Spintronics Meets Nonadiabatic Molecular Dynamics: Geometric Spin Torque and Damping on Dynamical Classical Magnetic Texture due to an Electronic Open Quantum System

Utkarsh Bajpai and Branislav K. Nikolić
Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, USA

(Received 14 June 2020; accepted 23 September 2020; published 28 October 2020)

We analyze a quantum-classical hybrid system of steadily precessing around the fixed axis slow classical localized magnetic moments (LMMs), forming a head-to-head domain wall, surrounded by fast electrons driven out of equilibrium by LMMs and residing within a metallic wire whose connection to macroscopic reservoirs makes electronic quantum system an open one. The model captures the essence of dynamical noncollinear magnetic textures encountered in spintronics, while making it possible to obtain the exact time-dependent nonequilibrium density matrix of electronic systems and split it into four contributions. The Fermi surface contribution generates dissipative (or dampinglike in spintronics terminology) spin torque on LMMs, as the counterpart of electronic friction in nonadiabatic molecular dynamics (MD). Among two Fermi sea contributions, one generates geometric torque dominating in the adiabatic regime, which remains as the only nonzero contribution in a closed system with disconnected reservoirs. Locally geometric torque can have nondissipative (or fieldlike in spintronics terminology) component, acting as the counterpart of geometric magnetism force in nonadiabatic MD, as well as a much smaller dampinglike component acting as “geometric friction.” Such current-independent geometric torque is absent from widely used micromagnetics or atomistic spin dynamics modeling of magnetization dynamics based on the Landau-Lifshitz-Gilbert equation, while previous analyses of how to include our Fermi-surface dampinglike torque have severely underestimated its total magnitude.

DOI: 10.1103/PhysRevLett.125.187202

One of the most fruitful applications of geometric (or Berry) phase [1] concepts is encountered in quantum-classical hybrid systems where separation of timescales makes it possible to consider fast quantum degrees of freedom interacting with the slow classical ones [2,3]. The amply studied example of this kind are fast electrons interacting [4,5] with slow nuclei in molecular dynamics (MD) [6–10] problems of physics, chemistry, and biology. The parameters driving adiabatic evolution of quantum subsystems, with characteristic frequency smaller that its level spacing, are nuclear coordinates elevated to the status of dynamical variables. The electronic system then develops a geometric phase in states evolving out of an instantaneous energy eigenstate, while also acquiring shifts in the energy levels. Conversely, nuclei experience forces due to backaction from electrons. The simplest force is the adiabatic Born-Oppenheimer (BO) force [4,5] which depends only on the coordinates of the nuclei, and it is associated with electronic adiabatic potential energy surfaces [6–8]. Even small violation of BO approximation leads to additional forces—the first nonadiabatic correction generates forces linear in the velocity of the nuclei, and being Lorentz-like they are dubbed [2,11] “geometric magnetism.” The “magnetism” is not a real magnetic field, but an emergent geometrical property of the Hilbert space [12], and akin to the true Lorentz force, the emergent geometric force is nondissipative.

Additional forces appear upon making the quantum system open by coupling it to a thermal bath [11,13] (usually modeled as an infinite set of harmonic oscillators [14]) or to macroscopic reservoirs of particles [15]. In the latter case, one can also introduce chemical potential difference between the reservoirs to drive particle current through the quantum system and push it out of equilibrium [15–20]. In both equilibrium and nonequilibrium, the energy spectrum of an open quantum system is transformed into a continuous one, and frictional forces [9–11,15–20] linear in the velocity of the nuclei become possible. Also, due to continuous spectrum, the adiabaticity criterion has to be replaced by a different one [15]. Stochastic forces also appear, both in equilibrium and in nonequilibrium, where in the former case [11,13] they are due to fluctuations at finite temperature while in the latter case they include additional contributions from nonequilibrium noise [15–17]. Specific to nonequilibrium is the emergence of nonconservative force [15–17,19,20]. The derivation of these forces requires us to compute nonadiabatic corrections to the density matrix (DM) [11,13,15–17,19,20]. This yields a non-Markovian stochastic Langevin equation [21] as the most general [17,20] equation for nuclei in nonadiabatic MD.
The analogous problem exists in spintronics, where the fast quantum system is comprised of conduction electron spins and the slow classical system is comprised of localized-on-atoms spins and associated localized magnetic moments (LMMs) described by unit vectors $\mathbf{M}_i(t)$. The dynamics of LMMs is accounted for by the Landau-Lifshitz-Gilbert (LLG) type of equation [22].

\[
\frac{\partial \mathbf{M}_i}{\partial t} = -g\mathbf{M}_i \times \mathbf{B}^{\text{eff}}_i + \lambda \mathbf{M}_i \times \frac{\partial \mathbf{M}_i}{\partial t} + \frac{\gamma}{\mu_M} \left( \mathbf{T}_i[I^{\text{ex}}_i] + \mathbf{T}_i[\mathbf{M}_i(t)] \right). \tag{1}
\]

Here $g$ is the gyromagnetic ratio; $\mathbf{B}^{\text{eff}}_i = -(1/\mu_M)\partial \mathcal{H}/\partial \mathbf{M}_i$ is the effective magnetic field as the sum of an external field, field due to interaction with other LMMs, and magnetic anisotropy in the classical Hamiltonian $\mathcal{H}$ of LMMs; and $\mu_M$ is the magnitude of LMM [22]. Equation (1) includes phenomenological Gilbert damping, whose parameter $\lambda$ can be measured or calculated [23] by using an electronic Hamiltonian with spin-orbit or magnetic disorder scattering. It can also include a spin-transfer torque (STT) term $\mathbf{T}_i[I^{\text{ex}}_i]$ due to externally supplied spin current $I^{\text{ex}}_i$. The STT is a phenomenon [24] in which spin angular momentum of conduction electrons is transferred to local magnetization not aligned with electronic spin polarization. Finally, some analyses [25–27] also consider current-independent torque $\mathbf{T}_i[\mathbf{M}_i(t)]$ as a backaction of electrons pushed out of equilibrium by time-dependent $\mathbf{M}_i(t)$.

Nevertheless, such effects have been deemed negligible [25,28] except possibly for short enough noncollinear magnetic textures [29–32].

The STT vector, $\mathbf{T}_i = \mathbf{T}^{\text{FL}}_i + \mathbf{T}^{\text{DL}}_i$, can be decomposed [Fig. (1)] into (i) even under time reversal or fieldlike (FL) torque, which affects precession of LMM around an external field, field due to interaction with other LMMs, and magnetic anisotropy in the classical Hamiltonian $\mathcal{H}$ of LMMs; and (ii) odd under time reversal or dampinglike (DL) torque, which either enhances the Gilbert damping by pushing LMM toward $\mathbf{B}^{\text{eff}}_i$ or competes with Gilbert term as “antidamping.” For example, negative value of $\mathbf{T}^{\text{DL}}_i = 0$ ensures perfectly adiabatic regime [33], while in panels (b) and (d) $J_{sd} = 20\gamma$ ensures perfectly adiabatic regime [33], $J_{sd}/\hbar\omega \gg 1$, for the chosen precession frequency $\hbar\omega = 0.001\gamma$.

