Nonequilibrium spin noise and noise of susceptibility

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(Received 8 August 2014; published 17 November 2014)

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 noninteracting spins coupled to a dissipative (Ohmic) bath.

DOI: 10.1103/PhysRevB.90.205419 PACS number(s): 05.40.−a, 85.25.Dq

I. INTRODUCTION

Noise in electronic circuits provides information about the microscopic structure of the system complementary to that obtained from linear response transport measurements [1,2]. For electronic circuits, the standard Johnson-Nyquist noise is intimately related to dissipative processes with typical time scales of the order of picoseconds. At low frequencies it is "white," i.e., frequency independent. In contrast, the ubiquitous $1/f$ noise is related to slow processes, e.g., to slow rearrangements of impurities or the internal dynamics of two-level systems [3]. Its power spectrum is commonly described by Hooge's law [4,5], $S_V(f) \propto \overline{V}^2/f$, where $\overline{V}$ is the average observed voltage. This suggests that this noise could only be observed out of equilibrium. But this was shown not to be the case by Voss and Clarke [6,7], who measured the low-frequency fluctuations of the mean-square Johnson voltage in equilibrium (i.e., the second noise, or noise of noise) and showed that these fluctuations possess a $1/f$-like spectrum.

Motivated by these experiments, Beck and Spruit [8] calculated the variance of the Johnson-Nyquist noise and showed that it comprised two contributions. The first one could be interpreted as arising from resistance fluctuations with a $1/f$ spectrum. The second, with a white spectrum, is intrinsic to any Gaussian fluctuating quantity. Consequently the equilibrium $1/f$ noise could only be observed at very low frequencies.

From a technical point of view the variance of noise is described by a four-point correlation function [1,8]. Such objects appear also in other physical contexts. For example, Weissman [9,10] has proposed to distinguish the droplet and hierarchical models of spin glasses by the properties of the second noise, which can be expressed in terms of a particular four-spin correlation function.

More recently, the problem of $1/f$ noise has attracted much attention in the field of superconducting quantum devices. Flux noise measurements initially performed with relatively large superconducting quantum interference devices (SQUIDs) showed the $1/f$ behavior [11]. In the past decade a similar effect has been observed in nanoscale quantum circuits [12–23]. 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 [24–27]. Indeed, evidence for the existence of surface spins was obtained in several dedicated experiments [28,29]. All these experimental findings are consistent with a surface spin density of $\sigma_S \sim 5 \times 10^{17}\ \mathrm{m}^{-2}$.

Sendelbach *et al.* [30] 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 [15] 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 [31,32]. This relation seems to be justified in cases where the system is controlled by a slowly fluctuating parameter, always remaining in a quasiequilibrium. 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 [33–37] based on the Majorana-fermion representation of spin-1/2 systems [38–41]; and (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 are widely used to describe $1/f$-noise (the Dutta-Horn model [42]). 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 noninteracting 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 [43,44].

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 nonvanishing white noise of susceptibility that scales as N/T^2, where N is the number of spins and T is 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 noninteracting system.

II. 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 $\langle\langle x^n\rangle\rangle$. For a symmetric distribution, $P(x) = P(-x)$ such that $\langle x\rangle = 0$, the noise of x is defined as [1]

$$S_1 \equiv 2\langle x^2\rangle. \tag{1}$$

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 [1]

$$S_2 \equiv 2(\langle x^4\rangle - \langle x^2\rangle^2), \tag{2}$$

which is neither a higher moment nor a cumulant. This particular definition is motivated by the measurement protocol [1], in which the time fluctuations of x^2 are recorded (see also the Appendix). For a Gaussian random quantity (U is quadratic) one finds

$$\langle\langle x^4\rangle\rangle = 0, \quad \langle x^4\rangle = 3\langle x^2\rangle^2, \quad S_2 = 4\langle x^2\rangle^2. \tag{3}$$

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

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

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

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

where

$$a = \tfrac{1}{2}(\langle x^4\rangle - 3\langle x^2\rangle^2). \tag{6}$$

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 fourth cumulant of x and, therefore, is inequivalent to the second noise S_2.

In typical experiments [6,7,30], the fluctuating quantity is time-dependent and instead of the averages (5) one has to consider correlation functions (see below). The noise is characterized by the spectral power [1,2], 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 χ 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.] By 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 [30]. 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 [2,45,46] that out-of-equilibrium the noise may be lower than the equilibrium noise at the same temperature.

As an illustration for such a negative nonequilibrium 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 nonequilibrium spin fluctuations to be described by the negative quantity (6) 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. [30].

