Nonequilibrium Spin Noise and Noise of Susceptibility

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We analyze out-of-equilibrium fluctuations in a driven spin system and relate them to the noise of spin susceptibility. In the spirit of the linear response theory we further relate the noise of susceptibility to a 4-spin correlation function in equilibrium. We show that, in contrast to the second noise (noise of noise), the noise of susceptibility is a direct measure of non-Gaussian fluctuations in the system. We develop a general framework for calculating the noise of susceptibility using the Majorana representation of spin-1/2 operators. We illustrate our approach by a simple example of non-interacting spins coupled to a dissipative (Ohmic) bath.

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In the last decade similar effect has been observed in nanoscale quantum circuits. Remarkably, the noise magnitude appears to be “universal”, i.e., of the same order of magnitude for a wide range of device sizes. This noise is believed to originate from an assemblage of spins localized at the surface or interface layers. Indeed evidence for the existence of surface spins was obtained in several dedicated experiments. All these experimental findings are consistent with a surface spin density of $\sigma_S \sim 5 \cdot 10^{17}$ m$^{-2}$.

Sendelbach et al. recently developed an alternative method to measure properties of flux noise, namely by observing fluctuations in the inductance of SQUIDs. As the spin contribution to the inductance is determined by the spin susceptibility, this experiment essentially amounts to measuring the noise of the susceptibility. Technically this quantity also corresponds to a four-spin correlation function, which however is distinct from the correlation function describing the second noise. To the best of our knowledge, there is no consensus in the literature on how to define noise of susceptibility. Some authors employ the fluctuation-dissipation theorem and, thus, relate the noise of noise and the noise of susceptibility. This relation seems to be justified in cases where the system is controlled by a slowly fluctuating parameter, always remaining in a quasi-equilibrium. Nevertheless, there is a need for a more general definition of the noise of susceptibility at the microscopic level.

In this paper we focus on spin systems where we pursue the following issues: (i) we give a general definition of noise of susceptibility in terms of four-spin correlation functions and emphasize its distinction from the second noise; (ii) as an illustration, we compute the noise of susceptibility in a simple model of a single spin 1/2 immersed in a dissipative environment; (iii) in order to perform the above calculation, we further develop a powerful technique based on the Majorana-fermion representation of spin-1/2 systems; (iv) we use the above results to...
estimate the noise of susceptibility for the model of independent spins with a distribution of the relaxation rates that widely used to describe 1/f-noise (the Dutta-Horn model). The results of the latter calculation are incompatible with the experimental results of Ref. [30] and we conclude that the observed surface spins cannot be described by non-interacting models.

We find that the four-spin correlation function corresponding to the noise of susceptibility vanishes if evaluated for Gaussian fluctuating quantities. Hence a system of harmonic oscillators (photons or phonons) would show no fluctuations of susceptibility. Therefore the noise of susceptibility constitutes a direct measure of non-Gaussian fluctuations. In contrast, the second noise is present in Gaussian systems, where it is independent of frequency. Furthermore, in non-Gaussian systems the second noise always contains this trivial Gaussian contribution, which often masks the interesting non-Gaussian noise making the latter very hard to observe.

As an illustration of our general arguments, we consider a model of independent spins. The spin degrees of freedom are intrinsically non-Gaussian even in the absence of spin-spin interactions. Indeed, in this model we find a non-vanishing white noise of susceptibility, that scales as $N/T^2$, where $N$ is the number of spins and $T$ the temperature. At the same time the average susceptibility scales as $N/T$, i.e., the fluctuations of susceptibility are small, as expected for a non-interacting system.

I. QUALITATIVE ARGUMENTS

Let us illustrate the commonly used statistical concepts devoted to noise by a simple example. Consider a random quantity $x$ with probability distribution $P(x) = Z^{-1} \exp[-U(x)]$. It provides complete information about $x$, which alternatively can be expressed by specifying all moments $\langle x^n \rangle$ or all cumulants $\langle x^n \rangle$. For a symmetric distribution, $P(x) = P(-x)$ such that $\langle x \rangle = 0$, the noise of $x$ is defined as

$$S_1 = 2 \langle x^2 \rangle. \quad (1)$$

Up to the factor 2, the noise is equal to the second cumulant of $x$ (which is here also equal to the second moment since $\langle x \rangle = 0$).

The second noise (or noise of noise) of $x$ is defined as

$$S_2 = 2 \left( \langle x^4 \rangle - \langle x^2 \rangle^2 \right), \quad (2)$$

which is neither a higher moment nor a cumulant. This particular definition is motivated by the measurement protocol in which the time fluctuations of $x^2$ are recorded (see also Appendix). For a Gaussian random quantity ($U$ is quadratic) one finds

$$\langle x^4 \rangle = 0, \quad \langle x^4 \rangle = 3 \langle x^2 \rangle^2, \quad S_2 = 4 \langle x^2 \rangle^2. \quad (3)$$

Now we perturb our system by a weak external field $B$, so that the new distribution function reads $P_B(x) = Z_B^{-1} \exp[-U(x) + Bx]$. Then the random quantity $x$ acquires a nonzero average value

$$\langle x \rangle_B = Z_B^{-1} \frac{\partial Z_B}{\partial B} = \chi B + O(B^3), \quad (4)$$

where $\chi = \langle x^2 \rangle$ is the corresponding linear susceptibility. The second moment of $x$ acquires an additional field dependent term, i.e.,

$$\langle x^2 \rangle_B = Z_B^{-1} \frac{\partial^2 Z_B}{\partial B^2} = \langle x^2 \rangle + (\chi^2 + a)B^2 + O(B^4), \quad (5)$$

where

$$a = \frac{1}{2} \left( \langle x^4 \rangle - 3 \langle x^2 \rangle^2 \right). \quad (6)$$

For a Gaussian distribution one has $a = 0$. Thus, the quantity $a$ given by Eq. (6) is a measure of the non-Gaussian nature of fluctuations. At the same time, $a$ is proportional to the 4-th cumulant of $x$ and, therefore, is inequivalent to the second noise $S_2$.

