Magnetization damping in a local-density approximation

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The linear response of itinerant transition-metal ferromagnets to transverse magnetic fields is studied in a self-consistent adiabatic local-density approximation. The susceptibility is calculated from a microscopic Hamiltonian, including spin-conserving impurities, impurity-induced spin-orbit interaction, and magnetic impurities using the Keldysh formalism. The Gilbert damping constant in the Landau-Lifshitz-Gilbert equation is identified, parametrized by an effective transverse spin dephasing rate, and is found to be inversely proportional to the exchange splitting. Our results justify the phenomenological treatment of transverse spin dephasing in the study of current-induced magnetization dynamics in weak, itinerant ferromagnets by Tserkovnyak *et al.* [Phys. Rev. B **74**, 144405 (2006)]. We show that neglect of gradient corrections in the quasiclassical transport equations leads to incorrect results when the exchange potential becomes of the order of the Fermi energy.

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I. INTRODUCTION

The drive to miniaturize and reduce power demands of electronic appliances motivates research in nanoscale magnetoelectronics, i.e., the science and technology that exploits additional functionalities offered by ferromagnets integrated into electronic circuits and devices. Spectacular advances have been realized already in the last decade, mainly in the area of magnetic disk and magnetic random access memories. Itinerant transition metals and its alloys are the materials of choice for magnetoelectronic applications due to their high electric conductivity and Curie temperatures. Increasing speed and reducing energy demands of switching a bit of information encoded by the magnetization direction of a ferromagnetic grain is one of the key problems in the field. A thorough understanding of the dynamics of the magnetization order parameter in transition metals is necessary to make progress in this direction.

Phenomenologically, the low-temperature magnetization dynamics in ferromagnets is well described by the Landau-Lifshitz-Gilbert (LLG) equation.[2,3] Ferromagnetic resonance (FMR) experiments can be fitted to obtain accurate values for the parameters of the LLG equation, viz., the Gilbert constant that parametrizes viscous damping and the effective (demagnetization and crystal anisotropy) fields. The LLG phenomenology has been successfully applied to explain a rich variety of dynamic magnetic phenomena.[4,5] Some progress has been made in predicting magnetic crystal anisotropies by first-principles calculations.[6] However, in spite of being a crucial device parameter that governs the switching time of magnetic memory elements, the material dependence of the intrinsic magnetization damping has not yet been understood. The Gilbert damping parameter also plays an important role in current-induced magnetization excitations and domain-wall motion.[7,1]

Deriving a microscopic description of the dynamics of transition-metal ferromagnets is a formidable task; even the nature of the ground state is still under debate. Two different viewpoints can be distinguished. On one hand, ferromagnetism can be seen to be caused by the atomic correlations in partially filled and essentially localized *d* orbitals. The *s* electrons that are responsible for electron transport are in this picture affected by the magnetic order only indirectly via local exchange interactions. Such physics is expressed by the so-called *s-d* model, in which the spin of localized *d* electrons, $\boldsymbol{S}_i$, interacts with free *s*-electron spins $\boldsymbol{s}(\boldsymbol{r}_i,t)$ through a Heisenberg exchange term.

In the opposite point of view, the *d* electrons are not only broadened into bands but are also strongly hybridized with neighboring *s-p* orbitals. A separate treatment of states with different orbital symmetries is then not warranted for the description of low-energy properties at long time scales. The Stoner model represents the essence of this itinerant magnetism in terms of two (minority and majority) parabolic energy bands that are split by a constant exchange potential. Spin density-functional theory in a local-density approximation is the modern version of itinerant magnetism, forming the basis of most band-structure calculations to date. The nature of the real wave function of 3*d* ferromagnets that combines features of both extremes is presumably captured by sophisticated many-body frameworks such as the dynamical mean-field model. It is at present not obvious, however, how to compute the low-energy collective dynamics of ferromagnets taking disorder as a well as local correlations into account.

The Gilbert damping coefficient in the LLG equation, usually denoted by α, has attracted quite some theoretical attention. Incoherent scattering of electron-hole pair excitations by phonons and magnons is a possible mechanism by which energy and angular momentum can be dissipated. Heinrich *et al.*[8] suggested a model in which conduction electron spins become polarized by scattering with magnons. The spin angular momentum is subsequently transferred to the lattice by spin-orbit mediated relaxations. The resulting

damping coefficient was found to be proportional to the electronic scattering rate, $\alpha \sim \tau^{-1}$. We will return to this result in Sec. IV B. More recently, a phenomenological treatment of the Gilbert damping has also been reported in Ref. 9.

A different relaxation process was proposed in Ref. 10 and was further elaborated in Refs. 11–14: In the presence of spin-orbit interaction, the electronic energy levels depend on a time-dependent magnetization direction, giving rise to the notion of a "breathing Fermi surface." The time lag of the electronic distribution response to a moving magnetization vector is equivalent to dissipation. In this model, the Gilbert damping coefficient is proportional to the scattering time, $\alpha \sim \tau$. Extrinsic contributions to the FMR linewidth such as eddy currents excited by time-dependent magnetic fields,[15] sample inhomogeneities, or two-magnon scattering processes[16–18] have been suggested as well.

In diluted magnetic semiconductors such as (Ga, Mn)As, the magnetism originates mainly from the local spins of the half-filled spin-5/2 Mn *d* shells. The spins are coupled by a local exchange interaction to the valence-band holes and nonlocally, via the holes, ferromagnetically to each other. The holes contribute only slightly to the magnetization but are exclusively responsible for the finite conductivity. The *s-d* model is therefore appropriate for understanding the magnetization damping in ferromagnetic semiconductors.[19,20] Magnetization damping in the *s-d* model can be understood in terms of the so-called spin-pumping mechanism.[21–24] The motion of the localized spins pumps a spin current into the conduction-electron bath, in which the thus created spin accumulation is dissipated by spin-flip scattering. Reference 20 reported a nonmonotonous dependence of the damping on the scattering rate, i.e., $\alpha \sim \tau^{-1}$ for clean and $\alpha \sim \tau$ for dirty samples. As mentioned above, the *s-d* model does not necessarily give a good description of transport and dynamical properties of transition-metal ferromagnets. The notion of *d* electrons pumping spins into an *s*-electron system becomes doubtful when the hybridization is very strong. Recently it has been demonstrated that the magnetization dynamics in the *s-d* model and in an itinerant Stoner model can be quite different indeed.[1] For example, for a given spin-flip relaxation mechanism, the Gilbert damping is significantly suppressed in the *s-d* description by a factor of the (usually small) fraction of the total magnetization carried by the delocalized *s* electrons.

The Gilbert damping and the nonadiabatic current-induced spin torque term β, postulated by Zhang and Li,[7] have been derived in Ref. 1 under the assumption that the exchange splitting is small compared to the Fermi energy. Recently, Kohno *et al.*[25] reported a diagrammatic derivation of these parameters, which was not restricted to weak ferromagnets. The differences in these results turned out to be very small for transition metals.[1]

In the present paper, we generalize the treatment of the transverse spin dephasing of Ref. 1 beyond the relaxation time approximation. We relax the previous limitation to weak ferromagnets and derive the corresponding Gilbert damping. We use a self-consistent adiabatic local-density approximation (ALDA) model in the presence of a dilute concentration of scalar and magnetic impurities, as well as spin-orbit interaction originating from impurities, and we demonstrate how to generalize the previous treatment to strong ferromagnets. The generalization of the Keldysh approach is nontrivial and introduces subtle but important gradient corrections that do not play a role in normal metals. We work out the details for a model Hamiltonian. In order to make connections with real experiments, we would have to make at least educated guesses about the disorder potentials and compute scattering matrix elements for realistic band structures. This is beyond the scope of the present paper; however, we do hope to stimulate experiments in which transport and magnetization dynamics are measured as a function of controlled disorder, which, in turn, would stimulate a quantitative theoretical study.