III. NONEQUILIBRIUM SPIN FLUCTUATIONS AND NOISE OF THE SUSCEPTIBILITY

We now generalize the above arguments to the case of a quantum spin system. Assume a coupling $\hat{H}_I = -\hat{\boldsymbol{S}}\boldsymbol{B}(t)$, where $\hat{\boldsymbol{S}}$ is the spin operator and $\boldsymbol{B}$ 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 [47]

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

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 [1]

$$S_M(\omega) \equiv \langle \hat{S}_z(t)\hat{S}_z(t') + \hat{S}_z(t')\hat{S}_z(t)\rangle_\omega = 2\coth\frac{\omega}{2T}\mathrm{Im}\chi(\omega). \quad (8)$$

The quantity (8) 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 [48]. In this formalism, the spin susceptibility (7) has the form

$$\chi_{ij}(t,t') = i\big\langle \mathcal{T}_K \hat{S}_i^{cl}(t)\hat{S}_j^{q}(t')\big\rangle_0 = -i\frac{\delta^2 \mathcal{Z}[\lambda^{cl},\lambda^q]}{\delta\lambda_i^q(t)\delta\lambda_j^{cl}(t')}\bigg|_{\lambda=0}, \quad (9)$$

where $\mathcal{T}_K$ denotes time ordering along the Keldysh contour, and $\mathcal{Z}[\lambda^{cl},\lambda^q]$ is the Keldysh partition function with the source terms $\lambda^{cl(q)}$ included. The subscripts q and cl on both the spin operators and source fields refer to the so-called "quantum" and "classical" variables [48]. They are related to the fields belonging to the upper (u) and lower (d) branches of the Keldysh contour according to

$$\hat{S}_i^{cl} = \frac{1}{\sqrt{2}}(\hat{S}_i^u + \hat{S}_i^d), \quad \hat{S}_i^q = \frac{1}{\sqrt{2}}(\hat{S}_i^u - \hat{S}_i^d),$$
$$\lambda_i^{cl} = \frac{1}{\sqrt{2}}(\lambda_i^u + \lambda_i^d), \quad \lambda_i^q = \frac{1}{\sqrt{2}}(\lambda_i^u - \lambda_i^d).$$

The "classical" source term defined in this way describes the physical probing field, $\lambda_i^{cl} \equiv \sqrt{2}B_i$, 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 the z direction. The symmetrized second moment of the z component of the physical spin is then given by

$$\begin{aligned}\langle \hat{S}_z(t_1)\hat{S}_z(t_2) + \hat{S}_z(t_2)\hat{S}_z(t_1)\rangle_B &= \big\langle \mathcal{T}_K \hat{S}_z^{cl}(t_1)\hat{S}_z^{cl}(t_2)\big\rangle_B \\ &= -\frac{\delta^2 Z\big[\lambda_z^q,B\big]}{\delta\lambda_z^q(t_1)\delta\lambda_z^q(t_2)}\bigg|_{\lambda_z^q=0} \\ &= \big\langle \mathcal{T}_K \hat{S}_z^{cl}(t_1)\hat{S}_z^{cl}(t_2)e^{i\int dt'\sqrt{2}B\hat{S}_z^q}\big\rangle. \end{aligned} \quad (10)$$

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,

$$\big\langle \hat{T}_K S_z^{cl}(t_1)\hat{S}_z^{cl}(t_2)\big\rangle_B = S_M(t_1 - t_2) + \int dt_1' dt_2' C_\chi(t_1,t_1',t_2,t_2') \times B(t_1')B(t_2') + \mathcal{O}(B^4). \quad (11)$$

The first term in Eq. (11) corresponds to the equilibrium noise (8) as it should be: The noise is characterized by the second cumulant of the fluctuating quantity in the absence of the applied field, similar to Eqs. (1) and (8). For the spin-1/2, it obeys the "sum rule" $S_M(0) = 1/2$.

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

$$C_\chi(t_1,t_1',t_2,t_2') = -\frac{\delta^4 Z[\lambda^{cl},\lambda^q]}{\delta\lambda_z^q(t_1)\delta\lambda_z^{cl}(t_1')\delta\lambda_z^q(t_2)\delta\lambda_z^{cl}(t_2')}\bigg|_{\lambda=0} = -\big\langle \mathcal{T}_K \hat{S}_z^{cl}(t_1)\hat{S}_z^q(t_1')\hat{S}_z^{cl}(t_2)\hat{S}_z^q(t_2')\big\rangle, \quad (12)$$