The current-driven STT $\mathbf{T}_i[I^{\text{ex}}_i]$ acts as the counterpart of nonconservative force in nonadiabatic MD. The Gilbert damping plus current-independent torque $\mathbf{T}_i[\mathbf{M}_i(t)]$ appear as the counterpart of electronic friction [9,10,15–20], but Gilbert damping requires agents [23] other than electrons alone considered in nonadiabatic MD. Thus, geometric spin torque, as the counterpart of geometric magnetism force [2], is conspicuously absent from standard modeling of classical magnetization dynamics. Geometric torque has been added ad hoc into the LLG equation applied to specific problems, such as spin waves within bulk magnetic

![FIG. 2. The FL and DL components [Fig. (1)] of three contributions to spin torque in Eq. (3) exerted by nonequilibrium spin density of electrons onto a single localized precessing magnetic moment in the setup of Fig. 1(a) as a function of coupling to the leads. Black dotted line is the sum of the three torques. In panels (a) and (c) $J_{sd} = 0.1\gamma$, while in panels (b) and (d) $J_{sd} = 20\gamma$ ensures perfectly adiabatic regime. $J_{sd}/\hbar\omega \gg 1$, for the chosen precession frequency $\hbar\omega = 0.001\gamma$.Nevertheless, such effects have been deemed negligible [25,28] except possibly for short enough noncollinear magnetic textures [29–32].](image-url)
materials [34–36]. A recent study [33] of a single classical LMM embedded into a closed electronic quantum system finds that nonequilibrium electronic spin density always generates geometric torque, even in the perfectly adiabatic regime where electron-spin-LMM interaction $J_{sd}$ is orders of magnitude larger than the characteristic frequency $\hbar \omega$ of LMM dynamics. It acts as a purely FL torque causing an anomalous frequency of precession that is higher than the Larmor frequency. By retracing the same steps [15,16] in the derivation of the stochastic Langevin equation for the electron-nuclei system connected to macroscopic reservoirs, Ref. [37] derived the stochastic LLG equation [38–41] for a single LMM embedded into an open electronic system out of equilibrium. The novelty in this derivation is damping, present even in the absence of traditional spin-flip relaxation mechanisms [25,27], while the same conclusion about geometric torque changing only the precession frequency of LMM has been reached (in some regimes, geometric phase can also affect the stochastic torque [42]). However, single LMM is a rather special case, which is illustrated in Fig. 1(a) and revisited in Fig. 2, and the most intriguing situations in spintronics involve dynamics of noncollinear textures of LMMs [26–32]. This is exemplified by current- or magnetic-field driven dynamics of domain walls (DWs) and skyrmions [27,41,43–47], where a much richer panoply of backaction effects from fast electronic system can be expected.

In this Letter, we analyze an exactly solvable model of LMMs steadily precessing with the frequency $\hbar \omega = 0.001 \gamma$, such as $\mathbf{M}_i(t) - \mathbf{M}_7(t)$ in Fig. 1(c), which are noncollinear and noncoplanar and embedded into a one-dimensional (1D) infinite wire hosting conduction electrons. The model can be viewed as a segment of dynamical noncollinear magnetic texture, and it directly describes magnetic field-driven [46,47] head-to-head Bloch DW [48], but without allowing it to propagate [46,47]. Its simplicity makes it exactly solvable—we find the exact time-dependent DM via the nonequilibrium Green’s functions (NEGF) [49] and analyze its contributions for different $J_{sd}/\hbar \omega$. In both Figs. 1(a) and 1(c), the electronic subsystem is an open quantum system and, although no bias voltage is applied between the macroscopic reservoirs, it is pushed into nonequilibrium by the dynamics of LMMs. For example, electronic quantum Hamiltonian becomes time dependent due to $\mathbf{M}_1(t)$ [Fig. 1(a)] or $\mathbf{M}_1(t) - \mathbf{M}_7(t)$ [Fig. 1(c)], which leads to pumping [27,50,51].

![Spatial profile of FL and DL components of $T_{geo}^i$, $T_{sea}^i$, and $T_{surf}^i$ spin torques on precessing LMMs depicted in Fig. 1(c) for closed or open electronic quantum system and for two different values of $J_{sd}$. Insets on the top of each row mark positions and static configuration of LMMs within the Bloch DW, with their $x$ component depicted by the color bar next to panel (a).](image)

**FIG. 3.** Spatial profile of FL and DL components of $T_{geo}^i$, $T_{sea}^i$, and $T_{surf}^i$ spin torques on precessing LMMs depicted in Fig. 1(c) for closed or open electronic quantum system and for two different values of $J_{sd}$. Insets on the top of each row mark positions and static configuration of LMMs within the Bloch DW, with their $x$ component depicted by the color bar next to panel (a).

![The $z$ component of total DL torque acting on DW in Fig. 1(c) as a function of $J_{sd}$ for $\gamma_c = \gamma$. Circles show that the sum of spin currents pumped into the leads matches $(\sum_i T_{surf,DL}^i)_z = I_{Lz}^\uparrow + I_{Rz}^\uparrow$. Panel (b) and (c), which correspond to Fig. 3(g), show spatial profile of local spin currents $I_{i,j}$ pumped between sites $i$ and $j$ for $J_{sd} = 0.1 \gamma$, with their sum being identically zero in panel (c). Dashed black lines in panels (a) and (b) are obtained from pumped local spin current by SMF [26,28], $I_{SMF}^z(z) = (\mu_B h G_0/4e^2) \partial M(z,t)/\partial t \times \partial M(z,t)/\partial z]$, where $G_0 = G^\uparrow + G^\downarrow$ is the total conductivity.](image)

**FIG. 4.** (a) The $z$ component of total DL torque acting on DW in Fig. 1(c) as a function of $J_{sd}$ for $\gamma_c = \gamma$. Circles show that the sum of spin currents pumped into the leads matches $(\sum_i T_{surf,DL}^i)_z = I_{Lz}^\uparrow + I_{Rz}^\uparrow$. Panel (b) and (c), which correspond to Fig. 3(g), show spatial profile of local spin currents $I_{i,j}$ pumped between sites $i$ and $j$ for $J_{sd} = 0.1 \gamma$, with their sum being identically zero in panel (c). Dashed black lines in panels (a) and (b) are obtained from pumped local spin current by SMF [26,28], $I_{SMF}^z(z) = (\mu_B h G_0/4e^2) \partial M(z,t)/\partial t \times \partial M(z,t)/\partial z]$, where $G_0 = G^\uparrow + G^\downarrow$ is the total conductivity.
[Figs. 4(b),4(c)] of spin current locally within the DW region, as well as into the leads [Fig. 4(a)].

The electrons are modeled on an infinite tight-binding (TB) chain, without impurities of spin-orbit coupling, whose Hamiltonian in the lab frame is $\hat H_{\text{lab}}(t) = \sum_{i,j} \left( \hat c_i^\dagger \hat c_j \right) J_{i,j} \sigma_{\alpha} \left( \hat c_i \right)$, where $\hat c_i^\dagger = (\hat c_{i,\uparrow}, \hat c_{i,\downarrow})$ and $\hat c_i = (\hat c_{i,\uparrow}, \hat c_{i,\downarrow})$ creates (annihilates) an electron of spin $\sigma = \uparrow, \downarrow$ at site $i$. The nearest-neighbor hopping is $\gamma = 1$ eV and $\hat \sigma$ is the vector of the Pauli matrices. The LMM region in Figs. 1(a) or 1(c) consists of one or seven sites, respectively, while the rest of the infinite TB chain is taken into account as the left ($L$) and the right ($R$) semi-infinite lead described by the first term in $\hat H_{\text{lab}}(t)$. The hopping between the leads and the LMM region is $\gamma$. The leads terminate at infinity into the macroscopic particle reservoirs with identical chemical potentials $\mu_L = \mu_R = E_F$ due to assumed absence of bias voltage, and $E_F = 0$ is chosen as the Fermi energy.

The spatial profile of noncoplanar Bloch DW in equilibrium is given by $M_i^z = -\text{sech}\left[\left( \frac{Z_{DW} - z_i}{W} \right) \text{tanh}\left( \frac{Z_{DW} - z_i}{W} \right) \right]$ and $M_i^z = \text{tanh}\left[\left( \frac{Z_{DW} - z_i}{W} \right) \text{tanh}\left( \frac{Z_{DW} - z_i}{W} \right) \right]$, where $Z_{DW} = 4$ and $W = 0.9$ (for an additional case of coplanar Néel DW, as well as different DW widths $W$ see the Supplemental Material [52]). Instead of solving LLG Eq. (1) for $M_i(t)-M_f(t)$ self-consistently coupled to DM calculations [43–45], we impose a solution where LMMs precess steadily around the $z$-axis: $M_i^z(t) = \sin \theta_i \cos(\omega t + \phi_i)$; $M_i^y(t) = \sin \theta_i \sin(\omega t + \phi_i)$; and $M_i^x(t) = \cos \theta_i$. Using a unitary transformation into the rotating frame (RF), $\hat H_{\text{lab}}(t)$ is transformed into time-independent [27,50], $\hat H_{\text{RF}} = \hat U(t) \hat H_{\text{lab}}(t) \hat U(t) - \left( \frac{\hbar \omega}{2 \pi} \right) \int_{-\infty}^{+\infty} dE \text{Im} \hat G(t,E) \hat U(t) + \hat \sigma_{\alpha} \left( \hat c_i^\dagger \right) + \hat \sigma_{\alpha} \left( \hat c_i \right)$. With LMMs frozen at $t = 0$ configuration from the lab. The unitary operator is $\hat U(t) = \exp(-i\omega t \hat \sigma_{\alpha}/2)$ for the $x$ axis of rotation. In the RF, the original two-terminal Landauer setup for quantum transport in Figs. 1(a) and 1(c) is mapped to $\hbar \omega \hat \sigma_{\alpha}/2$ term, onto an effective four-terminal setup [50], as illustrated for single LMM in Fig. 1(b). Each of its four leads is an effective half-metal ferromagnet which accepts only one spin species, $\uparrow$ or $\downarrow$, along the $\alpha$ axis, while an effective dc bias voltage $\hbar \omega/e$ acts between $L$ or $R$ pair of leads.