Our main result is that for spatially homogeneous itinerant ferromagnets, the Gilbert damping constant is given by

$$\alpha = \frac{\hbar}{\Delta \tau_{\perp}}, \tag{1}$$

where Δ is the modulus of the local-density exchange-correlation potential and $\tau_{\perp}^{-1}$ is a transverse (Bloch) spin dephasing rate caused by spin-orbit interaction and magnetic disorder. This appears to be at variance with Kohno *et al.*,[25] who found a Gilbert damping constant that depends on both the longitudinal and the transverse scattering rates. Except for this issue, we obtain the same detailed expression for α. This is gratifying since these theoretical machineries are completely different.

This paper is organized in the following way: The microscopic model as well as the simplifying ALDA are presented in Sec. II, while the linear-response formalism, microscopically and phenomenologically, will be treated in Sec. III. The detailed derivation of the linear-response function, starting from the Keldysh Green's function formalism, is the topic of Sec. IV. Conclusions are summarized in Sec. V.

II. TIME-DEPENDENT ADIABATIC LOCAL-DENSITY APPROXIMATION

Density-functional theory (DFT) is a successful and widely used method in the study of electronic structure and magnetism in transition-metal ferromagnets.[26] In the Kohn-Sham implementation, noninteracting pseudoparticles are introduced, which exhibit the same ground-state density as the interacting many-electron system. This is realized by introducing a fictitious exchange-correlation potential that has to be determined self-consistently by energy minimization.[27] DFT can be expanded to handle time-dependent phenomena in systems out of equilibrium.[28]

We study the magnetization dynamics in a simplified time-dependent DFT in the local spin-density approximation, in which noninteracting Kohn-Sham particles are treated as free electrons. We realize that the transition metals have complex energy bands and wave functions also in the local-density approximation and that even integrated properties such as conductivities are not necessarily well described by free electrons. However, since we are interested in dirty systems, in which an additional scrambling occurs by elastic impurity scattering between different bands, we are confident that our treatment is a good starting point for more sophisti-

cated computations that take the full band structure into account.

Our model for itinerant ferromagnetism is described by the Hamiltonian

$$\hat{\mathcal{H}} = \hat{1}[\mathcal{H}_0 + V(\boldsymbol{r})] + \frac{1}{2}\gamma\hbar H\hat{\sigma}_z + \frac{1}{2}\gamma\hbar \boldsymbol{H}_{\mathrm{xc}}[\hat{\rho}(\boldsymbol{r},t)]\cdot\hat{\boldsymbol{\sigma}} + \frac{1}{2}\gamma\hbar \boldsymbol{h}(\boldsymbol{r},t)\cdot\hat{\boldsymbol{\sigma}} + \hat{V}_{\mathrm{so}}(\boldsymbol{r}) + \hat{V}_m(\boldsymbol{r}). \quad (2)$$

Matrices in 2×2 spin space are denoted by a hat ($\hat{}$). Spin independence is indicated by the unit matrix $\hat{1}$, and $\hat{\boldsymbol{\sigma}}$ is a vector of the Pauli matrices. Here, $\mathcal{H}_0$ is the translationally invariant Hamiltonian of noninteracting electrons, and $V(\boldsymbol{r})$ is the elastic spin-conserving impurity potential. $H>0$ is an effective magnetic field in the z direction consisting of internal anisotropy fields and externally applied contributions, and $-\gamma<0$ is the electronic gyromagnetic ratio. Electron-electron interactions are described by the exchange-correlation (vector) field $\boldsymbol{H}_{\mathrm{xc}}$. The weak, transverse magnetic driving field is denoted by $\boldsymbol{h}(\boldsymbol{r},t)$, and the potentials due to impurity-induced spin-orbit interaction and a magnetic disorder configuration are denoted by $\hat{V}_{\mathrm{so}}(\boldsymbol{r})$ and $\hat{V}_m(\boldsymbol{r})$, respectively.

The published approximation schemes for time-dependent exchange-correlation functionals are still rather crude and/or untested. The simplest approximation is known as the adiabatic local-density approximation (ALDA). Here, the conventional local-density exchange-correlation potential is adopted for the instantaneous time-dependent density.[29,30] The ALDA potential is therefore local in both spatial and temporal degrees of freedom and reduces for the current problem to

$$\gamma\hbar \boldsymbol{H}_{\mathrm{xc}}[\hat{\rho}](\boldsymbol{r},t) \approx \Delta \boldsymbol{m}(\boldsymbol{r},t), \quad (3)$$

where Δ is an effective exchange splitting constant and $\boldsymbol{m}(\boldsymbol{r},t)$ is the local magnetization direction of the ferromagnet. By construction, the exchange field is always parallel to the magnetization direction and thus automatically satisfies the zero-torque theorem.[31] Since we are interested in the low-energy transverse magnetization dynamics, the (atomic-scale) position dependence of Δ is disregarded.

In the ALDA, the exchange-correlation potential is released from a possible functional dependence on the history of the system. The ALDA should therefore be valid only when the system is close to the equilibrium configuration, i.e., for a slowly varying magnetization direction in both space and time. This is the case when $\hbar\partial_t \ll \Delta$ and $\partial_{\boldsymbol{r}} \ll k_F$, with k_F being a characteristic Fermi wave vector. Improved descriptions of the exchange potential have been proposed (e.g., the generalized gradient approximation), but for slowly varying uniform perturbations, such corrections are believed to be small.[32]

III. LINEAR RESPONSE

For a weak magnetic driving field, the response of the ferromagnet can be formulated within the linear-response theory. An expression for the response to the perturbative field $\boldsymbol{h}(\boldsymbol{r},t)$ is derived quantum mechanically from the ALDA Hamiltonian, which is defined by Eqs. (2) and (3), in Sec. III A. The response derived from the phenomenological Landau-Lifshitz-Gilbert equation is presented in Sec. III B. These results are then used in Sec. IV to find a microscopic expression for the Gilbert damping coefficient.

A. Quantum linear response

The Kubo formalism provides expressions for the linear response to a time-dependent perturbation. The response functions can be derived by considering the time evolution of the nonequilibrium density matrix. Starting from the effective Hamiltonian (2) in the ALDA of Eq. (3), the small time-dependent perturbation operator should include the self-consistent exchange as

$$H_{\mathrm{int}}(t) = \int d\boldsymbol{r}\left[\frac{\Delta}{\hbar}\delta\boldsymbol{m}(\boldsymbol{r},t) + \gamma\boldsymbol{h}(\boldsymbol{r},t)\right]\cdot\boldsymbol{s}(\boldsymbol{r}),$$

where $\boldsymbol{s}(\boldsymbol{r})$ is the spin-density operator. The emphasis of this paper is on the transverse, nonequilibrium components of the spin density. We denote $s_0=|\boldsymbol{s}_0|$ and $\langle\boldsymbol{s}\rangle=-s_0\boldsymbol{e}_z+\langle\delta\boldsymbol{s}\rangle$, where $\langle\delta\boldsymbol{s}\rangle\perp\boldsymbol{e}_z$. Hence, $|\boldsymbol{s}|=s_0$ and $\boldsymbol{m}=-\langle\boldsymbol{s}\rangle/s_0$ in the ALDA.