which one can split into Gaussian and non-Gaussian parts

$$C_\chi = C_\chi^G + C_\chi^{NG}.$$

The Gaussian part is readily obtained by a pairwise averaging of the spin operators:

$$\begin{aligned}C_\chi^G(t_1,t_1',t_2,t_2') &= -\big\langle \mathcal{T}_K \hat{S}_z^{cl}(t_1)\hat{S}_z^q(t_1')\big\rangle\big\langle \mathcal{T}_K \hat{S}_z^{cl}(t_2)\hat{S}_z^q(t_2')\big\rangle \\ &\quad - \big\langle \mathcal{T}_K \hat{S}_z^{cl}(t_1)\hat{S}_z^q(t_2')\big\rangle\big\langle \mathcal{T}_K \hat{S}_z^{cl}(t_2)\hat{S}_z^q(t_1')\big\rangle \\ &= \chi_{zz}(t_1,t_1')\chi_{zz}(t_2,t_2') + \chi_{zz}(t_1,t_2')\chi_{zz}(t_2,t_1'). \end{aligned} \quad (13)$$

Note the absence of a contribution involving two "quantum" fields. Such terms vanish since the correlator of the two "quantum" fields is always zero.

As mentioned above, for the spin-1/2 the moment (10) at equal times $t_1 = t_2 = t$ is equal to 1/2 independently of the magnetic field and therefore $C_\chi(t,t_1',t,t_2') = 0$. Clearly, the Gaussian contribution (13) does not satisfy this "sum rule" and thus there must be a non-Gaussian contribution C_χ^{NG} as well. More generally, 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 [1,30], 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:

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

The measurement is performed for a time period T_χ centered around τ_n. This defines the measurement bandwidth $\Delta\omega \equiv 2\pi/T_\chi \ll \omega_0$, chosen to be much smaller that ω_0. The phase φ 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 [30]), 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

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

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 second cumulant, which is obtained by subtracting two times the product of averages, i.e., $2\langle\hat{S}(t_1)\rangle\langle\hat{S}(t_2)\rangle$, as in Eq. (15).

We now use the expansion (11) for the symmetrized average (10) 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:

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

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

$$\chi^{(2)}_{eq,\varphi_1,\varphi_2}(\nu|\omega_0,\Delta\omega) = \frac{1}{4B_0^2} f\left(\frac{\pi\nu}{\Delta\omega}\right)\{\cos(\varphi_1-\varphi_2)[S_M(\omega_0+\nu)+S_M(\omega_0-\nu)] - i\sin(\varphi_1-\varphi_2)[S_M(\omega_0+\nu)-S_M(\omega_0-\nu)]\} + \mathcal{O}\left(\frac{\Delta\omega}{\omega_0}\right), \quad (18)$$

where $f(x) \equiv \sin^2(x)/x^2$ restricts the frequency ν to be small, $\nu \lesssim \Delta\omega \ll \omega_0$. The appearance of the imaginary part in the noise spectrum (18) 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 τ_i 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 nonequilibrium 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_χ^{NG} of the correlation function (12) 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

$$\chi^{(2)}_{ne,\varphi_1,\varphi_2}(\tau_1,\tau_2|\omega_0,\Delta\omega) = \frac{1}{T_\chi^2}\int_{\tau_1-\frac{T_\chi}{2}}^{\tau_1+\frac{T_\chi}{2}} dt_1 \int_{\tau_2-\frac{T_\chi}{2}}^{\tau_2+\frac{T_\chi}{2}} dt_2 \cos(\omega_0 t_1 - \varphi_1)\cos(\omega_0 t_2 - \varphi_2) \times \int dt_1' dt_2' C_\chi^{NG}(t_1,t_1',t_2,t_2')\cos(\omega_0 t_1')\cos(\omega_0 t_2'). \quad (20)$$

The time integrals in Eq. (20) can be simplified with the help of the Fourier transform defined as follows:

$$C_\chi^{NG}(t_1,t_1',t_2,t_2') = \int \frac{d\nu d\omega_1 d\omega_2}{(2\pi)^3} C_\chi^{NG}(\nu,\omega_1,\omega_2) \times e^{-i\nu(t_1-t_2)} e^{-i\omega_1(t_1-t_1')} e^{-i\omega_2(t_2-t_2')}. \quad (21)$$

