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Quantifying non-Markovianity in magnetization dynamics via entropy production rates

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Magnetization dynamics is commonly described by the stochastic Landau-Lifshitz-Gilbert (LLG) equation. However, on picosecond timescales, inertial and open-system extensions of the LLG equation are necessary to interpret recent experiments. While it has been anticipated that these extensions make the magnetization dynamics non-Markovian, to date, the question of the extent of non-Markovianity has remained unanswered. Here, we provide a systematic quantification of the degree of non-Markovianity in these magnetization models using established measures. We find that, while the standard LLG equation exhibits strictly positive entropy production rates, inertial and open-system LLG dynamics display temporarily negative entropy production rates, which signal non-Markovian dynamics, as we demonstrate both analytically and numerically. Our numerical calculations show that experiments requiring the open-system LLG equation are more non-Markovian than those described by classical or inertial LLG equations.

Translational Brownian motion is one of the core concepts to describe fluids¹, gases^{2,3}, and a wide range of processes in statistical physics⁴, e.g., the random motion of micro-sized particles^{5,6} and molecules⁷ dispersed in aqueous solutions. Generally speaking, translational Brownian motion is modeled via a generalized Langevin equation, which includes memory effects, i.e., time-dependent friction, and colored noise fluctuations. These memory effects and fluctuations are linked by the fluctuation-dissipation theorem and lead to non-Markovian dynamics⁸. However, if the bath relaxes significantly faster than the system, $\tau_B \ll \tau_S$, where τ_B and τ_S are the bath and system relaxation times, respectively, the assumption of time-independent friction is sufficient^{9,10}, and the dynamics becomes memoryless, i.e., Markovian. When time-scale separation is broken, $\tau_B \sim \tau_S$, and energy from the bath may flow back into the system, the dynamics is non-Markovian^{9,11,12}. This is the case for systems that (strongly) interact with a structured or small thermal heat bath^{13–15}.

Here we consider a different kind of Brownian motion, namely rotational Brownian motion. In this case, the motion is constrained to the surface of a sphere rather than occurring in unconstrained Euclidean space as in translational Brownian motion^{16–18}. The rotational generalized Langevin equation¹⁶ is non-linear¹⁹ and the noise enters in a multiplicative way, making its analytical treatment more challenging^{20–22}. Such rotational motion occurs in nature, e.g. in the magnetization dynamics in ferromagnets.

For magnetization dynamics, the most commonly used stochastic equation is the Landau-Lifshitz-Gilbert (LLG) equation^{23,24}. It describes

damped precessional motion in an effective magnetic field²⁵ and has formed the theoretical basis for modeling experiments on spin dynamics for many decades.

Theoretical studies have proposed to extend the LLG equation by an inertial term^{26–35} to the so-called inertial LLG (iLLG) equation³⁶, which leads to an additional fast nutation oscillation on top of precessional motion. Time-resolved pump-probe experiments have started to detect and measure this nutation frequency on ultrafast timescales^{37,38}.

Further experiments on crystalline cobalt thin films have, however, measured dynamic properties that go beyond the theoretical description provided even by the iLLG equation^{39,40}. Specifically, ref. 40 shows that the frequency spectrum of crystalline cobalt thin films is well captured by a *generalized* (here referred to as “general” in the sense of the generalized Langevin equation for translational Brownian motion) magnetization dynamics equation, which takes the coupling between the magnetic system and the phononic bath explicitly into account. Here, a full memory kernel governs the dissipation of the magnetic system. As the equation has been derived from an open-quantum system approach⁴¹, we refer to it as the open-system LLG equation (os-LLG).

The experimental signatures in ref. 40 arise from a broken time-scale separation between system and bath degrees of freedom on the measured ultrashort timescales, which makes a non-Markovian description necessary. What remains an open question is the quantification of the amount of non-Markovianity in the different magnetization dynamics, i.e., if an experiment

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shows signatures consistent with the iLLG equation, will it be more or less non-Markovian than an experiment consistent with the os-LLG equation?