For axially symmetric systems, the nonequilibrium spin-density response can be expressed conveniently in terms of $\delta s_\pm=\delta s_x\pm i\,\delta s_y$. The transverse part of the response to the magnetic field can then be written as

$$\langle\delta s_-(\boldsymbol{q},\omega)\rangle = -\chi_{-+}(\boldsymbol{q},\omega)\left[\frac{\Delta}{\gamma\hbar}\delta m_-(\boldsymbol{q},\omega) + h_-(\boldsymbol{q},\omega)\right], \quad (4)$$

where the retarded susceptibility tensor

$$\chi_{\mu\nu}(\boldsymbol{r},\boldsymbol{r}';t) = \frac{i\gamma}{2\hbar}\Theta(t)\langle[s_\mu(\boldsymbol{r},t),s_\nu(\boldsymbol{r}',0)]\rangle,$$

has been introduced. The brackets $[\cdots]$ indicate a commutator and the angular brackets $\langle\cdots\rangle$ a thermodynamical average. In the derivation of the above expression, we have made use of axial symmetry under which $\chi_{++}=\chi_{--}=0$. In the ALDA, Eq. (4) can be simplified to

$$\langle\delta s_-(\boldsymbol{q},\omega)\rangle = -\tilde{\chi}_{-+}(\boldsymbol{q},\omega)h_-(\boldsymbol{q},\omega), \quad (5)$$

where the self-consistent linear response to the driving field,

$$\tilde{\chi}_{-+}^{-1}(\boldsymbol{q},\omega) = \chi_{-+}^{-1}(\boldsymbol{q},\omega) - \frac{\Delta}{\gamma\hbar s_0} \quad (6)$$

has been introduced. Hence, in the ALDA, the linear response of an interacting system reduces itself to calculating the response χ_{-+} of a noninteracting system with a fixed (Stoner enhancement) exchange field.[33]

B. Landau-Lifshitz-Gilbert susceptibility

The phenomenological Landau-Lifshitz equation[2] is widely used to model transverse magnetization dynamics. The magnetization direction $\boldsymbol{m}(\boldsymbol{r},t)$ is treated as a classical field whose dynamics is governed by an effective magnetic

field $\boldsymbol{H}_{\text{eff}}(\boldsymbol{r},t)$, obtainable from the free-energy functional of the system, $F[\boldsymbol{M}]$:

$$\boldsymbol{H}_{\text{eff}}(\boldsymbol{r},t) = -\partial_{\boldsymbol{M}} F[\boldsymbol{M}].$$

The Landau-Lifshitz equation describes undamped (i.e., free-energy conserving) precessional motion about the local effective magnetic field:

$$\partial_t \boldsymbol{m}(\boldsymbol{r},t) = -\gamma \boldsymbol{m}(\boldsymbol{r},t) \times \boldsymbol{H}_{\text{eff}}(\boldsymbol{r},t),$$

preserving the magnitude of the magnetization. The field $\boldsymbol{H}_{\text{eff}}(\boldsymbol{r},t)$ includes contributions from external, exchange, demagnetization, and crystal-anisotropy magnetic fields.

The Landau-Lifshitz equation does not dissipate energy, since the effective magnetic field always points normal to the instantaneous constant-free-energy surfaces. However, the electronic degrees of freedom do not respond infinitely fast to the magnetization dynamics, which means that in reality the effective field is a functional of the time-dependent magnetization at previous times. A finite lag in the response of the dynamics corresponds to energy dissipation. In a magnetic system, the energy-loss implies a lowering of the Zeeman energy by a torque in the direction of the cross product of magnetization and its time derivative; the energy loss can be parametrized by the phenomenological Gilbert damping constant α.[3] Hence, we arrive at the following Landau-Lifshitz-Gilbert (LLG) equation:

$$\partial_t \boldsymbol{m} = -\gamma \boldsymbol{m} \times \boldsymbol{H}_{\text{eff}} + \alpha \boldsymbol{m} \times \partial_t \boldsymbol{m}.$$

Here, $\boldsymbol{H}_{\text{eff}}$ only depends on the instantaneous magnetic configuration of the ferromagnet. Generally, the damping is a tensor quantity with symmetries reflecting the crystal structure,[18] but in practice anisotropic corrections to damping are small compared to those in the free energy.[34]

By assuming an external field of the form $\gamma \boldsymbol{H}_{\text{eff}}(\boldsymbol{r},t) = \omega_0(\boldsymbol{r},t)\boldsymbol{e}_z$ and a small rf driving field $\boldsymbol{h}(\boldsymbol{r},t)$, the excited small-angle transverse magnetization dynamics can be computed easily by the linearized LLG equation

$$m_-(\boldsymbol{q},\omega) = \frac{\gamma h_-(\boldsymbol{q},\omega)}{\omega_0(\boldsymbol{q},\omega) - \omega - i\alpha(\boldsymbol{q},\omega)\omega},$$

which corresponds to a susceptibility

$$\tilde{\chi}_{-+}(\boldsymbol{q},\omega) = \frac{\gamma s_0}{\omega_0(\boldsymbol{q},\omega) - \omega - i\alpha(\boldsymbol{q},\omega)\omega}$$

that can be directly compared with the microscopic response function χ_{-+} by Eq. (6). Assuming that $\Delta/\hbar \gg (\omega, \omega_0)$, one obtains[19,20]

$$\alpha(\boldsymbol{q},\omega \to 0) = \frac{\Delta^2}{\gamma\hbar^2 s_0} \lim_{\omega\to 0} \partial_\omega \operatorname{Im} \chi_{-+}(\boldsymbol{q},\omega). \tag{7}$$

Hence, finding a microscopic expression for the Gilbert damping is equivalent to determining the quantum-mechanical transverse susceptibility.

It is worth noting that, in general, the damping coefficient may depend on the spin-wave wave vector $\boldsymbol{q}$. A damping of the form $\alpha(\boldsymbol{q},\omega\to 0)\sim q^2$ will introduce an additional dissipative term in the LLG equation $\partial_t \boldsymbol{m} \propto -\alpha \boldsymbol{m} \times \nabla^2 \partial_t \boldsymbol{m}$, which is similar in form to the exchange field $\propto \boldsymbol{m} \times \nabla^2 \boldsymbol{m}$ for anisotropic ferromagnets. The main emphasis in this paper, however, is on the isotropic part of the Gilbert damping.

IV. MICROSCOPIC DERIVATION OF THE SUSCEPTIBILITY

In this section, we determine the susceptibility function according to time-dependent spin DFT in the ALDA for a disordered ferromagnet. The Keldysh Green's function formalism is used, and the assumption of weak and slowly varying perturbations in space and time allows us to use the simplifying gradient expansion. Finally, Eq. (7) is invoked to obtain the Gilbert damping coefficient.

A. Kinetic equation

We proceed from the ALDA Hamiltonian (see Sec. II):

$$\hat{\mathcal{H}} = \hat{1}[\mathcal{H}_0 + V(\boldsymbol{r})] + \frac{1}{2}(\Delta + \gamma\hbar H)\hat{\sigma}_z + \frac{1}{2}\gamma\hbar \boldsymbol{h}(\boldsymbol{r},t)\cdot\hat{\boldsymbol{\sigma}} + \hat{V}_{\text{so}}(\boldsymbol{r}) + \hat{V}_m(\boldsymbol{r}). \tag{8}$$

In the following discussion, we assume a homogeneous static magnetic field H, define $\Delta' = \Delta + \gamma\hbar H$, and drop the prime for brevity.