As stated above, in this paper we are only interested in low-frequency noise [30] $\nu \ll \Delta\omega \ll \omega_0$. Focusing on contributions that are slow functions of $\tau_1 - \tau_2$, we retain only the lowest harmonics and find

$$\chi^{(2)}_{ne,\varphi_1,\varphi_2}(\nu|\omega_0,\Delta\omega) = \frac{1}{16} f\left(\frac{\pi\nu}{\Delta\omega}\right)\left[\sum_{\epsilon_1,\epsilon_2=\pm1} e^{-i\epsilon_1\varphi_1} e^{-i\epsilon_2\varphi_2} C_\chi^{NG}(\nu,\epsilon_1\omega_0,\epsilon_2\omega_0) + \sum_{\epsilon=\pm1} e^{i\epsilon(\varphi_1-\varphi_2)} C_\chi^{NG}(\nu - 2\epsilon\omega_0,\epsilon\omega_0,-\epsilon\omega_0)\right]. \quad (22)$$

Thus, the nonequilibrium contribution to the noise of the susceptibility is a probe of non-Gaussian fluctuations in the system, in contrast to the second noise [1]. Below, we illustrate our general considerations by calculating C_χ^{NG} for the simplest model system, i.e., a single spin coupled to a dissipative environment.

IV. 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 $\hat{\vec{X}}$ coupled to the spin operator via the minimal Hamiltonian

$$H = \hat{\vec{s}} \cdot \hat{\vec{X}}. \quad (23)$$

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

$$\hat{\Pi}^{ab}_{\alpha\beta}(t,t') = \delta_{\alpha\beta}\Pi^{ab}(t,t') = \mathrm{Tr}\{\mathcal{T}_K \hat{X}^{\alpha,a}(t)\hat{X}^{\beta,b}(t')\}, \quad (24)$$

where Latin indices span the 2×2 Keldysh space $a,b = cl,q$ and Greek indices refer to the spin components $\alpha,\beta = x,y,z$.

The bath, being Ohmic, means that the following relation holds:

$$\Pi^R(\omega) - \Pi^A(\omega) = \lambda\omega, \tag{25}$$

where $\lambda \ll 1$ is the effective coupling constant and $\Pi^{R/A}$ are the retarded and the advanced components of Eq. (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

$$\Pi^K(\omega) = \coth\frac{\omega}{2T}[\Pi^R(\omega) - \Pi^A(\omega)]. \tag{26}$$

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 [33–41]

$$\hat{s}^\alpha = -(i/2)\epsilon_{\alpha\beta\gamma}\eta_\beta\eta_\gamma, \quad \eta_\alpha^\dagger = \eta_\alpha, \tag{27}$$

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

$$\{\eta_\alpha,\eta_\beta\} = \delta_{\alpha\beta}, \quad \eta_\alpha^2 = 1/2. \tag{28}$$

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 $\hat{s}^\alpha$.

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 [35,39,41] that can be resolved on the basis of the known [36] but not widely appreciated conjecture: The fact that the Hilbert space is bigger that 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 [34]. 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

$$H = -(i/2)\epsilon_{\alpha\beta\gamma}\hat{X}^\alpha\eta_\beta\eta_\gamma. \tag{29}$$

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

$$\langle\hat{s}^\alpha(t_1)\hat{s}^\beta(t_1')\hat{s}^\gamma(t_2)\hat{s}^\delta(t_1')\rangle = (1/4)\langle\eta_\alpha(t_1)\eta_\beta(t_1')\eta_\alpha(t_2)\eta_\beta(t_2')\rangle.$$

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 $\mathcal{T}_K$. 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 η_α the time ordering is different. Therefore, it is convenient to multiply every Majorana fermion in the above correlator by $-im$, 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 $-im\eta_\alpha$ behave as bosons similarly to the spin operators. The correlation function (12) then takes the form

$$\begin{aligned}C_\chi(t_1,t_1',t_2,t_2') &= -\langle\mathcal{T}_K\hat{s}^{cl}(t_1)\hat{s}^q(t_1')\hat{s}^{cl}(t_2)\hat{s}^q(t_1')\rangle\\ &= -\big\langle\mathcal{T}_K(\eta_x m)^{cl}_{t_1}(\eta_x m)^q_{t_1'}(\eta_x m)^{cl}_{t_2}(\eta_x m)^q_{t_2'}\big\rangle.\end{aligned} \tag{30}$$

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

$$\begin{aligned}G^R_{0,\alpha}(t,t') &= -i\big\langle\mathcal{T}_K\eta^{cl}_\alpha(t)\eta^q_\alpha(t')\big\rangle = -i\Theta(t-t'),\\ D^R(t,t') &= -i\langle\mathcal{T}_K m^{cl}(t)m^q(t')\rangle = -i\Theta(t-t').\end{aligned} \tag{31}$$

The coupling of the Majorana fermions η_α to the bath (23) is then described by means of a "self-energy," which can be obtained as a saddlepoint solution in the path-integral formulation of the theory [50]. At sufficiently high 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

$$\Sigma^R_\alpha = -i\Gamma = -i\lambda T. \tag{32}$$

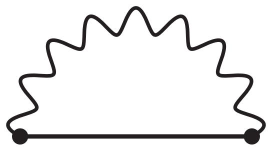

FIG. 1. The leading contribution to the self-energy. The wavy line refers to the bosonic correlator (26) and the solid line to $G_{0,\alpha}$; see Eq. (31).

