Ising-Glauber Spin Cluster Model for Temperature-Dependent Magnetization Noise in SQUIDs

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Clusters of interacting two-level-systems, likely due to Farbe + (F+) centers at the metal-insulator interface, are shown to self-consistently lead to 1/fα magnetization noise [with α(T) ≲ 1] in SQUIDs. Model calculations, based on a new method of obtaining correlation functions, explains various puzzling experimental features. It is shown why the inductance noise is inherently temperature dependent while the flux noise is not, despite the same underlying microscopics. Magnetic ordering in these systems, established by three-point correlation functions, explains the observed flux- inductance-noise cross correlations. Since long-range ferromagnetic interactions are shown to lead to a more weakly temperature dependent flux noise when compared to short-range interactions, the time reversal symmetry of the clusters is also not likely broken by the same mechanism which mediates surface ferromagnetism in nanoparticles and thin films of the same insulator materials.

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Superconducting quantum interference devices (SQUIDs) are key for quantum information as they can replicate natural qubits, such as electron and nuclear spins, using macroscopic devices. However, the performance of many superconducting qubits is severely impeded by the presence of 1/f magnetization noise which limits their quantum coherence. Though this type of noise was first observed in SQUIDs about three decades ago [1,2], its origins and many of its features remain unexplained. Recent activity in quantum computing has, however, revived tremendous interest in this subject [3–15].

Magnetic noise in SQUIDs has several puzzling features. While the flux noise (the first spectrum) is weakly dependent on temperature (T), the choice of the superconducting material and the SQUID’s area [1,8,16], the inductance noise (the second spectrum), surprisingly shows a strong T dependence, it decreases with increasing T and scales as 1/fα [8] where α is T dependent [13,17]. The flux noise is also weakly dependent on geometry [18]. This, along with recent experiments [8], suggests that flux noise arises from surface spins which reside at the superconductor-insulator interface in thin-film SQUIDs.

Experimental evidence also suggests that these surface spins are strongly interacting and that there is a net spin polarization. In Ref. [8], the 1/f inductance noise was shown to be highly correlated with the 1/f flux noise. This cross correlation is inversely proportional to T and is ∼1 roughly below 100 mK. Since inductance is even under time inversion and flux is odd, their three-point cross-correlation function must vanish unless time reversal symmetry is broken, which indicates the appearance of long range magnetic order. As this further implies that the mechanism producing both the flux- and inductance noise is the same, it is not clear why the associated spectrum should have a large T dependence [8].

Usually, 1/f noise is associated with the onset of a spin-glass phase and its kinetics at low T [19,20]. However, recent Monte Carlo simulations [21] have ruled this out for magnetization noise in SQUIDs. Though their [21] Ising-spin-glass-model with random nearest neighbor interactions (NNIs) qualitatively reproduced some experimental features, it did not show cross correlations between inductance noise and flux noise since spin glasses preserve time reversal symmetry.

Though, microscopically, this magnetization noise is not fully understood, phenomenologically, the 1/f noise arises from randomly fluctuating two-level-systems (TLSs). This picture has helped provide explanations in terms of metal induced gap states [22], random hopping between traps [23], other hopping conductivity models [24,25], insulator’s dangling bond states [26], and fractal spin structures [27]. However, a self-consistent comprehensive explanation of all the experimental features is still lacking.

Typically in the experiments, the dc SQUIDs have an amorphous Al2O3 insulating layer deposited on the surface...
of a metal (commonly Nb [8] or Al [7]). Al₂O₃ is likely to cluster on the surface before filling in and forming a homogeneous layer due to its higher binding energy which could lead to the Volmer-Weber growth mode. The lattice mismatch between the insulator and the metal could also lead to the formation of clusters. Near the metal surface, the clusters can host a number of point defects in the form of O vacancies that can capture one electron—Farbe + (F⁺) center, or two (F center).

In a related development, a few years ago, surface ferromagnetism (SFM) was reported in thin films and nanoparticles of a number of, otherwise, insulating metallic oxides [28] (including Al₂O₃) where the materials were not doped with any magnetic impurities. Further recent investigations attribute this room temperature SFM in Al₂O₃ nanoparticles [29] to F⁺ centers where it was found that amorphous Al₂O₃ is more likely to host the number of (including Al₂O₃) magnetism (SFM) was reported in thin films and nanoparticles (center, or two (F center).

Further recent investigations attribute this room temperature SFM in Al₂O₃ nanoparticles [29] to F⁺ centers where it was found that amorphous Al₂O₃ is more likely to host the number of F⁺ centers to cross the magnetic percolation threshold than the crystalline variant.