In the RF, the presence of the leads and reservoirs can be taken into account exactly using steady-state NEGFs [49] which depend on time difference $t - t'$, or energy $E$ upon Fourier transform. Using the retarded $\hat G(E)$ and the lesser, $\hat G^<(E)$, Green’s functions (GFs), we find the exact nonequilibrium DM of electrons in the RF, $\hat \rho_{\text{RF}} = \left( 1/2\pi \imath \right) \int dE \hat G^<(E)$. Here the two GFs are related by the Keldysh equation, $\hat G^<(E) = \hat G(E) \hat \Sigma^<(E) \hat G^<(E)$, where $\hat \Sigma^<(E)$ is the lesser self-energy [49] due to semi-infinite leads and $\hat G(E) = \left[ \hat E - \hat H_{\text{RF}} - \hat \Sigma(E,\hbar \omega) \right]^{-1}$ with $\hat \Sigma(E,\hbar \omega) = \sum_{\text{p},\text{l},\text{r},\sigma} \hat \Sigma_p^\text{p} \left( \hat c_{\text{p},\text{l},\sigma}^\dagger \hat c_{\text{p},\text{r},\sigma}^\dagger \right) \left( \hat c_{\text{p},\text{l},\sigma} \right) \left( \hat c_{\text{p},\text{r},\sigma} \right)$ being the sum of retarded self-energies for each of the four leads $p$, $\sigma$ in the RF. We use shorthand notation $Q_p^\text{p} = -1/2$ and $Q_p^\text{p} = +1/2$. Since typical frequency of magnetization dynamics is $\hbar \omega \ll E_F$, we can expand [53] $\hat \rho_{\text{RF}}$ in small $\hbar \omega/E_F$ and then transform it back to the lab frame, $\hat \rho_{\text{lab}}(t) = \hat U(t) \hat \rho_{\text{RF}} \hat U(t)^\dagger$ to obtain $\hat \rho_{\text{lab}}(t) = \hat \rho_{\text{geo}}(t) + \hat \rho_{\text{sea}}(t) + \hat \rho_{\text{surf}}(t)$, where (for additional details see Supplemental Material [52])

$$\hat \rho_{\text{geo}}(t) = \frac{1}{\pi^2} \int_{-\infty}^{+\infty} dE \text{Im} \left[ \hat G_0 \left( \hat \partial_t \right) \hat G_0 \right] f(E) \hat U(t),$$

(2a)

$$\hat \rho_{\text{sea}}(t) = \left( \frac{\hbar \omega}{2\pi} \right) \sum_p \int_{-\infty}^{+\infty} dE \text{Im} \left[ \hat G_0 \left( \frac{\partial \hat \Sigma_p^\text{p}}{\partial E} - \frac{\partial \hat \Sigma_p^\text{p}}{\partial E} \right) \hat G_0 \right] \times f(E) \hat U(t),$$

(2b)

$$\hat \rho_{\text{surf}}(t) = \frac{\hbar \omega}{4\pi} \sum_p \int_{-\infty}^{+\infty} dE \hat G_0 \left( \hat \rho_{\text{geo}}^\text{p} - \hat \rho_{\text{surf}}^\text{p} \right) \hat G_0 \left( \hat \partial_t \right) \hat U(t).$$

(2c)

We confirm by numerically exact calculations [43] that thus obtained $\hat \rho_{\text{lab}}(t)$ is identical to $\hbar g \hat G^<(t)/\hbar$ computed in the lab frame. Here $\hat G_0(E) = \left[ \hat E - \hat H_{\text{RF}} - \hat \Sigma(E,0) \right]^{-1}$ is $\hat G(E)$ with $\hbar \omega = 0$; $\hat G_0^p(E) = i \left[ \hat \Sigma_0^p(E) - \hat \Sigma_0^p(E)^\dagger \right]$ is the level broadening matrix due the leads; and $f_0^p(E) = f(E - E_F + Q_p^\text{p}) \hbar \omega)$ is the Fermi function of macroscopic reservoir $p$, $\sigma$ in the RF.

The total nonequilibrium spin density, $\langle \hat \sigma \rangle(t) = \text{Tr}[\hat \rho_{\text{lab}}(t) i \sigma \otimes \hat \sigma] = \langle \hat \sigma \rangle_\text{geo}(t) + \langle \hat \sigma \rangle_\text{sea}(t) + \langle \hat \sigma \rangle_\text{surf}(t)$, has the corresponding four contributions from DM contributions in Eq. (2). Here $\langle \hat \sigma \rangle_\text{geo}(t)$ is the equilibrium expectation value at an instantaneous time $t$ which defines adiabatic spin density [25,27,33,35,36]. It is computed using $\hat \rho_{\text{geo}}(t)$ as the grand canonical equilibrium DM expressed via the frozen (adiabatic) retarded GF [15,16,37], $\hat G_0(E) = \left[ \hat E - \hat H_{\text{RF}} - \hat \Sigma \right]^{-1}$, for instantaneous configuration of $\hat \rho_{\text{geo}}(t)$ while assuming $\partial \hat M_i^z(t)/\partial t = 0$ [subscript $t$ signifies parametric dependence on time through slow variation of $\hat \rho_{\text{geo}}(t)$]. The other three contributions—from $\hat \rho_{\text{geo}}(t)$ and $\hat \rho_{\text{sea}}(t)$ governed by the Fermi sea and $\hat \rho_{\text{surf}}(t)$ governed by the Fermi surface electronic states—contain first nonadiabatic correction [15,16,37] proportional to velocity $\partial \hat M_i^z(t)/\partial t$, as well as higher order terms due to $\hat \rho_{\text{lab}}(t)$ being exact. These three contributions define STT out of equilibrium [25,43,53]:

$$\hat T_i = J_{\text{STT}}(\langle \hat \sigma \rangle_\text{surf}) = \left( \hat T_i^\text{geo} + \hat T_i^\text{sea} + \hat T_i^\text{surf} \right)^\dagger.$$
To gain transparent physical interpretation of $T_{1,\text{geo}}^{\text{geo}}$ and $T_{1,\text{surf}}^{\text{geo}}$, we first consider the simplest case [33,37]—a single $M_1(t)$ in setup of Fig. 1(a). The STT contributions as a function of the coupling $\gamma_c$ to the leads (i.e., reservoirs) are shown in Fig. 2. We use two different values for $J_{sd}$, where large ratio of $J_{sd} = 20$ and $\hbar \omega = 0.001$ eV is perfect adiabatic limit [33,35,36]. Nevertheless, even in this limit and for $\gamma_c \to 0$ we find $T_{1,\text{geo}}^{\text{geo}} \neq 0$ in Fig. 2(b) as the only nonzero and purely FL torque. This is also found in closed system of Ref. [33] where $T_{1,\text{geo}}^{\text{geo}}$ was expressed in terms of the spin Berry curvature. As the quantum system becomes open for $\gamma_c > 0$, $T_{1,\text{geo}}^{\text{geo}}$ is slightly reduced while $T_{1,\text{surf}}^{\text{geo}}$ emerges with small FL [Fig. 2(b)] and large DL [Fig. 2(d)] components. The DL torque $T_{1,\text{surf}}^{\text{geo},\text{DL}}$ points toward the $z$ axis and, therefore, enhances the Gilbert damping. In the wide-band approximation [54], $\Sigma(E) = -i\mathcal{T}_2$ is energy independent which makes it possible to obtain (at zero temperature)