Consequently, the Green's functions of the Majorana fermions η_α in the model (23) take the simple form

$$G_\alpha^{R/A}(\omega) = \frac{1}{\omega \pm i\Gamma},$$
$$G_\alpha^K(\omega) = -\frac{2i\Gamma}{\omega^2+\Gamma^2}\tanh\frac{\omega}{2T}. \tag{33}$$

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 (33) of the Majorana fermions η_α and the dashed line refers to the Green's function D of the noninteracting Majorana fermion m; see Eq. (31). The noise of the real part of susceptibility is identical with that of the imaginary part

$$\chi^{(2)}_{eq,00}(\nu|\omega_0,\Delta\omega) = \chi^{(2)}_{eq,\frac{\pi}{2},\frac{\pi}{2}}(\nu|\omega_0,\Delta\omega) = \frac{1}{4B_0^2} f\left(\frac{\pi\nu}{\Delta\omega}\right)\left[\frac{\Gamma}{\Gamma^2+(\omega_0+\nu)^2} + \frac{\Gamma}{\Gamma^2+(\omega_0-\nu)^2}\right], \tag{34a}$$

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

$$\chi^{(2)}_{eq,0,\frac{\pi}{2}}(\nu|\omega_0,\Delta\omega) = \frac{i}{4B_0^2} f\left(\frac{\pi\nu}{\Delta\omega}\right)\left[\frac{\Gamma}{\Gamma^2+(\omega_0+\nu)^2} - \frac{\Gamma}{\Gamma^2+(\omega_0-\nu)^2}\right]. \tag{34b}$$

The result (34b) is an odd function of ν, 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.

FIG. 2. The Majorana representation of the two-spin correlation function. This representation is exact, as the additional Majorana fermion m does not enter the Hamiltonian. The double solid line corresponds to the "dressed" Green's functions (33) of the Majorana fermions η_α and the dashed line refers to the Green's function D of the noninteracting Majorana fermion m; see Eq. (31).

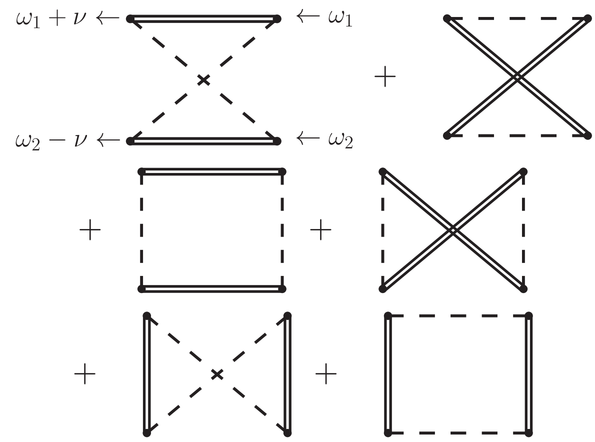

FIG. 3. The leading contribution to nonequilibrium noise of susceptibility.

Now we turn to the description of the more interesting, nonequilibrium noise of susceptibility. For this purpose, we need to evaluate the correlation function (30). Within the path-integral formulation of the theory [50], we may integrate out the bosonic bath and obtain the effective action for the Majorana fermions. This action can be formally expanded around the saddlepoint 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 sufficiently high 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

$$C_\chi^{NG}(\nu,\omega_1,\omega_2) = \frac{i\Gamma^2}{8T^2}\frac{\omega_1+\omega_2+2i\Gamma}{(\omega_1+i\Gamma)(\omega_2+i\Gamma)(\omega_1+\nu+i\Gamma)(\omega_2-\nu+i\Gamma)}. \tag{35}$$

Higher order diagrams (see, e.g., Fig. 4) may be neglected as long as the coupling constant λ remains small. We have ruled out the possibility that ladder diagrams might contribute to the lowest order in λ. The details of this calculation will be presented in a separate publication [50].