The impurities are assumed to be randomly distributed over positions $\boldsymbol{r}_i$ with short-range, scalar disorder potentials

$$V(\boldsymbol{r}) = \sum_i v_0(\boldsymbol{r}_i)\delta(\boldsymbol{r} - \boldsymbol{r}_i).$$

The scattering potentials are Gaussian distributed with zero average and a white noise correlator

$$\langle V(\boldsymbol{r})V(\boldsymbol{r}')\rangle = \xi\delta(\boldsymbol{r} - \boldsymbol{r}').$$

We define a characteristic scattering time τ by $\xi^{-1} = \pi(\nu_\uparrow + \nu_\downarrow)\tau/\hbar$, with ν_s being the density of states at the Fermi level for electrons with spin s. The spin-orbit interaction associated with impurities is described by the potential

$$\hat{V}_{\text{so}}(\boldsymbol{r}) = i\beta\hat{\boldsymbol{\sigma}}\cdot(\nabla V(\boldsymbol{r}) \times \nabla),$$

where β is a spin-orbit interaction strength given by $-\hbar^2/4m_e^2c^2$, in terms of the electron mass m_e and the speed of light c. The magnetic disorder in the ferromagnet is modeled as

$$\hat{V}_m(\boldsymbol{r}) = \sum_i v_m(\boldsymbol{r}_i)\delta(\boldsymbol{r} - \boldsymbol{r}_i)\mathcal{S}(\boldsymbol{r}_i)\cdot\hat{\boldsymbol{\sigma}},$$

where $\mathcal{S}(\boldsymbol{r}_i)$ denotes the spin of an impurity at position $\boldsymbol{r}_i$. The internal degrees of freedom of the magnetic impurities are assumed to be frozen. Also, the vector impurity exchange field $\boldsymbol{V}(\boldsymbol{r}) = \frac{1}{2}\text{Tr}[\hat{V}_m(\boldsymbol{r})\hat{\boldsymbol{\sigma}}]$ is taken to be distributed according to Gaussian white noise characteristics, i.e., $\langle V_\alpha(\boldsymbol{r})\rangle = 0$ and

$$\langle V_\alpha(\boldsymbol{r})V_\beta(\boldsymbol{r}')\rangle = \xi_m^{(\alpha)}\delta_{\alpha\beta}\delta(\boldsymbol{r} - \boldsymbol{r}'),$$

where α and β denote spatial components of the vector field, and the strength of the second moment is given by

$$\xi_m^{(\alpha)} = \begin{cases} \xi_\perp, & \alpha = x,y \\ \xi_\parallel, & \alpha = z, \end{cases}$$

similar to Ref. 25.

We employ the Keldysh Green's function formalism[35] to calculate the spin susceptibility defined earlier. This method has distinct advantages over the equilibrium formalism when it comes to describing nonequilibrium phenomena but also has some drawbacks. However, as we will see in Sec. IV B, in particular, whereas the equilibrium formalism requires tedious calculations of vertex corrections, the Keldysh formalism requires that one carefully accounts for subtle gradient corrections in order to obtain the correct dynamics.

The Green's function in Keldysh space, denoted by an inverted caret ($\check{}$) takes the form[36]

$$\check{G}(1,2) = \begin{pmatrix} \hat{G}^R(1,2) & \hat{G}^K(1,2) \\ 0 & \hat{G}^A(1,2) \end{pmatrix}.$$

The retarded, advanced, and Keldysh Green's functions are given by

$$\hat{G}^R(1,2) = -i\Theta(t_1 - t_2)\langle\{\Psi(1),\Psi^\dagger(2)\}\rangle,$$

$$\hat{G}^A(1,2) = +i\Theta(t_2 - t_1)\langle\{\Psi(1),\Psi^\dagger(2)\}\rangle,$$

and

$$\hat{G}^K(1,2) = -i\langle[\Psi(1),\Psi^\dagger(2)]\rangle,$$

respectively. The brackets $[\cdots]$ indicate a commutator and the curly brackets $\{\cdots\}$ an anticommutator, while $\Psi^{(\dagger)}$ is a fermion annihilation (creation) operator. In this notation, all field variables (position, time, and spin) are contained in the numerical indices 1 and 2.

In the presence of slowly varying perturbations, the two-point propagator variables can be transformed into the Wigner representation, viz., the center-of-mass coordinates and the Fourier transform of the Green's function with respect to the relative coordinates:

$$\check{G}(X,k) = \int dx\, e^{-ikx}\check{G}(X + x/2, X - x/2).$$

Here, a four-vector formulation has been introduced, where the vector for the center-of-mass coordinates is $X=(\boldsymbol{R},T)$, the corresponding relative coordinates are given by $x=(\boldsymbol{r},t)$, and finally $k=(\boldsymbol{k},\varepsilon)$. The four-vector product is defined as $k\cdot x = -\varepsilon t + \boldsymbol{k}\cdot\boldsymbol{r}$. The Wigner representation is particularly convenient when the variation of the Green's function on center coordinates is slow on the scale of the Fermi wavelength, since this allows us to perform a gradient expansion in these coordinates. Subtracting the Dyson equation and its conjugate, one finds the relation

$$[\check{G}_0^{-1} - \check{\Sigma} \otimes \check{G}] = 0, \tag{9}$$

where the symbol $\otimes$ denotes a convolution (in position, time, and spin), the commutator corresponds to the 2×2 Keldysh matrix structure, and $\check{G}_0^{-1}$ is the inverse of the Green's function in the absence of any impurities. It is diagonal in Keldysh space with the inverse of the retarded and advanced Green's function as elements. In the Wigner representation each of them has the structure

$$\hat{G}_0^{-1}(\boldsymbol{R},T;\boldsymbol{k},\varepsilon) = \hat{1}(\varepsilon - \varepsilon_k) - \frac{1}{2}\Delta\hat{\sigma}_z - \frac{1}{2}\gamma\hbar\boldsymbol{h}(\boldsymbol{R},T)\cdot\hat{\boldsymbol{\sigma}} \tag{10}$$

in spin space, with ε_k denoting the free-electron energy measured with respect to the chemical potential. The final component in Eq. (9), $\check{\Sigma}$, is the self-energy due to the impurity configurations and spin-orbit interaction. One can show by a formal Taylor expansion that the convolution can be represented by

$$(A \otimes B)(X,k) = e^{i(\partial_X^A\cdot\partial_k^B - \partial_k^A\cdot\partial_X^B)/2}A(X,k)B(X,k) \tag{11}$$

in the Wigner representation.[36]

Physical quantities such as occupation probabilities and densities are expressible in terms of the distribution Green's function $\hat{G}^<$, which is given by the combination

$$\hat{G}^< = \frac{1}{2}(\hat{G}^K + i\hat{A}),$$

where we have introduced the spectral function $\hat{A} = i(\hat{G}^R - \hat{G}^A)$. To derive a kinetic equation for $\hat{G}^<$, we subtract the diagonal components of Eq. (9) and combine the result with the Keldysh component of the same equation. In summary, one finds the kinetic equation

$$[\hat{G}^R]^{-1} \otimes \hat{G}^< - \hat{G}^< \otimes [\hat{G}^A]^{-1} = \hat{\Sigma}^< \otimes \hat{G}^A - \hat{G}^R \otimes \hat{\Sigma}^<.$$

Assuming slowly varying perturbations, we now use the gradient expansion, in which the exponential in Eq. (11) is expanded and only the first two terms of the expansion are kept.[37] This results in a simplified kinetic equation for the distribution Green's function, viz.,

$$\begin{aligned} &[\hat{G}_0^{-1},\hat{G}^<] + \frac{i}{2}[\hat{G}_0^{-1},\hat{G}^<]_p - \frac{i}{2}[\hat{G}^<,\hat{G}_0^{-1}]_p \\ &- (\hat{\Sigma}^R\hat{G}^< - \hat{G}^<\hat{\Sigma}^A) + (\hat{G}^R\hat{\Sigma}^< - \hat{\Sigma}^<\hat{G}^A) \\ &= \frac{i}{2}([\hat{\Sigma}^R,\hat{G}^<]_p - [\hat{G}^<,\hat{\Sigma}^A]_p) - \frac{i}{2}([\hat{G}^R,\hat{\Sigma}^<]_p - [\hat{\Sigma}^<,\hat{G}^A]_p). \end{aligned} \tag{12}$$

All terms to first order in the generalized Poisson bracket are kept, $[\hat{X},\hat{Y}]_p = \partial_X\hat{X}\cdot\partial_k\hat{Y} - \partial_k\hat{X}\cdot\partial_X\hat{Y}$, where the four-vector notation implies $\partial_X\cdot\partial_k = \partial_{\boldsymbol{R}}\cdot\partial_{\boldsymbol{k}} - \hbar\partial_T\partial_\varepsilon$. We see that the gradient expansion reduces the complex convolution of the Dyson equation (9) to the 2×2 matrix multiplication of Eq. (12).