In typical experiments, the fluctuating quantity is time-dependent and instead of the averages one has to consider correlation functions (see below). The noise is characterized by the spectral power, which is the Fourier transform of the corresponding correlation function evaluated in the presence of the external field. Such an analysis of the experimental data is usually performed over a reasonably long, but necessarily limited time interval. Repeating the analysis over a large number of such intervals one may find that the susceptibility $\chi$ itself takes different values at different times [30]. [Note, that this averaging is no longer described by the above model distribution $P(x)$. Within this simple model the susceptibility defined in Eq. (4) does not fluctuate.] Averaging over the fluctuating values of the susceptibility one finds its mean value. It is then tempting to use this averaged susceptibility in Eq. (5) and interpret the quantity $a$ as the noise of the susceptibility. At this point one has to be careful, as there is no guarantee that $a$ is positive. In fact, it is well-known in the theory of shot noise [31,32,33] that out of equilibrium the noise may be lower than the equilibrium noise at the same temperature.

As an illustration for such a negative non-equilibrium contribution to the noise we consider a single spin 1/2 subject to an external magnetic field. If one is interested in equal-time correlators one can use the above arguments with $x$ replaced by $\hat{S}_z$. Now, the square of the spin operator is simply proportional to the identity operator independent of whether the field is applied or not. Consequently, $\langle \hat{S}_z^2 \rangle_B = \langle \hat{S}_z^2 \rangle = 1/4$ and $a = -\chi^2 = -1/16$.

Thus one might expect the non-equilibrium spin fluctuations to be described by the negative quantity which appears to be inconsistent with its interpretation as noise of the susceptibility. In what follows, we provide a proper microscopic definition of the noise of susceptibility corresponding to the experimental protocol proposed in Ref. [34].
II. NON-EQUILIBRIUM SPIN FLUCTUATIONS AND NOISE OF THE SUSCEPTIBILITY

We now generalize the above arguments to the case of a quantum spin system. Assume a coupling \( H_I = -SB(t) \), where \( S \) is the spin operator and \( B \) is a magnetic field. A traditional way of describing the response of the system to a weak external perturbation is the spin susceptibility, which relates the applied field to the resulting magnetization, \( M_i \equiv \langle \hat{S}_i(t) \rangle = \int dt' \chi_{ij}(t,t')B_j(t') \), with the susceptibility given by the Kubo formula\(^1\)

\[
\chi_{ij}(t,t') = i \theta(t-t') \left \langle \left[ \hat{S}_i(t), \hat{S}_j(t') \right] \right \rangle. \tag{7}
\]

Here the spin operators must be in the Heisenberg representation with respect to the Hamiltonian of the system in the absence of the field. In isotropic systems, the susceptibility tensor is diagonal \( \chi_{ij} = \chi \delta_{ij} \).

Time-dependent magnetization fluctuations can then be described by a power spectrum (or spectral density) that in the simplest case can be related to the imaginary part of the susceptibility\(^7\) with the help of the fluctuation-dissipation theorem

\[
S_M(\omega) = \left \langle \langle \hat{S}_z(t) \hat{S}_z(t') + \hat{S}_z(t') \hat{S}_z(t) \rangle \right \rangle_{\omega} = 2 \coth \frac{\omega}{2T} \text{Im} \chi(\omega). \tag{8}
\]

The quantity \( S_M(\omega) \) is a generalization of Eq. \(1\).

Systematic calculations are often facilitated by using field-theoretical techniques. Real-time fluctuations, especially in the presence of an external field, can be conveniently described within the Keldysh formalism\(^13\). In this formalism, the spin susceptibility\(^7\) has the form

\[
\chi_{ij}(t,t') = i \left \langle T_K \hat{S}_i(t) \hat{S}_j(t') \right \rangle_0 \tag{9}
\]

\[
= -i \left. \frac{\delta^2 Z[\chi^d, \chi^q]}{\delta \partial \chi_i(t) \delta \partial \chi_j(t')} \right |_{\lambda=0},
\]

where \( T_K \) denotes time ordering along the Keldysh contour, and \( Z[\chi^d, \chi^q] \) is the Keldysh partition function with the source terms \( \chi^{d(q)} \) included. The subscripts \( d \) and \( cl \) on both the spin operators and source fields refer to the so-called “quantum” and “classical” variables\(^13\). They are related to the fields belonging to the upper (\( u \)) and lower (\( d \)) branch of the Keldysh contour according to

\[
\hat{S}_i^d = \frac{1}{\sqrt{2}} \left( \hat{S}_i^u + \hat{S}_i^q \right), \quad \hat{S}_i^q = \frac{1}{\sqrt{2}} \left( \hat{S}_i^u - \hat{S}_i^d \right)
\]

\[
\chi_i^d = \frac{1}{\sqrt{2}} \left( \chi_i^u + \chi_i^q \right), \quad \chi_i^q = \frac{1}{\sqrt{2}} \left( \chi_i^u - \chi_i^d \right).
\]

The “classical” source term defined in this way describes the physical probing field, \( \chi_i^d \equiv \sqrt{2}B_i \), while the “quantum” term is only needed to construct the correlation function and is set to zero at the end of the calculation.

Once the susceptibility is obtained, we can use Eq. \(8\) to find the noise spectrum.

Alternatively, we can characterize fluctuations of the magnetization by directly evaluating the second moment of the spin in the presence of the perturbation, generalizing Eq. \(5\). Without loss of generality, we can assume that the external field is applied along \( z \) direction. The symmetrized second moment of the \( z \)-component of the physical spin is then given by

\[
\left \langle \hat{S}_z(t_1)\hat{S}_z(t_2) + \hat{S}_z(t_2)\hat{S}_z(t_1) \right \rangle_B = \left \langle T_K \hat{S}_z^d(t_1)\hat{S}_z^d(t_2) \right \rangle_B = - \left. \frac{\delta^2 Z[\chi^d, \chi^q]}{\delta \chi_z(t_1) \delta \chi_z(t_2)} \right |_{\lambda^z=0} = \left \langle T_K \hat{S}_z^d(t_1)\hat{S}_z^d(t_2)e^{i \int dt' \sqrt{2B} \hat{S}_z^q(t')} \right \rangle_B. \tag{10}
\]

Note, that for a spin \(1/2\) the moment \(10\) at equal times \( t_1 = t_2 \) is given by a \( B \)-independent constant (which is equal to \(1/2\)).