Substituting the result (35) into Eq. (22), we find the nonequilibrium noise spectrum of the spin susceptibility in the model (23). For the noise of the real part of the susceptibility we find

$$\chi^{(2)}_{ne,00}(\nu|\omega_0,\Delta\omega) = \frac{f\left(\frac{\pi\nu}{\Delta\omega}\right)}{32T^2}\frac{\Gamma^3\left[\omega_0^2-3(\Gamma^2+\nu^2)\right]}{(\Gamma^2+\omega_0^2)\left[(\Gamma^2+\nu^2)^2+2(\Gamma^2-\nu^2)\omega_0^2+\omega_0^4\right]}. \tag{36a}$$

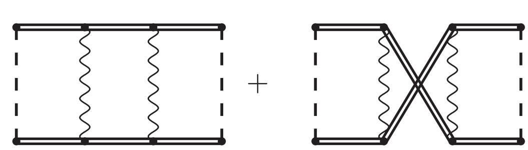

FIG. 4. Examples of higher order diagrams.

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

$$\chi^{(2)}_{ne,\frac{\pi}{2},\frac{\pi}{2}}(\nu|\omega_0,\Delta\omega) = -\frac{f\left(\frac{\pi\nu}{\Delta\omega}\right)}{32T^2}\frac{\Gamma^3\left(\Gamma^2+5\omega_0^2+\nu^2\right)}{\left(\Gamma^2+\omega_0^2\right)\left[(\Gamma^2+\nu^2)^2+2(\Gamma^2-\nu^2)\omega_0^2+\omega_0^4\right]}. \tag{36b}$$

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

$$\chi^{(2)}_{ne,0,\frac{\pi}{2}}(\nu|\omega_0,\Delta\omega) = -\frac{f\left(\frac{\pi\nu}{\Delta\omega}\right)}{32T^2}\frac{\omega_0\Gamma^2\left(3\Gamma^2-\omega_0^2+\nu^2-4i\Gamma\nu\right)}{\left(\Gamma^2+\omega_0^2\right)\left[(\Gamma^2+\nu^2)^2+2(\Gamma^2-\nu^2)\omega_0^2+\omega_0^4\right]}. \tag{36c}$$

V. 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 [28,51]. The main features of the experimental results are often explained with the help of the generic model of paramagnetic spins [14,15,42]. The model consists of an ensemble of noninteracting spins, each coupled to a dissipative environment. It is assumed that the corresponding relaxation rates Γ vary with the distribution function in a certain interval between two cutoff scales Γ_L and Γ_H:

$$p(\Gamma) = \frac{1}{\ln\left(\frac{\Gamma_H}{\Gamma_L}\right)}\frac{1}{\Gamma}. \tag{37}$$

As a single spin has the Lorentzian-shaped noise spectrum, by averaging over $p(\Gamma)$ one obtains a $1/f$-noise spectrum of the whole system within the frequency range $\Gamma_L < f < \Gamma_H$. The resulting noise is roughly independent of temperature [15] and may be used to fit the experimental data. Weak deviations of the exponent α of the measured noise spectra [11,20,21,52] $S_\phi \propto 1/f^\alpha$ can be accounted for by changing the distribution function [9] to $p(\Gamma) \propto 1/\Gamma^\alpha$.

The paramagnetic model does not include interactions between spins. Indeed [27], typical interaction scales seem to be small, of the order $J_{\text{typ}} \sim 50$ mK, justifying the approach of Refs. [14,15] in the high-temperature regime $T > J_{\text{typ}}$. This is also consistent with indications that the system of spins is in the classical high-temperature regime characterized by a Curie susceptibility [28,51] and an ohmic environment [53].

A system with a large number of noninteracting 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

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

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

$$\begin{aligned} C_\chi(t_1,t_1',t_2,t_2') &= -\langle \mathcal{T}_K \hat{S}_z^{cl}(t_1)\hat{S}_z^{q}(t_1')\hat{S}_z^{cl}(t_2)\hat{S}_z^{q}(t_2')\rangle \\ &= -\sum_i^N \langle \mathcal{T}_K \hat{s}_{z,i}^{cl}(t_1)\hat{s}_{z,i}^{q}(t_1')\hat{s}_{z,i}^{cl}(t_2)\hat{s}_{z,i}^{q}(t_2')\rangle \\ &\quad -\sum_{i\neq j}^N \langle \mathcal{T}_K \hat{s}_{z,i}^{cl}(t_1)\hat{s}_{z,i}^{q}(t_1')\rangle\langle \mathcal{T}_K \hat{s}_{z,j}^{cl}(t_2)\hat{s}_{z,j}^{q}(t_2')\rangle \\ &\quad -\sum_{i\neq j}^N \langle \mathcal{T}_K \hat{s}_{z,i}^{cl}(t_1)\hat{s}_{z,i}^{q}(t_2')\rangle\langle \mathcal{T}_K \hat{s}_{z,j}^{cl}(t_2)\hat{s}_{z,j}^{q}(t_1')\rangle. \end{aligned} \tag{38}$$

