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 $1/f^{\alpha}$ magnetization noise [with $\alpha(T) \lesssim 1$] 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 $1/f^{\alpha}$ [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 Al_2O_3 insulating layer deposited on the surface

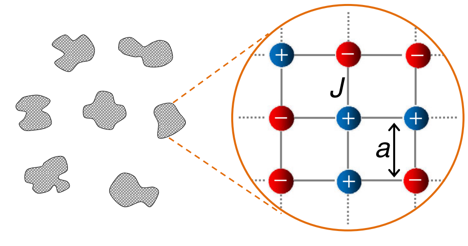

FIG. 1 (color online). Proposed $1/f^{\alpha}$ noise model consisting of interacting and fluctuating TLSs in a cluster. The clusters are assumed to form due to random defects at the SQUID's metal-insulator interface and are sufficiently far apart so that only spins within a single cluster interact. Number of TLSs within a cluster and the lattice constant a vary.

of a metal (commonly Nb [8] or Al [7]). Al_2O_3 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 Al_2O_3) where the materials were not doped with any magnetic impurities. Further recent investigations attribute this room temperature SFM in Al_2O_3 nanoparticles [29] to F^+ centers where it was found that amorphous Al_2O_3 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 F 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 $1/f^\alpha$ noise, with $\alpha(T) \lesssim 1$ 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 when compared to clusters with long range interactions. It is analytically shown why the inductance noise (second spectrum) will inherently have a huge T dependence even though the flux noise (first spectrum) may not. And time reversal symmetry is shown to be spontaneously broken at low T from three-point flux noise and inductance noise cross-correlation calculations.

An Ising-Glauber spin cluster model (see Fig. 1) is proposed here, and a method is introduced to systematically obtain arbitrary n-point correlation functions and subsequent noise spectral functions. These calculations are self-consistent; the usual heuristic assumption on the $1/\gamma$ distribution of switching rates required for $1/f$ noise [35]) is avoided.

There are also other indications of ferromagnetic spin clustering in experiments which best explain the integrated mean square flux noise's inverse T dependence [13] and why the measured spin paramagnetism follows a Curie T^{-1} scaling only up to a certain point and saturates thereafter [36].

Model and Method.—A schematic of our model is shown in Fig. 1 where each cluster comprises of an interacting 2D spin lattice. To model the randomness of the surface defects, randomly varying lattice constants are considered. Individual clusters are assumed to be sufficiently far apart so that there are no interactions across the clusters, but there could be an effective mean field. For the infinite range Ising-Glauber model, all N spins within a single cluster interact with every other via a RKKY-type mechanism.

The temporal dynamics for N interacting spins is governed by the master equation $\dot{\mathbf{W}}(t) = \mathbf{V}\mathbf{W}(t)$ [37], where $\mathbf{V}$ is a matrix of transition rates (such that the sum of each of its columns is zero) and $\mathbf{W}$ is the flipping probability matrix for the spins. Each spin's random fluctuation is a temperature (or interaction) driven process governed by Glauber dynamics. In this Markov process, the new spin distribution depends only on the current configuration and the new and old spin configurations agree everywhere except at a single site. Overall, the nonequilibrium spin dynamics for a system of correlated spins can be treated this way [38]. The matrix elements of $\mathbf{V}$, giving the conditional probability of a single spin flip, are

$$\mathbf{V}(\mathbf{s}\to\mathbf{s}') = \begin{cases} \dfrac{\gamma e^{-\beta H(\mathbf{s}')}}{e^{-\beta H(\mathbf{s})} + e^{-\beta H(\mathbf{s}')}} & \text{for } \mathbf{s}\neq\mathbf{s}' \text{ and } \\ & \sum_i (1 - s_i s_i') = 2 \\ -\sum_{\mathbf{s}\neq\mathbf{s}'} V(\mathbf{s}\to\mathbf{s}') & \text{for } \mathbf{s}=\mathbf{s}' \end{cases} \quad (1)$$

Here, $\mathbf{s}'(\mathbf{s})$ is a vector that denotes the present (earlier) spin configuration, and γ is the flipping rate of a spin (all $\gamma = 1$ in this Letter). The non-negative off-diagonal matrix elements satisfy the detailed balance condition and the negative sum of the off-diagonal column terms on the diagonal ensures the conservation of probability. The temporal dynamics requires evaluating the 2^N dimensional $\mathbf{W} = \exp(-\mathbf{V}t)$ matrix. The zero eigenvalues of $\mathbf{V}$ correspond to the equilibrium distribution, whereas the real and negative eigenvalues also eventually tend to the equilibrium distribution as $t \to \infty$ [37]. This model also provides the quasi-Hamiltonian method [39–41] with a connection to the underlying noise microscopics.

