Stochastic dynamics of magnetization in a ferromagnetic nanoparticle out of equilibrium

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(Received 22 September 2008; revised manuscript received 22 December 2008; published 26 February 2009)

We consider a small metallic particle (quantum dot) where ferromagnetism arises as a consequence of Stoner instability. When the particle is connected to electrodes, exchange of electrons between the particle and the electrodes leads to a temperature- and bias-driven Brownian motion of the direction of the particle magnetic moment. Under certain conditions this Brownian motion is described by the stochastic Landau-Lifshitz-Gilbert equation. As an example of its application, we calculate the frequency-dependent magnetic susceptibility of the particle in a constant external magnetic field, which is relevant for ferromagnetic resonance measurements.

DOI: 10.1103/PhysRevB.79.064418 PACS number(s): 73.23.-b, 73.40.-c, 73.50.Fq

I. INTRODUCTION

The description of fluctuations of the magnetization in small ferromagnetic particles pioneered by Brown¹ is based on the Landau-Lifshitz-Gilbert (LLG) equation^{2,3} with a phenomenologically added stochastic term. This approach has been widely used: just a few recent applications are studies of the dynamic response of the magnetization to the oscillatory magnetic field, 4-7 a numerical study of ferromagnetic resonance spectra, 8 a study of resistance noise in spin valves, 9 a study of the magnetization switching and relaxation in the presence of anisotropy and a rotating magnetic field, 10 and calculations of the power spectrum of magnetization fluctuations. 11,12 Typically, the treatment is based either on direct numerical integration of the stochastic LLG equation^{5,7,8,10,13} or on solution of the associated Fokker-Planck equation for the magnetization probability distribution. 1,4,6,10,11

In equilibrium the statistics of stochastic term in the LLG equation can be simply written from the fluctuationdissipation theorem. However, out of equilibrium a proper microscopic derivation is required. Microscopic derivations of the stochastic LLG equation out of equilibrium, available in the literature, use the model of a localized spin coupled to itinerant electrons, 14-17 or deal with noninteracting electrons. 18 In this model the derivation of the Fokker-Planck equation can be done using the standard density matrix formalism. 19 To put our work in the general context, we do this simple exercise in Appendix A. In contrast to the model of a localized spin, our main interest is a purely electronic system where the magnetization is a collective slow degree of freedom arising as a consequence of the Stoner instability. Our derivation has certain similarity with that of Ref. 20 for a bulk ferromagnet, where the direction of magnetization is fixed and cannot be changed globally, so its local fluctuations are small and their description by a Gaussian action is sufficient. The effect of electric current on itinerant electron ferromagnet near the Stoner instability was recently addressed in Refs. 21 and 22. The bulk situation should be contrasted to the case of a nanoparticle where the direction of the magnetization can be completely randomized by the fluctuations, so that the particle is not ferromagnetic in the strict sense (the long-time average of the magnetic moment vanishes). This situation is often called "superparamagnetism." From the formal point of view, as the overall variation in the magnetization vector is large, the effective action describing its slow motion is essentially non-Gaussian, and this is the situation we are interested in. The bias-driven Brownian motion of the magnetization with a fixed direction (due to an easy-axis anisotropy and ferromagnetic electrodes) has been also studied in Ref. 23 using rate equations, which corresponds to a fully quantum treatment of the problem. In contrast, our treatment is quasiclassical, which the most natural language when the total spin of the particle is large.

We assume that the single-electron spectrum of the particle, which is also called a quantum dot in the literature, to be chaotic and described by the random-matrix theory. 24,25 To take into account the electron-electron interactions in the dot we use the universal Hamiltonian,²⁶ with a generalized spin part corresponding to a ferromagnetic particle. Electrons occupy the quantum states of the full Hamiltonian and form a net spin of the particle of order of $S_0 \gg 1$; throughout the paper we use $\hbar = 1$. The dot is coupled to two leads, see Fig. 1, which we assumed to be nonmagnetic. The approach can be easily extended to the case of magnetic leads. The number $N_{\rm ch}$ of the transverse channels in the leads, which are well coupled to the dot, is assumed to be large, $N_{ch} \gg 1$. Equivalently, the escape rate $1/\tau$ of electrons from the dot into the leads is large compared to the single-electron mean level spacing δ_1 in the dot. In this situation one can disregard the electron-electron interaction in the charge channel, whose

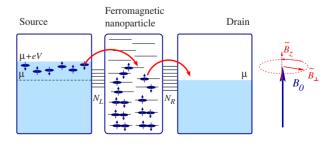


FIG. 1. (Color online) Device setup considered in this work: a small ferromagnetic particle (quantum dot) coupled to two nonmagnetic leads (see text for details).

effect (weak Coulomb blockade) is suppressed for $N_{\rm ch} \gg 1.^{25}$

The coupling to the leads is responsible for tunneling processes of electrons between states in the leads and in the dot with random spin orientation. As a result of such tunneling events, the net spin of the particle changes. We show that this exchange of electrons gives rise both to the Gilbert damping and the magnetization fluctuations in the presented model, and under conditions specified below, the time evolution of the particle spin is described by the stochastic LLG equation. In equilibrium the magnitude of these classical fluctuations is proportional to the system temperature T, by virtue of the fluctuation-dissipation theorem. Out of equilibrium (i.e., when a finite bias voltage V is applied between the contacts), the fluctuations are determined by the effective temperature $T_{\rm eff}$, the characteristic energy scale of the electronic distribution function given by a combination of T and V(the Boltzmann constant $k_B=1$ throughout the paper).

We study in detail the conditions for applicability of the stochastic approach. We find that these limits are set by three independent criteria. First, the contact resistance should be low compared to the resistance quantum, which is equivalent to $N_{\rm ch} > 1$. If this condition is broken, the statistics of the noise cannot be considered Gaussian. Physically, this condition means that each channel can be viewed as an independent source of noise, so the contribution of many channels results in the Gaussian noise by virtue of the central limit theorem if $N_{\rm ch} \gg 1$. Second, the system should not be too close to the Stoner instability: the mean-field value of the total spin $S_0 \gg \sqrt{N_{\rm ch}}$. If this condition is violated, the fluctuations of the absolute value of the magnetic moment become of the order of its average. Third, $S_0^2 \gg T_{\rm eff}/\delta_1$. Otherwise, the separation of the degrees of freedom into slow (the direction of the magnetic moment) and fast (the electron dynamics and the fluctuations of the absolute value of the magnetic moment) is not possible. This third condition, however, turns out to be weaker than $T_{\rm eff} \ll S_0 \delta_1$, which ensures that the magnetic moment is not destroyed by thermal fluctuations (i.e., the effective temperature must be below the Curie temperature).

Effects of spin-orbit interaction are beyond the scope of the present work. Spin-orbit interaction on the atomic length scale may lead to an intrinsic magnetic anisotropy of the particle; this effect can be easily incorporated by introducing the corresponding term in the universal Hamiltonian. At longer scales (of the order of the particle size), the spin-orbit interaction can modify the character of electron collisions with the walls as well as collisions between electrons, which can be an additional source of magnetization fluctuations and Gilbert damping. Incorporation of these effects is not trivial, but seems feasible within the technical framework presented here. These effects deserve a separate study.

Finally, as an application of the formalism, we consider the magnetic susceptibility in the ferromagnetic resonance measurements, which is a standard characteristic of magnetic samples. Recently, progress was reported in measurements of the magnetic susceptibility on small spatial scales in response to high-frequency magnetic fields.²⁷ Measurements of the ferromagnetic resonance were also reported for nanoparticles, connected to leads for a somewhat different setup in Ref. 28.

The paper is organized as follows. In Sec. III we introduce the model for electrons in a small metallic particle subject to Stoner instability. In Sec. IV we analyze the effective bosonic action for the magnetization of the particle. In Sec. V we obtain the equation of motion for the magnetization with the stochastic Langevin term, which has the form of the stochastic Landau-Lifshitz-Gilbert equation, and derive the associated Fokker-Planck equation. In Sec. VI we discuss the conditions for the applicability of the approach. In Sec. VII we calculate the magnetic susceptibility from the stochastic LLG equation.

II. QUALITATIVE PICTURE AND MAIN RESULTS

We consider a metallic nanoparticle containing a large number of electrons. When the Stoner criterion is satisfied, i.e., the electron-electron interaction in the spin channel is ferromagnetic and sufficiently strong, the ground state of the electrons in the particle corresponds to a nonzero total spin $S_0 \gg 1$. In the magnetically isotropic case, which we focus on, the spin can point in any direction, defined by a unit vector \boldsymbol{n} . The nonzero total spin can be seen to be due to an internal magnetic field (which can be also called magnetization field or exchange field) which shifts the energy levels of individual electrons with spins parallel/antiparallel to \boldsymbol{n} down/up by an amount h_0 . If δ_1 is the average spacing between the single-electron levels, there are $2h_0/\delta_1$ more electrons with spins along \boldsymbol{n} than those with opposite spins. Thus,

$$h_0 = S_0 \delta_1. \tag{2.1}$$

The large but finite degeneracy of the ground state, $2S_0+1 \gg 1$, is reminiscent of the Goldstone mode for an infinite bulk ferromagnet.

At a finite temperature T (not too high so that the average of the electron spin is still large, $T \leq h_0$), the electronic state will evolve. It is clear that even at lowest temperatures the system is energetically allowed to explore $(2S_0+1)$ -degenerate ground-state manifold. Representing the large spin quasiclassically, this dynamics can be viewed as that of the direction of the magnetic moment, n(t). We assume that the magnetic state of the particle is completely described by the total spin, the energy of spatially inhomogeneous spin configurations significantly exceeding all other relevant energy scales in the problem, such as the mean energy spacing in the particle, temperature or electric bias between the leads. In other words, we restrict our attention to the spin wave with zero wave vector, the modes with finite wave vectors having very high frequencies due to the small size of the particle.

The single-electron dynamics is assumed to be dominated by the exchange of electrons between the particle and the leads. This exchange is quantified by τ , the average time during which electron stays inside the particle before escaping into the leads. Separating the contributions of the left (τ_L) and right (τ_R) leads, we express them in terms of the transparency coefficients T_n for each channel of the leads connected to the particle, $0 < T_n < 1$:

$$\frac{1}{\tau} = \frac{1}{\tau_L} + \frac{1}{\tau_R}, \quad \frac{1}{\tau_{L(R)}} = \sum_{n \in L(R)} \frac{T_n \delta_1}{2\pi}.$$
 (2.2)

Our main assumption is $1/\tau \gg \delta_1$, which is the case when many channels, $N_{\rm ch} \gg 1$, have transparencies $T_n \sim 1$. This assumption allows us to neglect the electron-electron interaction in the charge channel, whose main effect, Coulomb blockade, is suppressed as $\sim 1/N_{\rm ch}$. We also neglect electron-electron collisions inside the particle, whose rate²⁹ is much smaller than the electron escape rate $1/\tau$. Under these assumptions the dynamics of the particle magnetic moment is governed by the exchange of electrons between the particle and the leads.

The analysis of this paper does not assume the system to be thermal equilibrium and is valid for arbitrary distribution functions of electron in the leads, which determine the distribution function in the particle. Our focus is when two leads have the same temperature but different chemical potentials, corresponding to a finite voltage bias U. In this case the electron distribution in the particle is strongly different from the equilibrium Fermi-Dirac form and cannot be characterized by a definite value of temperature. Nevertheless, it turns out that there exists a single energy scale determining the fluctuations, which can thus be called an effective temperature:

$$T_{\text{eff}} \equiv T + \Xi \left(\frac{eU}{2} \coth \frac{eU}{2T} - T \right),$$
 (2.3)

where Ξ is the "Fano factor" of the particle

$$\Xi = \frac{\tau^2}{\tau_L \tau_R} + \frac{\tau^3 \delta_1}{2 \pi \tau_R^2} \sum_{n \in L} T_n (1 - T_n) + \frac{\tau^3 \delta_1}{2 \pi \tau_L^2} \sum_{n \in R} T_n (1 - T_n).$$
(2.4)

At U=0 this expression gives $T_{\rm eff}$ =T and all our findings recover the results for equilibrium. At strong bias eU \gg T, when the spin fluctuations originate from the shot noise of electric current, it gives $T_{\rm eff}$ = $(\Xi/2)eU$.

Since each electron enters or leaves the particle in a random manner, and in each event the total spin change is much smaller than the spin itself, the dynamics of the spin will be diffusive. The corresponding diffusion coefficient can be estimated from the following simple argument. Significant fluctuations in the occupations of single-electron levels occur only within energy window $T_{\rm eff}$ near the Fermi energy. The number of single-electron levels in this window is $T_{\rm eff}/\delta_1$. Since a single electron changes the direction of the total spin typically by an angle $\Delta \theta \sim 1/S_0$, the variance of the deviation angle for the total spin is $\langle \Delta \theta^2 \rangle \sim (1/S_0)^2 (T_{\rm eff}/\delta_1)$. This deviation is accumulated during the typical lifetime of electrons in the particle, τ . The corresponding diffusion coefficient can be estimated as $\langle \Delta \theta^2 \rangle / \tau \sim T_{\text{eff}} / (S_0^2 \delta_1 \tau)$. At $T_{\rm eff} \lesssim \delta_1$, the same estimate is valid, but the ratio $T_{\rm eff}/\delta_1$ has the meaning of the probability of tunneling.

If an external magnetic field \mathbf{B} is applied, the ground-state degeneracy is lifted, the additional energy being $-g\mu_BS_0\mathbf{B}\cdot\mathbf{n}$, where g is the Lande factor for electrons in the nanoparticle, and $\mu_B = e/2m_ec$ is the Bohr magneton. In this case electron exchange between the particle and the leads results in the

relaxation of the spin direction toward the lowest energy state with n along B, referred to as the Gilbert damping. The relaxation rate can be estimated as follows. Suppose that initially $n \perp B$. This means that there are as many electrons with spins along B as with spins opposite to B. At the same time, the single-electron levels are shifted by $\pm g\mu_B B/2$ for the two spin projections, which results in a nonequilibrium state with the difference of the Fermi levels equal to $g\mu_BB$. Because electrons in the leads are not polarized, the difference in the number of incoming and outgoing electrons can be estimated as $g\mu_B B/\delta_1$. The orientation of the total spin changes by an angle $\Delta\theta \sim 1/S_0$ per electron, or by $\Delta\theta \sim g\mu_B B/(2\delta_1 S_0)$ during time τ . Thus, the damping rate can be estimated as $g\mu_B B/(2S_0\delta_1\tau)$. The investigation of the damping of magnetic moment of ferromagnetic particles due to electron exchange with the leads was presented previously in Ref. 30 and provided the expression for the Gilbert damping rate, consistent with our estimate.