Thermodynamic entropy production rates (EPR) are a tool to investigate and quantify non-Markovianity in ultrafast magnetization dynamics^{42–44}. In quantum systems, the entropy production rate $\dot{\Sigma}(t)$ can be negative due to quantum fluctuations, entanglement and correlations between system and bath, and backflow of energy^{45–48}. In classical systems, negative entropy production rates indicate a non-Markovian evolution if the system is undriven and local detailed balance holds⁴⁹. The entropy production rate can be defined via the relative entropy D (often also referred to as Kullback-Leibler divergence)

$$D[p_\alpha(t)||\pi_\beta] = \sum_\alpha p_\alpha(t) \ln \frac{p_\alpha(t)}{\pi_\beta}, \quad (1)$$

where k_B is the Boltzmann constant. It is a measure of the distinguishability between two probability distributions p_α and here the thermal equilibrium state π_β , which is the fixed point of the dynamics and necessary for the calculation of the entropy production rate⁴⁹. It is defined as $\pi_\beta = e^{-\beta E_\alpha} / \mathcal{Z}$, where β and $\mathcal{Z} = \sum_\alpha e^{-\beta E_\alpha}$ are the inverse temperature and the partition function, respectively⁴⁹. The energy of the system in state α is denoted by E_α . The negative time derivative of the relative entropy defines then the entropy production rate $\dot{\Sigma}$ ^{49,50}

$$\dot{\Sigma}(t) = -k_B \partial_t D[p_\alpha(t)||\pi_\beta]. \quad (2)$$

The following theorem linking entropy production rates and non-Markovianity was proven by Strasberg and Esposito⁴⁹: If a classical system is undriven, local detailed balance holds, and the total system is initialized within the set of conditional steady states, then the occurrence of negative entropy production rates indicates that the system undergoes a non-Markovian evolution. Note that positive entropy production rates do not necessarily imply purely Markovian dynamics.

In this work, we determine the entropy production rates^{45,49,51,52} of the LLG, iLLG, and os-LLG equations and use them to detect and quantify the non-Markovianity of each magnetization dynamics, thereby establishing a direct link between magnetism and concepts from stochastic thermodynamics and statistical physics. We analytically and numerically show that the LLG equation only leads to positive entropy production rates⁵³. In contrast, we show that the entropy production rates of the iLLG and os-LLG equations can be negative, signaling a non-Markovian evolution⁴⁹. By employing two different non-Markovianity measures, “negative EPR window”⁵⁴ and “relative EPR window”, we quantitatively compare the differences in the LLG, iLLG, and os-LLG equation dynamics.

We consistently find that the magnitude of non-Markovianity is largest for the os-LLG equation, which indicates that it is the most non-Markovian of the magnetization dynamics compared here. While the LLG equation exhibits zero non-Markovianity regardless of the initial conditions, the iLLG equation exhibits non-Markovianity for a canted orientation between the external field and initial magnetization. This indicates a dependence of the non-Markovianity on the initial conditions.

Results

Ultrafast magnetization dynamics

Stochastic magnetization dynamics on ultrafast timescales is commonly described by the LLG equation

$$\dot{\mathbf{m}} = -\gamma \mathbf{m} \times (\mathbf{B}_{\text{eff}} + \mathbf{b}_{\text{th}}(t) + \alpha \mathbf{m}) \quad (3)$$

where $\mathbf{m}$ is the unit-free, normalized magnetization vector, which precesses because the torque interaction with the effective magnetic field $\mathbf{B}_{\text{eff}}$ scaled by the electronic gyromagnetic ratio γ . This field can consist of any combination of a constant external bias field, a possibly time-dependent external field, and any anisotropy field arising from the material and magnetization itself. The damping in the third term is scaled by the Gilbert damping

parameter α . The thermal noise is denoted by $\mathbf{b}_{\text{th}}$ and is Gaussian white noise with

$$\begin{aligned} \langle \mathbf{b}_{\text{th}}(t) \rangle &= 0 \\ \langle b_{\text{th},i}(t) b_{\text{th},j}(t') \rangle &= \eta \delta_{ij} \delta(t - t'), \end{aligned}$$

where the correlation factor is $\eta = \alpha k_B T$.

The stochastic LLG Eq. (3) can be extended by an inertial term^{26–33} which is scaled by the angular momentum relaxation time τ and describes an additional nutation motion

$$\dot{\mathbf{m}} = -\gamma \mathbf{m} \times (\mathbf{B}_{\text{eff}} + \mathbf{b}_{\text{th}}(t) + \alpha \mathbf{m} + \alpha \tau \ddot{\mathbf{m}}). \quad (4)$$

