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Modeling flux noise in SQUIDs due to hyperfine interactions

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Recent experiments implicate spins on the surface of metals as the source of flux noise in SQUIDs, and indicate that these spins are able to relax without conserving total magnetization. We present a model of $1/f$ flux noise in which electron spins on the surface of metals can relax via hyperfine interactions. Our results indicate that flux noise would be significantly reduced in superconducting materials where the most abundant isotopes do not have nuclear moments such as zinc and lead.

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Although there have been significant advances in superconducting qubits, they continue to be plagued by noise and decoherence. Low frequency $1/f$ flux noise [1] in superconducting quantum interference devices (SQUIDs) is one of the dominant sources of noise in superconducting flux [2, 3] and phase [4] qubits. Recent experiments indicate this flux noise arises from the fluctuations of spins residing on the surface of normal metals [5] and superconductors [6]. These spins have a high density ($\sim 5 \times 10^{17}$ m^{-2}), and may arise from local electron moments in localized states at the metal-insulator interface [7].

One early model of flux noise due to spins proposed that the spin of an electron in a surface trap is fixed, but that the orientation of the spin can change when the electron hops to a different trap [8]. However, the density of defect traps needed to explain the experiments was orders of magnitude larger than what is estimated to exist in a typical glassy material [4]. Another model suggested that spin flips of paramagnetic dangling bonds occurred as a result of interactions with tunneling two-level systems mediated by phonons [9]. However, to obtain $1/f$ flux noise, the maximum two level system energy splitting would have to be a few mK which is orders of magnitude smaller than accepted values.

There is some experimental indication of interactions between the spins [6] leading to the theoretical suggestion that flux noise is the result of spin diffusion via Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions [10] between the spins [11]. RKKY interactions between randomly placed spins produce spin glasses, and Monte Carlo simulations of Ising spin glass systems show that interacting spins produce 1/f flux and inductance noise [12].

In addition, RKKY interactions conserve the total spin and magnetization, so the total magnetization should not change. However, the Stanford group measured the total magnetization of small isolated gold rings and found that the total magnetization is not conserved since the magnetization follows the externally applied ac magnetic field [5]. While this does not rule out magnetization conserving interactions such as RKKY, it does imply that the spins must (also) be involved in interactions that do not conserve total magnetization. (The angular momenta contained in the external magnetic field and in the electric current induced in the gold ring are orders of magnitude too small to conserve total angular momentum by accomodating the change in angular momentum of the spins associated with the change in total magnetization.)

The 3 possible interactions that do not conserve total magnetization are spin-orbit, magnetic dipole-dipole, and hyperfine interactions. The orbital angular momentum of a neutral gold atom is zero, so we can ignore spin-orbit interactions. The dipole-dipole interaction between 2 electrons is of order 1 mK if we use a separation of 1.4 nm corresponding to a spin density of 5×10^{17} m^{-2}. This is much smaller than the hyperfine contact interaction which is of order 70 mK in hydrogen, for example. This implies that the hyperfine interaction dominates. In support of this is the fact that scanning SQUID microscope experiments [5] found that the magnetic susceptibility of spins on silicon is 5 to 20 times smaller than that of metals and insulating AlO_x. This is consistent with hyperfine interactions since the only isotope of silicon that has a nuclear spin and hence hyperfine interactions is ^{29}Si which has a natural isotopic abundance of 5%. Note that spin angular momentum is conserved in hyperfine interactions where there is a spin flip exchange between the nuclear spin and the electron spin, but the magnetic moment is not conserved since the gyromagnetic ratios of the nuclear and electron spins differ by about 3 orders of magnitude. Previous authors [8, 11] have pointed out that flux noise cannot be directly due to fluctuating nuclear spins because the frequency range and magnitude of nuclear flux noise would be much lower than what is seen experimentally. However, this does not rule out the possibility that the electron spins that are responsible for flux noise can relax via hyperfine interactions with nearby nuclear spins.

