



Relativistic theory of magnetic inertia in ultrafast spin dynamics

Ritwik Mondal,^{*} Marco Berritta, Ashis K. Nandy, and Peter M. Oppeneer

Department of Physics and Astronomy, Uppsala University, P.O. Box 516, SE-75120 Uppsala, Sweden

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The influence of possible magnetic inertia effects has recently drawn attention in ultrafast magnetization dynamics and switching. Here we derive rigorously a description of inertia in the Landau-Lifshitz-Gilbert equation on the basis of the Dirac-Kohn-Sham framework. Using the Foldy-Wouthuysen transformation up to the order of $1/c^4$ gives the intrinsic inertia of a pure system through the second order time derivative of magnetization in the dynamical equation of motion. Thus, the inertial damping \mathcal{I} is a higher order spin-orbit coupling effect, $\sim 1/c^4$, as compared to the Gilbert damping Γ that is of order $1/c^2$. Inertia is therefore expected to play a role only on ultrashort timescales (subpicoseconds). We also show that the Gilbert damping and inertial damping are related to one another through the imaginary and real parts of the magnetic susceptibility tensor, respectively.

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I. INTRODUCTION

The foundation of contemporary magnetization dynamics is the Landau-Lifshitz-Gilbert (LLG) equation which describes the precession of spin moment and a transverse damping of it, while keeping the modulus of magnetization vector fixed [1–3]. The LLG equation of motion was originally derived phenomenologically and the damping of spin motion has been attributed to relativistic effects such as the spin-orbit interaction [1,4–6]. In recent years there has been a flood of proposals for the fundamental microscopic mechanism behind the Gilbert damping: the breathing Fermi surface model of Kamberský, where the damping is due to magnetization precession and the effect of spin-orbit interaction at the Fermi surface [4], the extension of the breathing Fermi surface model to the torque-torque correlation model [5,7], scattering theory description [8], effective field theories [9], linear response formalism within relativistic electronic structure theory [10], and the Dirac Hamiltonian theory formulation [11].

For practical reasons it was needed to extend the original LLG equation to include several other mechanisms [12,13]. To describe, e.g., current induced spin-transfer torques, the effects of spin currents have been taken into account [14–16], as well as spin-orbit torques [17] and the effect of spin diffusion [18]. A different kind of spin relaxation due to the exchange field has been introduced by Bar'yakhtar *et al.* [19]. In the Landau-Lifshitz-Bar'yakhtar equation nonlocal spin dissipations originate from the spatial dispersion of exchange effects through the second order space derivative of the effective field [20,21]. A further recent work predicts the existence of extension terms that contain spatial as well as temporal derivatives of the local magnetization [22].

Another term, not discussed in the above investigations, is the magnetic inertial damping that has recently drawn attention [23–26]. Originally, magnetic inertia was discussed following the discovery of earth's magnetism [27]. Within the LLG framework, inertia is introduced as an additional term [9,24,28,29] leading to a modified LLG equation,

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} + \mathbf{M} \times \left(\Gamma \frac{\partial \mathbf{M}}{\partial t} + \mathcal{I} \frac{\partial^2 \mathbf{M}}{\partial t^2} \right), \quad (1)$$

where Γ is the Gilbert damping constant [1–3], γ the gyromagnetic ratio, \mathbf{H}_{eff} the effective magnetic field, and \mathcal{I} is the inertia of the magnetization dynamics, similar to the mass in Newton's equation. This type of motion has the same classical analog as the nutation of a spinning symmetric top. The potential importance of inertia is illustrated in Fig. 1. While Gilbert damping slowly aligns the precessing magnetization to the effective magnetic field, inertial dynamics causes a trembling or nutation of the magnetization vector [24,30,31]. Nutation could consequently pull the magnetization toward the equator and cause its switching to the antiparallel direction [32,33], while depending crucially on the strength of the magnetic inertia. The parameter \mathcal{I} that characterizes the nutation motion is in the most general case a tensor and has been associated with the magnetic susceptibility [29,31,33]. Along a different line of reasoning, Fähnle *et al.* extended the breathing Fermi surface model to include the effect of magnetic inertia [9,34]. The technological importance of nutation dynamics is thus its potential to steer magnetization switching in memory devices [23–25,32] and also in skyrmionic spin textures [35]. Magnetization dynamics involving inertial dynamics has been investigated recently, and it was suggested that its dynamics belongs to the faster time scales [24,26], i.e., the femtosecond regime. However, the origin of inertial damping from a fundamental framework is still missing, and, moreover, although it is possible to vary the size of the inertia in spin-dynamics simulations, it is unknown what the typical size of the inertial damping is.

Naturally the question arises whether it is possible to derive the extended LLG equation including inertia while starting from the fully relativistic Dirac equation. Hickey and Moodera [36] started from a Dirac Hamiltonian and obtained an intrinsic Gilbert damping term which originated from spin-orbit coupling. However they started from only a part of the spin-orbit coupling Hamiltonian which was anti-Hermitian [37,38]. A recent derivation based on Dirac Hamiltonian theory formulation [11] showed that the Gilbert damping depends strongly on both interband and intraband transitions (consistent with Ref. [39]) as well as the magnetic susceptibility response function, χ_m . This derivation used the relativistic expansion to the lowest order $1/c^2$ of the Hermitian Dirac-Kohn-Sham (DKS) Hamiltonian including the effect of exchange field [40].

^{*}ritwik.mondal@physics.uu.se

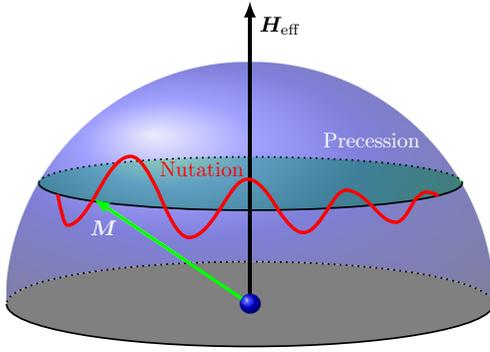


FIG. 1. Schematic illustration of magnetization dynamics. The precessional motion of \mathbf{M} around \mathbf{H}_{eff} is depicted by the blue solid-dashed curve, and the nutation is shown by the red curve.