The origin of SFM in these, otherwise, nonmagnetic metal oxides is itself somewhat controversial [30]. Some of the suggested mechanisms include exchange coupling from F⁺ center induced impurity bands [31], F⁻ center mediated superexchange [32], and spin triplets at the Farbe center [33]. In addition to this, in the SQUID geometry, because of the proximity to the metal, these local magnetic moments can spin polarize the metal’s conduction band electrons which can lead to a RKKY-type long range interaction mechanism, which was first pointed out by Faoro and Ioffe [34]. This can likely lead to competing interaction mechanisms.

This Letter shows that 1/f noise, with αT ≤ 1 at low T, arises naturally from a spin-cluster defect model with interacting TLSs and different cluster size distributions. It is shown that ferromagnetic short-range-mechanisms will lead to flux noise that varies considerably more with T than those local magnetic moments can spin polarize the metal’s conduction band electrons which can lead to a RKKY-type long range interaction mechanism, which was first pointed out by Faoro and Ioffe [34]. This can likely lead to competing interaction mechanisms.

The general system Hamiltonian is

\[ H(s) = -\frac{1}{2} \sum_{i,j} J_{ij} s_i s_j - B \sum_i s_i, \]

where \( B \) is the magnetic field (\( B = 0 \) here) and \( J_{ij} \) is the interaction between the \( i \)th and \( j \)th Ising spins. Now, for \( N \) interacting spins, any \( n \)th order correlation function can be calculated as follows:

\[ \langle s_1(t_1) s_2(t_2), \ldots, s_k(t_k) \rangle = \langle \mathbf{f} | \sigma_z^{(s_j)} W(t_2), \ldots, \sigma_z^{(s_j)} W(t_1) | \mathbf{i} \rangle, \]

where the spin indices \{\( i, j, \ldots, k \)\} \( \in \{1, 2, \ldots, N\} \), \( | \mathbf{i} \rangle = | \mathbf{f} \rangle \) are the initial and final state vectors that correspond to the equilibrium distribution (i.e., \( | \mathbf{W} | = | \mathbf{i} \rangle \)). It is implied that \( \sigma_z^{(s_j)} = \sigma_1^{(s_j)} \otimes \sigma_2^{(s_j)} \otimes \ldots \otimes \sigma_N^{(s_j)} \), where \( \sigma_z \) is the
\( z \)-Pauli matrix and \( \sigma_n \) is the identity. For just two spins, if all \( \gamma_i = 1 \) (which is subsequently followed for all calculations) the two-point correlation functions are

\[
\langle s_i(0)s_j(t) \rangle = \frac{1}{2} e^{-2\Gamma_{\pm}|t|} + \left( \delta_{ij} - \frac{1}{2} \right) e^{i\text{Re}e^{-2\Gamma_{\pm}|t|}},
\]

where \( \Gamma_{\pm} = [1 + \exp(\pm 2\mu J)]^{-1} \). Whereas, if \( \gamma_i \) is retained, then \( \lim_{\mu \to 0} \langle s_i s_j \rangle = \delta_{ij} e^{-\gamma_i |t|} \) is obtained from this model.

Within a single cluster, the spins interact via an oscillatory RKKY-like form with a ferromagnetic \( J_o \), \( J_{ij} = J_o k_F R_{ij} \cos(k_F R_{ij}) - \sin(k_F R_{ij}) \) \( \gamma \) \( J_{ij} \) is the separation between two spins (on a lattice of lattice constant \( a \), \( k_F \) is a Fermi wave-vector-type parameter. For the calculations here \( J_o \approx 10^{11} \text{ Hz} / \text{m} \) is taken as a fitting parameter independent of \( k_F \).

**Discussion.**—Because of the high estimated areal spin density, the spontaneous magnetization of the spins strongly flux couples to the SQUID. The fluctuation-dissipation theorem relates the magnetization noise spectrum to the imaginary part of the susceptibility [16,42,43]. If all the NNIs randomly vary for each cluster. Now, \( 1 / f \) noise (see Fig. 3) only can be obtained in the spin-cluster model if the NNIs have a 1/\( J \)-type distribution. If the RKKY \( J_{ij} \) is expanded for small \( k_F \), then \( J_{ij} \propto 1/R_{ij} \); hence, a uniform distribution of \( R_{ij} \) results in a 1/\( J_{ij} \) distribution of interaction strengths at a certain crossover length 1/\( k_F \).

Though this short-range interaction model also self-consistently produces 1/\( f \)-like flux noise, and the integrated \( \alpha \) also increases with decreasing \( T \) (seen in both models and in agreement with experiment [13]), the flux noise’s variations with \( T \), here, are quite large (see Fig. 3). Moreover, the noise is 1/\( f \)-like only over a short range. Hence, in view of the experiments [8], it is unlikely that the short range interactions are the dominant cause of SFM as in the case of metal oxide nanoparticles [31,32,46].