$$T_{1,\text{geo}}^{\text{geo}}(t) = \frac{\hbar \omega}{2\pi} \left[ \pi - 2\tan^{-1}\left(\frac{\Gamma}{J_{sd}}\right) \right] \sin \theta e_y(t), \quad (4)$$

where $e_y(t) = -\sin \omega t e_x + \cos \omega t e_y$. Thus, in the perfect adiabatic limit, $J_{sd}/\hbar \omega \to \infty$, or in a closed system, $\Gamma \to 0$, $T_{1,\text{geo}}^{\text{geo}}$ is independent of microscopic parameters as expected from its geometric nature [34]. The always present $T_{1,\text{geo}}^{\text{geo}} \neq 0$ means that electron spin is never parallel [33] to the adiabatic direction of $(\mathbf{S})_{\text{sd}}$. Switching to DW [Fig. 1(c)] embedded into a closed quantum system ($\gamma_c = 0$) shows in Figs. 3(a)–3(d) that only $T_{1,\text{geo}}^{\text{geo}} \neq 0$, which also acquires DL component locally with damping or antidamping action depending on the position of LMM. At first sight, $T_{1,\text{geo},\text{DL}}^{\text{geo}} \neq 0$ violates Berry and Robbins original analysis [2] according to which an isolated quantum system, with discrete energy spectrum, cannot exert friction onto the classical system. This is resolved by noting that, in the absence of external driving, the total energy of electrons plus LMMs system remains conserved [55]. Upon opening the quantum system ($\gamma_c = \gamma$), Figs. 3(e)–3(h) shows emergence of additional $T_{1,\text{surf}}^{\text{geo}} \neq 0$ which, however, becomes negligible [Fig. 3(f), 3(h)] in the perfectly adiabatic limit $J_{sd}/\hbar \omega \gg 1$. Figure 4(a) confirms that total $(\sum_i T_{1,\text{surf},\text{DL}})^{\text{geo}}_x = \hat{I}_L^x + \hat{I}_R^x$ is identical to net spin current pumped into the leads via which the conduction electrons carry away excess angular momentum of precessing LMMs [51]. Such identity underlies physical picture where spin current pumped by time-dependent magnetization becomes DL torque [26,51]. Note that pumped spin current $\hat{I}_L^x$ due to $\hat{\rho}_{\text{geo}}$ or $\hat{\rho}_{\text{geo}}$ in Fig. 4(c) can be nonzero locally, but they sum to zero. Although $(\sum_i T_{1,\text{geo},\text{DL}})^{\text{geo}}_x = 0$ in Fig. 4(a), small $(\sum_i T_{1,\text{geo},\text{DL}})^{\text{geo}}_y \neq 0$ exists and contributes [dash-dotted line in Fig. 4(a)] to pumped $\hat{I}_L^y + \hat{I}_R^y$ which then acts as “geometric friction” [11]. The nonuniform pumped spin current due to spatially and time varying magnetization has prompted proposals [26,28] to amend the LLG equation by adding the corresponding DL torque $\mathbf{M} \times \mathbf{D} \cdot \partial \mathbf{M}/\partial t$ with $3 \times 3$ damping tensor $\mathbf{D}$ whose spatial dependence is given by the so-called spin-motive force formula. However, SMF correction was estimated to be small [28] in the absence of spin-orbit coupling in the band structure. We confirm its smallness in Figs. 4(a), 4(b) for our DW case, but this actually reveals that SMF formula produces incorrectly an order of magnitude smaller total torque than obtained from our exact $\hat{\rho}_{\text{surf}}(t)$. Because of the possibly complex [44,47] time and spatial dependence of $T_{1,\text{surf}}^{\text{geo}}$ and $T_{1,\text{geo}}^{\text{geo}}$, the accurate path to incorporate them is offered by self-consistent coupling of electronic DM and LLG calculations, as proposed in Refs. [43,45,55]. This is because electrons can be “integrated out” [38–41] only in the perturbative limit of small $J_{sd}$ [44,55] and small $\gamma_c$ [44] to obtain LMM-only equation that is akin to nuclei-only equations with universal [9] electronic friction term in nonadiabatic MD [8,10,15–20].

This research was supported by the U.S. National Science Foundation (NSF) under Grant No. CHE 1566074.

[1] M. V. Berry, Quantal phase factors accompanying adiabatic changes, Proc. R. Soc. A 392, 45 (1984).
[2] M. V. Berry and J. M. Robbins, Chaotic classical and half-classical adiabatic reactions: geometric magnetism and deterministic friction, Proc. R. Soc. A 442, 659 (1993).
[3] Q. Zhang and B. Wu, General Approach to Quantum-Clasical Hybrid Systems and Geometric Forces, Phys. Rev. Lett. 97, 190401 (2006).
[4] S. K. Min, A. Abedi, K. S. Kim, and E. K. U. Gross, Is the Molecular Berry Phase an Artifact of the Born-Oppenheimer Approximation?, Phys. Rev. Lett. 113, 263004 (2014).
[5] R. Requist, F. Tandetzky, and E. K. U. Gross, Molecular geometric phase from the exact electron-nuclear factorization, Phys. Rev. A 93, 042108 (2016).
[6] L.G. Ryabinkin, L. Joubert-Doriol, and A.F. Izmaylov, Geometric phase effects in nonadiabatic dynamics near conical intersections, Acc. Chem. Res. 50, 1785 (2017).
[7] F. Agostini and B. F. E. Curchod, Different flavors of nonadiabatic molecular dynamics, WIREs Comput. Mol. Sci. 9, e1417 (2019).
[8] W. Dou and J.E. Subotnik, Nonadiabatic molecular dynamics at metal surfaces, J. Phys. Chem. A 124, 757 (2020).
[9] W. Dou and J. E. Subotnik, Perspective: How to understand electronic friction, J. Chem. Phys. 148, 230901 (2018).
[10] J.-T. Lü, B.-Z. Hu, P. Hedegård, and M. Brandbyge, Semi-classical generalized Langevin equation for equilibrium and nonequilibrium molecular dynamics simulation, Prog. Surf. Sci. 94, 21 (2019).
[11] M. Campisi, S. Denisov, and P. Hänggi, Geometric magnetism in open quantum systems, Phys. Rev. A 86, 032114 (2012).
[12] M. Kolodrubetz, D. Sels, P. Mehta, and A. Polkovnikov, Geometry and non-adiabatic response in quantum and classical systems, Phys. Rep. 697, 1 (2017).

[13] F. Gaitan, Berry’s phase in the presence of a stochastically evolving environment: A geometric mechanism for energy-level broadening, Phys. Rev. A 58, 1665 (1998).

[14] R. S. Whitney and Y. Gefen, Berry Phase in a Nonisolated System, Phys. Rev. Lett. 90, 190402 (2003).

[15] M. Thomas, T. Karzig, S. V. Kusminskiy, G. Zaránd, and F. von Oppen, Scattering theory of adiabatic reaction forces due to out-of-equilibrium quantum environments, Phys. Rev. B 86, 195419 (2012).

[16] N. Bode, S. V. Kusminskiy, R. Egger, and F. von Oppen, Scattering Theory of Current-Induced Forces in Mesoscopic Systems, Phys. Rev. Lett. 107, 036804 (2011).

[17] J.-T. Lü, M. Brandbyge, P. Hedegård, T. N. Todorov, and F. Gaitan, Berry phase in the presence of a stochastically evolving environment, Phys. Rev. B 98, 054405(R) (2018).

[18] W. Dou, G. Miao, and J. E. Subotnik, Born-Oppenheimer Dynamics, Electronic Friction, and the Inclusion of Electron-Electron Interactions, Phys. Rev. Lett. 119, 046001 (2017).

[19] M. Hopjan, G. Stefanucci, E. Perfetto, and C. Verdozzi, Molecular junctions and molecular motors: Including Coulomb repulsion in electronic friction using nonequilibrium Green’s functions, Phys. Rev. B 98, 041405(R) (2018).

[20] F. Chen, K. Miwa, and M. Galperin, Current-induced forces for nonadiabatic molecular dynamics, J. Phys. Chem. A 123, 693 (2019).