In this paper we follow an approach similar to that of Ref. [11], but we consider higher order expansion terms of the DKS Hamiltonian up to the order of $1/c^4$. This is shown to lead to the intrinsic inertia term in the modified LLG equation and demonstrates that it stems from a higher-order spin-orbit coupling term. A relativistic origin of the spin nutation angle, caused by Rashba-like spin-orbit coupling, was previously concluded, too, in the context of semiconductor nanostructures [41,42].

In the following, we derive in Sec. II the relativistic correction terms to the extended Pauli Hamiltonian up to the order of $1/c^4$, which includes the spin-orbit interaction and an additional term. Then the corresponding magnetization dynamics is computed from the obtained spin Hamiltonian in Sec. III, which is shown to contain the Gilbert damping and the magnetic inertial damping. Finally, we discuss the size of the magnetic inertia in relation to other earlier studies.

II. RELATIVISTIC HAMILTONIAN FORMULATION

We start our derivation with a fully relativistic particle, a Dirac particle [43] inside a material and in the presence of an external field, for which we write the DKS Hamiltonian:

$$\begin{aligned} \mathcal{H} &= c\boldsymbol{\alpha} \cdot (\mathbf{p} - e\mathbf{A}) + (\underline{\beta} - \underline{\mathbb{1}})mc^2 + V\underline{\mathbb{1}} \\ &= \mathcal{O} + (\underline{\beta} - \underline{\mathbb{1}})mc^2 + \mathcal{E}, \end{aligned} \quad (2)$$

where V is the effective crystal potential created by the ion-ion, ion-electron, and electron-electron interactions. In general, there can be an additional potential term for the DKS exchange field, however, we suppress to write it explicitly here (see Ref. [11] for details). $\mathbf{A}(\mathbf{r}, t)$ is the magnetic vector potential from the external field, c is the speed of light, m is the particle's mass, and $\underline{\mathbb{1}}$ is the 4×4 unit matrix. $\boldsymbol{\alpha}$ and $\underline{\beta}$ are the Dirac matrices that have the form

$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix}, \quad \underline{\beta} = \begin{pmatrix} \mathbf{1} & 0 \\ 0 & -\mathbf{1} \end{pmatrix},$$

where $\boldsymbol{\sigma}$ is the Pauli spin matrix vector and $\mathbf{1}$ is 2×2 unit matrix. The Dirac equation is then written as $i\hbar \frac{\partial \psi(\mathbf{r}, t)}{\partial t} = \mathcal{H}\psi(\mathbf{r}, t)$ for a Dirac bispinor ψ . The quantity $\mathcal{O} = c\boldsymbol{\alpha} \cdot (\mathbf{p} - e\mathbf{A})$ defines the off-diagonal or odd terms in the matrix formalism, and $\mathcal{E} = V\underline{\mathbb{1}}$ are the diagonal, i.e., even terms. The latter have

to be multiplied by a 2×2 block diagonal unit matrix in order to bring them in a matrix form. To obtain the nonrelativistic Hamiltonian and the relativistic corrections one can write down the Dirac bispinor in double two component form as

$$\psi(\mathbf{r}, t) = \begin{pmatrix} \phi(\mathbf{r}, t) \\ \eta(\mathbf{r}, t) \end{pmatrix},$$

and substitute those into the Dirac equation. The upper two components represent the particle, and the lower two components represent the antiparticle. However the question of separating the particle's and antiparticle's wave functions is not clear for any given momentum. As the part $\boldsymbol{\alpha} \cdot \mathbf{p}$ is off-diagonal in the matrix formalism, it retains the odd components and thus links the particle-antiparticle wave functions. One way to eliminate the antiparticle's wave function is by an exact transformation [44] which gives terms that require a further expansion in powers of $1/c^2$. Another way is to search for a representation where the odd terms become smaller and smaller, and one can ignore those with respect to the even terms and retain only the latter [45]. The Foldy-Wouthuysen (FW) transformation [46,47] was the very successful attempt to find such a representation.

It is a unitary transformation obtained by suitably choosing the FW operator,

$$U_{\text{FW}} = -\frac{i}{2mc^2} \underline{\beta} \mathcal{O}. \quad (3)$$

The minus sign in front of the operator is because $\underline{\beta}$ and \mathcal{O} anticommute with each other. The transformation of the wave function adopts the form $\psi'(\mathbf{r}, t) = e^{iU_{\text{FW}}} \psi(\mathbf{r}, t)$ such that the probability density remains the same, $|\psi|^2 = |\psi'|^2$. The time-dependent FW transformation can be expressed as [45,48]

$$\mathcal{H}_{\text{FW}} = e^{iU_{\text{FW}}} \left(\mathcal{H} - i\hbar \frac{\partial}{\partial t} \right) e^{-iU_{\text{FW}}} + i\hbar \frac{\partial}{\partial t}. \quad (4)$$

The first term can be expanded in a series as

$$\begin{aligned} e^{iU_{\text{FW}}} \mathcal{H} e^{-iU_{\text{FW}}} &= \mathcal{H} + i[U_{\text{FW}}, \mathcal{H}] + \frac{i^2}{2!} [U_{\text{FW}}, [U_{\text{FW}}, \mathcal{H}]] + \dots \\ &+ \frac{i^n}{n!} [U_{\text{FW}}, [U_{\text{FW}}, \dots [U_{\text{FW}}, \mathcal{H}] \dots]] + \dots \end{aligned} \quad (5)$$

The time dependency enters through the second term of Eq. (4) and for a time-independent transformation one works with $\frac{\partial U_{\text{FW}}}{\partial t} = 0$. It is instructive to note that the aim of the whole procedure is to make the odd terms smaller and one can notice that as it goes higher and higher in the expansion, the corresponding coefficients decrease of the order $1/c^2$ due to the choice of the unitary operator. After a first transformation, the new Hamiltonian will contain new even terms, \mathcal{E}' , as well as new odd terms, \mathcal{O}' of $1/c^2$ or higher. The latter terms can be used to perform a next transformation having the new unitary operator as $U'_{\text{FW}} = -\frac{i}{2mc^2} \underline{\beta} \mathcal{O}'$. After a second transformation the new Hamiltonian \mathcal{H}'_{FW} is achieved that has the odd terms of the order $1/c^4$ or higher. The transformation is a repetitive process, and it continues until the separation of positive and negative energy states is guaranteed.