For weak external fields, we may expand the quantity \(10\) in a power series in \( B \),

\[
\left \langle T_K \hat{S}_z^d(t_1)\hat{S}_z^d(t_2) \right \rangle_B = S_M(t_1-t_2) + \int dt'_1 dt'_2 C_N(t_1-t'_1, t_1-t'_2) B(t'_1) B(t'_2) + \mathcal{O}(B^4). \tag{11}
\]

The first term in Eq. \(11\) corresponds to the equilibrium noise \(S_M(0) = 1/2\).

The second term in Eq. \(11\) contains the four-point correlation function

\[
C_N(t_1-t'_1, t_1-t'_2) = - \left. \frac{\delta^4 Z[\chi^d, \chi^q]}{\delta \chi_z(t_1) \delta \chi_z(t'_1) \delta \chi_z(t_2) \delta \chi_z(t'_2)} \right |_{\lambda=0} = - \left \langle T_K \hat{S}_z^d(t_1)\hat{S}_z^d(t'_1)\hat{S}_z^d(t_2)\hat{S}_z^d(t'_2) \right \rangle, \tag{12}
\]

which one can split into Gaussian and non-Gaussian parts

\[
C_N = C_N^G + C_N^{NG}.
\]

The Gaussian part is readily obtained by a pair-wise averaging of the spin operators:

\[
C_N^G(t_1-t'_1, t_1-t'_2) = \left \langle T_K \hat{S}_z^d(t_1)\hat{S}_z^d(t'_1) \right \rangle \left \langle T_K \hat{S}_z^d(t_2)\hat{S}_z^d(t'_2) \right \rangle = \chi_{zz}(t_1, t'_1) \chi_{zz}(t_2, t'_2) + \chi_{zz}(t_1, t'_2) \chi_{zz}(t_2, t'_1). \tag{13}
\]
Using the explicit form (14), we find each time bin as a fluctuating quantity (as it is in the probability is obtained by averaging over the time bins. In practice, in every time bin one finds a different result and the average susceptibility, respectively. In general, the Wick’s theorem does not hold for spin operators, reflecting the fact that their algebra is non-Abelian. This quantity cannot be expressed in terms of the averaged susceptibilities and has to be evaluated specifically for each system.

In a typical experiment, the system is probed with a harmonic perturbation,

\[ B(t) = B_0 \cos(\omega_0 t). \]

The susceptibility is then measured using lock-in techniques, which amounts to obtaining the average of the following operator

\[ \hat{\chi}_\omega(\tau_n, \omega_0, \Delta \omega) = \frac{1}{B_0 T_\chi} \int_{\tau_n - \frac{T_\chi}{2}}^{\tau_n + \frac{T_\chi}{2}} dt \cos(\omega_0 t - \varphi) \hat{S}_{z,B}(t). \quad (14) \]

The measurement is performed for a time period \( T_\chi \) centered around \( \tau_n \). This defines the measurement bandwidth \( \Delta \omega \equiv 2\pi/T_\chi \ll \omega_0 \), chosen to be much smaller than \( \omega_0 \). The phase \( \varphi \) allows discriminating between the in-phase (\( \varphi = 0 \)) and the out-of-phase (\( \varphi = \pi/2 \)) response, corresponding to the real and imaginary parts of the susceptibility, respectively. In practice, in every time bin one finds a different result and the average susceptibility is obtained by averaging over the time bins.

Treating the result of susceptibility measurements in each time bin as a fluctuating quantity (as it is in the experiment), one can define its second moment or noise of susceptibility as follows

\[ \chi^{(2)}_{\varphi_1, \varphi_2}(\tau_1, \tau_2; \omega_0, \Delta \omega) = \langle \chi_{\varphi_1}(\tau_1) \chi_{\varphi_2}(\tau_2) + \chi_{\varphi_2}(\tau_2) \chi_{\varphi_1}(\tau_1) \rangle - 2 \langle \chi_{\varphi_1}(\tau_1) \rangle \langle \chi_{\varphi_2}(\tau_2) \rangle. \quad (15) \]

Using the explicit form (14), we find

\[ \chi^{(2)}_{\varphi_1, \varphi_2}(\tau_1, \tau_2; \omega_0, \Delta \omega) = \frac{1}{B_0^2 T_\chi} \int_{\tau_1 - \frac{T_\chi}{2}}^{\tau_1 + \frac{T_\chi}{2}} dt_1 \int_{\tau_2 - \frac{T_\chi}{2}}^{\tau_2 + \frac{T_\chi}{2}} dt_2 \times \cos(\omega_0 t_1 - \varphi_1) \cos(\omega_0 t_2 - \varphi_2) \times \left\langle \left\langle \hat{S}(t_1) \hat{S}(t_2) + \hat{S}(t_2) \hat{S}(t_1) \right\rangle_B \right\rangle. \quad (16) \]

In contrast to Eq. (14), the averaging in Eq. (16) has been already performed (as we are not interested in higher moments). The double angle brackets in Eq. (16) indicate the 2nd cumulant, which is obtained by subtracting two times the product of averages, i.e.,

\[ 2 \left\langle \hat{S}(t_1) \right\rangle \left\langle \hat{S}(t_2) \right\rangle, \]

as in Eq. (15).

We now use the expansion (11) for the symmetrized average and decompose the second moment of susceptibility Eq. (16) into two parts

\[ \chi^{(2)} = \chi^{(2)}_{eq} + \chi^{(2)}_{ne}. \]

The first term \( \chi^{(2)}_{eq} \) describes the equilibrium magnetization noise \( S_M \) in the absence of the external field:

\[ \chi^{(2)}_{eq, \varphi_1, \varphi_2}(\tau_1, \tau_2; \omega_0, \Delta \omega) = \frac{1}{B_0^2 T_\chi} \int_{\tau_1 - \frac{T_\chi}{2}}^{\tau_1 + \frac{T_\chi}{2}} dt_1 \int_{\tau_2 - \frac{T_\chi}{2}}^{\tau_2 + \frac{T_\chi}{2}} dt_2 \times \cos(\omega_0 t_1 - \varphi_1) \cos(\omega_0 t_2 - \varphi_2) S_M(t_1 - t_2). \quad (17) \]