In the main part of the paper these qualitative arguments are put on formal grounds. As a result, we arrive at the Fokker-Planck equation for the distribution function $\mathcal{P}(n)$ of the orientations n of the particle magnetic moment:

$$\frac{\partial \mathcal{P}}{\partial t} = g \mu_B \frac{\partial}{\partial \mathbf{n}} \cdot \left\{ -\left[\mathbf{n} \times \mathbf{B} \right] \mathcal{P} + \frac{\left[\mathbf{n} \times (\mathbf{n} \times \mathbf{B}) \right]}{2S_0 \delta_1 \tau} \mathcal{P} \right\} + \frac{1}{T_0} \frac{\partial^2 \mathcal{P}}{\partial \mathbf{n}^2}.$$
(2.5)

Here the first term on the right-hand side corresponds to the precession of magnetic moment in the direction perpendicular to the total magnetic field w+B, while the second term describes the Gilbert damping of magnetic moment, which tends to align the magnetic moment along the magnetic field. The last term represents diffusion, with the time constant \mathcal{T}_0 defined as

$$\mathcal{T}_0 = \frac{2S_0^2 \delta_1 \tau}{T_{\text{eff}}}.$$
 (2.6)

In the literature $T_0/2$ is often called the Néel time.

The Fokker-Planck equation, Eq. (2.5), is equivalent to the following Langevin equation (\dot{n} stands for the time derivative):

$$\dot{\mathbf{n}} = g \mu_B [\mathbf{n} \times (\mathbf{w} + \mathbf{B})] - \frac{g \mu_B}{2S_0 \delta_1 \tau} \{\mathbf{n} \times [\mathbf{n} \times (\mathbf{w} + \mathbf{B})]\}.$$
(2.7)

This equation corresponds to the LLG equation^{2,3} for the direction $\bf n$ of the macroscopic magnetic moment in a magnetic field: the first term on the right-hand side of Eq. (2.7) describes the precession, the second term describes the Gilbert damping. The magnetic field on the right-hand side of Eq. (2.7) is represented as a combination of an external deterministic field $\bf B$ and a stochastic field $\bf w$. The stochastic field is due to the interaction in the spin channel between electrons entering/leaving the particle and the rest of electrons, $\bf w(t)$ can be considered as a δ -correlated Gaussian field distributed isotropically in the tangent plane to a unit sphere. Its statistics is fully determined by the pair correlator which

depends on the direction of magnetic moment n(t) and has the form

$$\langle w_i(t)w_j(t')\rangle = \tau T_{\rm eff} \delta_1 \frac{\delta_{ij} - n_i(t)n_j(t)}{(2h_0\tau)^2 + 1} \delta(t - t'). \tag{2.8}$$

The conditions of applicability for the stochastic LLG equation, Eq. (2.7), and the Fokker-Planck equation, Eq. (2.5), can be summarized as follows:

(i) The fluctuations of the absolute value of the total spin are assumed to be small compared to its average S_0 . This is valid when

$$S_0^2 \gg \frac{1}{\tau \delta_1} \ln(E_T \tau),$$
 (2.9)

where E_T is the Thouless energy of the particle. For a particle of the size L and such that electron motion inside it is ballistic with the Fermi velocity v_F , $E_T \sim v_F/L$.

(ii) The Gaussian approximation for the stochastic field is effectively a consequence of the central limit theorem. Namely, the noise is contributed by many independent processes occurring in each conduction channel between the particle and the leads. This requirement can be written as

$$\frac{1}{\delta_1 \tau} \gg 1. \tag{2.10}$$

(iii) The Markovian approximation for the stochastic field w(t) is valid when its correlation time, $1/T_{\rm eff}$, is shorter than the typical time scale of the evolution:

$$\frac{1}{T_{\text{eff}}} \ll T_0. \tag{2.11}$$

This inequality can be rewritten as $S_0^2 \gg 1/(\delta_1 \tau)$, which is weaker than Eq. (2.9). Notice that the regular precession in the magnetic field with frequency $g\mu_B B$ is removed by a choice of the rotating coordinate system and does not restrict the applicability of the Markovian approximation.

(iv) The time scale of the evolution, \mathcal{T}_0 , must be longer than τ . This requirement translates into

$$S_0^2 \gg \frac{T_{\text{eff}}}{\delta_1}.\tag{2.12}$$

This condition is weaker than $T_{\rm eff} \lesssim S_0 \delta_1$, necessary to have a nonzero magnetic moment at all.

In the final part of the paper we apply the Fokker-Planck equation to calculate the linear magnetic susceptibility of the nanoparticle, describing the response to a weak probe magnetic field $\tilde{\boldsymbol{B}}(t)$ oscillating with the frequency ω in the presence of a constant magnetic field \boldsymbol{B}_0 . Since the only reference direction in the problem is determined by the field \boldsymbol{B}_0 , we can assume it to be along the z axis, and represent the probe field as a combination of the longitudinal and the two circularly polarized components:

$$\widetilde{B}(t) = [B_{\parallel}^{\omega} e_z + B_{+}^{\omega} (e_x + ie_y) + B_{-}^{\omega} (e_x - ie_y)] e^{-i\omega t} + \text{c.c.},$$
(2.13)

where "c.c." stands for the complex conjugate. The dimensionless longitudinal and transverse susceptibilities $\chi_{\parallel}(\omega)$ and $\chi_{\perp}(\omega)$ are defined by writing the linear response magnetic moment as

$$\begin{split} \widetilde{\boldsymbol{M}}(t) &= \frac{(g\mu_{B}S_{0})^{2}}{T_{\text{eff}}} \chi_{\parallel}(\omega) B_{\parallel}^{\omega} \boldsymbol{e}_{z} e^{-i\omega t} \\ &+ \frac{(g\mu_{B}S_{0})^{2}}{T_{\text{eff}}} \chi_{\perp}(\omega) B_{+}^{\omega}(\boldsymbol{e}_{x} + i\boldsymbol{e}_{y}) e^{-i\omega t} \\ &+ \frac{(g\mu_{B}S_{0})^{2}}{T_{\text{eff}}} \chi_{\perp}^{*}(-\omega) B_{-}^{\omega}(\boldsymbol{e}_{x} - i\boldsymbol{e}_{y}) e^{-i\omega t} + \text{c.c.} \end{split}$$

The susceptibilities depend also on the constant field B_0 (more precisely, on the dimensionless combination $b=g\mu_BB_0S_0/T_{\rm eff}$), and are obtained by solving the linearized Fokker-Planck equation; see Sec. VII. We have not been able to find an analytical expression for the susceptibilities, valid in the whole range of b and ω . However, from the numerical solution we deduce that they are well described (within a few percent) by the following approximate expressions:

$$\chi_{\parallel}^{\text{app}}(\omega, b) = \chi_{\parallel}^{\text{dc}}(b) \frac{1}{1 - i\omega \mathcal{T}_0 / \Gamma_{\text{app}}^{\parallel}(b)}, \qquad (2.15a)$$

$$\chi_{\perp}^{\text{app}}(\omega,b) = \chi_{\perp}^{\text{dc}}(b) \frac{1 - ig\mu_B B_0 S_0 \mathcal{T}_0 / \Gamma_{\text{app}}^{\perp}(b)}{1 - i(g\mu_B B_0 S_0 + \omega) \mathcal{T}_0 / \Gamma_{\text{app}}^{\perp}(b)},$$
(2.15b)

where

$$\chi_{\parallel}^{\text{dc}}(b) = \frac{1}{b^2} - \frac{1}{\sinh^2 b},$$
(2.16a)

$$\chi_{\perp}^{\text{dc}}(b) = \frac{b \coth b - 1}{b^2}$$
 (2.16b)

are the differential susceptibilities in a constant magnetic field, and

$$\Gamma_{\text{app}}^{\parallel}(b) = \frac{2}{h} \frac{\coth b - 1/b}{1/h^2 - 1/\sinh^2 h},$$
(2.17a)

$$\Gamma_{\text{app}}^{\perp}(b) = \frac{b^2}{b \coth b - 1} - 1.$$
(2.17b)

The algorithm for numerical calculation of the susceptibilities as well as their asymptotic behavior in various limiting cases is described in Sec. VII.

III. MODEL AND BASIC FORMALISM

Within the random matrix theory framework, electrons in a closed chaotic quantum dot are described by the following fermionic action:

$$S[\psi, \psi^*] = \oint dt \sum_{n,n'=1}^{N} \psi_n^{\dagger}(t) (\delta_{nn'} i \partial_t - H_{nn'}) \psi_{n'}(t)$$
$$- \oint dt \{ E[S(t)] - 2B(t) \cdot S(t) \}, \qquad (3.1a)$$

$$S_i \equiv \sum_{n=1}^{N} \psi_n^{\dagger} \frac{\hat{\sigma}^i}{2} \psi_n. \tag{3.1b}$$

Here ψ_n, ψ_n^{\dagger} are two-component Grassmann spinors, where the index $n=1,\ldots,N$ labels the orbital single-electron states inside the dot. The integration over time is performed along the forward-backward Keldysh contour, 31,32 as marked by \oint . The 2×2 Pauli matrices $\hat{\sigma}^i$, where i=x,y,z, act in the space of the two components of the spinors ψ_n . Throughout the paper we use the hat to indicate matrices in the spin space and use the notation $\hat{\sigma}_0$ for the 2×2 unit matrix. $\boldsymbol{B}(t)$ is the external magnetic field, measured in the energy units (such that the Zeeman splitting in a constant field is equal to $2\boldsymbol{B}$). The single-electron Hamiltonian of the dot is represented by an $N\times N$ random matrix $H_{nn'}$ from a Gaussian orthogonal ensemble, described by the pair correlators:

$$\overline{H_{mn}H_{m'n'}} = \frac{N\delta_1^2}{\pi^2} (\delta_{mn'}\delta_{nm'} + \delta_{mm'}\delta_{nn'}). \tag{3.2}$$

The constant δ_1 , which parametrizes the correlators, is the mean single-electron level spacing in the dot.

As mentioned in Sec. I, we do not include electronelectron interaction in the charge channel, whose effect is suppressed when the dot is well coupled to the leads. The electron-electron interaction in the spin channel is accounted for by the magnetization energy E(S), where S is the total spin of the dot. E(S) is the generalization of the J_sS^2 term in the universal Hamiltonian for the electron-electron interaction in a chaotic quantum dot.²⁶ Since we are going to describe a ferromagnetic state we must go beyond the quadratic term. Since the system is assumed to be far from the Stoner critical point, $S \ge 1$, all terms $(S^4, S^6, ...)$ should be included. E(S) can be viewed as the sum of all irreducible manyparticle vertices in the spin channel, obtained after integrating out degrees of freedom with high energies (above Thouless energy); the corresponding term in the action is thus local in time, and can be written as the time integral of an instantaneous function E[S(t)].

This functional can be decoupled using the Hubbard-Stratonovich transformation with a real vector field h(t), which we call below the internal magnetic field:

$$\exp\left[-i\oint dt E(S)\right] = \int \mathcal{D}\boldsymbol{h}(t) \exp\left\{i\oint dt \left[2\boldsymbol{h}\cdot\boldsymbol{S} - \widetilde{E}(\boldsymbol{h})\right]\right\}.$$
(3.3)

As a result, the action $S[\psi, \psi^*]$ can be rewritten in the form

$$S[\psi, \psi^*, \boldsymbol{h}] = \oint dt \sum_{n,n'=1}^{N} \psi_n^{\dagger}(t) (\hat{G}^{-1})_{nn'} \psi_{n'}(t) - \oint dt \widetilde{E}[\boldsymbol{h}(t)],$$
(3.4)

where the inverse Green's function,

$$(\hat{G}^{-1})_{nn'} = (i\hat{\sigma}_0 \partial_t + \boldsymbol{h} \cdot \hat{\boldsymbol{\sigma}} + \boldsymbol{B} \cdot \hat{\boldsymbol{\sigma}}) \delta_{nn'} - H_{nn'} \hat{\sigma}_0, \quad (3.5)$$

is a matrix in the time variables t,t', in the orbital indices $n,n'=1,\ldots,N$, in the spin indices, and in the forward (+) and backward (-) directions on the Keldysh contour. Integration over the fermionic fields ψ_n,ψ_n^{\dagger} yields the purely bosonic action:

$$S[\boldsymbol{h}] = -i \operatorname{Tr}[\ln(-i\hat{G}^{-1})] - \oint dt \widetilde{E}[\boldsymbol{h}(t)], \qquad (3.6)$$

where the trace is taken over *all* indices of the Green's function, listed above.

In the space of the forward and backward directions on the Keldysh contour, we perform the standard Keldysh rotation, 33,34 introducing the retarded (G^R) , advanced (G^A) , Keldysh (G^K) , and zero (G^Z) components of the Green's function:

$$\begin{pmatrix} \hat{G}^{R} & \hat{G}^{K} \\ \hat{G}^{Z} & \hat{G}^{A} \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix} \begin{pmatrix} \hat{G}^{++} & \hat{G}^{+-} \\ \hat{G}^{-+} & \hat{G}^{--} \end{pmatrix} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix},$$
(3.7)

as well as the classical (\mathbf{h}^{cl}) and quantum (\mathbf{h}^{q}) components of the internal magnetic field:

$$\begin{pmatrix} \mathbf{h}^{\text{cl}} & \mathbf{h}^{q} \\ \mathbf{h}^{q} & \mathbf{h}^{\text{cl}} \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ 1 & 1 \end{pmatrix} \begin{pmatrix} \mathbf{h}^{+} & 0 \\ 0 & -\mathbf{h}^{-} \end{pmatrix} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}. \quad (3.8)$$

We will also write this matrix as $h = h^{cl} \tau^{cl} + h^q \tau^q$, where τ^{cl} and τ^q are 2×2 matrices in the Keldysh space coinciding with the unit 2×2 matrix and the first Pauli matrix τ_x , respectively.

The saddle point of the bosonic action Eq. (3.6) is found by the first order variation with respect to $h^{cl,q}(t)$, which gives the self-consistency equation

$$\boldsymbol{h}^{q}(t) = 0, \quad -\frac{\partial \widetilde{E}[\boldsymbol{h}^{\text{cl}}(t)]}{\partial h_{i}^{\text{cl}}(t)} = \frac{i}{2} \text{Tr}[\hat{\sigma}^{j} \hat{G}_{nn}^{K}(t,t)]. \quad (3.9)$$

We also note that the right-hand side of this equation is proportional to the expectation value of the total spin of the particle for a given trajectory of the internal field h(t):

$$\mathbf{S}(t) = \frac{i}{4} \text{Tr} \{ \hat{\boldsymbol{\sigma}} \hat{\boldsymbol{G}}^K(t, t) \}. \tag{3.10}$$

In Eqs. (3.9) and (3.10), the trace is taken over orbital and spin indices only.