After a fourth transformation we derive the new transformed Hamiltonian with all the even terms that are correct up

to the order of $\frac{1}{m^3 c^6}$ as [48–50]

$$\begin{aligned} \mathcal{H}_{\text{FW}}''' = & (\underline{\beta} - \underline{1})mc^2 + \underline{\beta} \left(\frac{\mathcal{O}^2}{2mc^2} - \frac{\mathcal{O}^4}{8m^3 c^6} \right) + \mathcal{E} \\ & - \frac{1}{8m^2 c^4} [\mathcal{O}, [\mathcal{O}, \mathcal{E}] + i\hbar \partial_t \mathcal{O}] \\ & + \frac{\underline{\beta}}{16m^3 c^6} \{\mathcal{O}, [[\mathcal{O}, \mathcal{E}], \mathcal{E}]\} + \frac{\underline{\beta}}{8m^3 c^6} \{\mathcal{O}, [i\hbar \partial_t \mathcal{O}, \mathcal{E}]\} \\ & + \frac{\underline{\beta}}{16m^3 c^6} \{\mathcal{O}, (i\hbar)^2 \partial_{tt} \mathcal{O}\}. \end{aligned} \quad (6)$$

Here $\partial_t \equiv \partial/\partial t$ stands for the first-order time derivative. Note that $[A, B]$ defines the commutator, while $\{A, B\}$ represents the anticommutator for any two operators A and B . A similar Foldy-Wouthuysen transformation Hamiltonian up to an order of $1/m^3 c^6$ was derived by Hirschberger and Hervieux in their recent work [51], however there are some differences, for example, the first and second terms in the third line of Eq. (6) were not given. Once we have the transformed Hamiltonian as a function of odd and even terms, the final form is achieved by substituting the correct form of odd terms \mathcal{O} and calculating term by term.

Evaluating all the terms separately, we derive the Hamiltonian for only the positive energy solutions, i.e., the upper components of the Dirac bispinor as a 2×2 matrix formalism [40,51,52]:

$$\begin{aligned} \mathcal{H}_{\text{FW}}''' = & \frac{(\mathbf{p} - e\mathbf{A})^2}{2m} + V - \frac{e\hbar}{2m} \boldsymbol{\sigma} \cdot \mathbf{B} - \frac{(\mathbf{p} - e\mathbf{A})^4}{8m^3 c^2} \\ & - \frac{e\hbar^2}{8m^2 c^2} \nabla \cdot \mathbf{E}_{\text{tot}} + \frac{e\hbar}{8m^3 c^2} \{(\mathbf{p} - e\mathbf{A})^2, \boldsymbol{\sigma} \cdot \mathbf{B}\} \\ & - \frac{e\hbar}{8m^2 c^2} \boldsymbol{\sigma} \cdot [\mathbf{E}_{\text{tot}} \times (\mathbf{p} - e\mathbf{A}) - (\mathbf{p} - e\mathbf{A}) \times \mathbf{E}_{\text{tot}}] \\ & - \frac{e\hbar^2}{16m^3 c^4} \{(\mathbf{p} - e\mathbf{A}), \partial_t \mathbf{E}_{\text{tot}}\} - \frac{i e \hbar^2}{16m^3 c^4} \boldsymbol{\sigma} \\ & \cdot [\partial_t \mathbf{E}_{\text{tot}} \times (\mathbf{p} - e\mathbf{A}) + (\mathbf{p} - e\mathbf{A}) \times \partial_t \mathbf{E}_{\text{tot}}]. \end{aligned} \quad (7)$$

The higher order terms ($1/c^6$ or more) will involve similar formulations and more and more time derivatives of the magnetic and electric fields will appear that stem from the time derivative of the odd operator \mathcal{O} [48,51]. The fields in the last Hamiltonian (7) are defined as $\mathbf{B} = \nabla \times \mathbf{A}$, the external magnetic field, $\mathbf{E}_{\text{tot}} = \mathbf{E}_{\text{int}} + \mathbf{E}_{\text{ext}}$ are the electric fields where $\mathbf{E}_{\text{int}} = -\frac{1}{c} \nabla V$ is the internal field that exists even without any perturbation and $\mathbf{E}_{\text{ext}} = -\frac{\partial \mathbf{A}}{\partial t}$ is the external field (only the temporal part is retained here because of the Coulomb gauge).

A. The spin Hamiltonian

The aim of this work is to formulate the magnetization dynamics on the basis of this Hamiltonian. Thus, we split the Hamiltonian into spin-independent and spin-dependent parts and consider from now on electrons. The spin Hamiltonian is straightforwardly given as

$$\begin{aligned} \mathcal{H}^S(t) = & -\frac{e}{m} \mathbf{S} \cdot \mathbf{B} + \frac{e}{4m^3 c^2} \{(\mathbf{p} - e\mathbf{A})^2, \mathbf{S} \cdot \mathbf{B}\} \\ & - \frac{e}{4m^2 c^2} \mathbf{S} \cdot [\mathbf{E}_{\text{tot}} \times (\mathbf{p} - e\mathbf{A}) - (\mathbf{p} - e\mathbf{A}) \times \mathbf{E}_{\text{tot}}] \end{aligned}$$

$$\begin{aligned} & - \frac{i e \hbar}{8m^3 c^4} \mathbf{S} \cdot [\partial_t \mathbf{E}_{\text{tot}} \times (\mathbf{p} - e\mathbf{A}) \\ & + (\mathbf{p} - e\mathbf{A}) \times \partial_t \mathbf{E}_{\text{tot}}], \end{aligned} \quad (8)$$

where the spin operator $\mathbf{S} = (\hbar/2) \boldsymbol{\sigma}$ has been used. Let us briefly explain the physical meaning behind each term that appears in $\mathcal{H}^S(t)$. The first term defines the Zeeman coupling of the electron's spin with the externally applied magnetic field. The second term defines an indirect coupling of light to the Zeeman interaction of spin and the optical \mathbf{B} field, which can be shown to have the form of a relativistic Zeeman-like term. The third term implies a general form of the spin-orbit coupling that is gauge invariant [53], and it includes the effect of the electric field from an internal as well as an external field. The last term is the new term of relevance here that has only been considered once in the literature by Hirschberger *et al.* [51]. Note that, although the last term in Eq. (8) contains the total electric field, only the time derivative of the external field plays a role here, because the time derivative of internal field is zero as the ionic potential is time independent. In general if one assumes a plane-wave solution of the electric field in Maxwell's equation as $\mathbf{E} = \mathbf{E}_0 e^{i\omega t}$, the last term can be written as $\frac{e\hbar\omega}{8m^3 c^4} \mathbf{S} \cdot (\mathbf{E} \times \mathbf{p})$ and thus adopts the form of a higher-order spin-orbit coupling for a general E field.