In this paper we present a model of flux noise in which electrons residing in harmonic traps undergo spin exchange with nearby nuclei via the hyperfine contact interaction. The relaxation time T_1 of a given electron spin is dominated by exchange with the nearest nonzero nuclear moment. In materials where not all the isotopes have a nuclear moment, the distance to the nearest nucleus

with a nuclear spin could be quite large. For example, the only isotope of Pb with a nuclear moment is ^{209}Pb which has a 22% natural isotopic abundance. The distribution of distances between trapped electrons and the nearest nucleus with a magnetic moment gives rise to a distribution of electron spin relaxation times T_1, which in turn results in 1/f noise up to 10 MHz. Unlike the model [11] of spin diffusion via RKKY that found white noise at low frequencies in contradiction to experiment, we find that 1/f flux noise extends down to 10^{-5} Hz.

The Hamiltonian of an electron spin $\mathbf{S}$ that is in an external field $\mathbf{H}_{\rm ext}$ and that has a contact hyperfine coupling to nearby nuclear spins $\mathbf{I}_i$ is given by [13]

$$\begin{aligned}\mathcal{H} &= \mathcal{H}_0 + \mathcal{H}_{hyp}\\ \mathcal{H}_0 &= -g\mu_B\ \mathbf{H}_{\rm ext}\cdot\mathbf{S}\\ &\equiv -\hbar\omega_0 S_z\\ \mathcal{H}_{hyp} &= \sum_i \frac{8\pi}{3}\frac{\mu_0}{4\pi} g_0\mu_B\gamma_n\hbar\mathbf{I}_i\cdot\mathbf{S}\delta(\mathbf{r}-\mathbf{r_i}),\end{aligned} \tag{1}$$

where $I_z = \pm 1$ and $S_z = \pm 1$. We ignore the dipolar interaction between the electron and the nuclear spins because it is much smaller than the contact hyperfine interaction [14]. We choose $\mathbf{H}_{\rm ext}$ parallel to $\hat{z}$. μ_0 is the permeability constant. The external field could be due to an applied external magnetic field or to the magnetic field produced by local electric currents. $\mathbf{r}_i$ and $\mathbf{r}$ are the coordinates of the i-th nuclei and the electron, g_0 is the free-electron g-factor and γ_n is the nuclear gyromagnetic ratio. μ_B is the Bohr magneton of the electron. Taking the expectation value of $\mathcal{H}_{hyp}$ with respect to the electron wavefunction $\psi(r)$ yields

$$\langle\mathcal{H}_{\rm hyp}\rangle_e = \frac{2}{3}\mu_0 g_0\mu_B\gamma_n\hbar\sum_i \mathbf{I}_i\cdot\mathbf{S}|\psi(\mathbf{r_i})|^2, \tag{2}$$

where $\psi(\mathbf{r_i})$ is the wavefunction of the electron at the position of the ith nucleus. $\langle\rangle_e$ indicates the expectation value of the Hamiltonian $\mathcal{H}_{\rm hyp}$ with respect to the electron wavefunction. This can be written in the standard form of a hyperfine interaction:

$$\langle\mathcal{H}_{\rm hyp}\rangle_e = \sum_i (A^i_{\rm hf}\ \mathbf{I_i})\cdot\mathbf{S} \tag{3}$$

$A^i_{\rm hf}$ is the hyperfine coupling constant between the ith nuclear spin and the electron spin. A typical hyperfine frequency, e.g., for hydrogen, is $f_{hf} = A_{\rm hf}/h \sim 1.4$ GHz where h is Planck's constant. We can also express this in terms of an effective random field $\mathbf{H}^i_{\rm R}$ produced by the ith nuclear spin:

$$\langle\mathcal{H}_{\rm hyp}\rangle_e = \sum_i g_0\mu_B\mathbf{H}^i_{\rm R}\cdot\mathbf{S} \tag{4}$$

where

$$\mathbf{H}^i_{\rm R} = \frac{2}{3}\mu_0\gamma_n\hbar|\psi(\mathbf{r_i})|^2\mathbf{I_i} \tag{5}$$