In the limit $N \rightarrow \infty$, one can obtain a closed equation for the Green's function traced over the orbital indices:³⁵

$$\hat{g}(t,t') = \frac{i\delta_1}{\pi} \sum_{n=1}^{N} \hat{G}_{nn}(t,t').$$
 (3.11)

The matrix $\hat{g}(t,t')$ satisfies the following constraint:

$$\int \hat{g}(t,t'')\hat{g}(t'',t')dt'' = \tau^{cl}\hat{\sigma}_0\delta(t-t'), \qquad (3.12)$$

where the right-hand side is just the direct product of unit matrices in the spin, Keldysh, and time indices. The Wigner transform of $\hat{g}^K(t,t')$ is related to the spin-dependent distribution function $\hat{f}(\epsilon,t)$ of electrons in the dot:

$$\int_{-\infty}^{\infty} \hat{g}^{K}(t+\tilde{t}/2,t-\tilde{t}/2)e^{i\epsilon\tilde{t}}d\tilde{t} = 2\hat{f}(\epsilon,t). \tag{3.13}$$

In equilibrium, $\hat{f}(\epsilon) = \hat{\sigma}_0 \tanh(\epsilon/2T)$. The self-consistency condition (3.9) takes the form

$$-\frac{\partial \widetilde{E}[\boldsymbol{h}^{\text{cl}}(t)]}{\partial h_{i}^{\text{cl}}(t)} = 2\boldsymbol{S}(t), \qquad (3.14a)$$

$$S(t) = \frac{\pi}{4 \delta_1} \lim_{t' \to t} \operatorname{Tr} \{ \hat{\boldsymbol{\sigma}} \hat{\boldsymbol{g}}^K(t, t') \} - \frac{\boldsymbol{h}^{\text{cl}}(t) + \boldsymbol{B}(t)}{\delta_1}. \quad (3.14b)$$

The second of these equations provides the relation between the internal magnetic field, a fictitious field used in the Hubbard-Stratonovich transformation, and the spin of the dot, which is an observable quantity. The last term in Eq. (3.14) takes care of the anomaly arising from noncommutativity of the limits $N{\longrightarrow}\infty$ and $t'{\longrightarrow}t$. As we will see in Sec. IV A, the anomalous term actually gives the dominant contribution to the value of the observable, as in the leading order of the gradient expansion (expansion in the "slowness" of spin fluctuations), Eq. (4.8), ${\rm Tr}_{\sigma}\{\hat{\pmb{\sigma}}\hat{g}^K\}$ vanishes, so one can simply set

$$S(t) \approx \frac{h^{\rm cl}}{\delta_1}.\tag{3.15}$$

In this paper we consider the dot coupled to two leads, identified as left (L) and right (R). The leads have N_L and N_R transverse channels, respectively, see Fig. 1. For nonmagnetic leads and spin-independent coupling between the leads and the particle, we can characterize each channel by its transmission T_n with $0 < T_n \le 1$ and by the distribution function of electrons in the channel $\mathcal{F}_n(t-t')$, assumed to be stationary. We consider the limit of strong coupling between the leads and the particle, $\sum_{n=1}^{N_{\rm ch}} T_n \gg 1$.

The coupling to the leads gives rise to a self-energy term, which should be included in the definition of the Green's function, Eq. (3.5). Without going into details of the derivation, presented in Ref. 35, we give the final form of the equation for the Green's function traced over the orbital states, Eq. (3.11):

$$\begin{aligned} \left[\partial_{t} - i\boldsymbol{h} \cdot \hat{\boldsymbol{\sigma}} - i\boldsymbol{B} \cdot \hat{\boldsymbol{\sigma}}, \hat{g} \right] \\ &= \sum_{n=1}^{N_{\text{ch}}} \frac{T_{n} \delta_{1}}{2\pi} \begin{pmatrix} -\mathcal{F}_{n} \hat{g}^{Z} & \hat{g}^{R} \mathcal{F}_{n} - \mathcal{F}_{n} \hat{g}^{A} - \hat{g}^{K} \\ \hat{g}^{Z} & -\hat{g}^{Z} \mathcal{F}_{n} \end{pmatrix} \\ &\times \left[\hat{1} + \frac{T_{n}}{2} \begin{pmatrix} \hat{g}^{R} - \hat{1} + \mathcal{F}_{n} \hat{g}^{Z} & \hat{g}^{R} \mathcal{F}_{n} + \mathcal{F}_{n} \hat{g}^{A} \\ 0 & -\hat{g}^{A} - \hat{1} + \hat{g}^{Z} \mathcal{F}_{n} \end{pmatrix} \right]^{-1}. \end{aligned}$$

$$(3.16)$$

Here the products of functions include convolution in time variables. This equation is analogous to the Usadel equation used in the theory of dirty superconductors.³⁶

To conclude this section, we discuss the dependence $\widetilde{E}(h)$. Deep in the ferromagnetic state, i.e., far from the Stoner critical point, we expect the mean-field approach to give a good approximation for the total spin of the dot. Namely, the mean field acting on the electron spins is given by $2h_0=dE(S)/dS\equiv E'(S)$. We then require that the response of the system to this field gives the same average value for the spin:

$$S_0 = \frac{2h_0}{2\delta_1} = \frac{E'(S_0)}{2\delta_1}. (3.17)$$

Here we evaluated S_0 from Eq. (3.10) and applied the self-consistency equation, Eq. (3.14), to equilibrium state with $\hat{g}^K \propto \hat{\sigma}_0$, when the contribution of the first term in the right-hand side of Eq. (3.14) vanishes.

Not expecting strong deviations of the magnitude of the spin from the mean-field value, we focus on the form of $\tilde{E}(h)$ when $|h| \approx h_0$. The inverse Fourier transform of Eq. (3.3) and angular integration for the isotropic E(S) gives

$$e^{-i\tilde{E}(h)\Delta t} = \text{const} \int_0^\infty \frac{\sin 2Sh\Delta t}{2Sh\Delta t} e^{-iE(S)\Delta t} S^2 dS,$$
 (3.18)

where Δt is the infinitesimal time increment used in the construction of the functional integral in Eq. (3.3).

Expanding E(S) near the mean-field value S_0 ,

$$E(S) \approx E(S_0) + E'(S - S_0) + \frac{E''}{2}(S - S_0)^2,$$
 (3.19)

performing the integration in the stationary phase approximation and using $S_0 = h_0 / \delta_1 = -E' / (2 \delta_1)$, we obtain

$$\widetilde{E}(h) = -2\frac{(h - h_0 - E''S_0/2)^2}{E''} + \widetilde{E}_0,$$
 (3.20)

where \tilde{E}_0 is *h*-independent term. This expression for $\tilde{E}(h)$ defines the action $\mathcal{S}[h]$, Eq. (3.6).

The energy E(S) does not contain the energy $E_{\rm orb}(S)$, associated with the orbital motion of electrons in the particle. Namely, to form a total spin S of the particle, we have to redistribute S electrons over orbital states, which changes the orbital energy of electrons by $E_{\rm orb}(S) \simeq \delta_1 S^2$. The total energy $E_{\rm tot}(S)$ of the particle is the sum of two terms: $E_{\rm tot}(S) = E(S) + E_{\rm orb}(S)$. Similarly, we obtain the total energy of the system in terms of internal magnetic field

$$\widetilde{E}_{\text{tot}}(h) = \widetilde{E}(h) - \frac{h^2}{\delta_1} = -2\left(\frac{1}{E''} + \frac{1}{2\delta_1}\right)(h - h_0)^2 + \widetilde{E}_1,$$
(3.21)

where \widetilde{E}_1 does not depend on h. We notice that the extremum of $\widetilde{E}_{\text{tot}}(h)$ corresponds to $h\!=\!h_0$ and describes the expectation value of the internal magnetic field in an isolated particle. The energy cost of fluctuations of the magnitude of the internal magnetic field is characterized by the coefficient $1/E''+1/2\delta_1$.

IV. EFFECTIVE ACTION

In this section we analyze the action Eq. (3.6) for the internal magnetic field h^{α} . We expect that the classical component $h^{cl}(t)$ of this field contains fast and small oscillations of its magnitude around the mean-field value h_0 . We further expect that the orientation of $h^{cl}(t)$ changes slowly in time but is not restricted to small deviations from some specific direction. Based on this picture, we introduce a unit vector n(t), assumed to depend slowly on time, and write

$$\boldsymbol{h}^{\text{cl}}(t) = [h_0 + h_{\parallel}^{\text{cl}}(t)]\boldsymbol{n}(t), \qquad (4.1)$$

where $h_{\parallel}^{\text{cl}}(t)$ is assumed to be fast and small. The external magnetic field $\boldsymbol{B}(t)$ is also assumed to be much smaller the internal one h_0 , and is treated on equal footing with $h_{\parallel}^{\text{cl}}(t)$. We expand action (3.6) to the second order in the quantum component $\boldsymbol{h}^q(t)$ and in the fluctuations of the radial classical component $h_{\parallel}^{\text{cl}}(t)$:

$$S[\boldsymbol{h}] \approx -\frac{2\pi}{\delta_{l}} \int dt \boldsymbol{g}^{K}(t,t) \cdot \boldsymbol{h}^{q}(t) + \frac{8}{E''} \int dt h_{\parallel}^{cl}(t) \boldsymbol{n}(t) \cdot \boldsymbol{h}^{q}(t)$$

$$- \int dt dt' \Pi_{ij}^{R}(t,t') h_{i}^{q}(t) [h_{\parallel}^{cl}(t') n_{j}(t') + B_{j}(t')]$$

$$- \int dt dt' \Pi_{ij}^{A}(t,t') [h_{\parallel}^{cl}(t) n_{i}(t) + B_{i}(t)] h_{j}^{q}(t')$$

$$- \int dt dt' \Pi_{ij}^{K}(t,t') h_{i}^{q}(t) h_{j}^{q}(t'). \tag{4.2}$$

The applicability of this quadratic expansion is discussed in Sec. VI B.

In the first term of Eq. (4.2) we introduced the vector part \mathbf{g}^K of the Keldysh component of the Green's function $\hat{\mathbf{g}}^K = \hat{\boldsymbol{\sigma}}_0 \mathbf{g}_0^K + \hat{\boldsymbol{\sigma}} \cdot \mathbf{g}^K$. (Indeed, an arbitrary matrix in the 2×2 spin space can be expressed as a linear combination of the unit matrix and the three Pauli matrices.)

In the last three terms of Eq. (4.2) we introduced the polarization operator, defined as the kernel of the quadratic part of the action of the fluctuating bosonic fields:

$$\begin{pmatrix} \Pi^{Z} & \Pi^{A} \\ \Pi^{R} & \Pi^{K} \end{pmatrix} \equiv \begin{pmatrix} \Pi^{\text{cl,cl}} & \Pi^{\text{cl,q}} \\ \Pi^{q,\text{cl}} & \Pi^{q,q} \end{pmatrix}, \tag{4.3a}$$

$$\Pi_{ij}^{\alpha\beta}(t,t') = \frac{i}{2} \frac{\delta^2 \operatorname{Tr}\{\ln G^{-1}\}}{\delta h_i^{\beta}(t') \, \delta h_i^{\alpha}(t)}, \tag{4.3b}$$

where $\alpha, \beta = \text{cl}, q$ and i, j = x, y, z. The $t' \rightarrow t$ anomaly is explicitly taken into account in the definition of the polarization operators; see Eq. (4.14) below.

We emphasize that the Green's function and the polarization operator appearing in Eq. (4.2) are calculated at $h_{\parallel}^{\rm cl}(t) = 0$ and $h^q(t) = 0$ for a given trajectory of the classical field $h_0 n(t)$. Their explicit calculation is performed in the following two subsections.

A. Keldysh component of the Green's function

Here we use the Usadel equation, Eq. (3.16), with $h^{cl}(t)=h_0 n(t)$, $h^q(t)=0$. For the Green's function we have

$$\hat{g}^{R}(t,t') = -\hat{g}^{A}(t,t') = \hat{\sigma}^{0}\delta(t-t'),$$
 (4.4)

while the Keldysh component satisfies the equation

$$\left[\partial_t - ih_0 \boldsymbol{n} \cdot \hat{\boldsymbol{\sigma}}, \hat{g}^K\right] = \sum_{n=1}^{N_{\text{ch}}} \frac{T_n \delta_1}{2\pi} (2\mathcal{F}_n - \hat{g}^K). \tag{4.5}$$

We introduce the notation

$$\frac{1}{\tau} = \sum_{n=1}^{N_{\text{ch}}} \frac{T_n \delta_1}{2\pi} = \frac{1}{\tau_L} + \frac{1}{\tau_R}.$$
 (4.6)

Then the scalar g_0^K and vector \mathbf{g}^K components of $\hat{\mathbf{g}}^K = \hat{\boldsymbol{\sigma}}_0 \mathbf{g}_0^K + \hat{\boldsymbol{\sigma}} \cdot \mathbf{g}^K$ satisfy two coupled equations:

$$\label{eq:def_equation} \begin{split} \left[\partial_t + \partial_{t'} + \frac{1}{\tau} \right] & g_0^K(t,t') = \sum_{n=1}^{N_{\rm ch}} \frac{T_n \delta_1}{2\pi} 2 \mathcal{F}_n(t-t') \\ & + i h_0 [\boldsymbol{n}(t) - \boldsymbol{n}(t')] \cdot \boldsymbol{g}^K(t,t'), \end{split} \tag{4.7a}$$

$$\left[\partial_t + \partial_{t'} + \frac{1}{\tau}\right] \mathbf{g}^K(t,t') + h_0[\mathbf{n}(t) + \mathbf{n}(t')] \mathbf{g}^K(t,t')$$

$$= ih_0[\mathbf{n}(t) - \mathbf{n}(t')] \mathbf{g}_0^K(t,t'). \tag{4.7b}$$

In the zero approximation, we can consider the stationary situation: $g_0^K(t,t')=g_0^K(t-t')$ and n(t)=const. In this case, we have

$$g_0^K(t,t') = \frac{\tau}{\tau_L} 2\mathcal{F}_L(t-t') + \frac{\tau}{\tau_R} 2\mathcal{F}_R(t-t'), \quad \mathbf{g}^K = 0. \eqno(4.8)$$

For an arbitrary time dependence n(t) Eqs. (4.7a) and (4.7b) cannot be solved analytically. However, if the variation of n(t) is slow enough, $|n|\tau \ll 1$, we can do the gradient expansion [expansion in the time derivatives of n(t)], restricting ourselves to the first derivative:

$$\left(\partial_{\bar{t}} + \frac{1}{\tau}\right) \mathbf{g}^K + 2h_0 \mathbf{n} \times \mathbf{g}^K = i\tilde{t}h_0 \dot{\mathbf{n}} g_0^K. \tag{4.9}$$

Here we introduced $\bar{t}=(t+t')/2$, $\tilde{t}=t-t'$, $\partial_t + \partial_{t'} = \partial_{\bar{t}}$. The dependence on \tilde{t} is split off and remains unchanged, while for

the dependence on \overline{t} the solution is determined by a linear integral operator \mathcal{L}_n^+ , conveniently determined in the Fourier

$$\mathcal{L}_{n}^{\pm} = \left(\frac{1}{\tau} \pm \partial_{\overline{t}} \pm 2h_{0}n \times\right)^{-1},\tag{4.10a}$$

$$\mathcal{L}_{n}^{+}(\omega)X(\omega) = \frac{n[n \cdot X(\omega)]}{-i\omega + 1/\tau} + \frac{1}{2} \sum_{\pm} \frac{-n \times [n \times X(\omega)] \pm i[n \times X(\omega)]}{-i(\omega \pm 2h_{0}) + 1/\tau}.$$
(4.10b)

Thus, all perturbations of g^{K} decay with the characteristic time τ . In particular, the solution of Eq. (4.9) has the form

$$\mathbf{g}^{K} = i\widetilde{t}h_{0}\mathcal{L}_{n}^{+}\dot{\mathbf{n}}g_{0}^{K} \approx i\widetilde{t}h_{0}\tau g_{0}^{K}\frac{\dot{\mathbf{n}} - 2h_{0}\boldsymbol{\tau}\mathbf{n} \times \dot{\mathbf{n}}}{(2h_{0}\tau)^{2} + 1}.$$
 (4.11)

Now, an explicit expression for the first term in Eq. (4.2) can be easily obtained from Eq. (4.11) by taking the limit $\tilde{t} \rightarrow 0$ and taking into account that any fermionic distribution function in the time representation has the following equaltime asymptotics:

$$g_0^K(t,t') \approx \frac{2}{i\pi} \frac{1}{t-t'}, \quad (t \to t').$$
 (4.12)

This asymptotics follows from Eq. (3.13) and the simple fact that the distribution function of a Fermi system in the energy representation always has the limits $f(\epsilon) \to \pm 1$ at $\epsilon \to \pm \infty$. We have

$$\mathbf{g}^{K}(t,t) = \frac{2h_{0}\tau \dot{\mathbf{n}} - 2h_{0}\tau \mathbf{n} \times \dot{\mathbf{n}}}{\pi (2h_{0}\tau)^{2} + 1}.$$
 (4.13)

We notice that $\mathbf{n} \cdot \mathbf{g}^{K}(t,t) = 0$, and therefore the first term in the action Eq. (4.2) is coupled only to the tangential fluctuations of $h^q(t) \perp n(t)$.