The corresponding noise spectrum is given by the Fourier transform of \( \chi^{(2)}_{eq} \):

\[ \chi^{(2)}_{eq, \varphi_1, \varphi_2}(\nu; \omega_0, \Delta \omega) = \frac{1}{4B_0^2} \int \frac{d\nu}{\Delta \omega} \times \{ \cos(\varphi_1 - \varphi_2) [S_M(\omega_0 + \nu) + S_M(\omega_0 - \nu)] - i \sin(\varphi_1 - \varphi_2) [S_M(\omega_0 + \nu) - S_M(\omega_0 - \nu)] \} + O\left(\frac{\Delta \omega}{\omega_0^2}\right), \quad (18) \]

where \( f(x) \equiv \sin^2(x)/x^2 \) restricts the frequency \( \nu \) to be small, \( \nu \ll \Delta \omega \ll \omega_0 \). The appearance of the imaginary part in the noise spectrum reflects the fact that cross-correlations between the real and imaginary parts of the susceptibility do not possess any symmetry as functions of time. Indeed, according to the definition (15), the noise of susceptibility obeys the symmetry

\[ \chi^{(2)}_{\varphi_1, \varphi_2}(\tau_1, \tau_2) = \chi^{(2)}_{\varphi_2, \varphi_1}(\tau_2, \tau_1) \quad (19) \]

and is a symmetric function of the two times \( \tau_1 \) only if \( \varphi_1 = \varphi_2 \). As a result, the noise of the real (or imaginary) part of susceptibility is characterized by a real spectrum, while the Fourier transform of the cross-correlator may contain an imaginary part.

Turning to the non-equilibrium contribution \( \chi^{(2)}_{ne} \), composed of the second term of the expansion (11) substituted into Eq. (16), we note that only the non-Gaussian part \( C^{NG}_\chi \) of the correlation function contributes. This is because the Gaussian part (13) corresponds exactly to the subtracted product of the averages, i.e., the
last term in Eq. (15). Thus we obtain
\[ \chi^{(2)}_{\nu e, \varphi_1, \varphi_2}(\tau_1, \tau_2 | \omega_0, \Delta \omega) = \frac{1}{T^2} \] (20)
\[ \tau_1 + \frac{T}{\omega_0} \quad \tau_2 + \frac{T}{\omega_0} \]
\[ \times \int d t_1 \int d t_2 \cos(\omega_0 t_1 - \varphi_1) \cos(\omega_0 t_2 - \varphi_2) \]
\[ \tau_1 - \frac{T}{\omega_0} \quad \tau_2 - \frac{T}{\omega_0} \]
\[ \times \int d t'_1 d t'_2 C^{NG}_x(t_1, t_1', t_2, t_2') \cos(\omega_0 t_1') \cos(\omega_0 t_2'). \]

The time integrals in Eq. (20) can be simplified with the help of the Fourier transform defined as follows
\[ C^{NG}_x(t_1, t_1', t_2, t_2') = \int \frac{d \nu d \omega_1 d \omega_2}{(2 \pi)^3} C^{NG}_x(\nu, \omega_1, \omega_2) \]
\[ \times e^{-i \nu (t_1 - t_2)} e^{-i \omega_1 (t_1 - t_1')} e^{-i \omega_2 (t_2 - t_2')}. \] (21)

As stated above, in this paper we are only interested in low-frequency noise, \( \nu \ll \Delta \omega \ll \omega_0 \). Focusing on contributions that are slow functions of \( \tau_1 - \tau_2 \), we retain only the lowest harmonics and find
\[ \chi^{(2)}_{\nu e, \varphi_1, \varphi_2}(\nu, \omega_0, \Delta \omega) = \frac{1}{16 T^2} \left( \frac{\pi \nu}{\Delta \omega} \right) \]
\[ \times \sum_{\epsilon_1, \epsilon_2 = \pm 1} e^{-i \epsilon_1 \varphi_1} e^{-i \epsilon_2 \varphi_2} C^{NG}_x(\nu, \epsilon_1 \omega_0, \epsilon_2 \omega_0) \]
\[ + \sum_{\epsilon = \pm 1} e^{i \epsilon (\varphi_1 - \varphi_2)} C^{NG}_x(\nu - 2 \epsilon \omega_0, \epsilon \omega_0, - \epsilon \omega_0) \]. (22)

Thus, the non-equilibrium contribution to the noise of the susceptibility is a probe of non-Gaussian fluctuations in the system, in contrast to the second noise.\[ B. \] Below, we will illustrate our general considerations by calculating \( C^{NG}_x \) for the simplest model system, i.e. a single spin coupled to a dissipative environment.

III. SUSCEPTIBILITY NOISE OF A SINGLE SPIN

A. The model

Let us now illustrate our general arguments using a simple example of a single spin-1/2 coupled to a dissipative bath in the presence of a magnetic field. In this Section, we calculate the four-spin correlation functions \[ 17 \] and \[ 22 \] leaving the discussion of the results and their relation to experiments for the subsequent Section.

We model the bath by an isotropic, bosonic vector degree of freedom \( \vec{X} \) coupled to the spin operator via the minimal Hamiltonian
\[ H = \vec{s} \cdot \vec{X}. \] (23)

The physical properties of the bath can be encoded in the bosonic correlation function. Here we have also chosen to incorporate the coupling constant into the definition of the bosonic correlator. Within the frame of the Keldysh formalism, the bosonic correlation function is defined as
\[ \tilde{\Gamma}_{\alpha \beta}^{ab}(t, t') = \delta_{\alpha \beta} \Gamma^{ab}(t, t') = \text{Tr} \left\{ \mathcal{T}_{K} \chi^{\alpha-a}(t) \chi^{\beta-b}(t') \right\}, \]
(24)
where Latin indices span the 2 \( \times 2 \) Keldysh space \( a, b = c, l, q \) and Greek indices refer to the spin components \( \alpha, \beta = x, y, z \). The bath being Ohmic means that the following relation holds:
\[ \Pi^{R}(\omega) - \Pi^{A}(\omega) = \lambda \omega, \]
(25)
where \( \lambda \ll 1 \) is the effective coupling constant, and \( \Pi^{R/A} \) are the retarded and the advanced components of \[ 24 \]. We assume the bath to be in thermal equilibrium such that the Keldysh component of the correlation function \[ 24 \] is given by the standard expression
\[ \Pi^{K}(\omega) = \coth \frac{\omega}{2T} \left[ \Pi^{R}(\omega) - \Pi^{A}(\omega) \right]. \] (26a)
Note that the model \[ 23 \] is similar to the Kondo model \[ 33,39,49 \] in the high temperature regime, \( T \gg T_{K} \), where the latter effectively describes a spin coupled to an Ohmic bath \[ 25 \].