To find the effective field $\mathbf{H}^i_{\rm R}$ on the electron due to the ith nucleus, we use Eq. (5) and assume that the electron is in a harmonic trap with a ground state wavefunction

$$\psi(r) = \frac{1}{\sqrt{\pi\xi^2}}e^{-r^2/2\xi^2} \tag{6}$$

where $\xi^2 = \hbar/m_e\omega$, m_e is electron mass and Ω is the frequency of the harmonic oscillator. (If we assume that the localized wavefunction decays exponentially as $\exp(-r/\xi_\ell)$ where ξ_ℓ is the localization length, then we still obtain $1/f$ noise up to logarithmic corrections in the frequency.) This gives

$$\begin{aligned}\mathbf{H}^i_{\rm R} &= \frac{2}{3}\frac{\mu_0\gamma_n\hbar}{\pi\xi^2}\mathbf{I}_i e^{-r_i^2/\xi^2}\\ &\equiv \frac{A_0}{\xi^2}\mathbf{I}_i e^{-r_i^2/\xi^2}\end{aligned} \tag{7}$$

where A_0 is a constant.

The nuclear spin dynamics can be characterized by a correlation time τ_0 that is roughly the time scale over which the nuclear spins keep their orientation [15]

$$\begin{aligned}C^i_j(t) &\equiv \left\langle H^i_{\rm R,j}(t+\tau)H^i_{\rm R,j}(t)\right\rangle\\ &= \left\langle (H^i_{\rm R,j})^2\right\rangle\exp(-|\tau|/\tau_0),\end{aligned} \tag{8}$$

where $j = x, y, z$ are the components of the random magnetic fields. The Fourier transform of this correlation function is

$$C^i_j(f) = \left\langle (H^i_{\rm R,j})^2\right\rangle\frac{\tau_0}{1+(2\pi f)^2\tau_0^2}, \tag{9}$$

We will regard τ_0 as a constant. If τ_0 is determined by spin diffusion via nuclear dipole-dipole interactions, then $\tau_0 \sim 1/Dq^2$ where D is the spin diffusion constant and $q = \pi/a$ where a is the typical distance between nuclear spins.

The electron spin dynamics is given by the Bloch equation [15, 16]:

$$\frac{d}{dt}\langle\mathbf{S}\rangle = -\mu_B\,\mathbf{H}_{\rm ext}\times\langle\mathbf{S}\rangle - \frac{1}{T_1}\langle S_z\rangle\,\hat{z} - \frac{1}{T_{2x}}\langle S_x\rangle\,\hat{x} - \frac{1}{T_{2y}}\langle S_y\rangle\,\hat{y}, \tag{10}$$

where T_1 is the spin-lattice relaxation time and T_2 is the spin-spin relaxation time defined by [15, 16]

$$\frac{1}{T_1} = \left(\left\langle (H^i_{\rm R,x})^2\right\rangle + \left\langle (H^i_{\rm R,y})^2\right\rangle\right)\frac{\tau_0}{1+\omega_0^2\tau_0^2}, \tag{11}$$

$$\frac{1}{T_{2x}} = \left\langle (H^i_{\rm R,z})^2\right\rangle\tau_0 + \left\langle (H^i_{\rm R,y})^2\right\rangle\frac{\tau_0}{1+\omega_0^2\tau_0^2}, \tag{12}$$

$$\frac{1}{T_{2y}} = \left\langle (H^i_{\rm R,z})^2\right\rangle\tau_0 + \left\langle (H^i_{\rm R,x})^2\right\rangle\frac{\tau_0}{1+\omega_0^2\tau_0^2}. \tag{13}$$

Here we used Eq. (9).

Each nuclear spin produces an electron relaxation time $T_1(\mathbf{r}_i)$ that we can obtain by plugging Eq. (7) into