It is important to carefully consider all contributions to first order in the Poisson brackets. The correct dynamics is captured only when gradient corrections to the spectral function are kept and, in the case of anisotropically distributed magnetic impurities, those to the self-energies as well. Such gradient corrections are caused by a nonuniform driving field in magnetic metals. For simple systems such as normal metals, such corrections are irrelevant. We address this point in

more detail in Sec. IV B, and we also refer the reader to the Appendix, where one can explicitly see from, e.g., Eq. (A1) that the gradient corrections to the retarded and/or advanced Green's functions are proportional to Δ for weak disorder. The corrections vanish when the normal metal limit $\Delta \to 0$ is taken. Boltzmann equations that disregard certain Poisson brackets[36] should therefore not be generalized naively to *strong* ferromagnets.

The final ingredient that transforms Eq. (12) into a useful kinetic equation is an expression for the self-energy. For weak impurity scattering, the self-consistent Born approximation is appropriate:

$$\check{\Sigma}(1,2) = \langle \hat{V}_{\text{tot}}(1)\check{G}(1,2)\hat{V}_{\text{tot}}(2)\rangle,$$

where $\hat{V}_{\text{tot}}$ is the total potential (which is diagonal in Keldysh space), angular brackets $\langle\cdots\rangle$ denotes impurity potential averaging, and $\check{G}(1,2)$ is already an impurity averaged Green's function. This expression can be separated into four different self-energy contributions. The spin-conserving impurity scattering is described by

$$\check{\Sigma}_{\text{imp}}(\boldsymbol{R},T;\boldsymbol{k},\varepsilon) = \xi \int \frac{d\boldsymbol{k}'}{(2\pi)^3}\check{G}(\boldsymbol{R},T;\boldsymbol{k}',\varepsilon).$$

Introducing $\boldsymbol{n}=\boldsymbol{k}\times\boldsymbol{k}'$, the terms arising from the spin-orbit interaction can be written as

$$\check{\Sigma}^{(1)}_{\text{so}}(\boldsymbol{R},T;\boldsymbol{k},\varepsilon) = -i\xi\beta \int \frac{d\boldsymbol{k}'}{(2\pi)^3}[\check{G}(\boldsymbol{R},T;\boldsymbol{k}',\varepsilon)\hat{\boldsymbol{\sigma}}\cdot\boldsymbol{n} - \boldsymbol{n}\cdot\hat{\boldsymbol{\sigma}}\check{G}(\boldsymbol{R},T;\boldsymbol{k}',\varepsilon)]$$

and

$$\check{\Sigma}^{(2)}_{\text{so}}(\boldsymbol{R},T;\boldsymbol{k},\varepsilon) = \xi\beta^2 \int \frac{d\boldsymbol{k}'}{(2\pi)^3}\boldsymbol{n}\cdot\hat{\boldsymbol{\sigma}}\check{G}(\boldsymbol{R},T;\boldsymbol{k}',\varepsilon)\hat{\boldsymbol{\sigma}}\cdot\boldsymbol{n}.$$

The magnetic impurity configuration results in

$$\check{\Sigma}_m(\boldsymbol{R},T;\boldsymbol{k},\varepsilon) = \sum_{i=x,y,z}\xi^{(i)}_m \int \frac{d\boldsymbol{k}'}{(2\pi)^3}\hat{\sigma}_i\check{G}(\boldsymbol{R},T;\boldsymbol{k}',\varepsilon)\hat{\sigma}_i.$$

Finally, to make connection with the spin density, one can use that

$$\langle \boldsymbol{s}(\boldsymbol{R},T)\rangle = \frac{\hbar}{4i\pi}\int_{-\infty}^{\infty} d\varepsilon \int \frac{d\boldsymbol{k}}{(2\pi)^3}\text{Tr}\{\hat{\boldsymbol{\sigma}}\hat{G}^<(\boldsymbol{R},T;\boldsymbol{k},\varepsilon)\}.$$

All necessary quantities are now defined, and a kinetic equation for the distribution Green's function can be derived. In the next section, the details are worked out for a bulk, single-domain ferromagnet.

B. Homogeneous ferromagnet

We concentrate in the following on the Gilbert damping constant in the limit of vanishing spin-wave wave vector, $\boldsymbol{q} \to 0$, as measured in FMR experiments. In this limit (and the ALDA), only spin-orbit interaction and magnetic disorder can transfer angular momentum out of the spin dynamics into the lattice. Without it, spin and orbital degrees of freedom are completely decoupled and the Gilbert constant vanishes. Spin waves with finite wavelengths may decay also by spin-conserving scattering, which is likely to dominate magnetic impurity or spin-orbit interaction scattering when q becomes larger.[38]

We simplify the notation by defining the time- and energy-dependent density matrix

$$\hat{\rho}(T,\varepsilon) = \int \frac{d\boldsymbol{k}}{(2\pi)^3}\hat{G}^<(T;\boldsymbol{k},\varepsilon)$$

and solve Eq. (12) to obtain a diffusion equation for this quantity. In detail, we find that

$$\begin{aligned} i\hbar\partial_T\hat{\rho} - \frac{1}{2}\Delta[\hat{\sigma}_z,\hat{\rho}] - \frac{1}{2}\gamma\hbar\boldsymbol{h}(T)\cdot[\hat{\boldsymbol{\sigma}},\hat{\rho}] + \frac{i}{4}\gamma\hbar^2\{\partial_T(\boldsymbol{h}\cdot\hat{\boldsymbol{\sigma}}),\partial_\varepsilon\hat{\rho}\} \\ = \sum_i \xi^{(i)}_m \int \frac{d\boldsymbol{k}}{(2\pi)^3}(\hat{\sigma}_i\hat{G}^R\hat{\sigma}_i\hat{\rho} - \hat{G}^R\hat{\sigma}_i\hat{\rho}\hat{\sigma}_i - (\hat{\rho}\hat{\sigma}_i\hat{G}^A\hat{\sigma}_i \\ - \hat{\sigma}_i\hat{\rho}\hat{\sigma}_i\hat{G}^A)) + \sum_i \frac{i\xi^{(i)}_m}{2}\int \frac{d\boldsymbol{k}}{(2\pi)^3}([\hat{\sigma}_i\hat{G}^R\hat{\sigma}_i,\hat{\rho}]_p \\ - [\hat{G}^R,\hat{\sigma}_i\hat{\rho}\hat{\sigma}_i]_p - ([\hat{\rho},\hat{\sigma}_i\hat{G}^A\hat{\sigma}_i]_p - [\hat{\sigma}_i\hat{\rho}\hat{\sigma}_i,\hat{G}^A]_p)) \\ + \sum_{i,j}\xi\beta^2\int\frac{d\boldsymbol{k}'}{(2\pi)^3}\int\frac{d\boldsymbol{k}}{(2\pi)^3}n_in_j(\hat{\sigma}_i\hat{G}^R\hat{\sigma}_j\hat{G}^< \\ - \hat{G}^R\hat{\sigma}_i\hat{G}^<\hat{\sigma}_j - (\hat{G}^<\hat{\sigma}_i\hat{G}^A\hat{\sigma}_j - \hat{\sigma}_i\hat{G}^<\hat{\sigma}_j\hat{G}^A)) \\ + \sum_{i,j}\frac{\xi\beta^2}{2}\int\frac{d\boldsymbol{k}'}{(2\pi)^3}\int\frac{d\boldsymbol{k}}{(2\pi)^3}n_in_j([\hat{\sigma}_i\hat{G}^R\hat{\sigma}_j,\hat{G}^<]_p \\ - [\hat{G}^R,\hat{\sigma}_i\hat{G}^<\hat{\sigma}_j]_p - ([\hat{G}^<,\hat{\sigma}_i\hat{G}^A\hat{\sigma}_j]_p - [\hat{\sigma}_i\hat{G}^<\hat{\sigma}_j,\hat{G}^A]_p)). \end{aligned} \tag{13}$$

Here, the arguments of $\hat{\rho}(T,\varepsilon)$, $\hat{G}^{R/A}(T;k,\varepsilon)$, and $\hat{G}^<(T;k',\varepsilon)$ are not written out explicitly for the sake of notation. Summation indices i and j run over Cartesian components x, y, and z. On the left-hand side of the equation, we recognize precession around the fixed exchange field and the driving field, as well as a gradient term due to the nonuniformity of the driving field. On the right-hand side, we find collision integrals due to spin-orbit interaction and magnetic impurities. We also see that there are gradient corrections to the collision integrals in the above equation. These corrections are often neglected but are important for strong ferromagnets to be discussed below. As explained, scalar disorder does not affect the uniform spin dynamics and drops out of the kinetic equation.