The general system Hamiltonian is

$$H(\mathbf{s}) = -\frac{1}{2}\sum_{i,j} J_{i,j} s_i s_j - B\sum_i s_i, \quad (2)$$

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

$$\langle s_i(t_1) s_j(t_2), \ldots, s_\kappa(t_n)\rangle = \langle \mathbf{f}|\sigma_z^{(\kappa)}\mathbf{W}(t_n), \ldots, \sigma_z^{(i)}\mathbf{W}(t_1)|\mathbf{i}\rangle, \quad (3)$$

where the spin indices $\{i, j, \ldots, \kappa\} \in \{1, 2, \ldots, N\}$, $|\mathbf{i}\rangle = |\mathbf{f}\rangle$ are the initial and final state vectors that correspond to the equilibrium distribution (i.e., $\mathbf{W}|\mathbf{i}\rangle = |\mathbf{i}\rangle$). It is implied that $\sigma_z^{(\kappa)} = \underset{1}{\sigma_o} \otimes \underset{2}{\sigma_o} \ldots \underset{\kappa-1}{\sigma_o} \otimes \underset{\kappa}{\sigma_z} \otimes \ldots \underset{N}{\sigma_o}$ where σ_z is the

z-Pauli matrix and σ_o 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

$$\langle s_i(0)s_j(t)\rangle = \frac{1}{2}e^{-2\Gamma_-|t|} + \left(\delta_{ij} - \frac{1}{2}\right)e^{4\beta J}e^{-2\Gamma_+|t|}, \qquad (4)$$

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

Within a single cluster, the spins interact via an oscillatory RKKY-like form with a ferromagnetic J_o, $J_{ij} = J_o[k_F R_{ij}\cos(k_F R_{ij}) - \sin(k_F R_{ij})]/(k_F R_{ij})^4$ where R_{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 $J_o \approx 10^{11}$ Hz$/\hbar$ is taken as a fitting parameter independent of k_F.

Discussion.—Because of the high estimated areal spin density, the coherent 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 surface spins couple to the SQUID equally, the flux noise from the ℓth spin cluster is

$$P_\phi^{(\ell)}(\omega) = 2\mu_o^2\mu_B^2\frac{\rho}{\pi}\frac{R}{r}\int_0^\infty \sum_{i,j=1}^{N}\langle s_i(0)s_j(t)\rangle e^{\iota\omega t}dt, \qquad (5)$$

where R is the radius of the loop, r is the radius of the wire, $R/r = 10$ [16], and ρ is the surface spin density. Using Eq. (3), $\sum\langle s_i(0)s_j(t)\rangle$ is calculated considering all combinations of two-point autocorrelation ($i=j$) and cross correlation ($i \neq j$) functions for a given cluster. Each cluster is assumed to be sufficiently far apart and noninteracting, and the total flux noise power is $P_\phi(\omega) = \sum_\ell P_\phi^{(\ell)}(\omega)$.

The T-dependent flux noise and the respective noise slopes are shown in Fig. 2. Four hundred spin clusters were considered with 6–9 spins each. Each cluster is assigned a random $k_F a$ with a uniform distribution [see Fig. 2(d)]. Note that, from the estimated areal surface spin density of $\rho \sim 5\times 10^{17} m^{-2}$ [8,36], one can estimate k_F and the average spin separation $\langle a\rangle$.

The $1/f^\alpha$ flux noise ($\alpha \sim 1$) appears at an intermediate range of frequencies. At high frequencies, the noise slope of 2 corresponds to the Lorentzian tail. The upper and lower cutoff frequencies for the $1/f$ noise depends on the distribution of $k_F a$ and J_o (or T). Eventually, for all T, $\alpha \to 0$ at very low frequencies. Note that, in the absence of interactions in this model, the noise spectrum is just a simple Lorentzian.

The flux noise's weak T dependence, shown in these calculations, agrees with experiment [8]. However, in Ref. [2], P_ϕ was not necessarily independent of T under all circumstances. Above 1 K, depending on the construction for the same set of materials, P_ϕ could sometimes vary with T. In the present model, these types of spectral features can originate from the interacting random spin clusters. Note that conflicting T dependencies of $1/f$ noise are known to exist for glassy systems [44,45].