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

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

Averaging over the distribution (37) one obtains [9,15,24]

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

Using our results (34) and (36) we can now obtain the noise of the susceptibility in the model of noninteracting spins. In the limit, where the probing frequency ω_0 is much smaller than the slowest relaxation rate of the spins $\omega_0 \ll \Gamma_L$ we find

$$\begin{aligned} X^{(2)}_{0,0} &\approx \frac{N}{4\Gamma_L}\left[\frac{2}{B_0^2}-\frac{3}{8T^2}\right] f\left(\frac{\pi\nu}{\Delta\omega}\right)\ln^{-1}\frac{\Gamma_H}{\Gamma_L}, \\ X^{(2)}_{\frac{\pi}{2},\frac{\pi}{2}} &\approx \frac{N}{4\Gamma_L}\left[\frac{2}{B_0^2}-\frac{1}{8T^2}\right] f\left(\frac{\pi\nu}{\Delta\omega}\right)\ln^{-1}\frac{\Gamma_H}{\Gamma_L}, \\ X^{(2)}_{0,\frac{\pi}{2}} &\approx -\frac{N\omega_0}{4\Gamma_L^2}\left[\frac{3}{16T^2}+\frac{i\nu}{\Gamma_L}\left(\frac{4}{3B_0^2}-\frac{1}{6T^2}\right)\right] \\ &\quad\times f\left(\frac{\pi\nu}{\Delta\omega}\right)\ln^{-1}\frac{\Gamma_H}{\Gamma_L}. \end{aligned} \tag{41}$$

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

$$\begin{aligned} X^{(2)}_{0,0} &\approx \frac{\pi N}{4\omega_0}\left[\frac{1}{B_0^2}-\frac{1}{16T^2}\right] f\left(\frac{\pi\nu}{\Delta\omega}\right)\ln^{-1}\frac{\Gamma_H}{\Gamma_L}, \\ X^{(2)}_{\frac{\pi}{2},\frac{\pi}{2}} &\approx \frac{\pi N}{4\omega_0}\left[\frac{1}{B_0^2}-\frac{1}{16T^2}\right] f\left(\frac{\pi\nu}{\Delta\omega}\right)\ln^{-1}\frac{\Gamma_H}{\Gamma_L}, \\ X^{(2)}_{0,\frac{\pi}{2}} &\approx -\frac{N}{4\omega_0}\left[\frac{1}{16T^2}+\frac{i\pi\nu}{\omega_0}\left(\frac{1}{B_0^2}-\frac{1}{16T^2}\right)\right] \\ &\quad\times f\left(\frac{\pi\nu}{\Delta\omega}\right)\ln^{-1}\frac{\Gamma_H}{\Gamma_L}. \end{aligned} \tag{42}$$

The above results do not show the $1/\nu$ behavior observed in Ref. [30]. Moreover, the experiment shows nonvanishing 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, Atalaya, Clarke, and the two of us [54] 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.

To conclude, in this paper we have (i) given a general definition of noise of susceptibility and distinguished it from the second noise; (ii) computed the noise of susceptibility of a single spin-1/2 immersed in a dissipative environment; (iii) further developed the powerful technique [33–37] based on the Majorana-fermion representation of a spin-1/2 system [38–41]; and (iv) estimated the noise of susceptibility for the Dutta-Horn [42] model of independent spins. The Dutta-Horn model appears to be insufficient to account for all features observed in Ref. [30]. We conjecture that strong spin-spin interactions, leading to cluster formation and glassy behavior, are the key ingredients needed to explain the experiment.

ACKNOWLEDGMENTS

We acknowledge discussions with A. Mirlin, Yu. Makhlin, and J. Clarke. We acknowledge support of the DFG under Grants No. SCHO 287/7-1 and No. SH 81/2-1.

APPENDIX: NOISE OF NOISE

Here we briefly review the four-point correlation function corresponding to the second spectrum or noise of noise [1,9]. For more details the reader is referred to the book by Kogan [1].