B. Majorana representation

Our goal is to calculate a 4-spin correlation function. In any standard fermionic representation of the spin operators \[ 39,47 \], \( N \)-point spin correlators correspond to \( 2N \)-point fermionic correlators. In our case, we would have to evaluate an 8-point fermionic correlation function which is generally not an easy task. Fortunately, we can substantially simplify calculations by introducing the so-called Martin’s Majorana-fermion representation \[ 33,43 \]
\[ \hat{s}^a = -(i/2) \epsilon_{a \beta} \gamma_3 \eta_\beta, \quad \eta_\alpha = \eta_\alpha, \]
(27)
or explicitly
\[ \hat{s}^x = -i \eta_y \eta_z, \quad \hat{s}^y = -i \eta_z \eta_x, \quad \hat{s}^z = -i \eta_x \eta_y. \]
The Majorana fermions obey the Clifford algebra
\[ \{ \eta_\alpha, \eta_\beta \} = \delta_{\alpha \beta}, \quad \eta_\alpha^2 = 1/2. \] (28)
This representation is convenient since it explicitly preserves the spin-rotation symmetry and allows for a straightforward formulation of the field theory, while perfectly reproducing the \( SU(2) \) algebra of the operators \( \hat{s}^a \).

At the same time, the above representation is not “exact” as the Hilbert space of the Majorana triplet is ill-defined \[ 33,37,39,41 \]. In order to build the proper fermionic Hilbert space, one can increase the number of Majorana fermions in the theory making it even. Adding
an additional Majorana fermion, we may build a four-dimensional Hilbert space, twice as large as the original Hilbert space of the spin. Thus the Majorana representation possesses extra states, not present in the original model. This is a well-known problem[33,34,50] that can be resolved on the basis of the known[29] but not widely appreciated conjecture: The fact that the Hilbert space is bigger than the 2-dimensional spin-1/2 Hilbert space has no effect on the spin correlation functions. This can be understood as follows. The Majorana Hilbert space can be roughly thought of as consisting of two copies of the physical spin[33]. Any operator of any physical quantity operates only within a two-dimensional subspace corresponding to one of the two spin copies. The remaining subspace does contribute to the partition function, but this contribution is limited to a multiplicative factor that cancels out from any correlation function. A rigorous proof of this statement will be published elsewhere.[50]

In the Majorana representation the Hamiltonian (23) takes the form

\[ H = -(i/2)\epsilon_{\alpha\beta\gamma}\hat{X}^\alpha\eta^\beta\eta^\gamma. \] (29)

Any spin correlation function can now be represented as a correlation function of the Majorana fermions[33,34,50]. For example, the four-point function is given by

\[ \langle \hat{s}^\alpha(t_1)\hat{s}^\beta(t_1')\hat{s}^\gamma(t_2)\hat{s}^\delta(t_2') \rangle = \]

\[ = (1/4) \langle \eta_{\alpha}(t_1)\eta_{\beta}(t_1')\eta_{\alpha}(t_2)\eta_{\beta}(t_2') \rangle. \]

This relation demonstrates the strength of the Majorana representation[27]: the four-point spin correlator is given by the four-point correlator of the Majorana fermions. However, in order to extend this correspondence to the time ordered correlations functions on the Keldysh contour, we need to take care of the time-ordering operator \( T_K \). In terms of spin operators, the time ordering is similar to that of bosons [see, e.g. Eqs. (9) and (12)]. Yet, for the fermionic operators \( \eta_\alpha \) the time ordering is different. Therefore, it is convenient to multiply every Majorana fermion in the above correlator by \(-i m\), where \( m \) is the fourth Majorana fermion (needed anyway to construct the Hilbert space). Given the algebra (28), this operation does not change the correlation function (which is effectively multiplied by 1/4). With respect to the time ordering the bilinear terms \(-i m\eta_\alpha \) behave as bosons similarly to the spin operators. The correlation function (12) then takes the form

\[ C_\chi(t_1,t_1',t_2,t_2') = -\langle T_K \hat{s}^\chi(t_1)\hat{s}^\chi(t_1')\hat{s}^\chi(t_2)\hat{s}^\chi(t_2') \rangle \] (30)

\[ = -\langle T_K (\eta_\chi m^\ell(t_1)\eta_\chi m^\ell(t_1')\eta_\chi m^\ell(t_2)\eta_\chi m^\ell(t_2') \rangle. \]

At this stage one might get an impression that we have achieved nothing, as we are back to an 8-fermion correlation function. However, the operator \( m \) commutes with the Hamiltonian of the system. The Green’s functions corresponding to \( m \) remain “bare”, which is a great simplification.

### C. Diagrammatic expansion

Having defined the model in the Majorana representation we can now proceed using the usual diagrammatic technique, which was not possible for the original spin operators. The peculiarity of the single-spin model is that the spin has no Hamiltonian in the absence of the bath (and the magnetic field). Therefore, the “free” Green’s functions of the Majorana fermions are

\[ G^{R}_{\alpha,\alpha}(t,t') = -i \langle T_K \eta^\ell_\alpha(t)\eta^\ell_\alpha(t') \rangle = -i\Theta(t-t'), \] (31)

\[ D^K(t,t') = -i \langle T_K m^{cl}(t)m^{cl}(t') \rangle = -i\Theta(t-t'). \]

The coupling of the Majorana fermions \( \eta_\alpha \) to the bath [23] is then described by means of a “self-energy”, which can be obtained as a saddle-point solution in the path-integral formulation of the theory[29]. At high enough temperatures, the leading contribution to the self-energy is graphically depicted in Fig. 1 [where the wavy line refers to the bosonic correlator (26) and the solid line to \( G_{0,\alpha} \)] and is given by

\[ \Sigma^R_\alpha = -i\Gamma = -i\lambda T. \] (32)

Consequently, the Green’s functions of the Majorana fermions \( \eta_\alpha \) in the model[23] take the simple form

\[ G^{R/A}_{\alpha}(\omega) = \frac{1}{\omega \pm i\Gamma} \] (33)

\[ G^K_{\alpha}(\omega) = -\frac{2i\Gamma}{\omega^2 + \Gamma^2} \tanh \frac{\omega}{2T}. \]