B. Polarization operator

For the polarization operator it is sufficient to take the leading (zeroth) order of the expansion in the time derivatives of n(t), i.e., to calculate $\prod_{ij}^{\alpha\beta}(t,t')$ for a fixed direction n, given by the instantaneous value of n(t). This calculation is done analogously to Ref. 35.

The polarization operator can be represented as the response of the Green's functions to a change in the field, as follows directly from definition (4.3) and expression (3.6) for the action

$$\Pi_{ij}^{\alpha\beta}(t,t') = \frac{\pi}{2\delta_{l}} \frac{\operatorname{Tr}_{4\times4} \{ \tau^{\alpha} \hat{\sigma}^{i} \delta \hat{g}(t,t) \}}{\delta h_{j}^{\beta}(t')} - \frac{2}{\delta_{l}} \tau_{\alpha\beta}^{q} \delta_{ij} \delta(t-t').$$

$$(4.14)$$

Here the Green's function $\delta \hat{g}(t,t)$ can be calculated as the first-order response of the solution of Eq. (3.16) to small arbitrary (in all three directions) increments of $\delta h^{cl}(t)$ and $\delta h^{q}(t)$. The zero-order solution of Eq. (3.16) in the field h^{cl} $=h_0\mathbf{n}$ and $\mathbf{h}^q=0$ is

$$\hat{g}(t,t) = \hat{\sigma}_0 \begin{pmatrix} \delta(t-t') & g_0^K(t-t') \\ 0 & -\delta(t-t') \end{pmatrix}. \tag{4.15}$$

First, we calculate $\delta \hat{g}^Z$, which responds only to δh^q :

$$\left[\partial_t + \partial_{t'} - \frac{1}{\tau} \right] \delta \hat{g}^Z(t, t') - ih_0 \hat{\boldsymbol{\sigma}} \cdot \boldsymbol{n}(t) \delta \hat{g}^Z(t, t')
+ \delta \hat{g}^Z(t, t') ih_0 \hat{\boldsymbol{\sigma}} \cdot \boldsymbol{n}(t') = 2i \hat{\boldsymbol{\sigma}} \cdot \delta \boldsymbol{h}^q(t) \delta(t - t').$$
(4.16)

Since $\partial_t + \partial_{t'} = \partial_{\overline{t}}$, the solution always remains proportional to $\delta(t-t')$:

$$\delta \hat{g}^{Z}(t,t') = -2i\hat{\boldsymbol{\sigma}} \cdot (\mathcal{L}_{\boldsymbol{n}}^{-} \delta \boldsymbol{h}^{q})(t) \,\delta(t-t'). \tag{4.17}$$

Given $\delta \hat{g}^Z$, components $\delta \hat{g}^{R,A}$ can be found either from Eq. (3.16), or, equivalently, using constraint (3.12):

$$\hat{g}\,\delta\hat{g} + \delta\hat{g}\,\hat{g} = 0 \quad \Rightarrow \quad \delta\hat{g}^R = -\frac{\hat{g}^K\delta\hat{g}^Z}{2}, \quad \delta\hat{g}^A = \frac{\delta\hat{g}^Z\hat{g}^K}{2}.$$
(4.18)

We notice that both $\delta \hat{g}^{R,A}$ respond only to $h^{q}(t)$ and, there-

$$\Pi_{ij}^{Z}(t,t') \propto \frac{\operatorname{Tr}\{\hat{\sigma}^{i}[\delta\hat{g}^{R}(t,t) + \delta\hat{g}^{A}(t,t)]\}}{\delta h_{i}^{cl}(t')} \equiv 0. \quad (4.19)$$

This equation ensures that the action along the Keldysh contour vanishes for $h^q \equiv 0$.

To evaluate the remaining three components of the polarization operator, we can apply the variational derivatives to the sum of $\delta \hat{g}^{K}(t,t) + \delta \hat{g}^{Z}(t,t)$ with respect to either classical $\delta h^{cl}(t')$ or quantum $\delta h^{q}(t')$ field, which give $\Pi_{ii}^{R}(t,t')$ and $\Pi_{ii}^{K}(t,t')$, respectively. Then, the advanced component $\Pi_{ii}^{A}(t,t') = [\Pi_{ii}^{R}(t',t)]^{*}.$

The equation for $\delta \hat{g}^K = \hat{\sigma} \cdot \delta \mathbf{g}^K$ reads as

$$\left[\partial_{t} + \partial_{t'} + \frac{1}{\tau} \right] \delta \mathbf{g}^{K}(t, t')
+ h_{0}[\mathbf{n}(t) \times \delta \mathbf{g}^{K}(t, t') - \delta \mathbf{g}^{K}(t, t') \times \mathbf{n}(t')]
= i \left[\delta \mathbf{h}^{\text{cl}}(t) - \delta \mathbf{h}^{\text{cl}}(t') \right] g_{0}^{K}(t - t') - 2i \delta \mathbf{h}^{q}(t) \delta(t - t')
- \mathbf{Q}(t, t'),$$
(4.20a)

where

$$Q = \sum_{n=1}^{N_{\text{ch}}} \frac{T_n \delta_1}{2\pi} \left(\frac{g_0^K}{2} \delta \mathbf{g}^Z \frac{g_0^K}{2} + \mathcal{F}_n \delta \mathbf{g}^Z \mathcal{F}_n \right)$$
$$- \sum_{n=1}^{N_{\text{ch}}} \frac{T_n (1 - T_n) \delta_1}{2\pi} \left(\frac{g_0^K}{2} - \mathcal{F}_n \right) \delta \mathbf{g}^Z \left(\frac{g_0^K}{2} - \mathcal{F}_n \right)$$

$$(4.20b)$$

and $\delta \mathbf{g}^Z = \text{Tr}\{\hat{\boldsymbol{\sigma}}\delta\hat{g}^Z\}/2$ with $\delta\hat{g}^Z$ given by Eq. (4.17). To calculate the retarded component Π^R_{ij} of the polarization operator, we calculate the response of $\delta \mathbf{g}^{K}(t,t')$ to $\delta \mathbf{h}^{q}$ in the limit $t' \rightarrow t$. Using the asymptotic behavior of the Fermi function, Eq. (4.12), we obtain

$$\delta \mathbf{g}^{K}(t,t) = \int \frac{d\omega}{2\pi} \frac{-2i\omega}{\pi} \mathcal{L}_{n}^{+}(\omega) \, \delta \mathbf{h}^{\text{cl}}(\omega) e^{-i\omega t}. \tag{4.21}$$

Substituting this expression for $\delta g^{K}(t,t)$ to Eq. (4.14), we obtain

$$\Pi_{ij}^{R}(\omega) = \Pi_{\parallel,ij}^{R}(\omega) + \Pi_{\perp,ij}^{R}(\omega), \qquad (4.22a)$$

with

$$\Pi_{\parallel,ij}^{R}(\omega) = -\frac{2}{\delta_1} \frac{n_i n_j}{1 - i\omega\tau},\tag{4.22b}$$

$$\Pi_{\perp,ij}^{R}(\omega) = -\frac{2}{\delta_{1}} \sum_{\pm} \frac{\delta_{ij} - n_{i}n_{j} \pm ie_{ijk}n_{k}}{2} \frac{(1 \pm 2ih_{0}\tau)}{1 - i(\omega \mp 2h_{0})\tau}.$$
(4.22c)

Here we represented the polarization operator $\Pi^R_{ij}(\omega)$ as a sum of the radial, $\Pi^R_{\parallel,ij}(\omega)$, and tangential, $\Pi^R_{\perp,ij}(\omega)$, terms. We note that the action Eq. (4.2) contains only the radial component of the retarded and advanced polarization operators because we do not perform expansion in terms of the tangential fluctuations of the classical component of the field $h^{\rm cl}(t)$.

In response to δh^q , both corrections $\delta g^K(t,t')$ and $\delta g^Z(t,t')$ contain terms $\propto \delta(t-t')$. However, their sum $\delta g^K(t,t') + \delta g^Z(t,t')$ remains finite in the limit $t \rightarrow t'$:

$$\delta \mathbf{g}^{K}(t,t') + \delta \mathbf{g}^{Z}(t,t') = -2i \int dt'' \int dt_{1} dt_{2} \mathcal{L}_{n}^{+}(\overline{t} - t_{1})$$

$$\times \mathcal{Q}(t_{1} - t_{2} + \overline{t}/2; t_{2} - t_{1} + \overline{t}/2)$$

$$\times \mathcal{L}_{n}^{-}(t_{2} - t'') \delta \mathbf{h}^{q}(t''), \qquad (4.23a)$$

$$Q(\tau_{1}; \tau_{2}) = \frac{2}{\tau} \delta(\tau_{1}) \delta(\tau_{2})$$

$$- \sum_{n=1}^{N_{ch}} \frac{T_{n} \delta_{1}}{2\pi} \left[\frac{g_{0}^{K}(\tau_{1})}{2} \frac{g_{0}^{K}(\tau_{2})}{2} + \mathcal{F}_{n}(\tau_{1}) \mathcal{F}_{n}(\tau_{2}) \right]$$

$$- \sum_{n=1}^{N_{ch}} \frac{T_{n} (1 - T_{n}) \delta_{1}}{2\pi} \left[\frac{g_{0}^{K}(\tau_{1})}{2} - \mathcal{F}_{n}(\tau_{1}) \right]$$

$$\times \left[\frac{g_{0}^{K}(\tau_{2})}{2} - \mathcal{F}_{n}(\tau_{2}) \right], \tag{4.23b}$$

with $\overline{t} = (t+t')/2$ and $\widetilde{t} = t-t'$.

From Eq. (4.22) we obtain the following expression for the Keldysh component of the polarization operator:

$$\Pi_{ij}^{K}(\omega) = \Pi_{\parallel,ij}^{K}(\omega) + \Pi_{\perp,ij}^{K}(\omega), \qquad (4.24a)$$

where

$$\Pi_{\parallel,ij}^{K}(\omega) = -i \frac{n_i n_j}{\omega^2 + 1/\tau^2} \mathcal{R}(\omega), \qquad (4.24b)$$

$$\Pi_{\perp,ij}^{K}(\omega) = -\frac{i}{2} \sum_{\pm} \frac{\delta_{ij} - n_i n_j \pm i e_{ijk} n_k}{(\omega \mp 2h_0)^2 + 1/\tau^2} \mathcal{R}(\omega). \quad (4.24c)$$

Here function $\mathcal{R}(\omega)$ coincides with the noise power of electric current through a metallic particle in the approximation of noninteracting electrons

$$\mathcal{R}(\omega) = \sum_{n=1}^{N_{\text{ch}}} \int \frac{d\epsilon}{8\pi} T_n \delta_1$$

$$\times \{ [8 - g_0^K(\epsilon) g_0^K(\epsilon + \omega) - 4\mathcal{F}_n(\epsilon) \mathcal{F}_n(\epsilon + \omega)] + (1 - T_n) [g_0^K(\epsilon) - 2\mathcal{F}_n(\epsilon)] [g_0^K(\epsilon + \omega) - 2\mathcal{F}_n(\epsilon + \omega)] \}.$$

$$(4.25)$$

In principle, electron-electron interaction in the charge channel can be taken into account. The interaction modifies the expression Eq. (4.25) for $\mathcal{R}(\omega)$ in higher orders³⁷ in $\tau \delta_1 \ll 1$ and we neglect this correction here.

In this paper we consider a particle connected to electron leads at temperature T with the applied voltage bias V/e. In this case, $\mathcal{F}_{L,R}(\epsilon) = \tanh(\epsilon - \mu_{L,R})/(2T)$ with $\mu_L - \mu_R = V$, and the integration over ϵ gives

$$2\pi\tau \mathcal{R}(\omega) = 4\omega \coth \frac{\omega}{2T} + \Xi Y_T(V, \omega), \qquad (4.26)$$

where

$$Y_T(V,\omega) \equiv \sum_{\pm} 2(\omega \pm V) \coth \frac{\omega \pm V}{2T} - 4\omega \coth \frac{\omega}{2T}$$
(4.27)

and Ξ is the "Fano factor" for a particle

$$\Xi = \frac{\tau^2}{\tau_L \tau_R} + \frac{\tau^3 \delta_1}{2\pi \tau_R^2} \sum_{n \in L} T_n (1 - T_n) + \frac{\tau^3 \delta_1}{2\pi \tau_{Ln \in R}^2} \sum_{n \in R} T_n (1 - T_n).$$
(4.28)

For low bias, $|V| \ll T$, the term $\Xi Y_T(V,\omega)$ represents just a small correction to the equilibrium (Nyquist) value of $\mathcal{R}(\omega)$. At $|V| \gg T$ the function $Y_T(V,\omega)$ has two scales of ω : (i) T smears the nonanalyticity at $\omega \to 0$, but the value of $Y_T(V,\omega)$ deviates from $Y_T(V,0)$ at $|\omega| \sim |V|$. Thus, the typical time scale above which one can approximate $\Pi^K(\omega)$ by a constant is at least $\omega \ll \max\{T, |V|\}$. In the limit $\omega \to 0$ we have

$$\Pi_{ij}^{K}(\omega=0) = -i\frac{8\tau T_{\text{eff}}}{\delta_{1}} \left(n_{i}n_{j} + \frac{\delta_{ij} - n_{i}n_{j}}{(2h_{0}\tau)^{2} + 1} \right), \quad (4.29)$$

where the effective temperature $T_{\rm eff}$ is given by

$$T_{\rm eff} \equiv T + \Xi \left(\frac{V}{2} \coth \frac{V}{2T} - T \right). \tag{4.30}$$

In the time domain the limit $\omega \rightarrow 0$ corresponds to approximating the polarization operator by

$$\Pi_{ij}^{K}(t,t') \approx \Pi_{ij}^{K}(\omega = 0) \delta(t - t'). \tag{4.31}$$

According to the abovesaid, this is valid for times $|t-t'| \gg 1/T_{\rm eff}$.