Both of these stochastic magnetization Eqs. (3) and (4) are contained in the os-LLG equation⁴¹, which has been shown to successfully describe higher-order oscillations in ultrafast dynamics⁴⁰ and to produce the correct equilibrium magnetization⁵⁵. The os-LLG equation arising from an isotropic coupling of the magnetic system with a thermal bath is given by

$$\dot{\mathbf{m}} = -\gamma \mathbf{m} \times \left(\mathbf{B}_{\text{eff}} + \tilde{\mathbf{b}}_{\text{th}}(t) + \int_{-\infty}^t dt' \mathcal{K}(t - t') \mathbf{m}(t') \right), \quad (5)$$

where $\tilde{\mathbf{b}}_{\text{th}}$ describes again the thermal fluctuations of the bath. However, these fluctuations are now colored (for better clarity, denoted by a tilde), and $\mathcal{K}(t - t')$ encodes the memory of the magnetization dynamics. The thermal fluctuations and the dissipative memory kernel are linked by the fluctuation-dissipation theorem, where the memory kernel is given by $\mathcal{K}(\tau) = 2\pi\Theta(\tau) \int_0^\infty d\nu I(\nu) \sin(2\pi\nu\tau)$ with the spectral density $I(\nu)$.

It is insightful to understand that the LLG (3) and inertial LLG (4) can be obtained from the open-system LLG (5) by expanding the kernel integral in powers of magnetization and its time derivatives⁴¹. The first expansion term then gives the Gilbert damping $\sim \mathbf{m} \times \dot{\mathbf{m}}$, while the second expansion term yields the inertial correction $\sim \mathbf{m} \times \ddot{\mathbf{m}}$ ^{33–35}. We remark that such an expansion needs to be treated carefully⁵⁶ and is only a good approximation for very short memory times. In the absence of a clear separation between spin and phonon timescales, as encountered in magnetic insulators^{56,57}, such an expansion is expected to fail. Whereas the memory kernel associated with electronic baths generally admits a time-local expansion because a sufficient separation of timescales is common^{58,59}. In this manuscript, simulations of Eq. (5) include the full memory kernel as parameterized below.

We note that all three LLG equations preserve the magnitude of magnetization based on their structure. Without loss of generality, we choose $|\mathbf{m}| = 1$ for all equations.

Analytical bounds for entropy production rates

We now analyze *contractivity*⁴⁹ for the entropy production rates of the different magnetization dynamics analytically. Contractivity states that two probability distributions will monotonically come closer in time for all times, implying

$$\dot{\Sigma}(t) \geq 0, \quad (6)$$

with $\dot{\Sigma}(t) = 0$ in thermal equilibrium. Contractivity is therefore directly linked to the theorem formulated by Strasberg and Esposito⁴⁹, when contractivity is broken, the dynamics is non-Markovian.

For the LLG Eq. (3), we prove that the entropy production rate is always positive, i.e., the magnetization dynamics is contractive. To show this, consider the definition of the Fokker-Planck equation for the stochastic LLG equation¹⁷

$$\begin{aligned} \frac{\partial p(\mathbf{m}, t)}{\partial t} &= -\nabla_{\mathcal{S}^2} \cdot (\mathbf{A}(\mathbf{m}) p(\mathbf{m}, t)) \\ &\quad + \frac{1}{2} \nabla_{\mathcal{S}^2} \cdot (\mathcal{D}(\mathbf{m}) \nabla_{\mathcal{S}^2} p(\mathbf{m}, t)), \end{aligned} \quad (7)$$

where $\mathbf{A}(\mathbf{m}) = -\gamma \mathbf{m} \times \mathbf{B}_{\text{eff}} - \gamma \alpha \mathbf{m} \times (\mathbf{m} \times \mathbf{B}_{\text{eff}})$ is the drift vector field of the deterministic part of the dynamics and $\mathcal{D}(\mathbf{m}) = 2\alpha k_B T (\mathbb{1} - \mathbf{m} \mathbf{m}^T) / \gamma$ is