The equilibrium noise of the susceptibility[18] is described by the two-point function corresponding to the diagram in Fig. 2, where the double solid line corresponds...
to the “dressed” Green’s functions \( \chi^{(2)} \) of the Majorana fermions \( \eta_a \) and the dashed line refers to the Green’s function \( D \) of the non-interacting Majorana fermion \( m \), see Eq. (31). The noise of the real part of susceptibility is identical with that of the imaginary part

\[
\chi^{(2)}_{\text{eq},00}(\nu|\omega_0, \Delta \omega) = \chi^{(2)}_{\text{eq},12}(\nu|\omega_0, \Delta \omega) = \frac{1}{4B_0^2} f \left( \frac{\pi \nu}{\Delta \omega} \right) \frac{\Gamma}{\Gamma^2 + (\omega_0 + \nu)^2} \frac{\Gamma}{\Gamma^2 + (\omega_0 - \nu)^2},
\]

and is purely real in accordance with the symmetry (19). In contrast, the cross-correlations are characterized by the purely imaginary noise spectrum

\[
\chi^{(2)}_{\text{eq},02}(\nu|\omega_0, \Delta \omega) = \frac{i}{4B_0^2} f \left( \frac{\pi \nu}{\Delta \omega} \right) \frac{\Gamma}{\Gamma^2 + (\omega_0 + \nu)^2} \frac{\Gamma}{\Gamma^2 + (\omega_0 - \nu)^2}.
\]

The result (34b) is an odd function of \( \nu \), such that integrating over the frequency would yield zero, corresponding to the absence of cross-correlations between the real and imaginary parts of susceptibility at equal times.

Now we turn to the description of the more interesting, non-equilibrium noise of susceptibility. For this purpose, we need to evaluate the correlation function (30). Within the path-integral formulation of the theory\(^{20}\), we may integrate out the bosonic bath and obtain the effective action for the Majorana fermions. This action can be formally expanded around the saddle-point solution. The result can be graphically presented in diagrammatic form, see Figs. 3 and 4. The external frequencies in these diagrams correspond to the Fourier transform (21).

At high enough temperatures, \( T \gg T_K, \omega_1, \omega_2, \nu, \Gamma \), the leading contribution to the correlation function (30) is described by the diagrams in Fig. 3 and is given by

\[
C_{\chi}^{\text{NG}}(\nu, \omega_1, \omega_2) = i\Gamma^2 \frac{\omega_1 + \omega_2 + 2i\Gamma}{8T^2 (\omega_1 + i\Gamma)(\omega_2 + i\Gamma)(\omega_1 + \nu + i\Gamma)(\omega_2 + \nu + i\Gamma)}.
\]

Higher-order diagrams (see, e.g., Fig. 4) may be neglected as long as the coupling constant \( \lambda \) remains small. We have ruled out the possibility that ladder diagrams might contribute to the lowest order in \( \lambda \). The details of this calculation will be presented in a separate publication\(^{20}\).

Substituting the result (35) into Eq. (22), we find the non-equilibrium noise spectrum of the spin susceptibility in the model\(^{23}\). For the noise of the real part of the susceptibility we find

\[
\chi^{(2)}_{\text{ne},00}(\nu|\omega_0, \Delta \omega) = f \left( \frac{\pi \nu}{\Delta \omega} \right) \frac{\Gamma}{32T^2} \left[ \omega_0^2 - 3(\Gamma^2 + \nu^2) \right] \frac{\omega_0^2}{(\Gamma^2 + \omega_0^2)[(\Gamma^2 + \nu^2)^2 + 2(\Gamma^2 - \nu^2)\omega_0^2 + \omega_0^4]}.
\]

FIG. 3. The leading contribution to non-equilibrium noise of susceptibility.

In contrast to the equilibrium contribution (34a), the noise of the imaginary part of the susceptibility is inequivalent to Eq. (36a) and is given by

\[
\chi^{(2)}_{\text{ne},02}(\nu|\omega_0, \Delta \omega) = -\frac{f \left( \frac{\pi \nu}{\Delta \omega} \right)}{32T^2} \Gamma^3 \left( \Gamma^2 + 5\omega_0^2 + \nu^2 \right) \frac{\omega_0^4}{(\Gamma^2 + \omega_0^2)[(\Gamma^2 + \nu^2)^2 + 2(\Gamma^2 - \nu^2)\omega_0^2 + \omega_0^4]].
\]

Finally, one can also compute the “cross-correlation” of the real and imaginary parts of the susceptibility:

\[
\chi^{(2)}_{\text{ne},02}(\nu|\omega_0, \Delta \omega) = -\frac{f \left( \frac{\pi \nu}{\Delta \omega} \right)}{32T^2} \Gamma^3 \left( \Gamma^2 - \omega_0^2 + \nu^2 - 4i\Gamma \nu \right) \frac{\omega_0^4}{(\Gamma^2 + \omega_0^2)[(\Gamma^2 + \nu^2)^2 + 2(\Gamma^2 - \nu^2)\omega_0^2 + \omega_0^4]}.
\]

IV. DISCUSSION

It is instructive to relate our approach to the existing literature on the experimentally observed flux noise and the noise of the spin susceptibility\(^{25,29,31}\). The main features of the experimental results are often explained with the help of the generic model of paramagnetic spins\(^{11,13,14,22}\). The model consists of an ensemble of non-interacting spins, each coupled to a dissipative environment. It is assumed that the corresponding relaxation rates \( \Gamma \) vary with the distribution function in a certain interval between two cutoff scales \( \Gamma_L \) and \( \Gamma_H \):

\[
p(\Gamma) = \frac{1}{\ln\left( \frac{\Gamma_H}{\Gamma_L} \right)} \frac{1}{\Gamma}.
\]
As a single spin has the Lorentzian-shaped noise spectrum, averaging over $p(\Gamma)$ one obtains a $1/f$-noise spectrum of the whole system within the frequency range $\Gamma_L < \omega < \Gamma_H$. The resulting noise is roughly independent of temperature and may be used to fit the experimental data. Weak deviations of the exponent $\alpha$ of the measured noise spectra $S_n \propto 1/\omega$ can be accounted for by changing the distribution function to $p(\Gamma) \propto 1/\Gamma^\alpha$.