C. Final form of the action

Summarizing the results of the previous two subsections, write the action for the classical and quantum components of the internal magnetic field $\mathbf{h}^{\text{cl}}(t)$, $\mathbf{h}^{q}(t)$, with $\mathbf{h}^{\text{cl}}(t)$ in the form of Eq. (4.1), as a sum of the radial and tangential terms:

$$S[\boldsymbol{h}] = S_{\parallel}[h_{\parallel}^{\text{cl}}, h_{\parallel}^{q}] + S_{\perp}[\boldsymbol{n}(t), \boldsymbol{h}_{\perp}^{q}]. \tag{4.32}$$

The radial term in the action has the form

$$S_{\parallel}[h_{\parallel}^{\text{cl}}, h_{\parallel}^{q}] = \int dt dt' (D_{\parallel}^{-1})^{\alpha\beta}(t, t') h_{\parallel}^{\alpha}(t) h_{\parallel}^{\beta}(t')$$
$$+ \frac{4}{\delta_{1}} \int dt \boldsymbol{B}(t) \cdot \boldsymbol{n}(t) h_{\parallel}^{\text{cl}}(t), \qquad (4.33)$$

where the inverse of the internal magnetic field propagator is given by

$$(D_{\parallel}^{-1})(t,t') = \frac{4}{E''} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \delta(t-t'), \tag{4.34}$$

$$-\begin{pmatrix} 0 & \Pi_{\parallel}^{R}(t,t') \\ \Pi_{\parallel}^{A}(t,t') & \Pi_{\parallel}^{K}(t,t') \end{pmatrix}$$
(4.35)

and $\Pi_{\parallel}^{\alpha\beta}(t,t')=n_i(t)n_j(t)\Pi_{\parallel,ij}^{\alpha\beta}(t,t')$ [we remind that the calculation of the polarization operator was done in the leading order of the expansion in time derivatives of $\boldsymbol{n}(t)$, i.e., neglecting the difference between $\boldsymbol{n}(t)$ and $\boldsymbol{n}(t')$]. The matrix inverse is both in the 2×2 space of the Keldysh indices α,β =cl,q, and in the space of time arguments t,t'. The latter inversion is conveniently done after the Fourier transform with respect to t-t':

$$D_{\parallel}^{R}(\omega) = D_{\parallel}^{q,\text{cl}}(\omega) = \frac{E''}{4} \frac{-i\omega + 1/\tau}{-i\omega + (\delta_1 + E''/2)/(\tau\delta_1)},$$
(4.36)

and $D_{\parallel}^{A}(\omega) = [D_{\parallel}^{R}(\omega)]^{*}$. The Keldysh component is

$$D_{\parallel}^{K}(\omega) = D_{\parallel}^{q,q}(\omega) = D_{\parallel}^{R}(\omega)\Pi_{\parallel}^{K}(\omega)D_{\parallel}^{A}(\omega), \qquad (4.37)$$

where $n_i n_i \Pi_{\parallel}^K(\omega)$ is given by Eq. (4.24b).

The most interesting for us is the tangential term in the action, which describes the slow dynamics of the direction n(t) of the internal field. This term is given by

$$S_{\perp}[\mathbf{n}(t), \mathbf{h}_{\perp}^{q}] = -\frac{4h_{0}\tau}{\delta_{1}} \int dt \frac{(\mathbf{n} - 2h_{0}\tau\mathbf{n} \times \mathbf{n}) \cdot \mathbf{h}_{\perp}^{q}}{(2h_{0}\tau)^{2} + 1}$$
$$-\frac{4}{\delta_{1}} \int dt [\mathbf{n} \times [\mathbf{n} \times \mathbf{B}]] \cdot \mathbf{h}_{\perp}^{q}$$
$$-\int dt dt' h_{\perp,i}^{q}(t) \Pi_{\perp,ij}^{K}(t,t') h_{\perp,j}^{q}(t'). \tag{4.38}$$

The Fourier transform of the polarization operator $\Pi_{\perp,ij}^K(t,t')$ with respect to t-t' is given by Eq. (4.24).

We have assumed the typical time scale of external field $\boldsymbol{B}(t)$ to be much longer than τ or $1/h_0$. This enabled us to take the polarization operators in the instantaneous approxi-

mation $[\Pi_{ij}^{\alpha\beta}(t,t) \propto \delta(t-t')$, equivalent to the $\omega \to 0$ limit in the denominator of Eq. (4.22)] in the corresponding terms of the action.

V. STOCHASTIC LANDAU-LIFSHITZ-GILBERT EQUATION

A. Langevin equation for the orientation of the internal magnetic field

In this section we consider evolution of the direction vector n(t), described by the tangential part of the action, Eq. (4.38). We neglect fluctuations of the magnitude of the internal magnetic field, h_{\parallel} , and the conditions, under which this can be done, are listed in the next section.

We decouple the quadratic in h_{\perp}^q component of the action in Eq. (4.38) by introducing an auxiliary transverse field $w(t) \perp n(t)$:

$$\exp\left[-i\int dt dt' h_{\perp,i}^{q}(t) \Pi_{\perp,ij}^{K}(t,t') h_{\perp,j}^{q}(t')\right]$$

$$= \int \mathcal{D} \boldsymbol{w}(t) \exp\left[\frac{4i}{\delta_{1}} \int dt \boldsymbol{w}(t) \cdot \boldsymbol{h}_{\perp}^{q}(t)\right]$$

$$\times \exp\left[\frac{4i}{\delta_{1}^{2}} \int dt dt' (\Pi_{\perp}^{K})_{ij}^{-1}(t,t') w_{i}(t) w_{j}(t')\right]. \quad (5.1)$$

After this operation, the tangential part of the action for $\mathbf{n}(t)$, $\mathbf{h}_{\perp}^{q}(t)$ takes the form

$$S'_{\perp}[\mathbf{n}(t), \mathbf{h}_{\perp}^{q}] = -\frac{4}{\delta_{1}} \int dt \mathbf{h}_{\perp}^{q}(t) \left[\frac{h_{0} \boldsymbol{\tau} \dot{\mathbf{n}} - 2(h_{0} \boldsymbol{\tau})^{2} [\mathbf{n} \times \dot{\mathbf{n}}]}{(2h_{0} \boldsymbol{\tau})^{2} + 1} + \dot{\mathbf{n}} \times [\mathbf{n} \times \mathbf{B}] - \mathbf{w} \right]. \tag{5.2}$$

Integration of $e^{iS_{\perp}}$ over $h_{\perp}^{q}(t)$ produces a functional δ function, whose argument determines the equation of motion

$$\frac{h_0 \tau \dot{\boldsymbol{n}} - 2(h_0 \tau)^2 [\boldsymbol{n} \times \dot{\boldsymbol{n}}]}{(2h_0 \tau)^2 + 1} + \boldsymbol{n} \times [\boldsymbol{n} \times \boldsymbol{B}] = \boldsymbol{w}. \tag{5.3}$$

Thus, the field w(t) plays the role of the Gaussian random Langevin force, and the last exponential factor in Eq. (5.1) can be interpreted as its probability distribution. Equivalently, one can specify its correlation function:

$$\langle w_i(t)w_j(t')\rangle = \frac{\delta_1^2}{8}i\Pi_{\perp,ij}^K(t,t'). \tag{5.4}$$

It is convenient to resolve Eq. (5.3) with respect to \vec{n} :

$$\dot{\mathbf{n}} = 2[\mathbf{n} \times (\mathbf{w} + \mathbf{B})] - \frac{1}{h_0 \tau} \{\mathbf{n} \times [\mathbf{n} \times (\mathbf{w} + \mathbf{B})]\}. \quad (5.5)$$

This is nothing but the Landau-Lifshitz-Gilbert equation for the direction n(t) of the total spin (we remind that the total spin and the internal field are proportional to each other; see Eq. (3.14b) and the discussion after it], in the presence of an external magnetic field B(t) and a stochastic field w(t).

As discussed in the end of Sec. IV B, for times $|t-t'| \gg 1/T_{\rm eff}$ the polarization operator $\Pi^K_{\perp,ij}(t,t')$ and thus

the random field correlator $\langle w_i(t)w_j(t')\rangle$, Eq. (5.4), can be approximated by a δ function (Markov approximation):

$$\langle w_i(t)w_j(t')\rangle = \tau \delta_1 T_{\text{eff}} \frac{\delta_{ij} - n_i n_j}{(2h_0 \tau)^2 + 1} \delta(t - t'). \tag{5.6}$$

As seen from Eq. (4.29), the strength of the stochastic term is determined by the effective temperature $T_{\rm eff}$, Eq. (4.30). At low bias, $|V| \ll T$ (almost equilibrium situation) we have $T_{\rm eff} \approx T$, so correlator (5.6) can be obtained from the fluctuation-dissipation theorem, so the magnetization fluctuations can be viewed as the spin Nyquist noise. In the opposite limit of low temperature, $T \ll |V|$, the effective temperature $T_{\rm eff} \approx \Xi |V|/2$, and thus the magnetization fluctuations correspond to spin shot noise. We also note that the fact that Eq. (5.6) represents a limit of Eq. (5.4) with a finite correlation time, automatically prescribes the Stratonovich resolution of the Itô-Stratonovich ambiguity for multiplicative noise (see Ref. 5 for a detailed discussion).

B. Fokker-Planck equation for the orientation probability distribution

Next, we follow the standard procedure of derivation of the Fokker-Planck equation for the distribution $\mathcal{P}(n)$ of the probability for the internal magnetic field to point in the direction n. The probability distribution satisfies the continuity equation:

$$\frac{\partial \mathcal{P}}{\partial t} + \frac{\partial}{\partial \mathbf{n}} \cdot \mathbf{J} = 0, \tag{5.7}$$

where the probability current is defined as

$$\boldsymbol{J} = \left(2\boldsymbol{n} \times \boldsymbol{B} - \frac{1}{h_0 \tau} \boldsymbol{n} \times [\boldsymbol{n} \times \boldsymbol{B}]\right) \mathcal{P} + \frac{1}{2} \left\langle \boldsymbol{\xi} \left(\boldsymbol{\xi} \cdot \frac{\partial \mathcal{P}}{\partial \boldsymbol{n}}\right) \right\rangle$$
(5.8)

and the stochastic velocity ξ is introduced in terms of the field w as

$$\boldsymbol{\xi} = 2[\boldsymbol{n} \times \boldsymbol{w}] - \frac{1}{h_0 \tau} [\boldsymbol{n} \times (\boldsymbol{n} \times \boldsymbol{w})]. \tag{5.9}$$

The derivative $\partial/\partial n$ is understood as the differentiation with respect to local Euclidean coordinates in the tangent space. Performing averaging over fluctuations of w in Eq. (5.8), we obtain

$$\frac{\partial \mathcal{P}}{\partial t} = \frac{\partial}{\partial \mathbf{n}} \cdot \left\{ -2[\mathbf{n} \times \mathbf{B}]\mathcal{P} + \frac{[\mathbf{n} \times (\mathbf{n} \times \mathbf{B})]}{h_0 \tau} \mathcal{P} + \frac{1}{\mathcal{T}_0} \frac{\partial \mathcal{P}}{\partial \mathbf{n}} \right\}.$$
(5.10)

where the time constant \mathcal{T}_0 is the Néel time

$$\mathcal{T}_0 = \frac{2(h_0 \tau)^2}{\tau T_{\text{eff}} \delta_1}.$$
 (5.11)

Below we use the spherical coordinates for the direction of the internal magnetic field, $n = (\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)$, in which the Fokker-Planck equation takes the form (this is precisely the form used in the original work of Brown¹):

$$\frac{\partial \mathcal{P}}{\partial t} = \frac{1}{\sin \theta} \frac{\partial}{\partial \varphi} \left[F_{\varphi} \mathcal{P} + \frac{1}{\mathcal{T}_0} \frac{1}{\sin \theta} \frac{\partial \mathcal{P}}{\partial \varphi} \right] + \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left[\sin \theta F_{\theta} \mathcal{P} + \frac{\sin \theta}{\mathcal{T}_0} \frac{\partial \mathcal{P}}{\partial \theta} \right], \quad (5.12)$$

where

$$F_{\varphi} = \frac{B_x \sin \varphi - B_y \cos \varphi}{h_0 \tau} - 2 \cos \theta (B_x \cos \varphi + B_y \sin \varphi) + 2 \sin \theta B_z, \tag{5.13}$$

$$F_{\theta} = -2(B_x \sin \varphi - B_y \cos \varphi) - \frac{\cos \theta}{h_0 \tau} (B_x \cos \varphi + B_y \sin \varphi) + \frac{\sin \theta}{h_0 \tau} B_z.$$
 (5.14)

It should be supplemented by the normalization condition:

$$\int d^2 \mathbf{n} \mathcal{P}(\mathbf{n}) = \int_0^{2\pi} d\varphi \int_0^{\pi} \sin \theta d\theta \mathcal{P}(\varphi, \theta) = 1, \quad (5.15)$$

which is preserved if the boundary conditions at θ =0, π are imposed:

$$\lim_{\theta \to 0, \pi} \sin \theta \int_{0}^{2\pi} d\varphi \frac{\partial \mathcal{P}}{\partial \theta} = 0.$$
 (5.16)

Below we apply the Fokker-Planck equation to the calculation of the average magnetic moment of an ensemble of particles (in the units of $g\mu_B S_0$):

$$\mathbf{M} = \int d^2 \mathbf{n} \mathbf{n} \mathcal{P}(\mathbf{n}). \tag{5.17}$$

VI. APPLICABILITY OF THE APPROACH

In this section we discuss the conditions of validity of the stochastic LLG equation, Eq. (5.5), for the model of a magnetic particle connected to leads under a finite bias. We briefly listed these conditions in Sec. I. Here we present their more detailed quantitative analysis.

A. Fluctuations of the radial component of the internal magnetic field

We represented the classical component of the internal magnetic field $\boldsymbol{h}^{\text{cl}}$ in terms of a slowly varying direction $\boldsymbol{n}(t)$ and fast oscillations $h_{\parallel}^{\text{cl}}$ of its magnitude around the average value h_0 . Now, we evaluate the amplitude of oscillations of the radial component $h_{\parallel}^{\text{cl}}$ of the field, using the radial term in the action; see Eqs. (4.32) and (4.33).

The typical frequencies for time evolution of small fluctuations of the internal magnetic field in the radial direction are of order of

$$\omega \sim \frac{\delta_1 + E''/2}{\delta_1} \frac{1}{\tau} \tag{6.1}$$

as one can conclude from the explicit form of the propagator $D_{\parallel}^{R}(\omega)$, Eq. (4.36), of these fluctuations. This scale has the

meaning of the inverse RC time in the spin channel. Deep in the ferromagnetic state (i.e., far from the Stoner critical point $E''+2\delta_1=0$) we estimate $\delta_1+E''/2\sim\delta_1$ (which is equivalent to $E''\sim h_0/S_0$), so this spin-channel RC time is of the same order as the escape time τ . This estimate for the frequency range is consistent with the simple picture, which describes the evolution of the internal magnetic field of the grain as a response to a changing value of the total spin of the particle due to random processes of electron exchange between the dot and the leads. The electron exchange happens with the characteristic rate $1/\tau$.