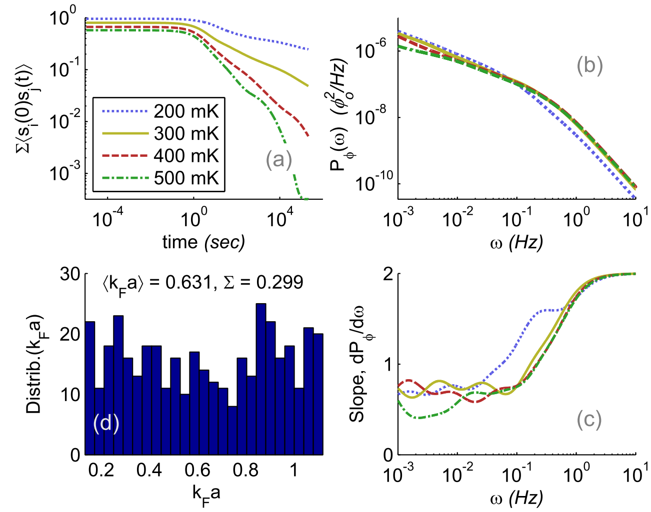

FIG. 2 (color online). (a) Temperature dependent net correlation function for 400 spin clusters, each with 6–9 spins with ferromagnetic($+|J_o|$) RKKY interactions. Note that the overall results are independent of cluster size [48]. (b) Corresponding flux noise spectrum showing $1/f^\alpha$ noise below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

Next, consider short range ferromagnetic NNIs $\mathcal{J}$ that randomly vary for each cluster. Now, $1/f$ noise (see Fig. 3) can only be obtained in the spin-cluster model if the NNIs have a $1/\mathcal{J}$-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 $1/f^\alpha$ flux noise, and the integrated α 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].

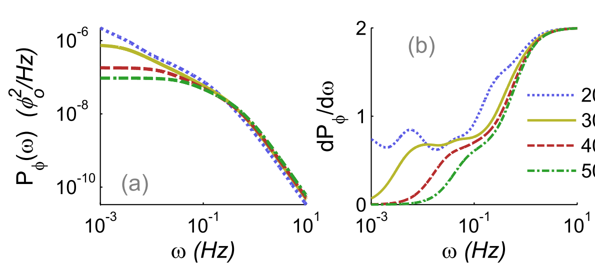

FIG. 3 (color online). (a) Sample flux noise power spectrum P_ϕ and (b) its slope for 400 spin clusters with nearest neighbor ferromagnetic interactions that vary randomly for each cluster.

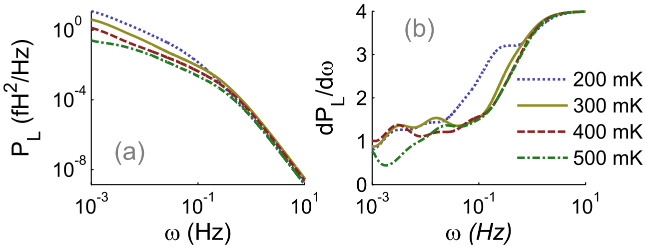

FIG. 4 (color online). (a) Power spectrum of the inductance noise, P_L (associated noise of P_ϕ of Fig. 2) and (b) its respective slope (α) for the 400 spin clusters with ferromagnetic ($+|J_o|$) RKKY interactions. The integrated $\langle\alpha\rangle$ between 0.001 and 0.05 Hz is $\{1.569, 1.415, 1.247, 1.242\}$ for the respective temperatures in ascending order.

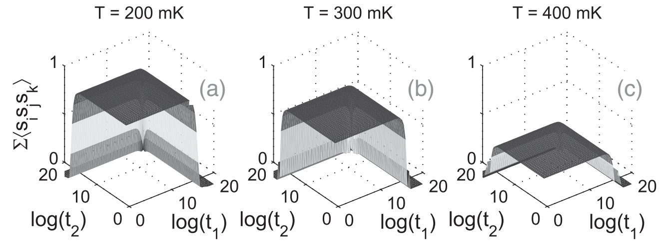

FIG. 5. Normalized sum of all three-point correlation functions, $\sum\langle s_i(0)s_j(t_1)s_k(t_2)\rangle/N^3$, indicating flux-inductance-noise cross correlation for a single cluster of $N=10$ spins with ferromagnetic ($+|J_o|$) RKKY interactions at temperatures of (a) $T=200$ mK (b) $T=300$ mK, and (c) $T=400$ mK.