For the response function $\chi_{-+}(\omega)$ introduced in Sec. III A, we need to find an expression for $\langle \delta s_-(\omega)\rangle$, the transverse part of the spin density. To this end, we extract the upper right matrix component of $\hat{\rho}(T,\varepsilon)$, a matrix component we simply denote $\delta s_-(T,\varepsilon)$. This is now related to the nonequilibrium spin density by

$$\langle \delta s_-(T)\rangle = \frac{\hbar}{2i\pi}\int_{-\infty}^{\infty} d\varepsilon\ \delta s_-(T,\varepsilon).$$

With $\nu(\varepsilon_k)$ being the density of states at energy ε_k, we find that

$$
\begin{aligned}
&i\hbar\partial_T\delta s_-(T,\varepsilon)-\Delta\delta s_-(T,\varepsilon)+\gamma\hbar h_-(T)\rho_z(\varepsilon)+\frac{i}{2}\gamma\hbar^2\partial_T h_-(T)\int d\varepsilon_k\nu(\varepsilon_k)\partial_\varepsilon G_d^<(k,\varepsilon)\\
&=-2i(\xi_\perp+\xi_\parallel)\int d\varepsilon_k\nu(\varepsilon_k)[A_d(k,\varepsilon)\delta s_-(T,\varepsilon)-\rho_d(\varepsilon)A_-(T;k,\varepsilon)]-\hbar(\xi_\perp-\xi_\parallel)\int d\varepsilon_k\nu(\varepsilon_k)[\partial_\varepsilon A_z(k,\varepsilon)\partial_T\delta s_-(T,\varepsilon)\\
&-\partial_\varepsilon\rho_z(\varepsilon)\partial_T A_-(T;k,\varepsilon)]-4(\xi_\perp-\xi_\parallel)\int d\varepsilon_k\nu(\varepsilon_k)[\mathrm{Re}\,G_z^R(k,\varepsilon)\delta s_-(T,\varepsilon)+\rho_z(\varepsilon)\mathrm{Re}\,G_-^R(T;k,\varepsilon)]+2i\hbar(\xi_\perp+\xi_\parallel)\int d\varepsilon_k\nu(\varepsilon_k)\\
&\times[\partial_\varepsilon\rho_d(\varepsilon)\partial_T\,\mathrm{Re}\,G_-^R(T;k,\varepsilon)+\partial_\varepsilon\,\mathrm{Re}\,G_d^R(k,\varepsilon)\partial_T\delta s_-(T,\varepsilon)]-\frac{8i}{9}\xi\beta^2\int d\varepsilon_k\nu(\varepsilon_k)k^2\int d\varepsilon_{k'}\nu(\varepsilon_{k'})\\
&\times k'^2[A_d(k,\varepsilon)G_-^<(T;k',\varepsilon)-G_d^<(k',\varepsilon)A_-(T;k,\varepsilon)]+\frac{8i\hbar}{9}\xi\beta^2\int d\varepsilon_k\nu(\varepsilon_k)k^2\int d\varepsilon_{k'}\nu(\varepsilon_{k'})\\
&\times k'^2[\partial_\varepsilon G_d^<(k',\varepsilon)\partial_T\,\mathrm{Re}\,G_-^R(T;k,\varepsilon)+\partial_\varepsilon\,\mathrm{Re}\,G_d^R(k,\varepsilon)\partial_T G_-^<(T;k',\varepsilon)],
\end{aligned}
\tag{14}
$$

where have used the convenient matrix notation

$$\hat{G}=\hat{1}G_d+\boldsymbol{G}\cdot\hat{\boldsymbol{\sigma}}. \tag{15}$$

We now need to Fourier transform and integrate this formidable equation over energy to obtain a diffusion equation for $\langle\delta s_-(\omega)\rangle$. Before we proceed with this calculation, we notice that the real part of the response function simply determines the resonance condition for the system and is thus unimportant for the determination of the Gilbert damping. With this in mind, we write

$$
\begin{aligned}
&(\hbar\omega-\Delta)\langle\delta s_-(\omega)\rangle\\
&=\frac{\hbar}{\pi}(\xi_\perp+\xi_\parallel)\int_{-\infty}^{\infty}d\varepsilon\int d\varepsilon_k\nu(\varepsilon_k)\\
&\times[\rho_d(\varepsilon)A_-(\omega;k,\varepsilon)-A_d(k,\varepsilon)\delta s_-(\omega,\varepsilon)]\\
&+\frac{\hbar^2\omega}{2\pi}(\xi_\perp-\xi_\parallel)\int_{-\infty}^{\infty}d\varepsilon\int d\varepsilon_k\nu(\varepsilon_k)\\
&\times[\partial_\varepsilon A_z(k,\varepsilon)\delta s_-(\omega,\varepsilon)-\partial_\varepsilon\rho_z(\varepsilon)A_-(\omega;k,\varepsilon)]\\
&+\frac{4\hbar}{9\pi}\xi\beta^2\int_{-\infty}^{\infty}d\varepsilon\int d\varepsilon_k\nu(\varepsilon_k)k^2\int d\varepsilon_{k'}\\
&\times\nu(\varepsilon_{k'})k'^2[G_d^<(k',\varepsilon)A_-(\omega;k,\varepsilon)\\
&-A_d(k,\varepsilon)G_-^<(\omega;k',\varepsilon)]+\mathcal{F}(\omega)h_-(\omega),
\end{aligned}
\tag{16}
$$

where $\mathcal{F}(\omega)$ is real and thus does not contribute to the damping. The dissipation is now determined by the above integrals over energy and momentum. Notice how the signs between the longitudinal and transverse magnetic impurity scattering strength enter in the above equation. For simple isotropic magnetic impurities, i.e., with $\xi_\perp=\xi_\parallel$, the second line of Eq. (16) does not contribute to the dissipative dynamics. This term is due to gradient corrections involving self-energies in the original kinetic equation (12). To correctly capture the dynamics when the magnetic impurities are anisotropically distributed, it is essential to include such gradient corrections as well.