The correlation function $\langle h_{\parallel}^{\rm cl}(t)h_{\parallel}^{\rm cl}(t')\rangle$ can be evaluated by performing the Gaussian integration with the quadratic action in $h_{\parallel}^{\rm cl}$ and h_{\parallel}^{q} . Using Eq. (4.37), we obtain the equal-time correlation function

$$\langle (h_{\parallel}^{\text{cl}})^2 \rangle = \frac{i}{2} \int \frac{d\omega}{2\pi} D_{\parallel}^K(\omega)$$

$$= \frac{(E'')^2}{32\tau\delta_1} \int \frac{d\omega}{2\pi} \frac{2\pi\mathcal{R}(\omega)}{\omega^2 + [1 + E''/(2\delta_1)]^2/\tau^2}. \quad (6.2)$$

This equation gives the value of fluctuations of the radial component of the internal magnetic field of the particle. These fluctuations survive even in the limit T=0 and V=0, when $\mathcal{R}(\omega)=2|\omega|/\pi\tau$. We have the following estimate:

$$\langle (h_{\parallel}^{\text{cl}})^2 \rangle = \frac{(E'')^2}{16\pi\tau\delta_1} \ln \frac{E_T\tau}{1 + E''/(2\delta_1)},$$
 (6.3)

the upper cutoff E_T is the Thouless energy, $E_T = v_F/L$ for a ballistic dot with diameter L and electron Fermi velocity v_F .

The separation of the internal magnetic field into the radial and tangential components is justified, provided that the fluctuations $\sqrt{\langle (h_{\parallel}^{\rm cl})^2 \rangle}$ of the radial component are much smaller than the average value of the field h_0 , i.e., $\langle (h_{\parallel}^{\rm cl})^2 \rangle \ll h_0^2$. Using the estimate Eq. (6.3), we obtain the necessary requirement for the applicability of equations for the slow evolution of the vector of the internal magnetic field of a particle:

$$S_0 \gg \sqrt{\frac{1}{\tau \delta_1} \ln(E_T \tau)},$$
 (6.4)

where S_0 is the spin of a particle in equilibrium and we again used the estimate $E'' \sim h_0/S_0$. Condition of Eq. (6.4) requires that the system is not close to the Stoner instability.

B. Applicability of the Gaussian approximation

Let us discuss the applicability of the Gaussian approximation for the action in h_{\parallel}^{cl} and h^q . The coefficients in front of terms $h^q(t)h_{\parallel}(t_1)\dots h_{\parallel}^{cl}(t_n)$ are obtained by taking the nth variational derivative of $\delta \mathbf{g}^K(t,t)+\delta \mathbf{g}^Z(t,t)$, or, equivalently, by iterating the Usadel equation n times. Since the typical frequencies of h_{\parallel} are $\omega \sim 1/\tau$, the left-hand side of the equation is $\sim \delta^{(n+1)} \mathbf{g}^K/\tau$, while the right-hand side is $h_{\parallel}^{cl} \delta^{(n)} \mathbf{g}^K$. Since the only time scale here is τ , all the coefficients of the expansion of the action in $h_{\parallel}^{cl}(\omega)$ at $\omega \sim 1/\tau$ are of the same order:

$$S_{n+1} \sim \frac{\tau^{n-1}}{E''} \int \frac{d\omega_1 \dots d\omega_n}{(2\pi)^n} \times h_{\parallel}^{\text{cl}}(\omega_1) \dots h_{\parallel}^{\text{cl}}(\omega_n) h^q(-\omega_1 - \dots - \omega_n). \quad (6.5)$$

At the same time, the typical value of $h_{\parallel}^{\rm cl}(\omega \sim 1/\tau)$, as determined by the Gaussian part of the action, was estimated in the previous subsection to be of the order of $\sqrt{\tau D_{\parallel}^{K}(\omega \sim \tau)} \sim \sqrt{\tau \delta_{1}} \ll 1$, so the higher-order terms are indeed not important.

For the quantum component of the field the quadratic and quartic terms in the action are estimated as

$$S_{n+1} \sim \frac{T_{\text{eff}} \tau^n}{\delta_1} \int \frac{d\omega_1 \dots d\omega_n}{(2\pi)^n} \times h^q(\omega_1) \dots h^q(\omega_n) h^q(-\omega_1 - \dots - \omega_n). \quad (6.6)$$

If $T_{\rm eff} \!\!>\! 1/\tau$, then the typical frequency scale is $\omega \sim 1/\tau$, so the quadratic term gives $h^q(\omega \sim 1/\tau) \sim \sqrt{\delta_1/T_{\rm eff}}$, and $S_n \sim (\delta_1/T_{\rm eff})^{n/2-1} \sim [\tau \delta_1/(\tau T_{\rm eff})]^{n/2-1}$. If $T_{\rm eff} \!\!<\! 1/\tau$, at the typical scale $\omega \sim T_{\rm eff}$ we obtain $h^q(\omega \sim T_{\rm eff}) \sim \sqrt{\delta_1/(T_{\rm eff}^2\tau)}$, so again $S_n \sim (\tau \delta_1)^{n/2-1} \ll 1$ for n > 2.

Physically, the parameter $1/(\tau\delta_1)=N_{\rm ch}$ (or $T_{\rm eff}/\delta_1$, if it is larger) can be identified with the number of the independent sources of the noise acting on the magnetization field. Thus, the smallness of the non-Gaussian part of the action is nothing but the manifestation of the central limit theorem.

C. Applicability of the Fokker-Planck equation

According to the Fokker-Planck equation, the typical scale of the time evolution of the direction n(t) is T_0 , defined in Eq. (5.11). According to Eq. (6.1), the time scale of the fluctuations of the magnitude of the internal magnetic field is of the order of τ . Thus, the separation into slow and fast variables is possible when $T_0 \gg \tau$. This is also the condition of validity of the gradient expansion of Sec. IV, $|\dot{n}|\tau \ll 1$. It can be equivalently presented as

$$\frac{T_{\text{eff}}}{\delta_1} \ll \left(\frac{h_0}{\delta_1}\right)^2 = S_0^2. \tag{6.7}$$

Note, however, that in order to have any magnetic moment at all, we need the effective temperature of the system to be lower than the Curie temperature, i.e.,

$$T_{\text{eff}} \leqslant h_0 = S_0 \delta_1. \tag{6.8}$$

This, in fact, represents a stronger condition than Eq. (6.7).

The Markovian approximation for the stochastic field w(t) is valid when its correlation time, $1/T_{\rm eff}$, is shorter than the typical scale of evolution: $1/T_{\rm eff} \ll T_0$. This can be rewritten as

$$S_0^2 \geqslant \frac{1}{\delta_1 \tau},\tag{6.9}$$

which is weaker than the condition of Eq. (6.4), and thus is redundant.

The regular precession in the magnetic field with frequency $g\mu_B B$ does not restrict the applicability of the Markovian approximation.

VII. MAGNETIC SUSCEPTIBILITY OF METALLIC PARTICLES OUT OF EQUILIBRIUM

The LLG equation derived in this paper for a ferromagnetic particle with finite bias between the leads can be applied to a number of experimental setups. In this paper we apply the stochastic equation for spin distribution function to the analysis of the magnetic susceptibility at finite frequency. The susceptibility is the basic characteristic of magnetic systems; it can often be measured directly and determines other measurable quantities.

Below, we calculate the susceptibility of an ensemble of particles placed in constant magnetic field of an arbitrary strength and oscillating weak magnetic field; see Fig. 1. We consider the oscillating magnetic field with its components in directions parallel and perpendicular to the constant magnetic field.

A. Solution at zero noise power

When w(t)=0, and at fixed direction of the field, $B(t)=e_zB(t)$ (thus corresponding to $\theta=0$), Eq. (5.5) is easily integrated for an arbitrary time dependence B(t):

$$\varphi = \varphi_0 - \int_0^t 2B(t')dt', \qquad (7.1)$$

$$\tan\frac{\theta}{2} = \tan\frac{\theta_0}{2} \exp\left[-\int_0^t \frac{B(t')}{h_0 \tau} dt'\right]. \tag{7.2}$$

Notice that the noise is absent for T=V=0 and at $\omega=0$, which requires a constant magnetic field in the above equations.

B. Constant magnetic field

At finite $T_{\rm eff}$ and constant magnetic field B_0 the Fokker-Planck equation has a simple solution

$$\mathcal{P}_0(\theta) = \frac{b}{\sinh b} \frac{e^{b\cos \theta}}{4\pi},\tag{7.3}$$

where the strength of constant magnetic field is written in terms of the dimensionless parameter

$$b = \frac{(2h_0\tau)B_0}{\tau\delta_1 T_{\text{eff}}} = \frac{B_0 T_0}{h_0 \tau}.$$
 (7.4)

Substituting this probability function into Eq. (5.17), we obtain the classical Langevin expression for the magnetic moment of a particle in a magnetic field

$$M_z(b) = \coth b - \frac{1}{b}, \quad M_x = M_y = 0.$$
 (7.5)

This expression for the magnetic moment coincides with that in thermal equilibrium, provided that the temperature is replaced by the effective temperature $T_{\rm eff}$ defined by Eq. (4.30).

The differential dc susceptibility along the field is equal to

$$\chi_{\parallel}^{\text{dc}}(b) = \frac{dM_z(b)}{db} = \frac{1}{b^2} - \frac{1}{\sinh^2 b}.$$
(7.6)

The dc susceptibility in the direction perpendicular to the field corresponds to a simple tilt of the magnetic field, producing a proportional tilt of the magnetic moment:

$$\chi_{\perp}^{\text{dc}}(b) = \frac{M_z(b)}{h} = \frac{b \coth b - 1}{h^2}.$$
 (7.7)

C. Longitudinal susceptibility

We now consider the response of the magnetization to weak oscillations $\tilde{B}_z(t)$ of the external magnetic field with frequency ω in direction parallel to the fixed magnetic field B_0 . We write the oscillatory component of the field in terms of the dimensionless field strength b_{\parallel} :

$$\widetilde{B}_{z}(t) = (b_{\parallel}e^{-i\omega t} + b_{\parallel}^{*}e^{i\omega t})\frac{B_{0}}{b}.$$
 (7.8)

The linear correction to the probability distribution can be cast in the form

$$\mathcal{P}(\theta,t) = \left[1 + b_{\parallel}u_{\parallel}(\theta)e^{-i\omega t} + b_{\parallel}^*u_{\parallel}^*(\theta)e^{i\omega t}\right]\mathcal{P}_0(\theta), \quad (7.9)$$

with $\mathcal{P}_0(\theta)$ defined by Eq. (7.3). The magnetic ac susceptibility can be evaluated from Eq. (7.9) using Eq. (5.17) as

$$\chi_{\parallel}(\omega,b) = 2\pi \int_0^{\pi} u_{\parallel}(\theta) \mathcal{P}_0(\theta) \cos \theta \sin \theta d\theta. \quad (7.10)$$

The equation for $u_{\parallel}(\theta)$ is obtained from Eq. (5.12) with $B_z = B_0 + \widetilde{B}_z(t)$:

$$\frac{\partial^2 u_{\parallel}}{\partial \theta^2} + \frac{\cos \theta - b \sin^2 \theta}{\sin \theta} \frac{\partial u_{\parallel}}{\partial \theta} + i\Omega u_{\parallel} = b \sin^2 \theta - 2 \cos \theta,$$
(7.11)

where we introduced the dimensionless frequency

$$\Omega = \omega \mathcal{T}_0, \tag{7.12}$$

and the time constant \mathcal{T}_0 is defined in Eq. (5.11).

Note the symmetry of Eq. (7.11) with respect to the simultaneous change $b \rightarrow -b$ and $\theta \rightarrow \pi - \theta$: the differential operator on the left-hand side is even, while the right-hand side is odd. This translates into the property $\chi_{\parallel}(\omega,b) = \chi_{\parallel}(\omega,-b)$. The normalization condition for $\mathcal{P}(\theta)$, Eq. (5.15), requires that

$$\int_0^{\pi} u_{\parallel}(\theta) \mathcal{P}_0(\theta) \sin \theta d\theta = 0. \tag{7.13}$$

The latter holds if the boundary conditions Eq. (5.16) are satisfied, which in the case of axial symmetry can be written as

$$\lim_{\theta \to 0, \pi} \left\{ \sin \theta \frac{\partial u_{\parallel}(\theta)}{\partial \theta} \right\} = 0. \tag{7.14}$$

The differential equation, Eq. (7.11), with the boundary condition Eq. (7.14) can be solved numerically and then the

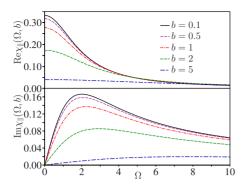


FIG. 2. (Color online) Plot of the real and imaginary parts of the susceptibility $\chi_{\parallel}(\Omega,b)$ as a function of the dimensionless frequency $\Omega = \omega T_0$. The oscillatory field at frequency ω is parallel to the constant magnetic field with strength b. The real part of the susceptibility decreases monotonically from its dc value, Eq. (7.6), as frequency increases, while the imaginary part increases linearly at small $\Omega \ll 1$, see Eq. (7.21), and decreases at higher frequencies.

susceptibility is evaluated according to Eq. (7.10). The result is shown in Figs. 2 and 3, where the susceptibility is shown as a function of frequency ω or magnetic field b, respectively. We also consider various asymptotes for the ac susceptibility, obtained from the solution of Eq. (7.11).

At zero constant magnetic field, b=0, we find the exact solution of Eq. (7.11) explicitly:

$$u_{\parallel}(\theta) = \frac{\cos \theta}{1 - i\Omega/2}.\tag{7.15}$$

This solution gives the ac susceptibility of a Debye form:

$$\chi_{\parallel}(\Omega, b=0) = \frac{1}{3} \frac{1}{1 - i\Omega/2}.$$
 (7.16)

For $b \ge 1$ only $\cos \theta \sim 1/b$ matter, and we can find a specific solution of the inhomogeneous equation:

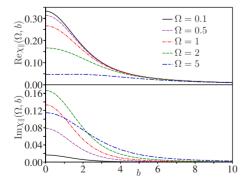


FIG. 3. (Color online) Plot of the real and imaginary parts of the ac susceptibility $\chi_{\parallel}(\Omega,b)$ at several values of the dimensionless frequency Ω of the oscillating magnetic field along the constant magnetic field with strength b. In general, magnetic field suppresses both real and imaginary parts of the susceptibility.

