi_0$  is the reduced inductance. For  $\beta_L = 1$  and  $S_L/L^2 =$ -120 dB/Hz at 1 Hz, the above relation predicts a flux noise that is 2 orders of magnitude lower than the noise that is typically observed. To explore the connection between the inductance noise and the flux noise, we have investigated the cross spectrum of inductance and flux fluctuations. In these experiments, the imaginary part of the SQUID inductance  $L''(t; f_0 = 100 \text{ Hz})$  and the quasistatic flux  $\Phi(t)$  threading the SQUID loop are monitored simultaneously as a function of time. From the two time series, we compute the normalized cross spectral density  $S_{L''\Phi}/(S_{L''}S_{\Phi})^{1/2}$ , shown in Fig. 4. The inductance and flux fluctuations are highly correlated at low temperature, indicating a common underlying physical mechanism. Moreover, the high degree of correlation provides further evidence for a small number of dominant fluctuators: as  $\Phi$ is odd under time reversal while L is even, we expect  $S_{L\Phi}$ to vanish in the limit of a large number of fluctuators, in the

absence of a symmetry breaking bias field. Detailed analysis of the phase of the cross spectrum reveals evidence for both correlated and anticorrelated fluctuators; this work will be the subject of a separate publication. Previously it was proposed that 1/f flux noise arises from equilibrium fluctuations of a static, dissipative spin system [14], but the strong correlation between low-frequency flux noise and inductance fluctuations points to a very different picture. This new evidence suggests that the 1/f flux noise is intimately connected to the complex, nonequilibrium dynamics of the surface spin system.

To conclude, we observe rich structure in the temperature-dependent complex inductance of dc SQUIDs cooled below 2 K. Fluctuations in the real part of the inductance indicate complex cooperative dynamics of spin clusters, while fluctuations in the imaginary part of the SQUID inductance point to temperature-driven evolution of the spin cluster relaxation time distribution and to significant fluctuations in spin cluster relaxation times. The data indicate an intimate connection between 1/f flux noise and the cooperative nonequilibrium dynamics of a surface spin system.

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