Eq. (11) to yield:

$$\frac{1}{T_1(\mathbf{r}_i)} = \frac{C^2\tau_0}{\xi^4(1+\omega_0^2\tau_0^2)}e^{-2r_i^2/\xi^2} = \frac{b}{\xi^4}e^{-2r_i^2/\xi^2} \quad (14)$$

where C and b are constants. To find an effective relaxation rate $1/T_1^{\rm eff}$, we sum or integrate over all the nuclei that lie within the electron wavefunction. The relaxation rate will be dominated by the nuclear spin that is closest to the electron. Let r_n denote the distance between the center of the electron wavefunction and nuclear spin closest to it. Then we obtain

$$\frac{1}{T_1^{\rm eff}} = \sum_i \frac{1}{T_1(\mathbf{r}_i)} \to \frac{1}{a^2}\int_{r_n}^{\infty} d^2\mathbf{r}\frac{1}{T_1(\mathbf{r})} = \frac{\pi b}{2\xi^2 a^2}e^{-2r_n^2/\xi^2}, \quad (15)$$

where a is the lattice constant. We now need to average over all the localized electron spins on the surface. Since the electrons are uniformly and randomly distributed on the surface, r_n has a distribution $P(r_n)$. For a 2D square lattice, the distribution of r_n is $P(r_n) = 2\pi r_n/a^2$ for $0 \le r_n \le a/2$. The surface on which the spins sit can be disordered, possibly resulting in an exponent for r_n that is different from unity. However, it is reasonable to assume $P(r_n) = A_1 r_n^{\gamma-1}/a^\gamma$ where A_1 is a constant and $\gamma \in (2,4)$ (γ is of order the dimension). Since r_n depends logarithmically on $T_1^{\rm eff}$ for both an electron wavefunction in a harmonic trap and an exponentially localized electron, the actual form of $P(r_n)$ is not important. The resulting distribution of $T_1^{\rm eff}$ is

$$P(T_1^{\rm eff}) = \frac{A_1}{2^{\gamma/2+1}}\left(\frac{\xi}{a}\right)^\gamma\left[\ln\left(\frac{\pi b T_1^{\rm eff}}{2\xi^2a^2}\right)\right]^{\frac{\gamma-2}{2}}\frac{1}{T_1^{\rm eff}}. \quad (16)$$

We can simplify the above formula by approximating the slowly varying function $\ln\left(\pi b T_1^{\rm eff}/2\xi^2a^2\right)$ by its average value. Then the distribution function is inversely proportional to $T_1^{\rm eff}$, and we can write $P(T_1^{\rm eff}) = D_1/T_1^{\rm eff}$ where D_1 is a normalization factor determined by

$$\int_{T_{1,min}^{\rm eff}}^{T_{1,max}^{\rm eff}} dT_1^{\rm eff}P(T_1^{\rm eff}) = 1 \quad (17)$$

$\left(T_{1,\rm max}^{\rm eff}\right)^{-1}$ and $\left(T_{1,\rm min}^{\rm eff}\right)^{-1}$ correspond to the minimum and maximum frequencies of the flux noise, and are determined by $(r_n)_{\rm max}$ and $(r_n)_{\rm min}$, respectively. Thus we find $D_1 = \xi^2/2\left[(r_n^2)_{\rm max} - (r_n^2)_{\rm min}\right] \equiv \xi^2/2\Delta r_n^2$.

According to the Wiener-Khintchine theorem, the spectral density $S(\omega)$ of the noise is given by twice the Fourier transform of the autocorrelation function of the spin fluctuations. From the fluctuation-dissipation theorem, the low frequency ($\hbar\omega \ll kT$) spin noise is proportional to the imaginary part of the spin susceptibility that we can derive from the Bloch equations in Eq. (10) [15]. The frequency dependence of the noise at low frequencies is determined by the z-component of the susceptibility [17]. The resulting spin noise power is [18]

$$\mathcal{S}_z(\omega) = 2\int_{T_{1,\rm min}^{\rm eff}}^{T_{1,\rm max}^{\rm eff}} dT_1^{\rm eff}P(T_1^{\rm eff})\mathrm{sech}^2\left(\frac{\hbar\omega_0}{k_BT}\right)\frac{1/T_1^{\rm eff}}{\omega^2+(1/T_1^{\rm eff})^2} \approx \frac{\xi^2}{\Delta r_n^2}\mathrm{sech}^2\left(\frac{\hbar\omega_0}{k_BT}\right)\frac{\pi}{\omega}, \quad (18)$$

where the limits of integration have a wide range with $\omega T_{1,\rm min}^{\rm eff} \ll 1 \ll \omega T_{1,\rm max}^{\rm eff}$.