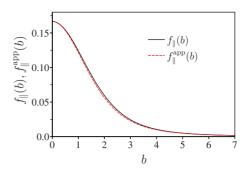


FIG. 4. (Color online) Dependence on magnetic field b of the slope of the imaginary part of the linear in frequency susceptibility $\chi_{\parallel}(\Omega,b)$ at low frequencies $\Omega \ll 1$, calculated according to Eq. (7.21). For comparison, we also plot function $f_{\parallel}^{app}(b)$; see Eq. (7.25)

$$u_{\parallel}(\theta) = \frac{1 - b(1 - \cos \theta)}{b - i\Omega/2}, \quad b \ge 1.$$
 (7.17)

The requirement of regularity at the opposite end can be replaced by the probability normalization condition, Eq. (7.13), which is satisfied by this solution. Substituting this solution to Eq. (7.10), we obtain the strong field asymptote for the ac susceptibility

$$\chi_{\parallel}(\Omega, b \gg 1) = \frac{1}{b(b - i\Omega/2)}.$$
(7.18)

For $\Omega \gg 1$ and $\Omega \gg b$, we can neglect the derivatives in Eq. (7.11) and find the solution in the form

$$u_{\parallel}(\theta) \approx \frac{b \sin^2 \theta - 2 \cos \theta}{i\Omega},$$
 (7.19)

This solution $u_{\parallel}(\theta)$ also satisfies Eq. (7.13). For the susceptibility, Eq. (7.10), we obtain

$$\chi_{\parallel}(\Omega \to \infty, b) = \frac{2i}{\Omega} \left(\frac{\coth b}{b} - \frac{1}{b^2} \right). \tag{7.20}$$

Finally, the low-frequency limit can be also analyzed analytically. The real part of the susceptibility coincides with the differential susceptibility in dc magnetic field, Eq. (7.6), for the imaginary part to the first order in the frequency we obtain, see Appendix B,

$$\operatorname{Im} \chi_{\parallel}(\Omega, b) = \Omega f_{\parallel}(b). \tag{7.21}$$

The function $f_{\parallel}(b)$ has a complicated analytical form and is not presented here, but its plot is shown in Fig. 4.

In all four limiting cases considered above, the asymptotic approximations hold regardless the order in which the limits are taken. Indeed, the asymptote of the expression for the susceptibility in the zero field, Eq. (7.16), has the asymptote at $\Omega \to \infty$ consistent with Eq. (7.20) at b=0. Similarly, the high-frequency limit of Eq. (7.18) coincides with the limit $b\to \infty$ of Eq. (7.20). Both limits of weak and strong magnetic field of the imaginary part of the susceptibility at low frequencies, Eq. (7.21), coincide with the imaginary part of $\chi_{\parallel}(\Omega,b)$, calculated from Eq. (7.16) and Eq. (7.20), respectively.

In general, the ac susceptibility is given by the following expression:

$$\chi_{\parallel}(\Omega, b) = \sum_{n} \frac{\chi_{n}^{\parallel}(b)}{1 - i\omega \mathcal{T}_{0}/\Gamma_{n}^{\parallel}(b)}, \qquad (7.22)$$

where functions $\chi_n^{\parallel}(b)$ and $\Gamma_n^{\parallel}(b)$ are real. This expression can be obtained by expanding the right-hand side of Eq. (7.11) in terms of the eigenfunctions of the linear differential operator on the left-hand side, analytically continued to real $i\Omega$. For the weak field, $b \le 1$, this expansion can be obtained by expanding $u_{\parallel}(\theta)$ in Legendre polynomials, which gives the poles $\Gamma_n^{\parallel}(b \to 0) = n(n+1) + O(b)$, and the residues $\chi_n^{\parallel}(b \to 0) = O(b^{(n-1)/2})$ for n odd and $\chi_n^{\parallel}(b \to 0) = O(b^{n/2+1})$ for n even.

For practical purposes, we found from a numerical analysis that even the simple Debye approximation,

$$\chi_{\parallel}^{\text{app}}(\omega, b) = \frac{\chi_{\parallel}^{\text{dc}}(b)}{1 - i\omega \mathcal{T}_0/\Gamma_{\text{app}}^{\parallel}(b)},$$
 (7.23)

with $\chi_{\parallel}^{\rm dc}(b)$ given by Eq. (7.6), and $\Gamma_{\rm app}^{\parallel}(b)$ determined from the high-frequency asymptotics, Eq. (7.20),

$$\Gamma_{\text{app}}^{\parallel}(b) = \frac{-i\mathcal{T}_0}{\chi_{\parallel}^{\text{dc}}(b)} \lim_{\omega \to \infty} \omega \chi_{\parallel}(\omega, b) = \frac{2}{b} \frac{\coth b - 1/b}{1/b^2 - 1/\sinh^2 b},$$
(7.24)

gives a very good estimate of the susceptibility for all values of ω (taken on the real axis) and b. The analysis shows that the susceptibility, Eq. (7.10), obtained from a numerical solution of Eq. (7.11), is within a few percent of the estimate given by Eq. (7.23). To illustrate the accuracy of the high-frequency approximation for $\Gamma^{\parallel}_{app}(b)$, Eq. (7.24), we apply it in the opposite limit of low frequencies, $\Omega \ll 1$, and compare the exact function $f_{\parallel}(b)$ appearing in Eq. (7.21), with

$$f_{\parallel}^{\text{app}}(b) = \frac{1}{i\mathcal{T}_0} \left. \frac{\partial \chi_{\parallel}^{\text{app}}(\omega, b)}{\partial \omega} \right|_{\omega=0} = \frac{\chi_{\parallel}^{\text{dc}(b)}}{\Gamma_{\text{app}}^{\parallel}(b)}. \tag{7.25}$$

For visual comparison of functions $f_{\parallel}(b)$ and $f_{\parallel}^{app}(b)$, we plot both functions in Fig. 4, where these curves are nearly indistinguishable. The difference between these two curves vanishes at $b \rightarrow 0$ and $b \rightarrow \infty$, and has a maximal difference at $b \approx 2$, which constitutes only a tiny fraction of $f_{\parallel}(b)$.

D. Transverse susceptibility

Next, we consider the response of the magnetization to weak oscillations $\tilde{B}_{\perp}(t)$ of the external magnetic field with frequency ω in the direction perpendicular to that of the fixed magnetic field B_0 . We write the oscillatory component of the field in the form

$$\widetilde{B}_{\perp}(t) = \left[b_{\perp}(e_x + ie_y)e^{-i\omega t} + b_{\perp}^*(e_x - ie_y)e^{i\omega t}\right] \frac{B_0}{b}.$$
(7.26)

This field represents a circular polarization of an ac magnetic field in the (x,y) plane, perpendicular to the fixed magnetic

field in the z direction: $\mathbf{B} = \{B_{\perp} \cos \omega t; B_{\perp} \sin \omega t; B_0\}$. We look for the linear correction to the probability distribution in the form

$$\mathcal{P}(\varphi,\theta,t) = \mathcal{P}_0(\theta) \big[1 + b_\perp u_\perp(\theta) e^{i\varphi - i\omega t} + b_\perp^* u_\perp^*(\theta) e^{-i\varphi + i\omega t} \big]. \tag{7.27}$$

We define the transverse susceptibility in response to the ac magnetic field, Eq. (7.26), as

$$\chi_{\perp}(\Omega, b) = 2\pi \frac{1}{2} \int_0^{\pi} u_{\perp}(\theta) \mathcal{P}_0(\theta) \sin^2 \theta d\theta.$$
 (7.28)

According to Eq. (5.17), such a definition translates in the following expression for the Cartesian components of the oscillating magnetic moment:

$$M_x(t) = 2 \text{ Re}(\chi_{\perp} b_{\perp} e^{-i\omega t}), \quad M_y(t) = 2 \text{ Im}(\chi_{\perp} b_{\perp} e^{-i\omega t}).$$
 (7.29)

The equation for $u_{\perp}(\theta)$ is obtained from the Fokker-Planck equation, Eq. (5.12), linearized in the parameter b_{\perp} :

$$\frac{\partial^{2} u_{\perp}}{\partial \theta^{2}} + \frac{\cos \theta - b \sin^{2} \theta}{\sin \theta} \frac{\partial u_{\perp}}{\partial \theta} + \left(i\Omega_{\perp} - \frac{1}{\sin^{2} \theta} \right) u_{\perp}$$

$$= -\sin \theta (2 - 2ih_{0}\tau b + b \cos \theta). \tag{7.30}$$

Here the dimensionless frequency Ω_{\perp} corresponds to the difference of the driving frequency ω and the precession frequency $-2B_0$ in the constant external field B_0 :

$$\Omega_{\perp} = (\omega + 2B_0)T_0 = \Omega + 2(h_0\tau)b,$$
 (7.31)

where \mathcal{T}_0 is defined in Eq. (5.11) and the second equality is written in terms of dimensionless variables Ω , Eq. (7.12), and b, Eq. (7.4).

Equation (7.30) is symmetric with respect to the simultaneous change $\theta \rightarrow \pi - \theta$, $b \rightarrow -b$, $i \rightarrow -i$, $\Omega_{\perp} \rightarrow -\Omega_{\perp}$; both sides are even. This translates into the property $\chi_{\perp}(\omega,b) = \chi_{\perp}^*(-\omega,-b)$ for real ω . The function $\mathcal{P}(\varphi,\theta,t)$ is single-valued at the poles $\theta = 0$ and $\theta = \pi$, only if

$$u_{\perp}(\theta=0) = 0, \quad u_{\perp}(\theta=\pi) = 0.$$
 (7.32)

The latter equations establish the boundary conditions for the differential equation, Eq. (7.30). We also note that the normalization condition (5.15) is satisfied for any function $u_{\perp}(\theta)$.

Solving numerically the differential equation, Eq. (7.30), with the corresponding boundary conditions, Eq. (7.32), we obtain the transverse susceptibility, Eq. (7.28), shown in Figs. 5 and 6. Below we analyze several limiting cases.

In zero fixed magnetic field, b=0, we have the exact solution of Eq. (7.30):

$$u_{\perp}(\theta) = \frac{\sin \theta}{1 - i\Omega_{\perp}/2}.$$
 (7.33)

This solution corresponds to the solution in the longitudinal case, rotated by 90°, cf. Eq. (7.15).

In strong fixed magnetic field, $b \ge 1$, we need to consider small angles $\theta \sim 1/\sqrt{b}$, therefore, we can approximate $\cos \theta \approx 1$ in Eq. (7.30) and obtain

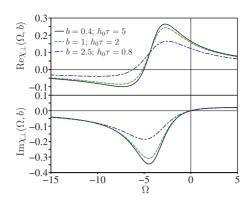


FIG. 5. (Color online) Plot of the real and imaginary parts of the transverse susceptibility $\chi_{\perp}(\Omega,b)$ as a function of the dimensionless frequency Ω . Negative frequency corresponds to the opposite sense of the circular polarization of the ac magnetic field in a plane, perpendicular to the constant magnetic field with strength b. The parameters of the three shown curves are chosen so that $h_0\tau b = 2$.

$$u_{\perp}(\theta) = \frac{b + 2 - 2ih_0\tau b}{b + 2 - i\Omega_{\perp}}\sin\theta.$$
 (7.34)

The susceptibility in the limit $b \ge 1$ is given by

$$\chi_{\perp}(\Omega, b \gg 1) = \frac{1 - 2ih_0\tau}{2b(b(1 - 2ih_0\tau) - i\Omega)}.$$
 (7.35)

At $\Omega_{\perp} \gg 1$, b we can disregard the terms in Eq. (7.30) with derivatives. Moreover, the contribution to the susceptibility, Eq. (7.28), from the vicinity of θ =0 and θ = π is suppressed as $\sin^2\theta$. This observation allows us to write the solution in the form

$$u_{\perp}(\theta) = \sin \theta \frac{2 + 2ih_0\tau b + b\cos \theta}{-i\Omega_{\perp}},$$
 (7.36)

Consequently, we obtain the following high frequency, $\Omega_{\perp} \gg 1$, asymptote for the susceptibility:

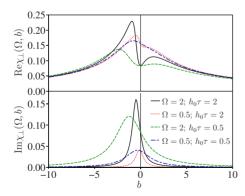


FIG. 6. (Color online) Plot of the real and imaginary parts of the transverse susceptibility $\chi_{\perp}(\Omega,b)$ as a function of the strength b of a constant magnetic field, shown for two values of frequency Ω and two values of the "damping factor" $h_0\tau$. Negative values of b corresponds to the opposite sense of the circular polarization of the ac magnetic field in a plane, perpendicular to the constant magnetic field. The real part of the susceptibility exhibits a strong nonmonotonic behavior at weak magnetic fields.

$$\chi_{\perp}(\Omega, b) = \frac{i}{2(\Omega + 2h_0\tau b)} \left[1 - \frac{b \coth b - 1}{b^2} (2ih_0\tau b + 1) \right].$$
(7.37)

Analogously to the previous subsection, we propose an approximate expression for the susceptibility in response to the transverse oscillating magnetic field:

$$\chi_{\perp}^{\text{app}}(\omega) = \chi_{\perp}^{\text{dc}}(b) \frac{1 - 2iB_0 \mathcal{T}_0 / \Gamma_{\text{app}}^{\perp}(b)}{1 - i(2B_0 + \omega)\mathcal{T}_0 / \Gamma_{\text{app}}^{\perp}(b)}, \quad (7.38)$$

with $\chi_{\perp}^{\text{dc}}(\omega)$ given by Eq. (7.7), and $\Gamma_{\text{app}}^{\perp}(b)$ found for any b from the asymptotic behavior of $\chi_{\perp}(\omega,b)$ at $\Omega_{\perp} \gg 1,b$:

$$\Gamma_{\text{app}}^{\perp}(b) = \frac{b^2}{b \coth b - 1} - 1.$$
(7.39)

Analogously to the case of the longitudinal susceptibility, Eq. (7.38) approximates the numerical solution within a few percent.

VIII. CONCLUSIONS

We have studied the slow dynamics of magnetization in a small metallic particle (quantum dot), where the ferromagnetism has arisen as a consequence of Stoner instability. The particle is connected to nonmagnetic electron reservoirs. A finite bias is applied between the reservoirs, thus bringing the whole electron system away from equilibrium. The exchange of electrons between the reservoirs and the particle results in the Gilbert damping³ of the magnetization dynamics and in a temperature- and bias-driven Brownian motion of the direction of the particle magnetic moment. Analysis of magnetization dynamics and transport properties of ferromagnetic nanoparticles is commonly performed^{5,8–10,17} within the stochastic Landau-Lifshitz-Gilbert (LLG) equation, ^{2,3} which is an analog of the Langevin equation written for a unit three-dimensional vector.

We derived the stochastic LLG equation from a microscopic starting point and established conditions under which the description of the magnetization of a ferromagnetic metallic particle by this equation is applicable. We concluded that the applicability of the LLG equation for a ferromagnetic particle is set by three independent criteria. (1) The contact resistance should be low compared to the resistance quantum, which is equivalent to $N_{\rm ch} \gg 1$. Otherwise the noise cannot be considered Gaussian. Each channel can be viewed as an independent source of noise and only the contribution of many channels results in the Gaussian noise by virtue of the central limit theorem for $N_{\rm ch} \gg 1$. (2) The system should not be too close to the Stoner instability: the mean-field value of the total spin $S_0^2 \gg N_{\rm ch}$. Otherwise, the fluctuations of the absolute value of the magnetic moment become of the order of its average. (3) $S_0^2 \gg T_{\text{eff}} / \delta_1$, where T_{eff} $\simeq \max\{T, |eV|\}$ is the effective temperature of the system, which is the energy scale of the electronic distribution function. Otherwise, the separation into slow (the direction of the magnetization) and fast (the electron dynamics and the magnitude of the magnetization) degrees of freedom is not possible. However, this condition is redundant, since at $T_{\rm eff} \gtrsim S_0 \delta_1$ the spin vanishes.