In the experiments of Ref. [8], the inductance noise was measured bellow $2K$ 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 $P(\omega)\approx 2k_BT\chi''/\omega$. Assuming all spins couple to the SQUID equally [40], the imaginary part of the inductance then relates to the spin susceptibility within a layer of thickness $d=\rho/\tilde{n}$ on the surface as $L''=\mu_o d\frac{R}{r}\chi''$; therefore,

$$P_L(\omega)=\left(\mu_o d\frac{R}{r}\right)^2\int_0^\infty\langle\chi(0)\chi(t)\rangle e^{\imath\omega t}dt, \quad (6)$$

and from the fluctuation dissipation theorem,

$$\chi''(\omega)=2\frac{\tilde{n}\mu_o\mu_B^2\omega}{k_BT}\sum_{i,j}\int_0^\infty\langle s_i(0)s_j(t)\rangle e^{\imath\omega t}dt. \quad (7)$$

It can be argued that, even with the interactions, the following sum can always be decomposed as $\sum\langle s_i(0)s_j(t)\rangle=\sum C_\nu e^{-2\Gamma_\nu t}$ [see e.g., Eq. (4)]. Hence,

$$\chi''(\omega)=2\tilde{n}\mu_o\mu_B^2\frac{\omega}{k_BT}\sum_\nu\frac{C_\nu\Gamma_\nu}{\Gamma_\nu^2+\omega^2}, \quad (8)$$

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

$$\chi(t)=\int_0^\infty\chi(\omega)e^{\imath\omega t}d\omega=\frac{2\tilde{n}\mu_o\mu_B^2}{k_BT}\sum_{i,j}\langle s_i(0)s_j(t)\rangle. \quad (9)$$

The inductance noise can then be explicitly expressed in terms of the spectral density of the dynamical four-point noise correlation functions [35],

$$P_L(\omega)=\left(2\rho\frac{\mu_o^2\mu_B^2}{k_BT}\frac{R}{r}\right)^2\iint_{\omega_a}^{\omega_b}S^{[2]}(\omega,\omega_1,\omega_2)d\omega_1d\omega_2, \quad (10)$$

where $S^{[2]}(\omega,\omega_1,\omega_2)=\iiint_0^\infty\sum\langle s_i(t_1)s_j(t_2)s_k(t_3)s_l(t_4)\rangle e^{\imath(\omega_1-\omega)\tau'}e^{\imath(\omega_2+\omega)\tau''}e^{\imath\omega\tau}d\tau'd\tau''d\tau$, here $\tau'=t_2-t_1$, $\tau''=t_4-t_3$ and $\tau=t_4+t_3-t_2-t_1$. $\Delta\omega=\omega_b-\omega_a$ is the bandwidth within which the second spectrum is observed. The T-dependent inductance noise spectrum and its slope are shown in Fig. 4, where $\Delta\omega$ covers the full spectrum. The noise spectrum now shows $1/f^\alpha$ behavior at intermediate frequencies where the average integrated α between 0.001 and 0.05 Hz varies from $\sim$1.57 (at 200 mK) to $\sim$1.24 (at 500 mK). For high T, the $1/f$ type feature disappears. The $\alpha=4$ at high ω is due to the square of the Lorentzian tail while at the lowest ω, $\alpha\to 0$, the onset of which is again T dependent. Overall the T-dependent inductance noise behavior for the spin cluster model agrees very well with experiment [8,17].

Finally, the SQUID's surface spins show a net polarization in the experiments [8] as the $1/f$ inductance noise was found to be highly correlated with the $1/f$ flux noise. The following expression gives the flux and inductance noise cross power spectrum:

$$P_{L\phi}(\omega)=\frac{1}{k_BT}\left(2\rho\mu_o^2\mu_B^2\frac{R}{r}\right)^{(3/2)}\int_{\omega_a}^{\omega_b}\iint_0^\infty\sum_{i,j,k}\langle s_i(t_1)s_j(t_2)s_k(t_3)\rangle e^{\imath\omega_-\tau}e^{\imath\omega_+\tau'}d\tau d\tau' d\omega', \quad (11)$$

where, $\tau=t_2-t_1$, $\tau'=t_3-t_2$, $\omega_\pm=\omega\pm\omega'$, and $\omega_b-\omega_a$ defines the bandwidth. In the experiments, $P_{L\phi}$ was found to be inversely proportional to T and $\sim$1 roughly below 100 mK and $P_{L\phi}$ 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 $\sum\langle s_i s_j s_k\rangle_{\max} \sim 1$ 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 $\sum\langle s_i s_j s_k\rangle_{\max} \sim 0$.

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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