The paramagnetic model does not include interactions between spins. Indeed, typical interaction scales seem to be small, of the order $J_{\text{typ}} \sim 50 \text{ mK}$, justifying the approach of Refs. [14] and [15] in the high-temperature regime $T > J$. This is also consistent with indications that the system of spins is in the classical high-temperature regime characterized by a Curie susceptibility [23] and an ohmic environment [23].

A system with a large number of non-interacting spins is a natural generalization of our approach. In this case, instead of the single spin-1/2 we need to consider the total spin of the system

$$\hat{S} = \sum_i^N \hat{s}_i.$$ 

The corresponding four-point correlation function [c.f. Eq. (12)] can be decomposed as follows:

$$C_{\chi}(t_1, t_1', t_2, t_2') = - \left\langle T_K \hat{s}_{i_1}^d(t_1) \hat{s}_{i_1}^q(t_1') \hat{s}^d_{j_2}(t_2) \hat{s}^q_{j_2}(t_2') \right\rangle$$

$$= - \sum_i \left\langle T_K \hat{s}_{i_1}^d(t_1) \hat{s}_{i_1}^q(t_1') \hat{s}_{j_2}^d(t_2) \hat{s}_{j_2}^q(t_2') \right\rangle$$

$$- \sum_{i \neq j} \left\langle T_K \hat{s}_{i_1}^d(t_1) \hat{s}_{i_1}^q(t_1') \hat{s}_{j_2}^d(t_2) \hat{s}_{j_2}^q(t_2') \right\rangle$$

$$- \sum_{i \neq j} \left\langle T_K \hat{s}_{i_1}^d(t_1) \hat{s}_{i_1}^q(t_1') \hat{s}_{j_2}^d(t_2) \hat{s}_{j_2}^q(t_2') \right\rangle.$$ (38)

Clearly, the last two lines of Eq. (38) do not contribute to Eq. (15) and therefore the noise of the susceptibility of the system of independent spins is given by the sum of the individual noises of each spin

$$X_{\varphi_1 \varphi_2}^{(2)} = \sum_i \chi^{(2)}_{\varphi_1 \varphi_2}(\Gamma_i).$$ (39)

Averaging over the distribution [37] one obtains [15]

$$X_{\varphi_1 \varphi_2}^{(2)} = N \int_{\Gamma_L}^{\Gamma_H} d\Gamma p(\Gamma) \chi^{(2)}_{\varphi_1 \varphi_2}(\Gamma).$$ (40)

Using our results [34] and [36] we can now obtain the noise of the susceptibility in the model of non-interacting spins. In the limit, where the probing frequency $\omega_0$ is much smaller than the slowest relaxation rate of the spins $\omega_0 \ll \Gamma_L$ we find

$$X_{0,0}^{(2)} \approx N \frac{3}{4 \Gamma_L} \left[ \frac{1}{B_0^2} - \frac{1}{8 T^2} \right] f \left( \frac{\pi \nu}{\Delta \omega} \right) \ln^{-1} \frac{\Gamma_H}{\Gamma_L},$$

$$X_{\pi, \pi}^{(2)} \approx N \frac{3}{4 \Gamma_L} \left[ \frac{1}{B_0^2} - \frac{1}{8 T^2} \right] f \left( \frac{\pi \nu}{\Delta \omega} \right) \ln^{-1} \frac{\Gamma_H}{\Gamma_L},$$

$$X_{0, \pi}^{(2)} \approx N \frac{3}{4 \Gamma_L} \left[ \frac{1}{B_0^2} - \frac{1}{8 T^2} \right] f \left( \frac{\pi \nu}{\Delta \omega} \right) \ln^{-1} \frac{\Gamma_H}{\Gamma_L},$$

$$X_{\pi, 0}^{(2)} \approx N \frac{3}{4 \Gamma_L} \left[ \frac{1}{B_0^2} - \frac{1}{8 T^2} \right] f \left( \frac{\pi \nu}{\Delta \omega} \right) \ln^{-1} \frac{\Gamma_H}{\Gamma_L}.$$ (41)

In the opposite limit $\Gamma_L \ll \omega_0 \ll \Gamma_H$ we obtain

$$X_{0,0}^{(2)} \approx \pi N \frac{1}{4 \omega_0} \left[ \frac{1}{B_0^2} - \frac{1}{16 T^2} \right] f \left( \frac{\pi \nu}{\Delta \omega} \right) \ln^{-1} \frac{\Gamma_H}{\Gamma_L},$$

$$X_{\pi, \pi}^{(2)} \approx \pi N \frac{1}{4 \omega_0} \left[ \frac{1}{B_0^2} - \frac{1}{16 T^2} \right] f \left( \frac{\pi \nu}{\Delta \omega} \right) \ln^{-1} \frac{\Gamma_H}{\Gamma_L},$$

$$X_{0, \pi}^{(2)} \approx \pi N \frac{1}{4 \omega_0} \left[ \frac{1}{B_0^2} - \frac{1}{16 T^2} \right] f \left( \frac{\pi \nu}{\Delta \omega} \right) \ln^{-1} \frac{\Gamma_H}{\Gamma_L},$$

$$X_{\pi, 0}^{(2)} \approx \pi N \frac{1}{4 \omega_0} \left[ \frac{1}{B_0^2} - \frac{1}{16 T^2} \right] f \left( \frac{\pi \nu}{\Delta \omega} \right) \ln^{-1} \frac{\Gamma_H}{\Gamma_L}.$$ (42)

The above results do not show the $1/\nu$ behavior observed in Ref. [30]. Moreover, the experiment shows non-vanishing correlations between the fluctuations of flux and susceptibility. As the model of independent spins remains invariant under time reversal, such correlations are excluded in this theory. It is therefore apparent that the model of independent spins misses the essential physics of the real noise sources affecting SQUIDs.