The spin-dependent part can be easily rewritten in a shorter format using the identities:

$$\begin{aligned} \mathcal{A} \times (\mathbf{p} - e\mathbf{A}) - (\mathbf{p} - e\mathbf{A}) \times \mathcal{A} = & 2\mathcal{A} \times (\mathbf{p} - e\mathbf{A}) \\ & + i\hbar \nabla \times \mathcal{A} \end{aligned} \quad (9)$$

$$\mathcal{A} \times (\mathbf{p} - e\mathbf{A}) + (\mathbf{p} - e\mathbf{A}) \times \mathcal{A} = -i\hbar \nabla \times \mathcal{A} \quad (10)$$

for any operator \mathcal{A} . This allows us to write the spin Hamiltonian as

$$\begin{aligned} \mathcal{H}^S = & -\frac{e}{m} \mathbf{S} \cdot \mathbf{B} + \frac{e}{2m^3 c^2} \mathbf{S} \cdot \mathbf{B} \left[p^2 - 2e\mathbf{A} \cdot \mathbf{p} + \frac{3e^2}{2} A^2 \right] \\ & - \frac{e}{2m^2 c^2} \mathbf{S} \cdot [\mathbf{E}_{\text{tot}} \times (\mathbf{p} - e\mathbf{A})] + \frac{i e \hbar}{4m^2 c^2} \mathbf{S} \cdot \partial_t \mathbf{B} \\ & + \frac{e\hbar^2}{8m^3 c^4} \mathbf{S} \cdot \partial_{tt} \mathbf{B}. \end{aligned} \quad (11)$$

Here, the Maxwell's equations have been used to derive the final form that the spatial derivative of the electric field will generate a time derivative of a magnetic field such that $\nabla \times \mathbf{E}_{\text{ext}} = -\frac{\partial \mathbf{B}}{\partial t}$, while the curl of an internal field results in zero as the curl of a gradient function is always zero. The final spin Hamiltonian (11) bears much importance for the strong laser field-matter interaction as it takes into account all the field-spin coupling terms. It is thus the appropriate fundamental Hamiltonian to understand the effects of those interactions on the magnetization dynamics described in Sec. III.

B. Single Dirac particle spin dynamics

Although the dynamics of a macroscopic magnetization is important for many technological applications, the dynamics of a single spin- $\frac{1}{2}$ Dirac particle is of fundamental value in its own right. Assuming that the electron's spin does not explicitly depend on time, the single spin dynamics in the Heisenberg

picture reduces to

$$\frac{\partial \mathbf{S}}{\partial t} = \frac{1}{i\hbar} [\mathbf{S}, \mathcal{H}^S(t)]. \quad (12)$$

The derivation of the ensuing spin dynamics is then straightforward, substituting the Hamiltonian terms given in (11) in the equation of motion and carrying out the different commutators. We use the commutator algebra of two spins $[S_j, S_k] = i\hbar \epsilon_{jkl} S_l$. The spin dynamics of a single Dirac particle becomes

$$\begin{aligned} \frac{\partial \mathbf{S}}{\partial t} = & \frac{e}{m} \mathbf{S} \times \mathbf{B} - \frac{e}{2m^3 c^2} \mathbf{S} \times \mathbf{B} \left[p^2 - 2e\mathbf{A} \cdot \mathbf{p} + \frac{3e^2}{2} A^2 \right] \\ & + \frac{e}{2m^2 c^2} \mathbf{S} \times [\mathbf{E}_{\text{tot}} \times (\mathbf{p} - e\mathbf{A})] \\ & - \frac{ie\hbar}{4m^2 c^2} \mathbf{S} \times \partial_t \mathbf{B} - \frac{e\hbar^2}{8m^3 c^4} \mathbf{S} \times \partial_{tt} \mathbf{B}. \end{aligned} \quad (13)$$

This is an insightful result describing the relativistic (up to the order of $1/c^4$) spin dynamics of a single electron. Even though this is given as spin angular momentum operator dynamics, it can be recognized, first, that the dynamics contains a spin precession (first two terms), stemming from the common nonrelativistic precession (first term) and a relativistic correction to it (second term). Note that there is no exchange field for a single spin- $\frac{1}{2}$ particle. The third term describes the dynamics due to the conventional spin-orbit interaction and the spin-orbit torque due to the applied electromagnetic field. The fourth term containing the first-order time derivative of the magnetic field causes the transversal relaxation of spins (reminiscent of the Gilbert damping). The last term, which contains the second-order time derivative of \mathbf{B} leads to nutation (see below) and thus establishes the existence of nutation even for a single spin. The obtained single spin dynamics thus contains precession, damping, electromagnetic field torque, and nutation and should be valid for any Dirac spin- $\frac{1}{2}$ particle under the influence of an external electromagnetic field (cf. Refs. [54,55]).