In order to calculate the integrals, we need expressions for $G_-^<$, δs_-, and the spectral function $\hat{A}$. Since these quantities enter the collision integrals, we can solve for δs_- and $\hat{A}$ to zeroth order in scattering rates. To this end, we solve Eq. (12) for the Fourier transform of $G_-^<$ and find that

$$
\begin{aligned}
G_-^<(\omega;k',\varepsilon)\approx&\frac{\gamma\hbar h_-(\omega)}{\Delta}\left(1+\frac{\hbar\omega}{\Delta}\right)G_z^<(k',\varepsilon)\\
&+\frac{\gamma\hbar^2\omega h_-(\omega)}{2\Delta}\partial_\varepsilon G_d^<(k',\varepsilon)+\mathcal{O}(\xi,\xi_m^{(i)}).
\end{aligned}
\tag{17}
$$

At this point, we take $G_d^<(k',\varepsilon)=in_F(\varepsilon)A_d(k',\varepsilon)$, with n_F the Fermi-Dirac distribution, so that

$$\partial_\varepsilon G_d^<(k',\varepsilon)=-i\delta(\varepsilon)A_d(k',\varepsilon)+in_F(\varepsilon)\partial_\varepsilon A_d(k',\varepsilon)$$

at low temperatures. Additionally, we use that

$$\delta s_-(\omega,\varepsilon)=\int\frac{d\boldsymbol{k}'}{(2\pi)^3}G_-^<(\omega;k',\varepsilon)$$

in combination with Eq. (17) to solve for the second quantity in question.

In the dilute limit, the Lorentzian shape of the spectral function approaches a Dirac delta function, and two quasiparticle spin bands, split by the exchange field Δ, are resolved. For a uniform, time-independent transverse magnetic field, one finds

$$\hat{A}_0(k,\varepsilon)=\pi\sum_s\delta(\varepsilon-\varepsilon_{ks})\left(\hat{1}+s\hat{\sigma}_z+s\frac{\gamma\hbar\boldsymbol{h}\cdot\hat{\boldsymbol{\sigma}}}{\Delta}\right). \tag{18}$$

The two spin bands are denoted by $s=\uparrow,\downarrow=\pm$, and the notation $\varepsilon_{ks}=\varepsilon_k+s\Delta/2$ has been introduced. A nonuniform driving will also introduce terms in the spectral function that are linear in gradients. A detailed derivation of the spectral

function to first order in the Poisson brackets for a time-dependent driving field $\boldsymbol{h}(T)$ is given in the Appendix, with the main result being

$$\hat{A}(T;k,\varepsilon) = \hat{A}_0(T;k,\varepsilon) + \frac{i\gamma\hbar^2}{\Delta^2} \times \left(A_z(k,\varepsilon) + \frac{\Delta}{2}\partial_\varepsilon A_d(k,\varepsilon) \right) \hat{\sigma}_z \partial_T (\boldsymbol{h}\cdot\hat{\boldsymbol{\sigma}}), \quad (19)$$

where $\hat{A}_0$ is the spectral function from Eq. (18). We see that the weak, transverse driving field induce off-diagonal gradient corrections to the "instantaneous" spectral function. The diagonal components are unchanged and are given by

$$A_d(k,\varepsilon) = \pi \sum_s \delta(\varepsilon - \varepsilon_{ks})$$

and

$$A_z(k,\varepsilon) = \pi \sum_s s\,\delta(\varepsilon - \varepsilon_{ks}).$$

We are now in a position to calculate the above energy integrals. To be more specific, considering the integrals due to magnetic disorder, we find that these terms become

$$\int d\varepsilon_k \nu(\varepsilon_k)[\rho_d(\varepsilon)A_-(\omega;k,\varepsilon) - A_d(k,\varepsilon)\delta s_-(\omega,\varepsilon)] = \frac{i\pi^2\gamma\hbar^2 h_-(\omega)\omega}{2\Delta}\delta(\varepsilon)(\nu_\uparrow + \nu_\downarrow)^2$$

and

$$\hbar\omega \int d\varepsilon_k \nu(\varepsilon_k)[\partial_\varepsilon A_z(k,\varepsilon)\delta s_-(\omega,\varepsilon) - \partial_\varepsilon \rho_z(\varepsilon)A_-(\omega;k,\varepsilon)] = \frac{i\pi^2\gamma\hbar^2 h_-(\omega)\omega}{\Delta}\delta(\varepsilon)(\nu_\uparrow - \nu_\downarrow)^2.$$

One also can derive analogous results for the spin-orbit contribution in Eq. (16). Equation (16) then becomes

$$(\hbar\omega - \Delta)\langle \delta s_-(\omega)\rangle \approx \mathcal{F}(\omega)h_- + \frac{2i\pi\gamma\hbar^3}{9\Delta} h_-\omega\xi\beta^2[(\nu_\uparrow^2 k_{F\uparrow}^4 + \nu_\downarrow^2 k_{F\downarrow}^4) + 2\nu_\uparrow\nu_\downarrow k_{F\uparrow}^2 k_{F\downarrow}^2] + \frac{i\pi\gamma\hbar^3}{\Delta} h_-\omega[\xi_\perp(\nu_\uparrow^2 + \nu_\downarrow^2) + 2\xi_\parallel \nu_\uparrow\nu_\downarrow].$$

Using Eq. (5) to identify (the low frequency) Im χ_{-+}, we find from Eq. (7) that the Gilbert damping is given by

$$\alpha = \frac{2\pi\hbar}{9s_0}\xi\beta^2[(\nu_\uparrow^2 k_{F\uparrow}^4 + \nu_\downarrow^2 k_{F\downarrow}^4) + 2\nu_\uparrow\nu_\downarrow k_{F\uparrow}^2 k_{F\downarrow}^2] + \frac{\pi\hbar}{s_0}[\xi_\perp(\nu_\uparrow^2 + \nu_\downarrow^2) + 2\xi_\parallel\nu_\uparrow\nu_\downarrow], \quad (20)$$

which agrees with the diagrammatic calculation of Kohno *et al.*[25]

Next, we would like to relate the Gilbert damping constant in Eq. (20) to other physical quantities. Comparing a Bloch-Bloembergen[1,39] equation of motion for the magnetization vector with the corresponding LLG equation, we find for weak driving fields and small-angle magnetization dynamics that

$$\alpha = \frac{\hbar}{\Delta\tau_\perp}, \quad (21)$$

where, in our case,

$$\frac{1}{\tau_\perp} = \frac{1}{\tau_{\rm so}} + \frac{1}{\tau_m}.$$

Above, we have defined the effective transverse scattering rates from spin-orbit interaction and magnetic impurities, viz.,

$$\frac{1}{\tau_{\rm so}} = \frac{2\pi\Delta}{9s_0}\xi\beta^2[(\nu_\uparrow^2 k_{F\uparrow}^4 + \nu_\downarrow^2 k_{F\downarrow}^4) + 2\nu_\uparrow\nu_\downarrow k_{F\uparrow}^2 k_{F\downarrow}^2]$$

and

$$\frac{1}{\tau_m} = \frac{\pi\Delta}{s_0}[\xi_\perp(\nu_\uparrow^2 + \nu_\downarrow^2) + 2\xi_\parallel\nu_\uparrow\nu_\downarrow].$$

By comparison, the longitudinal spin-relaxation rate obtained from, e.g., Fermi's golden rule reads

$$\frac{1}{\tau_\parallel} = \frac{4\pi}{\hbar}(\nu_\uparrow + \nu_\downarrow)\left[\xi_\parallel + \frac{2}{9}\xi\beta^2 k_{F\uparrow}^2 k_{F\downarrow}^2\right].$$

For weak ferromagnets, the density of states and momentum at the Fermi energy are not strongly spin dependent, i.e., $\nu_s \simeq \nu_F$ and $k_{Fs} \simeq k_F$. Therefore, $2s_0 \approx \hbar\Delta\nu_F$, which implies equal transverse and longitudinal scattering rates for impurity-induced spin-orbit interaction and *isotropic* magnetic impurity scattering, i.e., $\tau_\perp = \tau_\parallel$.[25]

Since we succeeded in reproducing the general diagrammatic result of Ref. 25, we also identified the necessary measure to transcend the semiclassical treatment of Ref. 1. Most important are the gradient corrections to the spectral function, but in the presence of anisotropically distributed magnetic impurities, gradient corrections to the self-energies should be included as well.