[21] R. L. S. Farias, R. O. Ramos, and L. A. da Silva, Stochastic Langevin equations: Markovian and non-Markovian dynamics, Phys. Rev. E 80, 031143 (2009).

[22] R. F. L. Evans, W. J. Fan, P. Chureemart, T. A. Ostler, M. O. A. Ellis, and R. W. Chantrell, Atomic spin model simulations of magnetic nanomaterials, J. Phys. Condens. Matter 26, 103202 (2014).

[23] A. A. Starikov, P. J. Kelly, A. Brataas, Y. Tserkovnyak, and G. E. W. Bauer, Unified First-Principles Study of Gilbert Damping, Spin-Flip Diffusion, and Resistivity in Transition Metal Alloys, Phys. Rev. Lett. 105, 236601 (2010).

[24] D. Ralph and M. Stiles, Spin transfer torques, J. Magn. Magn. Mater. 320, 1190 (2008).

[25] S. Zhang and Z. Li, Roles of Nonequilibrium Conduction Electrons on the Magnetization Dynamics of Ferromagnets, Phys. Rev. Lett. 93, 127204 (2004).

[26] S. Zhang and S. S.-L. Zhang, Generalization of the Landau-Lifshitz-Gilbert equation for conducting ferromagnets, Phys. Rev. Lett. 102, 086601 (2009).

[27] G. Tatara, Effective gauge field theory of spintronics, Physica (Amsterdam) 106E, 208 (2019).

[28] K.-W. Kim, J.-H. Moon, K.-J. Lee, and H.-W. Lee, Prediction of Giant Spin Motive Force due to Rashba Spin-Orbit Coupling, Phys. Rev. Lett. 108, 217202 (2012).

[29] J. Foros, A. Brataas, Y. Tserkovnyak, and G. E. W. Bauer, Current-induced noise and damping in nonuniform ferromagnets, Phys. Rev. B 78, 140402(R) (2008).

[30] Y. Tserkovnyak, E. M. Hankiewicz, and G. Vignale, Transverse spin diffusion in ferromagnets, Phys. Rev. B 79, 094415 (2009).

[31] E. M. Hankiewicz, G. Vignale, and Y. Tserkovnyak, Inhomogeneous Gilbert damping from impurities and electron-electron interactions, Phys. Rev. B 78, 020404(R) (2008).

[32] Z. Yuan, K. M. D. Hals, Y. Liu, A. A. Starikov, A. Brataas, and P. J. Kelly, Gilbert damping in noncollinear ferromagnets, Phys. Rev. Lett. 113, 266603 (2014).

[33] C. Stahl and M. Potthoff, Anomalous Spin Precession under a Geometrical Torque, Phys. Rev. Lett. 119, 227203 (2017).

[34] X. G. Wen and A. Zee, Spin Waves and Topological Terms in the Mean-Field Theory of Two-Dimensional Ferromagnets and Antiferromagnets, Phys. Rev. Lett. 61, 1025 (1988).

[35] Q. Niu and L. Kleinman, Spin-Wave Dynamics in Real Crystals, Phys. Rev. Lett. 80, 2205 (1998).

[36] Q. Niu, X. Wang, L. Kleinman, W.-M. Liu, D. M. C. Nicholson, and G. M. Stocks, Adiabatic Dynamics of Local Spin Moments in Itinerant Magnets, Phys. Rev. Lett. 83, 207 (1999).

[37] N. Bode, L. Arrachea, G. S. Lozano, T. S. Nunner, and F. von Oppen, Current-induced switching in transport through anisotropic magnetic molecules, Phys. Rev. B 85, 115440 (2012).

[38] M. Onoda and N. Nagaosa, Dynamics of Localized Spins Coupled to the Conduction Electrons with Charge and Spin Currents, Phys. Rev. Lett. 96, 066603 (2006).

[39] A. S. Núñez and R. A. Duine, Effective temperature and Gilbert damping of a current-driven localized spin, Phys. Rev. B 77, 054401 (2008).

[40] J. Fransson and J.-X. Zhu, Spin dynamics in a tunnel junction between ferromagnets, New J. Phys. 10, 013017 (2008).

[41] H. M. Hurst, V. Galitski, and T. T. Heikkilä, Electron-induced massive dynamics of magnetic domain walls, Phys. Rev. B 101, 054407 (2020).

[42] A. Shnirman, Y. Gefen, A. Saha, I. S. Burmistrov, M. N. Kiselev, and A. Altland, Geometric Quantum Noise of Spin, Phys. Rev. Lett. 114, 176806 (2015).

[43] M. D. Petrović, B. S. Popescu, U. Bajpai, P. Plecháč, and B. K. Nikolić, Spin and Charge Pumping by a Steady or Pulse-Current-Driven Magnetic Domain Wall: A Self-Consistent Multiscale Time-Dependent Quantum-Classical Hybrid Approach, Phys. Rev. Applied 10, 054038 (2018).

[44] U. Bajpai and B. K. Nikolić, Time-retarded damping and magnetic inertia in the Landau-Lifshitz-Gilbert equation self-consistently coupled to electronic time-dependent nonequilibrium Green functions, Phys. Rev. B 99, 134409 (2019).

[45] E. V. Boström and C. Verdozzi, Steering magnetic skyrmions with currents: A nonequilibrium Green’s functions approach, Phys. Status Solidi B 256, 1800590 (2019).

[46] S. Woo, T. Delaney, and G. S. D. Beach, Magnetic domain wall depinning assisted by spin wave bursts, Nat. Phys. 13, 448 (2017).

[47] M. D. Petrović, P. Plecháč, and B. K. Nikolić, Anihilation of topological solitons in magnetism with spin wave burst finale: The role of nonequilibrium electrons and their spin pumping over ultrabroadband frequencies, arXiv:1908.03194.

[48] R. D. McMichael and M. J. Donahue, Head to head domain wall structures in thin magnetic strips, IEEE Trans. Magn. 33, 4167 (1997).
[49] G. Stefanucci and R. van Leeuwen, *Nonequilibrium Many-Body Theory of Quantum Systems: A Modern Introduction* (Cambridge University Press, Cambridge, England, 2013).

[50] S.-H Chen, C.-R Chang, J. Q. Xiao, and B. K. Nikolić, Spin and charge pumping in magnetic tunnel junctions with precessing magnetization: A nonequilibrium Green function approach, *Phys. Rev. B* **79**, 054424 (2009).

[51] Y. Tserkovnyak, A. Brataas, G. E. W. Bauer, and B. I. Halperin, Nonlocal magnetization dynamics in ferromagnetic heterostructures, *Rev. Mod. Phys.* **77**, 1375 (2005).

[52] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.125.187202 for two additional figures, serving as counterparts of Figs. 3 and 4 but for the case of Néel DW where all LMMs are coplanar, as well as an additional figure showing dependence of pumped spin currents from Fig. 4 as a function of DW width for both the Bloch and Néel DWs. It also provides details of derivation leading to our central Eq. (2).

[53] F. Mahfouzi and B. K. Nikolić, How to construct the proper gauge-invariant density matrix in steady-state non-equilibrium: Applications to spin-transfer and spin-orbit torque, *SPIN* **03**, 02 (2013).

[54] A. Bruch, M. Thomas, S. Viola Kusminskiy, F. von Oppen, and A. Nitzan, Quantum thermodynamics of the driven resonant level model, *Phys. Rev. B* **93**, 115318 (2016).

[55] M. Sayad and M. Potthoff, Spin dynamics and relaxation in the classical-spin Kondo-impurity model beyond the Landau-Lifshitz-Gilbert equation, *New J. Phys.* **17**, 113058 (2015).
Supplemental Material for “Spintronics Meets Nonadiabatic Molecular Dynamics: Geometric Spin Torque and Damping on Dynamical Classical Magnetic Texture due to an Electronic Open Quantum System”

Utkarsh Bajpai and Branislav K. Nikolić
Department of Physics and Astronomy, University of Delaware, Newark, DE 19716, USA

This Supplemental Material provides three additional Figures [Sec. I]—for the case of coplanar Néel domain wall (DW) and different DW widths of both Bloch (from the main text) and Néel DWs—as well as additional details in the derivation [Sec. II] of the exact time-dependent nonequilibrium density matrix \( \hat{\rho}_{\text{lab}}(t) \) in Eq. (2) in the main text.