III. MAGNETIZATION DYNAMICS

In general, magnetization is given by the magnetic moment per volume element in a magnetic solid. Our next goal is to derive the dynamics of such a magnetization element. We work here in the framework of the DKS Hamiltonian that can be seen as an effective collective Hamiltonian describing all the electrons in a system. The general equation for the expectation value of an observable O is $\langle O \rangle = \text{Tr}[\rho O]$, where ρ is the density matrix. Considering that we are working in the Heisenberg picture, the density matrix does not evolve with time, so we can assume it to be the (diagonal) density matrix, which in the energy eigenstate representation adopts the form:

$$\rho_{nk;nk}(\mathbf{r}) = f(E_{nk}) \psi_{nk}^*(\mathbf{r}) \psi_{nk}(\mathbf{r}), \quad (14)$$

where $f(E_{nk})$ is the Fermi-Dirac distribution, $\psi_{nk}(\mathbf{r})$ are eigenstates of the DKS Hamiltonian in coordinate representation, and E_{nk} are the corresponding n th band electronic energies with momentum \mathbf{k} . Here we are focusing on the magnetization of some small volume, which can be written

as an expectation of the collective spin of the electrons, as

$$\mathbf{M}(t) = \frac{g\mu_B}{\Omega} \int_{\Omega} d\mathbf{r} \text{Tr}_{\sigma} [\mathbf{S} \rho(\mathbf{r})], \quad (15)$$

where Ω is a suitably chosen volume, e.g., the unit cell volume. At this point we can make a partition of the unit volume of the considered material, for instance taking volume elements Ω_j enclosing the individual atoms of the unit cell. In this way we can split the integral in Eq. (15) in different volume elements and obtain information on the magnetization localized on each individual atom of the unit cell. We can thus write:

$$\mathbf{M}(t) = \sum_j \frac{g\mu_B}{\Omega} \int_{\Omega_j} d\mathbf{r} \text{Tr}_{\sigma} [\mathbf{S} \rho(\mathbf{r})] = \sum_j \frac{g\mu_B}{\Omega} \langle \mathbf{S}^j \rangle. \quad (16)$$

Next, we introduce a coarse graining for the macroscopic material, where the spacial coordinate is associated with the position of one of the unit cells or atomic volumes at position \mathbf{R}_j , i.e., $\mathbf{M}(\mathbf{r}, t) = \mathbf{M}(t)|_{\mathbf{r}=\mathbf{R}_j}$.

To derive the dynamics, we take the time derivative on both sides of Eq. (16) and, employing the Heisenberg equation of motion, we arrive at the equation for the magnetization dynamics as [36,56,57]

$$\frac{\partial \mathbf{M}}{\partial t} = \sum_j \frac{g\mu_B}{\Omega} \frac{1}{i\hbar} \langle [\mathbf{S}^j, \mathcal{H}^S(t)] \rangle. \quad (17)$$

Now the task looks simple; one needs to substitute the spin Hamiltonian (11) and calculate the commutators in order to find the equation of motion. Note that the dynamics only considers the local dynamics as we have not taken into account the time derivative of the particle density operator (for details, see Ref. [11]). Incorporating the latter would give rise to local as well as nonlocal processes (i.e., spin currents) within the same footing.

The first term in the spin Hamiltonian produces the dynamics as

$$\frac{\partial \mathbf{M}^{(1)}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{B}, \quad (18)$$

where $\gamma = g|e|/2m$ defines the gyromagnetic ratio and the Landé g factor $g \approx 2$ for spins, the electronic charge $e < 0$. Using the linear relationship of magnetization with the magnetic field $\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M})$, the latter is replaced in Eq. (18) to get the usual form in the Landau-Lifshitz equation, $-\gamma_0 \mathbf{M} \times \mathbf{H}$, where $\gamma_0 = \mu_0 \gamma$ is the effective gyromagnetic ratio. This gives the Larmor precession of magnetization around an effective field \mathbf{H} . The effective field will always have a contribution from the exchange field (and the relativistic corrections to it), which has not been explicitly written out in this paper, as they are not in the focus here. For detailed calculations yet including the exchange field, see Ref. [11].

The second and third terms in the Hamiltonian (11) contain products of spin and orbital degrees of freedom. At this point it is important to notice that neither the spin nor the orbital degrees of freedom commute with the Hamiltonian due to the spin-orbit coupling and the $1/c^4$ corrections, which implies that the equilibrium density matrix ρ cannot be expressed exactly as $\rho = \rho_S \otimes \rho_O$ where ρ_S is the reduced density matrix for the spin degrees of freedom and ρ_O is the reduced density matrix of the orbital degrees of freedom. Considering

an observable O acting on the orbital degrees of freedom in Hilbert space (for instance the momentum or the orbital angular momentum or some function depending on them) and S the spin, due to the impossibility to separate orbital and spin parts of the density matrix we are not, in principle, allowed to write

$$\text{Tr}[\rho SO] = \text{Tr}[\rho_S S] \text{Tr}[\rho_O O] = \mathbf{M}\langle O \rangle. \quad (19)$$

It is important to realize that the nonseparability (entanglement) of the orbital and spin parts of the density matrix is due to the spin-orbit coupling and its corrections (since it prevents both quantities to be conserved). However, in ferromagnetic materials the energy separation of the spin states is mostly due to the exchange magnetic field which is orders of magnitude larger than the spin-orbit coupling and its corrections. As a consequence, the separation of the density matrix as a direct product of spin and orbital parts is a good approximation, therefore we can employ Eq. (19). Moreover, due to the large splitting of spin bands and the continuous smooth behavior of the energy levels as a function of momentum, the out-of-equilibrium dynamics on the latter degrees of freedom is faster than the dynamics of the spin degrees of freedom.

Using now the approximation (19), the second term in the spin Hamiltonian (11) will result in a relativistic correction to the magnetization precession. Within a uniform field approximation ($\mathbf{A} = \mathbf{B} \times \mathbf{r}/2$), the corresponding dynamics will take the form

$$\frac{\partial \mathbf{M}^{(2)}}{\partial t} = \frac{\gamma}{2m^2 c^2} \mathbf{M} \times \mathbf{B} \left(p^2 - e\mathbf{B} \cdot \mathbf{L} + \frac{3e^2}{8} (\mathbf{B} \times \mathbf{r})^2 \right), \quad (20)$$

with $\mathbf{L} = \mathbf{r} \times \mathbf{p}$ the orbital angular momentum. The presence of $\gamma/2m^2 c^2$ implies that the contribution of this dynamics to the precession is relatively small, while the leading precession dynamics is given by Eq. (18). For sake of completeness we note that a relativistic correction to the precession term of similar order $1/m^2 c^2$ was obtained previously for the exchange field [11].