In a metal, the longitudinal spin-orbit induced scattering time depends on the spin-conserving elastic scattering time. Experimentally, one typically finds that the ratio of spin-conserving to non-spin-conserving scattering events, $\epsilon = \tau/\tau_\parallel$, is not very sensitive to the concentration of impurities.[21] This means that in systems where spin-orbit induced dephasing dominates the Gilbert damping, α is proportional to the resistivity of the system,

$$\alpha = \frac{\hbar n e^2}{2\Delta m}\rho\epsilon,$$

where n is the electron concentration and ρ is the resistivity. A linear relation between Gilbert damping and resistivity was found in a recent experimental study of electronic transport in thin Permalloy films by Ingvarsson *et al.*[40]

The present discussion is mainly focused on the low-temperature regime, but the linear dependence on the scattering rates suggests that the damping should increase with in-

creasing temperatures. It is experimentally known that ϵ is not very sensitive to variations in temperature.[41] Consequently, we expect that the Gilbert damping constant is proportional to the resistivity also at higher temperatures.

Since the Gilbert damping coefficient is proportional to Δ^{-1}, dissipation is reduced in the strong ferromagnet limit. Since the damping in Eq. (21) depends on the transverse spin dephasing rate, Gilbert damping does not vanish in half metals in which the chemical potential falls below the band edge of one of the spin bands.

As mentioned in Sec. II, we introduced the simplifying assumption of free electrons with a parabolic band structure. Whereas our qualitative results are not affected by this choice, realistic band structures will introduce several complications such as wave vector and band-index dependent exchange splittings and scattering rates. Effectively, the splitting and scattering rates found here might have to be replaced by complicated Brillouin-zone integrals. Generalizations in this direction have our full attention.

The form of the damping coefficient found in Eqs. (20) and (21) agrees with previous studies on spin-flip mechanisms for magnetization damping. Nearly four decades ago, Heinrich *et al.*[8] suggested, based on the *s-d* exchange interaction between localized *d* electrons and itinerant *s* electrons, that electron-hole pairs could be excited by magnons. Assuming that the exchange splitting is much larger than the spin-flip rate and denoting the fraction of the total spin carried by the delocalized electrons by $\eta<1$, they found $\alpha = \eta\hbar^2\nu_F/2s_0\tau_{\mathrm{sf}}$ in the long wavelength limit. The result is expressed in terms of τ_{sf}, a phenomenological electron-hole pair lifetime. This result can be compared with Eqs. (20) and (21) by using the approximation $2s_0 \approx \hbar\Delta\nu_F$. We see that when the magnetization is mainly carried by the *d* electrons, which are not affected by spin-flip scattering, the predicted damping in the *s-d* model is much weaker than in our Stoner model.

In order to progress the field, the relation between, e.g., impurity doping species and densities with corresponding spin-flip diffusion lengths and Gilbert damping in ferromagnetic metals and semiconductors should be carried out. Systematic studies in this direction will be of great importance in order to verify any future theoretical predictions based on realistic disorder potentials and band structures.

V. CONCLUSION

In conclusion, we present a kinetic equation for the distribution matrix of itinerant ferromagnets in the adiabatic local-density approximation. The spin susceptibility and Gilbert damping constant are obtained microscopically for a homogeneous ferromagnet by the Keldysh Green's function formalism. Magnetization damping arises from magnetic disorder in the ferromagnet, and we have shown that it is important to keep all terms to linear order in the Poisson brackets to obtain the correct result in the presence of impurity-induced spin-orbit interaction magnetic disorder. The Gilbert coefficient can be expressed in terms of an effective transverse spin dephasing rate that has been introduced earlier as a phenomenological constant.[1] Our framework can be generalized to handle first-principles band-structure calculations for specific types of impurities and disorder. We hope that our work will stimulate more systematic studies of electron transport and Gilbert damping as a function of material parameters.

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APPENDIX: SPECTRAL FUNCTION

In this section, we derive the spectral function to first order in the Poisson brackets in the presence of a weak, time-dependent transverse driving field $\boldsymbol{h}(T)$. Since the spectral functions appearing in Eq. (16) are already proportional to scattering rates, we will not keep gradient terms involving self-energies in the following derivation.

To proceed, we consider $\hat{G}^R$, which is determined from the relation

$$\hat{G}^R \otimes [\hat{G}^R]^{-1} = \hat{1},$$

where $[\hat{G}^R]^{-1} = \hat{G}_0^{-1} - \hat{\Sigma}^R$ and $\hat{G}_0^{-1}$ is the inverse Green's function in the clean limit, viz., Eq. (10). To first order in the Poisson brackets, one can show that in the Wigner representation,

$$\hat{G}^R(T;\boldsymbol{k},\varepsilon) = \left(\hat{1} - \frac{i}{2}[\hat{G}_0^R,(\hat{G}^R)^{-1}]_p\right)\hat{G}_0^R + \mathcal{O}([\cdots]_p^2),$$

where now

$$\hat{G}_0^R(T;\boldsymbol{k},\varepsilon) = \frac{1}{\det[(\hat{G}^R)^{-1}]}\begin{pmatrix} \varepsilon - \varepsilon_k + \frac{1}{2}\Delta - \Sigma_{22}^R & \frac{1}{2}\gamma\hbar h_- + \Sigma_-^R \\ \frac{1}{2}\gamma\hbar h_+ + \Sigma_+^R & \varepsilon - \varepsilon_k - \frac{1}{2}\Delta - \Sigma_{11}^R \end{pmatrix},$$

with $\det[\cdots]$ denoting a matrix determinant, is simply the inverted retarded Green's function to zeroth order in the Poisson brackets. Matrix manipulations result in

$$\hat{G}^R(T;k,\varepsilon) \approx \hat{G}_0^R(T;k,\varepsilon) - \frac{i\gamma\hbar^2}{\Delta}G_{0,z}^R(k,\varepsilon)^2\hat{\sigma}_z\partial_T(\boldsymbol{h}\cdot\hat{\boldsymbol{\sigma}}), \tag{A1}$$

where once more we have used the convenient matrix notation introduced in Eq. (15).

A similar relation can also be found for $\hat{G}^A$, and we finally use that $\hat{A} = i(\hat{G}^R - \hat{G}^A)$ to obtain an expression for the spectral function linear in gradients, viz.,

$$\hat{A}(T;k,\varepsilon) = \hat{A}_0(T;k,\varepsilon) + \frac{\gamma\hbar^2}{\Delta}[G_{0,z}^R(k,\varepsilon)^2 - G_{0,z}^A(k,\varepsilon)^2]\hat{\sigma}_z\partial_T(\boldsymbol{h}\cdot\hat{\boldsymbol{\sigma}}),$$

where

$$\hat{A}_0(T;k,\varepsilon) = \pi \sum_s \delta(\varepsilon - \varepsilon_{ks}) \left(\hat{1} + s\hat{\sigma}_z + s\frac{\gamma\hbar \boldsymbol{h}\cdot\hat{\boldsymbol{\sigma}}}{\Delta} \right)$$

is the spectral function to zeroth order in gradients. Using that, we can rewrite

$$G^R_{0,z}(k,\varepsilon)^2 - G^A_{0,z}(k,\varepsilon)^2 = \frac{i}{\Delta} A_z(k,\varepsilon) + \frac{i}{2}\partial_\varepsilon A_d(k,\varepsilon),$$

and we find the spectral function

$$\hat{A}(T;k,\varepsilon) = \hat{A}_0(T;k,\varepsilon) + \frac{i\gamma\hbar^2}{\Delta^2} \times \left(A_z(k,\varepsilon) + \frac{\Delta}{2}\partial_\varepsilon A_d(k,\varepsilon) \right) \hat{\sigma}_z \partial_T (\boldsymbol{h}\cdot\hat{\boldsymbol{\sigma}}). \tag{A2}$$

Hence, the weak, transverse driving field induces off-diagonal gradient contributions to the spectral function. These prove to be essential in order to correctly capture the transverse magnetization dynamics.

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