I. ADDITIONAL FIGURES COMPLEMENTING FIGS. 3 AND 4 IN THE MAIN TEXT

Figures S1 and S2 show the case of coplanar Néel DW [1], as the counterparts of Figs. 3 and 4 (for the noncoplanar Bloch DW) in the main text, respectively. The coplanar spatial profile of Néel DW in equilibrium is given by

\[
M_i^z = 0, \quad M_i^x = \text{sech}\left[\frac{(Z_{DW} - z_i)}{W}\right], \quad M_i^y = \text{tanh}\left[\frac{(Z_{DW} - z_i)}{W}\right],
\]

where \( i = 1\ldots7, \) \( Z_{DW} = 4 \) and \( W = 0.9 \) in Figs. S1 and S2. Repeating the same analysis in the main text, we impose a solution where localized magnetic moments (LMMs), described by unit vectors \( \mathbf{M}_i(t) \), precess steadily around the \( z \)-axis—\( M_i^z(t) = \sin \theta_i \cos(\omega t + \phi_i) \); \( M_i^x(t) = \sin \theta_i \sin(\omega t + \phi_i) \); and \( M_i^y(t) = \cos \theta_i \)—instead of solving the Landau-Lifshitz-Gilbert equation for each LMM self-consistently coupled to quantum-mechanical calculations of the time-dependent nonequilibrium density matrix \( \hat{\rho}_{\text{lab}}(t) \). The chosen precession frequency is \( \hbar \omega = 0.001 \gamma \).

Interestingly, in the case of Néel DW, geometric spin torque is always field-like (FL) in Fig. S1. This leads to geometric contribution to local spin current in Fig. S2(c), as governed by \( \hat{\rho}_{\text{geo}}(t) \) in Eq. (2b) in the main text, being zero across each bond in order to ensure that difference of local spin currents flowing into and out of a given site, \( (I^{S_2}_{i \rightarrow i+1} - I^{S_2}_{i-1 \rightarrow i}) \), and the damping-like (DL) torque determined by it, \( T^\text{geo,DL}_i \), are zero. Since pumped local spin current \( I^{S_2}_{\text{SMF}} \) from phenomenological spin motive force (SMF) theory [4, 5] is the same in Fig. 4(b) in the main text and in Fig. S2(b), such phenomenological approach does not differentiate between Bloch and Néel DW.

Figure S3 shows local spin current and spin current outflowing into the right normal metal (NM), pumped by DW dynamics, as a function of DW width \( W \) where Bloch and Néel DWs composed of \( N = 21 \) LMMs are compared. As in Fig. S2(b), phenomenological SMF theory [4, 5] does not differentiate between Bloch and Néel DWs—dashed black lines in Figs. S3(b) and S3(c), or in Figs. S3(d) and S3(e), are identical. Also, pumped local spin current from the SMF theory [4, 5]

\[
I^{S_2}_{\text{SMF}}(z_i) \propto \frac{1}{W} \text{sech}^2\left(\frac{z_i - Z_{DW}}{W}\right) \text{tanh}\left(\frac{z_i - Z_{DW}}{W}\right),
\]

obtained analytically, decreases with \( W \) after reaching a maximum at some small \( W \approx 2 \) [Figs. S3(b) and S3(c)], thereby making it relevant only for very narrow DWs (in the absence of spin-orbit coupling [1, 5]). On the other hand, our exact result in Fig. S3 shows that both local spin currents [solid blue line in Figs. S3(b) and S3(c)] and outflowing into the right NM lead spin current [solid blue line in Figs. S3(d) and S3(e)] are enhanced in wider DWs, but they saturate with increasing \( W \).

II. ADDITIONAL DETAILS IN THE DERIVATION OF EQ. (2) IN THE MAIN TEXT

Using the same notation as in the main text, in this Section we provide a detailed derivation of the exact time-dependent nonequilibrium density matrix \( \hat{\rho}_{\text{lab}}(t) \) and its four contributions—\( \hat{\rho}_\text{ad}, \hat{\rho}_\text{geo}(t), \hat{\rho}_\text{sea}(t) \) and \( \hat{\rho}_\text{stat}(t) \)—in Eq. (2) in the main text. We first recall that \( \hat{\rho}_{\text{lab}}(t) = \hat{U}(t)\hat{\rho}_{\text{RF}}\hat{U}^\dagger(t) \) is obtained from steady-state nonequilibrium density matrix in the rotating frame (RF):

\[
\hat{\rho}_{\text{RF}} = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} dE \hat{G}^< (E) = \frac{1}{2\pi} \sum_{p,\sigma} \int_{-\infty}^{+\infty} dE \hat{G}(E) \hat{\tilde{G}}^\sigma_p (E, \hbar \omega) \hat{\tilde{G}}^\dagger (E) f^\sigma_p (E),
\]

(3)
FIG. S1. Spatial profile of FL and DL components of $T_i^{\text{geo}}$, $T_i^{\text{sea}}$ and $T_i^{\text{surf}}$ spin torques (defined in Eqs. 2 and 3 in the main text) as back-action of nonequilibrium electrons on precessing LMMs comprising Néel DW embedded into closed or open electronic quantum system and for two different values of $J_{sd}$. Insets on the top of each row mark positions and static configuration of LMMs in equilibrium, which are coplanar ($M_x = 0$) within the $yz$-plane.

FIG. S2. (a) The $z$-component of total DL torque acting on Néel DW as a function of $J_{sd}$ for $\gamma_c = \gamma$. Circles show that the sum of spin currents pumped into the leads matches $\left(\sum_i T_i^{\text{surf}, DL}\right)_z = I_{L}^{S_z} + I_{R}^{S_z}$. Panel (b) and (c), which correspond to Fig. S1(g), show spatial profile of local spin currents $I_{i\rightarrow j}^{S_z}$ pumped between sites $i$ and $j$ for $J_{sd} = 0.1\gamma$. Dashed black lines in panels (a) and (b) are obtained from pumped local spin current by SMF [4, 5], $I_{\text{SMF}}^{S_z}(z) = \frac{n_0 G_0 G^*}{4e^2}[\partial M(z,t)/\partial t \times \partial M(z,t)/\partial z]_z$, where $G_0 = G^* + G^2$ is the total conductivity.

Here we remind the reader that $\hat{G}(E) = [E - \hat{H}_{RF} - \hat{\Sigma}(E, \hbar \omega)]^{-1}$ where $\hat{H}_{RF} = \hat{H}_{\text{lab}}(t = 0) - i\hbar \hat{U}(\partial \hat{U}/\partial t) = \hat{H}_{\text{lab}}(t = 0) - \hbar \omega \hat{\sigma}_\alpha / 2$ (for rotation along $\alpha$-axis) while $\hat{U}(t) = \exp(-i\omega t \hat{\sigma}_\alpha)$ is used to perform unitary transformation into the RF. We also use $\hat{\Sigma}(E, \hbar \omega) = \sum_{p=L,R,\sigma=\uparrow,\downarrow} \hat{\Sigma}_p^\sigma (E - Q_p^\sigma \hbar \omega)$ as the self-energy matrix due to all leads in the RF; $\hat{\Gamma}_p^\sigma (E, \hbar \omega) = \frac{i}{2}[\hat{\Sigma}_p^\sigma (E, \hbar \omega) + \hat{\Sigma}_p^\sigma (E, \hbar \omega)^\dagger]$ as the corresponding level-broadening matrix; and $f_p^\sigma(E) = f(E - [E_F + Q_p^\sigma \hbar \omega])$.
FIG. S3. (a) Spatial profile of $M_i^y$ component of LMMs across Bloch DWs of width $W = 1$ (solid line) or $W = 15$ (dash-dot line). Dotted vertical line in panel (a) marks the position at which pumped local spin current $I_{12\rightarrow13}^{S_z}$ (between sites $i = 12$ and $i = 13$) is evaluated and plotted (solid lines) in panels (b) and (c) as a function of $W$ for Bloch and Néel DWs, respectively. Solid lines in panels (d) and (e) plot spin current $I_{R}^{S_z}$ pumped by DW dynamics which outflows into the right NM lead for both Bloch and Néel DWs, respectively. Dashed black lines in panels (b)–(e) are obtained from SMF formula [4, 5] for $I_{SMF}^{S_z}(z)$ displayed in the caption of Fig. S2.

is the Fermi function of the macroscopic reservoirs in the RF.