The next term in the Hamiltonian is a bit tricky to handle as the third term in Eq. (11) is not Hermitian, not even the fourth term which is anti-Hermitian. However *together* they form a Hermitian Hamiltonian [11,37,38]. Therefore one has to work together with those terms and cannot only perform the dynamics with an individual term. In an earlier work [11] we have shown that taking a uniform magnetic field along with the gauge $\mathbf{A} = \frac{\mathbf{B} \times \mathbf{r}}{2}$ will preserve the Hermiticity. This uniform-field condition is usually fulfilled for thin-film samples, where the skin depth of the electromagnetic field is longer than the film thickness. For thicker samples a field that is uniform over a part of the sample could alternatively be introduced. The dynamical equation of spin motion with the second and third terms can thus be written in a compact form for harmonic applied fields as [11]

$$\frac{\partial \mathbf{M}^{(3,4)}}{\partial t} = \mathbf{M} \times \left(\mathbf{A} \cdot \frac{\partial \mathbf{M}}{\partial t} \right), \quad (21)$$

with the intrinsic Gilbert damping parameter A that is a tensor defined by

$$A_{ij} = \frac{\gamma \mu_0}{4m^2 c^2} \sum_{n,k} [\langle r_i p_k + p_k r_i \rangle - \langle r_n p_n + p_n r_n \rangle \delta_{ik}] \times (\mathbb{1} + \chi_m^{-1})_{kj}. \quad (22)$$

Here χ_m is the magnetic susceptibility tensor of rank 2 (a 3×3 matrix), $\mathbb{1}$ is the 3×3 unit matrix, and $\langle \dots \rangle$ stands for the expectation value with respect to the DKS electronic states ψ_{nk} . Note that for diagonal terms, i.e., $i = k$ the contributions from the expectation values of $r_k p_i$ cancel each other. The damping tensor can be decomposed and shown to have contributions from isotropic Heisenberg-like, anisotropic Ising-like, and Dzyaloshinskii-Moriya-like tensors, as detailed in the following. First, the damping tensor A_{ij} is decomposed into a symmetric and an antisymmetric part defined as $A_{ij}^{\text{sym}} = \frac{1}{2}(A_{ij} + A_{ji})$ and $A_{ij}^{\text{anti}} = \frac{1}{2}(A_{ij} - A_{ji})$. The symmetric tensor can further be written as $A_{ij}^{\text{sym}} = \mathbb{I}_{ij} + \alpha \delta_{ij}$, where α defines the isotropic diagonal components, i.e., the Heisenberg-like contribution, and \mathbb{I}_{ij} are the Ising-like contributions. Note that if the Heisenberg contributions are such that $\alpha = \frac{1}{3} \text{Tr}\{A_{ij}^{\text{sym}}\}$, the trace of the Ising-like contributions becomes zero, $\text{Tr}\{\mathbb{I}_{ij}\} = 0$. The antisymmetric matrix can be decomposed into a vector multiplied by the antisymmetric Levi-Civita tensor, $A_{ij}^{\text{anti}} = \epsilon_{ijk} \mathbf{D}_k$, which gives the Dzyaloshinskii-Moriya-like contribution. The complete damping dynamics can then be written as [11]

$$\begin{aligned} \frac{\partial \mathbf{M}^{(3,4)}}{\partial t} = & \alpha \mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t} + \mathbf{M} \times \left[\mathbb{I} \cdot \frac{\partial \mathbf{M}}{\partial t} \right] \\ & + \mathbf{M} \times \left[\mathbf{D} \times \frac{\partial \mathbf{M}}{\partial t} \right]. \end{aligned} \quad (23)$$

The antisymmetric Dzyaloshinskii-Moriya term has been shown to lead to a chiral damping [11]; experimental observations of such damping have been reported recently [58]. The other cross term having the form $\mathbf{E} \times \mathbf{A}$ in Eq. (11) is related to the angular momentum of the electromagnetic field and thus provides a torque on the spin that has been at the heart of angular magnetoelectric coupling [53]. This relativistic Hamiltonian providing spin-photon coupling has been shown recently [59] to explain the coherent ultrafast magnetism observed in pump-probe experiments [60]. A possible effect in spin dynamics including the light's angular momentum has been investigated in the strong field regime, and it has been shown that one has to include this cross term in the dynamics in order to explain the qualitative and quantitative strong field dynamics [55].

For the last term in the spin Hamiltonian (11) it is rather easy to formulate the spin dynamics because it is evidently Hermitian. Working out the commutator with the spins gives a contribution to the dynamics as

$$\frac{\partial \mathbf{M}^{(5)}}{\partial t} = \delta \mathbf{M} \times \frac{\partial^2 \mathbf{B}}{\partial t^2}, \quad (24)$$

with the constant $\delta = \frac{\gamma \hbar^2}{8m^2 c^4}$.

Let us work explicitly with the second-order time derivative of the magnetic induction by the relation $\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M})$,

using a chain rule for the derivative:

$$\begin{aligned}\frac{\partial^2 \mathbf{B}}{\partial t^2} &= \frac{\partial}{\partial t} \left(\frac{\partial \mathbf{B}}{\partial t} \right) = \mu_0 \frac{\partial}{\partial t} \left(\frac{\partial \mathbf{H}}{\partial t} + \frac{\partial \mathbf{M}}{\partial t} \right) \\ &= \mu_0 \left(\frac{\partial^2 \mathbf{H}}{\partial t^2} + \frac{\partial^2 \mathbf{M}}{\partial t^2} \right).\end{aligned}\quad (25)$$

This is a generalized equation for the time derivative of the magnetic induction which can be used even for nonharmonic fields. The magnetization dynamics is then given by

$$\frac{\partial \mathbf{M}^{(5)}}{\partial t} = \mu_0 \delta \mathbf{M} \times \left(\frac{\partial^2 \mathbf{H}}{\partial t^2} + \frac{\partial^2 \mathbf{M}}{\partial t^2} \right).\quad (26)$$

Thus the extended LLG equation of motion will have these two additional terms: (1) a field-derivative torque and (2) magnetization-derivative torque, and they appear with their second order time derivative. It deserves to be noted that, in a previous theory we also obtained a similar term—a field-derivative torque in first order-time derivative appearing in the generalized Gilbert damping. Specifically, the extended LLG equation for a general time-dependent field $\mathbf{H}(t)$ becomes

$$\begin{aligned}\frac{\partial \mathbf{M}}{\partial t} &= -\gamma_0 \mathbf{M} \times \mathbf{H} + \mathbf{M} \times \left[\bar{\mathbf{A}} \cdot \left(\frac{\partial \mathbf{H}}{\partial t} + \frac{\partial \mathbf{M}}{\partial t} \right) \right] \\ &+ \mu_0 \delta \mathbf{M} \times \left(\frac{\partial^2 \mathbf{H}}{\partial t^2} + \frac{\partial^2 \mathbf{M}}{\partial t^2} \right),\end{aligned}\quad (27)$$

where $\bar{\mathbf{A}}$ is a modified Gilbert damping tensor (for details, see Ref. [11]).