Under the above conditions, the dynamics of the magnetization is described in terms of the stochastic LLG equation with the power of Langevin forces determined by the effective temperature of the system. The effective temperature is the characteristic energy scale of the electronic distribution function in the particle determined by a combination of the temperature and the bias voltage. In fact, for a considered here system with nonmagnetic contacts between nonmagnetic reservoirs and a ferromagnetic particle the power of the Langevin forces is proportional to the low-frequency noise of total charge current through the particle. We further reduced the stochastic LLG equation to the Fokker-Planck equation for a unit vector, corresponding to the direction of the magnetization of the particle. The Fokker-Planck equation can be used to describe time evolution of the distribution of the direction of magnetization in the presence of timedependent magnetic fields and voltage bias.

As an example of application of the Fokker-Planck equation for the magnetization, we have calculated the frequency-dependent magnetic susceptibility of the particle in a constant external magnetic field (i.e., linear response of the magnetic moment to a small periodic modulation of the field, relevant for ferromagnetic resonance measurements). We have not been able to obtain an explicit analytical expression for the susceptibility at arbitrary value of the applied external field and frequency; however, analysis of different limiting cases has led us to a simple analytical expression which gives a good agreement with the numerical solution of the Fokker-Planck equation.

ACKNOWLEDGMENTS

We acknowledge discussions with I. L. Aleiner, G. Catelani, A. Kamenev, and E. Tosatti. M.G.V. is grateful to the International Centre for Theoretical Physics (Trieste, Italy) for hospitality.

APPENDIX A: FROM SPIN DENSITY MATRIX TO FOKKER-PLANCK EQUATION

Here we show how the Fokker-Planck equation can be simply derived in the model of a localized large spin $S \gg 1$, coupled to a fermionic bath. The Hamiltonian of the spin reads as

$$\hat{H}_{S} = -2\boldsymbol{B}(t) \cdot \hat{\boldsymbol{S}} - \hat{\boldsymbol{h}} \cdot \hat{\boldsymbol{S}}, \tag{A1}$$

where \hat{S} is the operator of the localized spin, 2B(t) is the external classical magnetic field (the factor of 2 is introduced to be consistent with the rest of the paper), and \hat{h} is the fluctuating magnetic field produced by the fermionic bath:

$$\hat{H}_{b} = \sum_{k\sigma} \epsilon_{k} \hat{c}_{k\sigma}^{\dagger} \hat{c}_{k\sigma}, \quad \hat{\boldsymbol{h}} = \sum_{kk'} J_{kk'} \sum_{\sigma\sigma'} \hat{c}_{k\sigma}^{\dagger} \frac{\boldsymbol{\sigma}_{\sigma\sigma'}}{2} \hat{c}_{k'\sigma'}. \quad (A2)$$

Here the subscripts k and σ at the fermionic creation and annihilation operators $\hat{c}_{k\sigma}^{\dagger}, \hat{c}_{k\sigma}$ label the orbital states with

energies ϵ_k and the spin projections $\sigma = \uparrow, \downarrow$, respectively. $J_{kk'} = J_{k'k}^*$ are the matrix elements of the exchange coupling, and $\sigma = (\sigma^x, \sigma^y, \sigma^z)$ is the vector of the 2×2 Pauli matrices.

The bath field correlator is given by

$$\langle \hat{h}_i(t)\hat{h}_j(t')\rangle_b = \delta_{ij} \sum_{kk'} \frac{|J_{kk'}|^2}{2} \frac{(1 - f_k)(1 + f_{k'})}{4} e^{i(\epsilon_k - \epsilon_{k'})(t - t')},$$
(A3)

where the time dependence is determined by the Hamiltonian \hat{H}_b , and the average is taken over the bath density matrix, assumed to be stationary and nonmagnetic, so that

$$\langle \hat{c}_{k\sigma}^{\dagger} \hat{c}_{k'\sigma'} \rangle_b = \delta_{kk'} \delta_{\sigma\sigma'} \frac{1 - f_k}{2}. \tag{A4}$$

If the fermions are in equilibrium at temperature T and chemical potential μ , we have $f_k = \tanh[(\epsilon_k - \mu)/(2T)]$. If $f_k = f(\epsilon_k)$, the bath field correlator, Eq. (A3), can be rewritten in the frequency representation as

$$\langle \hat{h}_i(t)\hat{h}_j(0)\rangle_b = \delta_{ij} \int \frac{d\omega}{2\pi} C(\omega) e^{-i\omega t},$$
 (A5a)

$$C(\omega) = \int d\epsilon \mathcal{J}(\epsilon, \epsilon + \omega) [1 - f(\epsilon)] [1 + f(\epsilon + \omega)], \tag{A5b}$$

$$\mathcal{J}(\epsilon, \epsilon') = \frac{\pi}{4} \sum_{kk'} |J_{kk'}|^2 \delta(\epsilon_k - \epsilon) \delta(\epsilon_{k'} - \epsilon'). \tag{A5c}$$

Note that in equilibrium

$$C(-\omega) = e^{-\omega/T}C(\omega). \tag{A6}$$

Generally, we can expand $C(\omega)$ at low frequencies,

$$C(\omega) = 4\mathcal{J}_{\text{eff}}T_{\text{eff}} + 2\mathcal{J}_{\text{eff}}\omega + O(\omega^2). \tag{A7}$$

In equilibrium the effective temperature defined in this way coincides with the real temperature due to property (A6).

Let us assume the distribution function $f(\epsilon)$ to be a superposition of two steps with two different chemical potentials, corresponding to the bias V:

$$f(\epsilon) = \frac{1 - \sqrt{1 - 2\Xi}}{2} \tanh \frac{\epsilon}{2T} + \frac{1 + \sqrt{1 - 2\Xi}}{2} \tanh \frac{\epsilon + V}{2T},$$
(A8)

where the parameter $\Xi < 1/2$ simply parametrizes the relative magnitude of the two steps. If we neglect the energy dependence of $\mathcal{J}(\epsilon,\epsilon')$ on the energy scale of $\max\{V,T\}$, the energy integration gives

$$C(\omega) = \mathcal{J}\left[2\omega + 2\omega \coth\frac{\omega}{2T} + \frac{\Xi}{2}Y_T(V, \omega)\right], \quad (A9)$$

where $\Upsilon_T(V,\omega)$ is the function defined in Eq. (4.27).⁴⁰ Comparison to the low-frequency expansion, Eq. (A7), gives $\mathcal{J}_{\text{eff}} = \mathcal{J}$ and T_{eff} given by Eq. (4.30).

With the bath field correlator in our hands, we can follow the standard route to the master equation for the density matrix $\hat{\rho}(t)$ of the localized spin, where the bath degrees of freedom have been traced out.¹⁹ Neglecting the terms $O(\omega^2)$ and higher in expansion (A7), we arrive at the master equation in the quantum Brownian motion limit:

$$\frac{\partial \hat{\rho}}{\partial t} = i[2B_i \hat{S}_i, \hat{\rho}] - ie_{ijk} 2\mathcal{J}_{\text{eff}} B_i [\hat{S}_j, \{\hat{S}_k, \hat{\rho}\}] - 2\mathcal{J}_{\text{eff}} T_{\text{eff}} [\hat{S}_i, [\hat{S}_i, \hat{\rho}]]. \tag{A10}$$

Here [...] stands for the commutator of two operators, and $\{...\}$ for the anticommutator. This master equation is not of the Lindblad form and potentially may produce unphysical results (see Ref. 38 for a discussion). However, we will show below that in the limit $S \gg 1$ it reduces to the Fokker-Planck equation, Eq. (5.10).

To describe the quasiclassical dynamics of the spin, we employ the spin coherent states.³⁹ For an arbitrary unit vector \mathbf{n} one defines the coherent state $|\mathbf{n}\rangle$ as the state with the largest value of the projection of the spin on the direction \mathbf{n} , i.e., $(\mathbf{n} \cdot \hat{\mathbf{S}})|\mathbf{n}\rangle = S|\mathbf{n}\rangle$. This definition does not fix the phase, which can be arbitrary and \mathbf{n} dependent, so that it can be viewed as a gauge degree of freedom. We use the following explicit definition in the standard basis $|S_z\rangle$ of the eigenstates of \hat{S}_z :

$$|\mathbf{n}\rangle = \sum_{S_z = -S}^{S} \frac{\sqrt{(2S)!} e^{-iS_z \varphi}}{\sqrt{(S - S_z)! (S + S_z)!}} \left(\cos \frac{\theta}{2}\right)^{S + S_z} \left(\sin \frac{\theta}{2}\right)^{S - S_z} |S_z\rangle,\tag{A11}$$

where φ , θ are the spherical angles of the unit vector \mathbf{n} . The scalar product of coherent states,

$$\langle \boldsymbol{n}_1 | \boldsymbol{n}_2 \rangle = \left(\frac{1 + \boldsymbol{n}_1 \cdot \boldsymbol{n}_2}{2}\right)^S e^{2iS\phi} \approx e^{-S|\boldsymbol{n}_1 - \boldsymbol{n}_2|^2/4 - iSn_z(\varphi_1 - \varphi_2)},$$
(A12a)

$$\phi = \arctan\left[\frac{\cos(\theta_1 + \theta_2)/2}{\cos(\theta_1 - \theta_2)/2} \tan\frac{\varphi_1 - \varphi_2}{2}\right], \quad (A12b)$$

tends to the angular δ function as $S \rightarrow \infty$. The unit operator and the spin operator are represented as

$$(2S+1)\int \frac{d^2\mathbf{n}}{4\pi}|\mathbf{n}\rangle\langle\mathbf{n}| = \hat{1}, \qquad (A13a)$$

$$(2S+1)\int \frac{d^2\mathbf{n}}{4\pi}\mathbf{n}|\mathbf{n}\rangle\langle\mathbf{n}| = \frac{\hat{\mathbf{S}}}{S+1}.$$
 (A13b)

Also, it can be directly shown that

$$\hat{S}|\mathbf{n}\rangle = \frac{\mathbf{n} - n_z \mathbf{e}_z}{1 - n_z^2} S|\mathbf{n}\rangle + i\mathbf{n} \times \frac{\partial|\mathbf{n}\rangle}{\partial\mathbf{n}}.$$
 (A14)

The spin coherent states are used to construct the so-called P representation for the density matrix:

$$\hat{\rho}(t) = \int d^2 \boldsymbol{n} P(\boldsymbol{n}, t) |\boldsymbol{n}\rangle \langle \boldsymbol{n}|, \quad \int d^2 \boldsymbol{n} P(\boldsymbol{n}, t) = 1. \quad (A15)$$

In this representation we have

$$[\hat{\mathbf{S}}, \hat{\rho}] = -i \int d^2 \mathbf{n} \mathbf{n} \times \frac{\partial P(\mathbf{n})}{\partial \mathbf{n}} |\mathbf{n}\rangle \langle \mathbf{n}|,$$
 (A16a)

$$\{\hat{\mathbf{S}}, \hat{\boldsymbol{\rho}}\} = 2S \int d^2 \boldsymbol{n} \boldsymbol{n} P(\boldsymbol{n}) |\boldsymbol{n}\rangle \langle \boldsymbol{n}| \left[1 + O\left(\frac{1}{S}\right)\right].$$
 (A16b)

The first relation follows from Eq. (A14); the second one is obtained by representing the spin operator as in Eq. (A13) and using Eq. (A12). As a result, we arrive at the Fokker-Planck equation for the spin direction:

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial \mathbf{n}} \cdot \{-2[\mathbf{n} \times \mathbf{B}]P + 4S\mathcal{J}_{\text{eff}}[\mathbf{n} \times (\mathbf{n} \times \mathbf{B})]P\}$$

$$+2\mathcal{J}_{\text{eff}}T_{\text{eff}}\frac{\partial^2 P}{\partial \mathbf{n}^2}.$$
(A17)

This equation corresponds to Eq. (5.10) if one associates $4S\mathcal{J}_{\mathrm{eff}} \leftrightarrow 1/(h_0\tau)$ and $S \leftrightarrow h_0/\delta_1$. The latter is in agreement with Eq. (3.15).

APPENDIX B: LONGITUDINAL SUSCEPTIBILITY AT LOW FREQUENCIES

We find the linear in frequency $\Omega \leq 1$ correction to the dc susceptibility. For this purpose, we look for a solution to Eq. (7.11) in the form

$$u_{\parallel}(\theta) = u_{\parallel}^{(0)}(\theta) + u_{\parallel}^{(1)}(\theta),$$
 (B1)

where $u_{\parallel}^{(0)}(\theta)$ is the solution of Eq. (7.11) at $\Omega=0$ and $u_{\parallel}^{(1)}(\theta) \propto \Omega$. We choose

$$u_{\parallel}^{(0)}(\theta) = \frac{1}{b} - \coth b + \cos \theta, \tag{B2}$$

since this form of $u_{\parallel}^{(0)}(\theta)$ preserves the normalization condition (7.13). This function can be found directly as a solution of Eq. (7.11) with Ω =0 or as a variational derivative of function $\mathcal{P}_0(\theta)$, defined in Eq. (7.3), with respect to b.

The linear in Ω correction $u_{\parallel}^{(1)}(\theta)$ is the solution to the differential equation

$$\frac{\partial^{2} u_{\parallel}^{(1)}(\theta)}{\partial \theta^{2}} + \frac{\cos \theta - b \sin^{2} \theta}{\sin \theta} \frac{\partial u_{\parallel}^{(1)}(\theta)}{\partial \theta} = -i\Omega u_{\parallel}^{(0)}(\theta).$$
(B3)

From this equation, we can easily find

$$\frac{\partial u_{\parallel}^{(1)}(\theta)}{\partial \theta} = -\frac{i\Omega}{b \sin \theta} \left[\coth b - \cos \theta - \frac{e^{-b \cos \theta}}{\sinh b} \right]. \quad (B4)$$

We notice that the solution to the latter equation will automatically satisfy the boundary conditions, given by Eq. (7.14). Integrating Eq. (B4) once again, we obtain the following expression for function $u_{\parallel}^{(1)}(\theta)$:

$$u_{\parallel}^{(1)}(\theta) = C(b) - \frac{i\Omega}{b} \int_{0}^{\theta} \left[\coth b - \cos \theta' - \frac{e^{-b\cos \theta'}}{\sinh b} \right] \frac{d\theta'}{\sin \theta'}.$$
(B5)

Here the integration constant C(b) has to be chosen to satisfy the normalization condition, Eq. (7.13), which results in a complicated expression for the final form of the function $u_{\parallel}^{(1)}(\theta)$.

To obtain function $f_{\parallel}(b)$, introduced in Eq. (7.21), we have to perform the final integration

$$f_{\parallel}(b) = \frac{2\pi}{\Omega} \int_{0}^{\pi} u_{\parallel}^{(1)}(\theta) \mathcal{P}_{0}(\theta) \sin \theta \cos \theta d\theta.$$
 (B6)

The result of the integration is shown in Fig. 4.

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