Due to typical frequency of DW dynamics being $\hbar\omega \ll E_F$, we perform a Taylor expansion of each term in Eq. (3) that is linear order in $\hbar\omega$ to obtain an expression for $\hat{j}_{RF}$, akin to Ref. [6], where steady-state nonequilibrium density matrix in the presence of small external bias-voltage $eV_b \ll E_F$ was evaluated. In this spirit, the Fermi function of the macroscopic reservoirs in the RF expanded up to linear order in $\hbar\omega$ is given by

$$f_p^\sigma(\hat{E}) \approx f(\hat{E} - E_F) - Q_p^\sigma \frac{\partial f}{\partial \hat{E}} \hbar\omega, \quad (4)$$

while the level-broadening matrix is given by

$$\hat{\Gamma}_p^\sigma(\hat{E}, \hbar\omega) \approx \hat{\Gamma}_p^\sigma(\hat{E}) - Q_p^\sigma \frac{\partial \hat{\Gamma}_p^\sigma}{\partial \hat{E}} \hbar\omega. \quad (5)$$

To obtain such an expansion for $\hat{G}(E)$, we first define $\hat{G}_0(E)$ by setting $\hbar\omega = 0$ in $\hat{G}(E)$, i.e.,

$$\hat{G}_0(E) = \left[ E - \hat{H}_{lab}(t = 0) - \Sigma(E, 0) \right]^{-1}, \quad (6)$$
and note that $\hat{G}(E)$ can be expressed as

$$\hat{G}(E) = \left[ E - \hat{H}_{RF} - \hat{\Sigma}(E, \hbar \omega) \right]^{-1} = \left[ E - \hat{H}_{RF} - \hat{\Sigma}(E, \hbar \omega) + \hat{H}_{lab}(t = 0) - \hat{\Sigma}(E, 0) - \hat{\Sigma}(E, 0) \right]^{-1} = \left[ E - \hat{H}_{lab}(t = 0) - \hat{\Sigma}(E, 0) - \{ \hat{H}_{RF} - \hat{H}_{lab}(t = 0) \} - \{ \hat{\Sigma}(E, \hbar \omega) - \hat{\Sigma}(E, 0) \} \right]^{-1}. \quad (7)$$

We then rewrite $\hat{G}(E)$ using the Dyson equation

$$\hat{G}(E) = \hat{G}_0(E) + \hat{G}_0(E) \left[ \hat{H}_{RF} - \hat{H}_{lab}(t = 0) + \hat{\Sigma}(E, \hbar \omega) - \hat{\Sigma}(E, 0) \right] \hat{G}_0(E). \quad (8)$$

Note that $\hat{H}_{RF} - \hat{H}_{lab}(t = 0) = -\imath \hbar \hat{U}_t (\partial \hat{U}/\partial t) = -\hbar \omega \hat{\sigma}/2$ is already linear order in $\hbar \omega$ while the self-energy matrix can be expanded as

$$\hat{\Sigma}(E, \hbar \omega) - \hat{\Sigma}(E, 0) = \sum_{p, \sigma} \left[ \hat{\Sigma}_p^\sigma(E, \hbar \omega) - \hat{\Sigma}_p^\sigma(E, 0) \right] \approx -\sum_{p, \sigma} Q_p^\sigma \frac{\partial \hat{\Sigma}_p^\sigma}{\partial E} \hbar \omega. \quad (9)$$

By inserting this result into Eq. (8), we obtain expansion of $\hat{G}(E)$ to linear order in $\hbar \omega$ as

$$\hat{G}(E) \approx \hat{G}_0(E) + \hat{G}_0(E) \left[ ( -\imath \hbar \hat{U}_t \partial \hat{U} / \partial t ) + \sum_{p, \sigma} \left( -Q_p^\sigma \frac{\partial \hat{\Sigma}_p^\sigma}{\partial E} \hbar \omega \right) \right] \hat{G}_0(E). \quad (10)$$

By using Eq. (4), (5) and Eq. (10) in the integrand of Eq. (3), and by retaining only terms to linear order in $\hbar \omega$, we can decompose $\hat{\rho}_{RF}$ into a sum of $\hat{\rho}_{eq}$, $\hat{\rho}_{geo}$, $\hat{\rho}_{sea}$ and $\hat{\rho}_{surf}$ contributions

$$\hat{\rho}_{RF} = \frac{1}{2\pi} \sum_{p, \sigma} \int_{-\infty}^{+\infty} dE \hat{G}(E) \hat{\Gamma}_p^\sigma(E, \hbar \omega) \hat{G}^\dagger(E) f_p^\sigma(E) = \hat{\rho}_{eq} + \hat{\rho}_{geo} + \hat{\rho}_{sea} + \hat{\rho}_{surf}, \quad (11)$$

where

$$\hat{\rho}_{eq} = \frac{1}{2\pi} \sum_{p, \sigma} \int_{-\infty}^{+\infty} dE \hat{G}_0 \hat{\Gamma}_p^\sigma \hat{G}_0^\dagger f, \quad (12a)$$

$$\hat{\rho}_{geo} = \frac{1}{2\pi} \sum_{p, \sigma} \int_{-\infty}^{+\infty} dE \left[ \hat{G}_0 \left( -\imath \hbar \hat{U}_t \partial \hat{U} / \partial t \right) \hat{G}_0 \right] \hat{\Gamma}_p^\sigma \hat{G}_0^\dagger f + \frac{1}{2\pi} \sum_{p, \sigma} \int_{-\infty}^{+\infty} dE \hat{G}_0 \hat{\Sigma}_p^\sigma \left[ \hat{G}_0 \left( -\imath \hbar \hat{U}_t \partial \hat{U} / \partial t \right) \hat{G}_0 \right] \hat{\Gamma}_p^\sigma \hat{G}_0^\dagger f, \quad (12b)$$

$$\hat{\rho}_{sea} = \frac{\hbar \omega}{2\pi} \sum_{p, \sigma} \int_{-\infty}^{+\infty} dE \left[ \hat{G}_0 \left( -Q_p^\sigma \frac{\partial \hat{\Sigma}_p^\sigma}{\partial E} \right) \hat{G}_0 \right] \hat{\Gamma}_p^\sigma \hat{G}_0^\dagger f + \frac{\hbar \omega}{2\pi} \sum_{p, \sigma} \int_{-\infty}^{+\infty} dE \hat{G}_0 \hat{\Sigma}_p^\sigma \left[ \hat{G}_0 \left( -Q_p^\sigma \frac{\partial \hat{\Sigma}_p^\sigma}{\partial E} \right) \hat{G}_0 \right] \hat{\Gamma}_p^\sigma \hat{G}_0^\dagger f + \frac{\hbar \omega}{2\pi} \sum_{p, \sigma} \int_{-\infty}^{+\infty} dE \hat{G}_0 \left[ -Q_p^\sigma \frac{\partial f}{\partial E} \right] \hat{\Gamma}_p^\sigma \hat{G}_0^\dagger f \quad (12c)$$

$$\hat{\rho}_{surf} = \frac{\hbar \omega}{2\pi} \sum_{p, \sigma} \int_{-\infty}^{+\infty} dE \hat{G}_0 \hat{\Gamma}_p^\sigma \left[ -Q_p^\sigma \frac{\partial f}{\partial E} \right] \hat{G}_0^\dagger f. \quad (12d)$$

Here we suppress "function of $E$" notation on the right hand side for brevity.