To relate $\mathcal{S}_z(\omega)$ to the flux noise, we need to know how a spin couples magnetically to the SQUID. The effective flux $\Phi_{\rm eff}$ produced by the spin magnetization on a loop with current I is [11]

$$\Phi_{\rm eff} = g\mu_B\int\frac{\hat{S}(\mathbf{r})B(\mathbf{r})}{I}d\mathbf{r}, \quad (19)$$

where $\hat{S}(\mathbf{r})$ is the surface spin density operator and $B(r)$ denotes the probing magnetic field due to the current and, if applicable, an externally applied field. Consider a SQUID made from a strip conductor (where the width of the strip is d) circular in shape with radius R (measured from the center of the loop to the middle of the annulus). (For a square SQUID with circumference L and width W, we replace R and d by $L/2\pi$ and W, respectively.) If the penetration depth λ is much smaller than the width, the current density at x near the center of the strip is $J(x) = 2I/(\pi d)[1-(2x/d)^2]^{-1/2}$ for $(-d/2)+\lambda < x < (d/2)-\lambda$ [11, 19]. This current density produces the magnetic field $B(x) = \mu_0 J(x)/2$. Using this in Eq. (19), we obtain the flux autocorrelation function [11, 20]:

$$\langle\Phi(t)\Phi(0)\rangle = \frac{(g\mu_B)^2R}{I^2}\int_{-\frac{d}{2}}^{\frac{d}{2}}d\mathbf{r}d\mathbf{r}'\left\langle\hat{S}(\mathbf{r},t)B(\mathbf{r})\hat{S}(\mathbf{r}',0)B(\mathbf{r}')\right\rangle \quad (20)$$

If we assume the spins are isolated,

$$\langle\hat{S}(\mathbf{r},t)\hat{S}(\mathbf{r}',0)\rangle = \Theta(\sqrt{A}-|\mathbf{r}-\mathbf{r}'|)\langle\hat{S}(\mathbf{r},t)\hat{S}(\mathbf{r},0)\rangle = \sigma^2\mathcal{S}_z(t)\Theta(\sqrt{A}-|\mathbf{r}-\mathbf{r}'|)$$

where $\sigma = 1/A$ is the spin surface density, A is the average area per spin, $\Theta(x)$ is a step-function, and $\mathcal{S}_z(t)$ is the spin fluctuation autocorrelation function. After integrating over $\mathbf{r}'$ (using $\int\Theta(\sqrt{A}-|\mathbf{r}-\mathbf{r}'|)f(\mathbf{r}')d\mathbf{r}' \approx Af(\mathbf{r})$ for an arbitrary function $f(\mathbf{r})$), we obtain

$$\langle\Phi(t)\Phi(0)\rangle = \sigma\frac{(g\mu_B\mu_0)^2}{\pi}\frac{R}{d}\ln\left(\frac{d}{2\lambda}\right)\mathcal{S}_z(t) \quad (21)$$

The associated flux noise spectrum is

$$\mathcal{S}_\Phi(f) = \sigma(g\mu_B\mu_0)^2\mathrm{sech}^2\left(\frac{\hbar\omega_0}{k_BT}\right)\frac{R}{d}\ln\left(\frac{d}{2\lambda}\right)\frac{\xi^2}{\Delta r_n^2}\frac{1}{2\pi f}, \quad (22)$$

which give rise to $1/f$ flux noise. Notice that the flux noise is proportional to the density σ of electron spins. Note also that for materials with a low concentration of nuclei with magnetic moments, Δr_n^2 will be larger and the flux noise, which goes as $1/\Delta r_n^2$, will be smaller. This is consistent with the small susceptibility found on silicon samples [5] where ^{29}Si is the only isotope with a nuclear moment and its natural isotopic abundance is only 5%.