Recently, J. Atalaya, J. Clarke and the two of us [23] have performed a numerical analysis of interacting spin systems in the presence of disorder. It was found that the slow dynamics of the magnetization is dominated by spontaneously forming spin clusters, which give rise to $1/f$ noise of magnetization. We conjecture, that the observed $1/\nu$ noise of the spin susceptibility is due to slowly switching clusters, which affect the susceptibility of nearby spins. The apparent time-reversal symmetry breaking could then be attributed to the relatively short measurement times, during which some clusters never flip their magnetization.
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Appendix: Noise of Noise

Here we briefly review the four-point correlation function corresponding to the second spectrum or noise of a single measurement of the spectral density, which is the time to identify non-Gaussian contributions in the noise spectrum in the thermodynamic limit. For more details the reader is referred to the book by S.M. Kogan. The second spectrum was introduced in order to identify non-Gaussian contributions in 1/f noise. Let $x(t)$ be a classical fluctuating quantity, which is the signal to be measured. In a typical experimental protocol, the signal is bandwidth filtered and then squared. To facilitate the comparison of the second spectrum to the noise of susceptibility discussed in the main text, we assume (differing from Ref. 1) the filter output signal of having a form similar to Eq. \((14)\) the filter output signal of having a form similar to Eq. \((14)\), (differing from Ref. 1) the filter output signal of having a form similar to Eq. \((14)\) of susceptibility discussed in the main text, we assume (differing from Ref. 1) the filter output signal of having a form similar to Eq. \((14)\)

\[
\delta x(t|\omega_0, \Delta\omega) = \frac{1}{T_s} \int_{-T_s/2}^{T_s/2} dt e^{i\omega_0 t} \delta x(t). \quad (A.1)
\]

The time $T_s$ is similar to $T_1$ in Eq. \((14)\). This is the time of a single measurement of the spectral density, which defines the bandwidth $\Delta\omega = 2\pi/T_s$. The above defined $\delta x$ is related to the noise spectral density $S_x(t = t') = 2C^2(t = t') = 2\langle \delta x(t)\delta x(t') \rangle$ by

\[
2\langle |\delta x(t|\omega_0, \Delta\omega)|^2 \rangle = \int \frac{d\omega}{2\pi} f \left( \frac{\pi(\omega - \omega)}{\Delta\omega} \right) S_x(\omega) \approx \frac{\Delta\omega}{2\pi} S_x(\omega). \quad (A.2)
\]

The so-called second spectrum $S_x^{(2)}$ is a measure of fluctuations of the noise power. The definition reads

\[
S_x^{(2)}(\nu|\omega_0, \Delta\omega) = \frac{8}{T_t^2} \left\langle \int_{-T_t/2}^{T_t/2} d\tau e^{i\nu \tau} \left| \delta x(\tau|\omega_0, \Delta\omega) \right|^2 \right\rangle - \left\langle \left| \delta x(\tau|\omega_0, \Delta\omega) \right|^2 \right\rangle^2. \quad (A.3)
\]

The time $T_t$ is the total measurement time and one can safely use the limit $T_t \rightarrow \infty$. It is easy to show that the following relation holds

\[
\langle |\delta x(\tau_1|\omega_0, \Delta\omega)|^2 \rangle \langle |\delta x(\tau_2|\omega_0, \Delta\omega)|^2 \rangle = \frac{1}{T_s} \int_{-T_s/2}^{T_s/2} dt_1 \int_{-T_s/2}^{T_s/2} dt_2 \int_{-T_s/2}^{T_s/2} dt_3 \int_{-T_s/2}^{T_s/2} dt_4 e^{i\omega_0 (t_1 - t_3) - i\omega_0 (t_2 - t_4)} \times C_x^{(4)}(t_1, t_3, t_2, t_4). \quad (A.4)
\]

Here we introduced the classical four-point correlation function

\[
C_x^{(4)}(t_1, t_3, t_2, t_4) = \langle \delta x(t_1)\delta x(t_3)\delta x(t_2)\delta x(t_4) \rangle. \quad (A.5)
\]

This can be split into the Gaussian and the non-Gaussian parts $C_x^{(4)} = C_x^{(4,G)} + C_x^{(4,N,G)}$. The Gaussian part is obtained as

\[
C_x^{(4,G)}(t_1, t_3, t_2, t_4) = C_x^{(2)}(t_1, t_3)C_x^{(2)}(t_2, t_4) + C_x^{(2)}(t_1, t_2)C_x^{(2)}(t_3, t_4) + C_x^{(2)}(t_1, t_3)C_x^{(2)}(t_2, t_4). \quad (A.6)
\]

One can now use Eq. \((A.6)\) to find the Gaussian contribution to the second spectrum. One finds that the first term on the right-hand side of Eq. \((A.6)\) cancels the average $\langle |\delta x|^2 \rangle$ in Eq. \((A.3)\). The remaining two terms contribute yield the Gaussian contribution to the second noise spectrum

\[
S_x^{(2)}(\nu|\omega_0, \Delta\omega) = 8 \int \frac{d\Omega}{2\pi} C_x^{(2)}(\Omega)C_x^{(2)}(\Omega + \nu) \times f \left( \frac{\pi(\omega_0 - \Omega)}{\Delta\omega} \right) f \left( \frac{\pi(\omega_0 - \nu - \Omega)}{\Delta\omega} \right). \quad (A.7)
\]

In contrast, the noise of susceptibility does not contain any contribution of the Gaussian part of the 4-point correlation function. In the experimentally relevant regime $\nu \ll \Delta\omega \ll \omega_0$ we can approximate Eq. \((A.7)\) as

\[
S_x^{(2)}(\nu|\omega_0, \Delta\omega) \approx \frac{8\Delta\omega}{3\pi} C_x^{(2)}(\omega_0)^2. \quad (A.8)
\]

This spectrum is "white" as a function of $\nu$.

Now, in the system of $N$ non-interacting spins the noise spectrum scales linearly with the number of spins $S_N(\omega) \propto N$, while the Gaussian contribution to the second noise scales as $S_x^{(2)}(\nu|\omega, \Delta\omega) \propto N^2$, see Eq. \((A.6)\). In contrast, the non-Gaussian contribution is linear in $N$, similarly to Eq. \((A.6)\). Therefore, the Gaussian part of the second noise always dominates making it difficult to extract information about non-Gaussian fluctuations from the second noise measurements.

The above conclusion is not necessarily general. In context of Ising spin glasses, it has been shown that the infinite-range interaction may result in a $1/f$ second noise spectrum in the thermodynamic limit $N \rightarrow \infty$. 