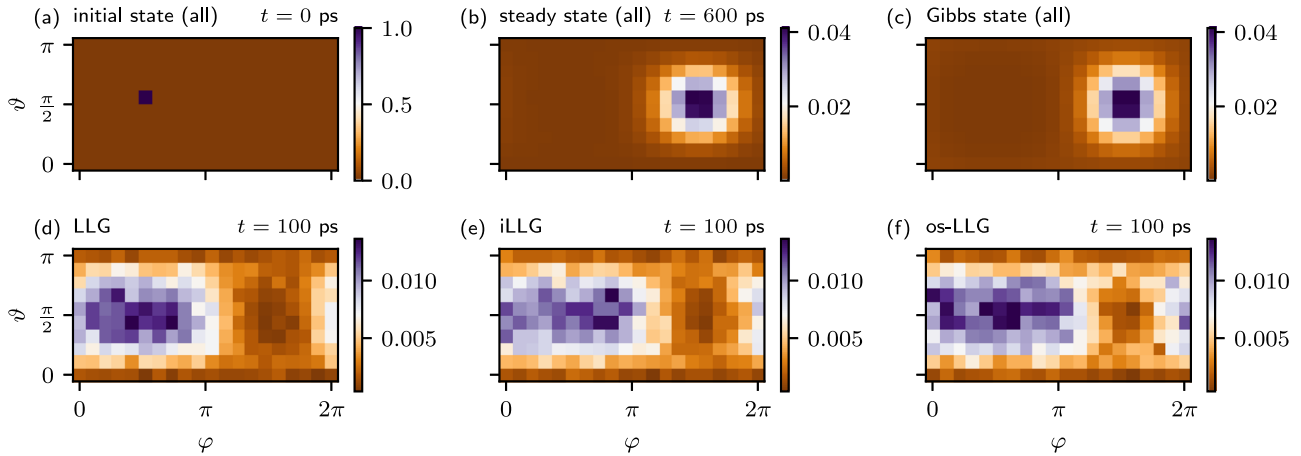


Fig. 1 | Probability distributions of the magnetization vector in spherical coordinates for LLG, iLLG, and os-LLG. **a** For all three equations, the magnetization is initialized in the parallel configuration. **b** The steady-state distribution is obtained by time-averaging the distribution once the system has equilibrated, here exemplarily shown for the LLG equation. **c** Analytically calculated Gibbs state $\pi_\beta(\mathbf{m})$, which

matches the numerically obtained steady-state of each of the magnetization equations. **d–f** show the magnetization distributions after $t = 100$ ps for the LLG, iLLG, and os-LLG equations, respectively, evidencing differences in the temporal evolution. All six panels show the distribution of the magnetization in spherical coordinates (φ, ϑ) for a fixed magnetization magnitude of $|\mathbf{m}| = 1.0$.

the diffusion tensor (for details see Supplementary Note 1). The operator ∇_{S^2} denotes differentiation with respect to the surface of the unit sphere S^2 , and $p(\mathbf{m}, t)$ is the probability distribution for magnetization $\mathbf{m}$ at time t .

The entropy production rate Eq. (2) in its continuous version with integration over the full phase-space S^2 reads

$$\frac{\dot{\Sigma}(t)}{k_B} = -\frac{\partial D(t)}{\partial t} = -\int_{S^2} \ln \left(\frac{p(\mathbf{m}, t)}{\pi_\beta(\mathbf{m})} \right) \frac{\partial p(\mathbf{m}, t)}{\partial t} d\Omega. \quad (8)$$

Plugging the Fokker-Planck equation (7) of the LLG equation into Eq. (8) yields a quadratic form, such that $\dot{\Sigma}(t) \geq 0$ for all times t . Details of this proof are given in Supplementary Note 1.

The iLLG Eq. (4), on the other hand, does not always lead to a contractive dynamics as defined in Eq. (6). This is because the full process is no longer described by the (marginal) probability density $p(\mathbf{m}, t)$ but by a probability density $P(\mathbf{m}, \mathbf{v}, t)$, which arises from the second-order time derivative in Eq. (4). By integrating over the “velocity” variable $\mathbf{v}$, using the Mori-Zwanzig formalism^{9,60} as done in Supplementary Note 2, the complete parameter space is reduced and information about the dynamics is lost. Therefore, the evolution of the marginal probability density $p(\mathbf{m}, t)$ is no longer time-local, and memory effects are introduced. Hence, the iLLG Eq. (4) describes a non-Markovian evolution of the magnetization vector $\mathbf{m}$ and, in general, $\dot{\Sigma}(t) \geq 0$ no longer holds.

Similar to the iLLG case, for the os-LLG Eq. (5), the full probability density is a higher-dimensional function $P(\mathbf{m}, \mathbf{v}, \mathbf{w}, t)$. Thus, a similar argument can be constructed to show that the os-LLG Eq. (5) leads to a non-Markovian evolution for the magnetization $\mathbf{m}$ alone (see Supplementary Note 2). Having established that both iLLG and os-LLG can break contractivity, we will quantify their degree of non-Markovianity below.