The contributions $\hat{\rho}_{eq}$, $\hat{\rho}_{geo}$, $\hat{\rho}_{sea}$ and $\hat{\rho}_{surf}$ obtained in the RF can be transformed back to their lab frame time-dependent counterparts, $\hat{\rho}_{eq}^{\text{lab}}$, $\hat{\rho}_{geo}^{\text{lab}}(t)$, $\hat{\rho}_{sea}^{\text{lab}}(t)$ and $\hat{\rho}_{surf}^{\text{lab}}(t)$, respectively, through a unitary transformation, such as $\hat{\rho}_{surf}^{\text{lab}}(t) = \hat{U}(t) \hat{\rho}_{surf} \hat{U}^\dagger(t)$. We proceed by showing how Eq. (12) leads to exact expressions for $\hat{\rho}_{eq}^{\text{lab}}$, $\hat{\rho}_{geo}^{\text{lab}}(t)$, $\hat{\rho}_{sea}^{\text{lab}}(t)$ and $\hat{\rho}_{surf}^{\text{lab}}(t)$ given in Eq. (2) in the main text. By introducing the property

$$\sum_{p, \sigma} \hat{G}_0 \hat{\Gamma}_p^\sigma \hat{G}_0^\dagger = i[\hat{G}_0 - \hat{G}_0^\dagger], \quad (13)$$
Eq. (12a) can be rewritten as the equilibrium density matrix (i.e., independent of $\hbar \omega$)

$$\dot{\rho}_{eq} = \frac{1}{2\pi} \int_{-\infty}^{+\infty} dE \{ \hat{G}_0 - \hat{G}_0^\dagger \} f = -\frac{1}{\pi} \int_{-\infty}^{+\infty} dE \text{Im} \hat{G}_0 f,$$

which reproduces $\dot{\rho}_{\text{eq}}^\text{ad} = \hat{U}(t)\dot{\rho}_{\text{eq}} \hat{U}^\dagger(t)$, shown in Eq. (2a) in the main text. To obtain the expression for $\dot{\rho}_{\text{geo}}(t)$, we insert Eq. (13) in Eq. (12b) to obtain

$$\dot{\rho}_{\text{geo}} = \frac{1}{2\pi} \int_{-\infty}^{+\infty} dE \hat{G}_0 \left( -i \hbar \dot{U}^\dagger \frac{\partial U}{\partial t} \right) \hat{G}_0^\dagger f + \frac{1}{2\pi} \int_{-\infty}^{+\infty} dE \hat{G}_0 \left( i \hbar \frac{\partial U^\dagger}{\partial t} \hat{U} \right) \hat{G}_0^\dagger f,$$

which leads to

$$\dot{\rho}_{\text{geo}} = \frac{1}{2\pi} \int_{-\infty}^{+\infty} dE \hat{G}_0 \left( -i \hbar \dot{U}^\dagger \frac{\partial U}{\partial t} \right) \hat{G}_0^\dagger f - \frac{1}{2\pi} \int_{-\infty}^{+\infty} dE \hat{G}_0 \left( i \hbar \frac{\partial U^\dagger}{\partial t} \hat{U} \right) \hat{G}_0^\dagger f + \frac{1}{2\pi} \int_{-\infty}^{+\infty} dE \hat{G}_0 \left( i \hbar \frac{\partial U^\dagger}{\partial t} \hat{U} \right) \hat{G}_0^\dagger f,$$

(16)

In the above result, if we use the property $\dot{U}^\dagger (\partial \dot{U} / \partial t) = -(\partial \dot{U}^\dagger / \partial t) \hat{U}$, then the second and third term cancel out. Furthermore, by using $\text{Im} \hat{O} = (\hat{O} - \hat{O}^\dagger) / 2i$, the sum of first and the fourth term furnishes

$$\dot{\rho}_{\text{geo}} = \frac{1}{2\pi} \int_{-\infty}^{+\infty} dE \text{Im} \left[ \hat{G}_0 \left( i \hbar \dot{U}^\dagger \frac{\partial U}{\partial t} \right) \hat{G}_0^\dagger f,\right]$$

which reproduces $\dot{\rho}_{\text{geo}}(t) = \hat{U}(t)\dot{\rho}_{\text{geo}} \hat{U}^\dagger(t)$, shown in Eq. (2b) in the main text. To obtain the expression for $\dot{\rho}_{\text{sea}}(t)$, we apply Eq. (13) on the first and second term of Eq. (12c), which yields

$$\dot{\rho}_{\text{sea}} = \frac{\hbar \omega}{2\pi} \int_{-\infty}^{+\infty} dE \sum_{p,q} \left( -Q_p^\dagger \frac{\partial \hat{\Sigma}_p}{\partial E} \right) \hat{G}_0^\dagger f + \frac{\hbar \omega}{2\pi} \int_{-\infty}^{+\infty} dE \sum_{p,q} \left( -Q_p^\dagger \frac{\partial \hat{\Sigma}_p}{\partial E} \right) \hat{G}_0^\dagger f,$$

and finally

$$\dot{\rho}_{\text{sea}} = \frac{\hbar \omega}{2\pi} \int_{-\infty}^{+\infty} dE \sum_{p,q} \left( -Q_p^\dagger \frac{\partial \hat{\Sigma}_p}{\partial E} \right) \hat{G}_0^\dagger f - \frac{\hbar \omega}{2\pi} \int_{-\infty}^{+\infty} dE \sum_{p,q} \left( -Q_p^\dagger \frac{\partial \hat{\Sigma}_p}{\partial E} \right) \hat{G}_0^\dagger f,$$

$$+ \frac{\hbar \omega}{2\pi} \sum_{p,q} \left[ -Q_p^\dagger \frac{\partial \hat{Q}_p}{\partial E} \right] \hat{G}_0^\dagger f,$$

(18)

In the above equation, the sum of second and third term cancels out the fifth term due to $\hat{Q}_p^\dagger = i(\hat{\Sigma}_p^\dagger - \hat{\Sigma}_p^\dagger)$). On the other hand, by using $\text{Im} \hat{O}$ and by substituting $Q_p^\dagger = -1/2$ and $Q_p^\dagger = +1/2$ in the sum of first and fourth term we obtain

$$\dot{\rho}_{\text{sea}} = -\frac{\hbar \omega}{2\pi} \int_{-\infty}^{+\infty} dE \text{Im} \left[ \hat{G}_0 \left( \frac{\partial \hat{\Sigma}_p^\dagger}{\partial E} - \frac{\partial \hat{Q}_p^\dagger}{\partial E} \right) \hat{G}_0^\dagger f \right],$$

(20)
which reproduces $\hat{\rho}_{\text{sea}}(t) = \hat{U}(t) \hat{\rho}_{\text{sea}} \hat{U}^\dagger(t)$, shown in Eq. (2c) in the main text. Lastly, we substitute $Q_p^\uparrow = -1/2$ and $Q_p^\downarrow = +1/2$ into Eq. (12d) to obtain

$$
\hat{\rho}_{\text{surf}} = \frac{\hbar \omega}{4\pi} \sum_p \int_{-\infty}^{+\infty} dE \hat{G}_0(\hat{\Gamma}_p^\uparrow - \hat{\Gamma}_p^\downarrow) \hat{G}_0 \frac{\partial f}{\partial E},
$$

which reproduces $\hat{\rho}_{\text{surf}}(t) = \hat{U}(t) \hat{\rho}_{\text{surf}} \hat{U}^\dagger(t)$, shown in Eq. (2d) in the main text.

[1] Z. Yuan, K. M. D. Hals, Y. Liu, A. A. Starikov, A. Brataas, and P. J. Kelly, Gilbert damping in noncollinear ferromagnets, Phys. Rev. Lett. 113, 266603 (2014).

[2] M. D. Petrović, B. S. Popescu, U. Bajpai, P. Plecháč, and B. K. Nikolić, Spin and charge pumping by a steady or pulse-current-driven magnetic domain wall: A self-consistent multiscale time-dependent quantum-classical hybrid approach, Phys. Rev. Applied 10, 054038 (2018).

[3] U. Bajpai and B. K. Nikolić, Time-retarded damping and magnetic inertia in the Landau-Lifshitz-Gilbert equation self-consistently coupled to electronic time-dependent nonequilibrium Green functions, Phys. Rev. B, 99, 134409 (2019).

[4] S. Zhang and S. S.-L. Zhang, Generalization of the Landau-Lifshitz-Gilbert equation for conducting ferromagnets, Phys. Rev. Lett. 102, 086601 (2009).

[5] K.-W. Kim, J.-H. Moon, K.-J. Lee, and H.-W. Lee, Prediction of giant spin motive force due to Rashba spin-orbit coupling, Phys. Rev. Lett. 108, 217202 (2012).

[6] F. Mahfouzi and B. K. Nikolić, How to construct the proper gauge-invariant density matrix in steady-state nonequilibrium: Applications to spin-transfer and spin-orbit torque, SPIN 03, 02 (2013).