The second spectrum was introduced in order to identify non-Gaussian contributions in $1/f$ spectra [1,9,55]. 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 has a form similar to Eq. (14):

$$\delta x(\tau|\omega_0,\Delta\omega) = \frac{1}{T_s}\int_{\tau-\frac{T_s}{2}}^{\tau+\frac{T_s}{2}} dt\, e^{i\omega_0 t}\,\delta x(t). \quad \text{(A1)}$$

The time T_s is similar to T_χ 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 δx 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 [1]

$$2\langle|\delta x(t|\omega_0,\Delta\omega)|^2\rangle = \int\frac{d\omega}{2\pi} f\left(\frac{\pi(\omega_0-\omega)}{\Delta\omega}\right)S_x(\omega) \approx \frac{\Delta\omega}{2\pi}\,S_x(\omega_0). \quad \text{(A2)}$$

The so-called second spectrum $S_x^{(2)}$ is a measure of fluctuations of the noise power. The definition reads [1]

$$S_x^{(2)}(\nu|\omega_0,\Delta\omega) = \frac{8}{T_t}\left\langle\left|\int_{-T_t/2}^{T_t/2} d\tau\, e^{i\nu\tau}(|\delta x(\tau|\omega_0,\Delta\omega)|^2 - \langle|\delta x(\tau|\omega_0,\Delta\omega)|^2\rangle)\right|^2\right\rangle. \quad \text{(A3)}$$

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

$$\langle|\delta x(\tau_1|\omega_0,\Delta\omega)|^2\,|\delta x(\tau_2|\omega_0,\Delta\omega)|^2\rangle = \frac{1}{T_s^4}\int_{\tau_1-\frac{T_s}{2}}^{\tau_1+\frac{T_s}{2}} dt_1 dt_1' \int_{\tau_2-\frac{T_s}{2}}^{\tau_2+\frac{T_s}{2}} dt_2 dt_2'\, e^{i\omega_0(t_1-t_1')}e^{i\omega_0(t_2-t_2')} \times C_x^{(4)}(t_1,t_1',t_2,t_2'). \quad \text{(A4)}$$

Here we introduced the classical four-point correlation function

$$C_x^{(4)}(t_1,t_1',t_2,t_2') = \langle\delta x(t_1)\delta x(t_1')\delta x(t_2)\delta x(t_2')\rangle. \quad \text{(A5)}$$

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

$$C_x^{(4,G)}(t_1,t_1',t_2,t_2') = C_x^{(2)}(t_1,t_1')C_x^{(2)}(t_2,t_2') + C_x^{(2)}(t_1,t_2)C_x^{(2)}(t_1',t_2') + C_x^{(2)}(t_1,t_2')\,C_x^{(2)}(t_1',t_2). \quad \text{(A6)}$$

One can now use Eq. (A6) to find the Gaussian contribution to the second spectrum. One finds that the first term on the right-hand side of Eq. (A6) cancels the average $\langle|\delta x|^2\rangle$ in Eq. (A3). The remaining two terms contribute yield the Gaussian contribution to the second noise spectrum

$$S_{x,G}^{(2)}(\nu|\omega_0,\Delta\omega) = 8\int\frac{d\Omega}{2\pi}\,C_x^{(2)}(\Omega)C_x^{(2)}(\Omega+\nu) \times f\left(\frac{\pi(\omega_0-\Omega)}{\Delta\omega}\right)f\left(\frac{\pi(\omega_0-\nu-\Omega)}{\Delta\omega}\right). \quad \text{(A7)}$$

In contrast, the noise of susceptibility does not contain any contribution of the Gaussian part of the 4-point correlation function (13).

In the experimentally relevant regime $\nu \ll \Delta\omega \ll \omega_0$ we can approximate Eq. (A7) as

$$S_{x,G}^{(2)}(\nu|\omega_0,\Delta\omega) \approx \frac{8\Delta\omega}{3\pi}\left(C_x^{(2)}(\omega_0)\right)^2. \quad \text{(A8)}$$

This spectrum is "white" as a function of ν.

Now, in the system of N noninteracting spins the noise spectrum scales linearly with the number of spins $S_s(\omega)\propto N$, while the Gaussian contribution to the second noise scales as $S_G^{(2)}(\nu|\omega,\Delta\omega)\propto N^2$; see Eq. (A6). In contrast, the non-Gaussian contribution is linear in N, similarly to Eq. (40). 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.

[1] In order to keep the discussion as simple as possible we use just one exponent in Eq. (A1) instead of the cosine in Eq. (14) in the main text. As a consequence, we have to add a factor of 2 to the expressions $|\delta x|^2$ compared to the notations in Ref. [1].

The above conclusion is not necessarily general. In context of Ising spin glasses, it has been shown [31,43] that the infinite-range interaction may result in a $1/f$ second noise spectrum in the thermodynamic limit $N \to \infty$.

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