Let us estimate the flux noise magnitude at 1 Hz for a Josephson junction. We can set the temperature factor to unity since $(\mu_B H_{\rm ext} = \hbar\omega_0) \ll k_B T$ for $H_{\rm ext}$ in the range of 1 to 100 G and T between 25 mK and 10 K. Since $(r_n)_{\rm min} \approx 0$ and we estimate that $(r_n)^2_{\rm max}/\xi^2 > 30$, we make the approximation that $\xi^2/\Delta r_n^2 = \xi^2/2\left[(r_n^2)_{\rm max} - (r_n^2)_{\rm min}\right] \approx 1/30$. Using $R/d = 10$, $\sigma = 5\times 10^{17}{\rm m}^{-2}$ [6], $\ln(d/2\lambda) \sim 8.5$, and $g\mu_0\mu_B \sim 11.3(\mu\Phi_0)({\rm nm})$, we estimate the amplitude of the flux noise to be $\mathcal{S}^{1/2}_{\Phi,{\rm hf}} \approx 5\ \mu\Phi_0/{\rm Hz}^{1/2}$. This agrees with experimental values which are typically in the range of 1 to 10 $\mu\Phi_0/{\rm Hz}^{1/2}$ [2, 21].

Eq. (22) gives the flux noise due to spins that are only on the surface of SQUIDs. However, paramagnetic spins have also been found on a diectric surface [5]. So if unpaired spins also reside on the substrate, these fluctuating spins will also contribute to the flux noise, reducing the dependence on d. Let L be the self-inductance of the SQUID. Then we can follow Wellstood [22] and use the expression for the electromagnetic energy $E = LI^2/2 = \int |B(r)|^2 d^2\mathbf{r}/(2\mu_0)$ to evaluate the integral in Eq. (20):

$$\mathcal{S}_\Phi(f) = \sigma\mu_0(g\mu_B)^2{\rm sech}^2\left(\frac{\hbar\omega_0}{k_B T}\right) L\frac{\xi^2}{\Delta r_n^2}\frac{1}{2\pi f}, \qquad (23)$$

The dependence of the flux noise on the geometry and the substrate are included in L. This result agrees with recent experiments [23] that found a nearly linear relationship between flux noise and L when the inductance of the SQUID was enhanced by inductor coils. Furthermore, our result also implies that flux noise and inductance noise should be correlated [24].

To summarize, we have presented a model of $1/f$ flux noise in which electron spins on the surface of metals relax via hyperfine interactions. Since the electron spin relaxation time depends exponentially on the distance between the electron and the nuclear spin, the nearest nuclear spin dominates the spin relaxation process. The distribution of distances results in a distribution $P(T_1^{\rm eff})$. Averaging over this distribution results in $1/f$ flux noise. Experimentally, the SQUIDs producing flux noise are in steady state equilibrium, so the noise is normally stationary and Gaussian. This is what we have assumed in our calculations. (Stationary means that the system, and hence the autocorrelation functions, are translationally invariant in time. For Gaussian processes, higher order correlation functions can be expressed as products of the two-point (lowest order) correlation functions [25].) Since the magnetization sums over individual spins, the magnetization noise, and hence the flux noise, is Gaussian if there are enough spins for the central limit theorem to apply. In both our model and experiment, non-Gaussian noise could arise in very small samples [26].

Our results indicate that flux noise would be significantly reduced in superconducting materials where the most abundant isotopes do not have nuclear moments such as zinc and lead. The only isotopes of zinc and lead that have nuclear moments are ^{67}Zn and ^{207}Pb which have natural isotopic abundances of 4% and 22%, respectively. Thus, compared to Nb SQUIDs, we would expect flux noise to be lower by roughly a factor of 25 and 5 in Zn and Pb SQUIDs since the relevant factor is $\xi^2/\Delta r_n^2$ in Eq. (22). This is assuming that Nb, Zn, and Pb have approximately the same atomic arrangement on their surface with approximately the same density of surface spins. For experimentally relevant values, the flux noise expression in Eq. (22) does not have any temperature dependence (${\rm sech}(\hbar\omega_0/{\rm k_B T}) \approx 1$ since $\hbar\omega_0 \ll k_B T$). This is consistent with a quantum process such as hyperfine exchange coupling, and with the plateau seen below 0.5 K in plots of the flux noise versus temperature [21]. The unusual temperature dependence of the flux noise that is experimentally found above 0.5 K [21] may involve thermal fluctuations of the spins.

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