However for harmonic fields, the response of the ferromagnetic materials is measured through the differential susceptibility, $\chi_m = \partial \mathbf{M} / \partial \mathbf{H}$, because there exists a net magnetization even in the absence of any applied field. With this, the time derivative of the harmonic magnetic field can be further written as:

$$\begin{aligned}\frac{\partial^2 \mathbf{H}}{\partial t^2} &= \frac{\partial}{\partial t} \left(\frac{\partial \mathbf{H}}{\partial \mathbf{M}} \frac{\partial \mathbf{M}}{\partial t} \right) = \frac{\partial}{\partial t} \left(\chi_m^{-1} \cdot \frac{\partial \mathbf{M}}{\partial t} \right) \\ &= \frac{\partial \chi_m^{-1}}{\partial t} \cdot \frac{\partial \mathbf{M}}{\partial t} + \chi_m^{-1} \cdot \frac{\partial^2 \mathbf{M}}{\partial t^2}.\end{aligned}\quad (28)$$

In general the magnetic susceptibility is a spin-spin response function that, in reciprocal space, is wave-vector and frequency dependent, $\chi_m = \chi_m(\mathbf{q}, \omega)$. Substituting this expression in Eq. (26), the dynamics assumes the form with the first and second order time derivatives as

$$\frac{\partial \mathbf{M}^{(5)}}{\partial t} = \mathbf{M} \times \left(\mathcal{K} \cdot \frac{\partial \mathbf{M}}{\partial t} + \mathcal{I} \cdot \frac{\partial^2 \mathbf{M}}{\partial t^2} \right),\quad (29)$$

where the parameters $\mathcal{I}_{ij} = \mu_0 \delta (\mathbb{1} + \chi_m^{-1})_{ij}$ and $\mathcal{K}_{ij} = \mu_0 \delta \partial_t (\chi_m^{-1})_{ij}$ are tensors. The dynamics of the second term is that of the magnetic inertia that operates on shorter time scales [25].

Having all the required dynamical terms, finally the full magnetization dynamics can be written by joining together all the individual parts. Thus the full magnetization dynamics becomes, for harmonic fields,

$$\frac{\partial \mathbf{M}}{\partial t} = \mathbf{M} \times \left(-\gamma_0 \mathbf{H} + \Gamma \cdot \frac{\partial \mathbf{M}}{\partial t} + \mathcal{I} \cdot \frac{\partial^2 \mathbf{M}}{\partial t^2} \right).\quad (30)$$

Note that the Gilbert damping parameter Γ has two contributions, one from the susceptibility itself, A_{ij} , which is of order $1/c^2$ and an other from the time derivative of it, \mathcal{K}_{ij} of order $1/c^4$. Thus, $\Gamma_{ij} = A_{ij} + \mathcal{K}_{ij}$. However we will focus on the first one only as it will obviously be the dominant contribution, i.e., $\Gamma_{ij} \approx A_{ij}$. Even though we consider only the Gilbert damping term of order $1/c^2$ in the discussions, we shall explicitly analyze the other term of the order $1/c^4$. For an ac susceptibility, i.e., $\chi_m^{-1} \propto e^{i\omega t}$ we find that $\mathcal{K}_{ij} \propto \mu_0 \delta \partial_t (\chi_m^{-1})_{ij} \propto i \mu_0 \omega \delta \chi_m^{-1}$, which suggests again that the Gilbert damping parameter of the order $1/c^4$ will be given by the imaginary part of the susceptibility, $\mathcal{K}_{ij} \propto -\mu_0 \omega \delta \text{Im}(\chi_m^{-1})$.

The last equation (30) is the central result of this work, as it establishes a rigorous expression for the intrinsic magnetic inertia. Magnetization dynamics including inertia has been discussed in a few earlier articles [24,30,31,61]. The very last term in Eq. (30) has been associated previously with the inertial magnetization dynamics [32,62,63]. As mentioned, it implies a magnetization nutation, i.e., a changing of the precession angle as time progresses. Without the inertia term we obtain the well-known LLG equation of motion that has already been used extensively in magnetization dynamics simulations (see, e.g., Refs. [64–68]).

IV. DISCUSSIONS

Magnetic inertia was discussed first in relation to the earth's magnetism [27]. From a dimensional analysis, the magnetic inertia of a uniformly magnetized sphere undergoing uniform acceleration was estimated to be of the order of $1/c^2$ [27], which is consistent with the here-obtained relativistic nature of magnetic inertia.

The spin dynamics derived for a single Dirac particle [Eq. (13)] is a general and fundamental result, which establishes the existence of nutation even for any Dirac particle. To describe the magnetization dynamics of a small volume element, we introduce a collective macroscopic variable \mathbf{M} , stemming from the spin degrees of freedom, where the other degrees of freedom (e.g., electronic orbitals, environments) are averaged out.

The derived magnetization dynamics, based on the fundamental Dirac-Kohn-Sham Hamiltonian, provides explicit expressions for both the Gilbert and inertial dampings. Thus, a comparison can be made between the Gilbert damping parameter and the magnetic inertia parameter of a pure system. As noticed above, both the parameters depend on the magnetic susceptibility tensor, however it should be noted that the quantity $\langle r_\alpha p_\beta \rangle$ is imaginary itself, because [11]

$$\langle r_\alpha p_\beta \rangle = -\frac{i\hbar}{2m} \sum_{n,n',k} \frac{f(E_{nk}) - f(E_{n'k})}{E_{nk} - E_{n'k}} p_{nn'}^\alpha p_{n'n}^\beta.\quad (31)$$