FIG. 2 (color online). (a) Temperature dependent net correlation function for 400 spin clusters, each with 6–9 spins with ferromagnetic(+)\( J_{ij} \) RKKY interactions. Note that the overall results are independent of cluster size [48]. (b) Corresponding flux noise spectrum showing 1/\( f^\alpha \) noise below \( \sim 0.1 \) Hz and (c) its respective slope \( \alpha \), where \( \alpha \approx 1 \) below about 0.1 Hz. (d) Distribution of \( k_F a \) for each cluster, with mean \( \langle k_F a \rangle \) and standard deviation (2) as indicated.

![FIG. 2](image-url)

FIG. 3 (color online). (a) Sample flux noise power spectrum \( P_\phi \) and (b) its slope for 400 spin clusters with nearest neighbor ferromagnetic interactions that vary randomly for each cluster.
In the experiments of Ref. [8], the inductance noise was measured below $2K$ where the inductance noise ($P_L$) was mostly dominated by the imaginary part of the susceptibility and varied considerably with $T$. $P_L$ is the associated noise spectrum or the second spectrum, which is a quantitative measure of the spectral wandering of the first spectrum and is interpreted as the noise of the noise [47]. The first spectrum (flux noise) is related to the imaginary part of the susceptibility via the fluctuation-dissipation theorem $P_L(\omega) \approx 2k_BT\chi''/\omega$. Assuming all spins couple to the SQUID equally [40], the imaginary part of the inductance within a layer of thickness $d = \rho/n$ on the surface as $L'' = \mu_d d\frac{\partial}{\partial \omega} \chi''$; therefore,

$$P_L(\omega) = \left(\mu_d d\frac{\partial}{\partial \omega} \chi''\right)^2 \int_0^\infty \langle \chi'(0)\chi'(t) \rangle e^{\omega t} dt,$$

and from the fluctuation dissipation theorem,

$$\chi''(\omega) = 2\frac{\tilde{\mu}_e\mu_B^2}{k_BT} \sum_{i,j} \int_0^\infty \langle s_i(0) s_j(t) \rangle e^{\omega t} dt.$$  

It can be argued that, even with the interactions, the following sum can always be decomposed as $\sum \langle s_i(0) s_j(t) \rangle = \sum C_{ij} e^{-\Delta T}$ [see e.g., Eq. (4)]. Hence,

$$\chi''(\omega) = 2\frac{\tilde{\mu}_e\mu_B^2}{k_BT} \sum_{i,j} \int_0^\infty \langle s_i(0) s_j(t) \rangle e^{\omega t} dt.$$  

while Kramers-Kronig relation gives $\chi'(\omega) = \frac{\chi''(\omega)}{\omega}$. Now, from the total $\chi = \chi' + i\chi''$, it explicitly follows that:

$$P_L(\omega) = \frac{1}{k_BT} \left(2\rho\mu_d^2\mu_B^2\right)^{(3/2)} \int_0^\infty \int_0^\infty \int_0^\infty \sum_{i,j,k} \langle s_i(t_1) s_j(t_2) s_k(t_3) \rangle e^{\omega t_1} e^{\omega t_2} e^{\omega t_3} dt_1 dt_2 dt_3.$$  

where $\tau = t_2 - t_1, \tau' = t_3 - t_2, \omega_\perp = \omega \pm \omega', \omega_\parallel = \omega_\perp \pm \omega$, and $\omega_\parallel - \omega_\perp$ defines the bandwidth. In the experiments, $P_L(\omega)$ was found to be inversely proportional to $T$ and $\sim 1$ roughly below 100 mK and $P_L(\omega)$ depends on the sum of all three-point correlation functions (TPCFs). As inductance is even under time inversion and magnetic flux is odd, the flux-inductance-TPCF can only be nonzero if time reversal symmetry is broken, indicating the appearance of long range magnetic order. This indicates that the interactions must be ferromagnetic. To show this, the TPCF (for all possible spin
combinations) is calculated by for a single cluster of 10 spins with ferromagnetic RKKY interactions, which gives $\sum (s_i \cdot s_j)^\text{max} \sim 1$ at low $T$ and keeps decreasing with increasing $T$ (see Fig. 5). This is in excellent agreement with experiment and verifies that the same mechanism produces both the flux noise and inductance noise. Whereas, if $J_o$ is antiferromagnetic, then $\sum (s_i \cdot s_j)^\text{max} \sim 0$.

In summary, a spin cluster model, with long range ferromagnetic RKKY interactions, explains various puzzling features of magnetization noise in SQUIDs. The method suggested here for obtaining $n$-point correlation functions was key. This model is fully self-consistent with no a priori assumptions on the distribution of fluctuation rates for $1/f^\alpha$ noise and the results are independent of cluster size [48]. While the inductance noise is shown to be inherently $T^{-2}$ dependent, the flux noise is not. The existence of a magnetically ordered phase in this system is further established.

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