Here the momentum matrix elements $p_{nn'}^\alpha$ are taken with respect to the states ψ_{nk} that follow from the DKS Hamiltonian (2) or (approximately) from Hamiltonian (6). The Gilbert damping parameter should consequently be given by the imaginary part of the susceptibility tensor [36,69]. On the other hand the magnetic inertia tensor must be given by the real part of the susceptibility [31]. This is in agreement with a recent

article where the authors also found the same dependence of real and imaginary parts of susceptibility to the nutation and Gilbert damping, respectively [33]. In our calculation, the Gilbert damping and inertia parameters adopt the following forms, respectively,

$$\begin{aligned}
\Gamma_{ij} &= \frac{i\gamma\mu_0}{4mc^2} \sum_{n,k} [\langle r_i p_k + p_k r_i \rangle - \langle r_n p_n + p_n r_n \rangle \delta_{ik}] \\
&\quad \times \text{Im}(\chi_m^{-1})_{kj} \\
&= -\frac{\mu_0\gamma\hbar}{4mc^2} \sum_{n,k} \left[\frac{\langle r_i p_k + p_k r_i \rangle - \langle r_n p_n + p_n r_n \rangle \delta_{ik}}{i\hbar} \right] \\
&\quad \times \text{Im}(\chi_m^{-1})_{kj} \\
&= -\zeta \sum_{n,k} \left[\frac{\langle r_i p_k + p_k r_i \rangle - \langle r_n p_n + p_n r_n \rangle \delta_{ik}}{i\hbar} \right] \\
&\quad \times \text{Im}(\chi_m^{-1})_{kj}, \tag{32} \\
\mathcal{I}_{ij} &= \frac{\mu_0\gamma\hbar^2}{8m^2c^4} [\mathbb{1} + \text{Re}(\chi_m^{-1})_{ij}] = \frac{\zeta\hbar}{2mc^2} [\mathbb{1} + \text{Re}(\chi_m^{-1})_{ij}], \tag{33}
\end{aligned}$$

with $\zeta \equiv \frac{\mu_0\gamma\hbar}{4mc^2}$. Note that the change of sign from damping tensor to the inertia tensor is also consistent with Ref. [33] and also that a factor of two is present in inertia. However, most importantly, the inertia tensor is \hbar/mc^2 times *smaller* than the damping tensor as is revealed in our calculations. Considering atomic units we can evaluate

$$\begin{aligned}
\zeta &\sim \frac{\mu_0}{4c^2} \sim \frac{0.00066}{4 \times 137^2} \sim 8.8 \times 10^{-9}, \\
\frac{\zeta\hbar}{2mc^2} &\sim \frac{\zeta}{2c^2} \sim \frac{8.8 \times 10^{-9}}{2 \times 137^2} \sim 2.34 \times 10^{-13}.
\end{aligned}$$

This implies that the intrinsic inertial damping is typically 4×10^4 times smaller than the Gilbert damping, and it is not an independently variable parameter. Also, because of its smallness magnetic inertial dynamics will be more significant on shorter timescales [24,26]. As recently outlined by Wegrowe and Olive the quite different time scales of Gilbert and inertial dampings can be exploited to study the effect of the fast inertial motion on the slower precessional motion [26].

A further analysis of the Gilbert and inertial parameters can be made. One can use the Kramers-Kronig transformation to relate the real and imaginary parts of a susceptibility tensor with one another. This suggests a relation between the two parameters that has been found previously by Föhnle *et al.* [34], namely $\mathcal{I} = -\Gamma\tau$, where τ is a relaxation time. We obtain here a similar relation, $\mathcal{I} \propto -\Gamma\bar{\tau}$, where $\bar{\tau} = \hbar/mc^2$ has time dimension.

Even though the Gilbert damping is c^2 times larger than the inertial damping, the relative strength of the two parameters also depends on the real and imaginary parts of the susceptibility tensor. In special cases, when the real part of the susceptibility is much higher than the imaginary part, their strength could be comparable to each other. We note in this context that there exist materials where the real part of the susceptibility is 10^2 – 10^3 times larger than the imaginary part.

Finally, we emphasize that our derivation provides the *intrinsic* inertial damping of a pure, isolated system. For the Gilbert damping it is already well known that environmental effects, such as interfaces or grain boundaries, impurities, film thickness, and even interactions of the spins with quasiparticles, for example, phonons, can modify the extrinsic damping (see, e.g., Refs. [70–72]). Similarly, it can be expected that the inertial damping will become modified through environmental influences. An example of environmental effects that can lead to magnetic inertia have been considered previously, for the case of a local spin moment surrounded by conduction electrons, whose spins couple to the local spin moment and affect its dynamics [31,32].

V. CONCLUSIONS

In conclusion, we have rigorously derived the magnetization dynamics from the fundamental Dirac Hamiltonian and have provided a solid theoretical framework for, and established the origin of, magnetic inertia in pure systems. For a single spin- $\frac{1}{2}$ Dirac particle under the influence of an electromagnetic field we have derived the relativistic spin dynamics and showed that it contains an inertia term. For the dynamics of a macroscopic magnetic volume element, we have derived expressions for the Gilbert damping and the magnetic inertial damping on the same footing and have shown that both of them have a relativistic origin. The Gilbert damping stems from a generalized spin-orbit interaction involving external fields, while the inertial damping is due to higher-order (in $1/c^2$) spin-orbit contributions in the external fields. Both have been shown to be tensorial quantities. For general time dependent external fields, a field-derivative torque with a first order time derivative appears in the Gilbert-type damping, and a second order time-derivative field torque appears in the inertial damping.

In the case of harmonic external fields, the expressions of the magnetic inertia and the Gilbert damping scale with the real part and the imaginary part, respectively, of the magnetic susceptibility tensor, and they are opposite in sign. Alike the Gilbert damping, the magnetic inertia tensor is also temperature dependent through the magnetic response function and also magnetic moment dependent. Importantly, we find that the intrinsic inertial damping is much smaller than the Gilbert damping, which corroborates the fact that magnetic inertia was neglected in the early work on magnetization dynamics [1–3,19]. This suggests, too, that the influence of magnetic inertia will be quite restricted, unless the real part of the susceptibility is much larger than the imaginary part. Another possibility to enhance the magnetic inertia would be to use environmental influences to increase its extrinsic contribution. Our theory based on the Dirac Hamiltonian leads to exact expressions for both the intrinsic Gilbert and inertial damping terms, thus providing a solid base for their evaluation within *ab initio* electronic structure calculations and giving suitable values that can be used in future LLG magnetization dynamics simulations.

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