Non-Markovianity measures

To do so, we need non-Markovianity measures. Here, we choose to quantify the degree of non-Markovianity using the entropy production rate. We employ the following, previously characterized and used measure⁵⁴, which we refer to as “negative EPR window”

$$\mathcal{N}_A = -\int_{\dot{\Sigma}(t) < 0} \dot{\Sigma}(t) dt, \quad (9)$$

where the time integration is over all intervals in which $\dot{\Sigma}(t) < 0$. Consequently, for a Markovian process, where $\dot{\Sigma}(t) \geq 0$ for all times, we have

$\mathcal{N}_A = 0$. This measure also reflects the absolute strength of the relative entropy, which can be of interest but makes comparison between different setups difficult. Therefore, we also introduce the normalized version as a non-Markovian measure, here called “relative EPR window”,

$$\overline{\mathcal{N}}_A = -\frac{\int_{\dot{\Sigma}(t) < 0} \dot{\Sigma}(t) dt}{\int |\dot{\Sigma}(t)| dt}, \quad (10)$$

where we have normalized by the total time integral of the absolute value of the entropy production rate, which is equal to the absolute area of the entire entropy production rate curve. This leads to a measure that is independent of the absolute scale of the relative entropy. By definition, $\overline{\mathcal{N}}_A$ takes values between $[0, 1]$ with $\overline{\mathcal{N}}_A > 0$ indicating non-Markovian dynamics.

There are other Markovianity measures⁶¹, which we leave as tools for future work. Two examples, counting timesteps with negative entropy production rate and counting the number of sign switches of the entropy production rate, are briefly discussed in Supplementary Note 5.

Entropy production rates and non-Markovianity for different magnetization dynamics

In what follows, we numerically investigate the relative entropy and the entropy production rate as defined in Eqs. (1) and (2) and quantify non-Markovianity via the negative and relative EPR windows. We simulate $N_{\text{traj}} = 50000$ trajectories of the LLG (3), iLLG (4) and os-LLG (5) equations with realistic parameter sets for cobalt thin films measured in ultrafast magnetization dynamics experiments^{39,40}. For the simulations, we make use of the SPIDY package⁶².

To specify the dynamics of the os-LLG equation, we need to choose a spectral density $I(\nu)$, which details the coupling of the system to the bath and relates to the memory kernel as detailed above. As in previous works, we use a Lorentzian spectral density^{40,63}, $I(\nu) = \mathcal{A} \nu / ((\nu_0^2 - \nu^2)^2 + \nu^2 \Gamma^2)$, where ν_0 denotes the central frequency and Γ the width of the Lorentzian. The amplitude $\mathcal{A}$ is proportional to the strength of the system-bath interaction and the dissipation. The Lorentzian spectral density describes, e.g., the coupling of the magnetization to modes of the phonon bath. The response in an ultrafast experiment⁴⁰ showed a Fourier spectrum with multiple peaks, which is matched by the os-LLG equation (5) with a specific set of Lorentzian parameters. We here use the same parameters to quantify the degree of non-Markovianity that occurred in the experiment. The experimentally required Lorentzian parameters for Eq. (5) are $\mathcal{A} = 242 \text{ THz}^3$, $\nu_0 = 4.2 \text{ THz}$, and $\Gamma = 0.2 \text{ THz}$, see ref. 40. They relate to the parameters of the LLG (3) and iLLG (4) via $\alpha = \mathcal{A} \Gamma / \nu_0^4$ and $\tau = (\nu_0^2 - \Gamma^2) / (2\pi \nu_0^2 \Gamma)$, which follows from

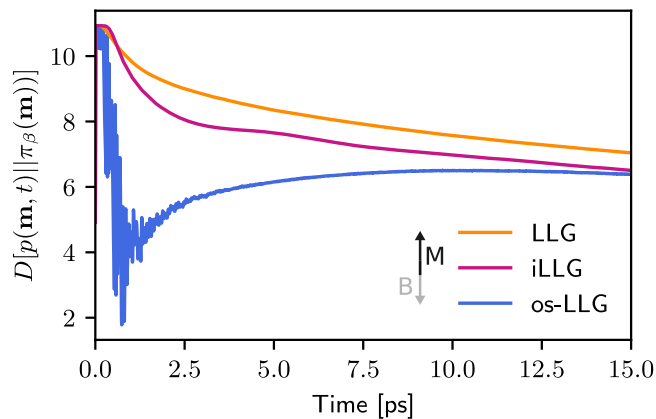


Fig. 2 | Relative entropy of the LLG, iLLG, and os-LLG equations. The three equations of motion are initialized in the parallel configuration. The damping parameter is always $\alpha = 0.15$ and the inertial parameter is $\tau = 0.8$ ps for the simulations of iLLG and os-LLG (with Lorentzian parameters fixed as $\nu_0 = 4.2$ THz, $\Gamma = 0.2$ THz and $\mathcal{A} = 242$ THz³, see ref. 40). For all $N_{\text{traj}} = 20000$ trajectories the temperature is $T_{\text{sim}} = 0.1$.

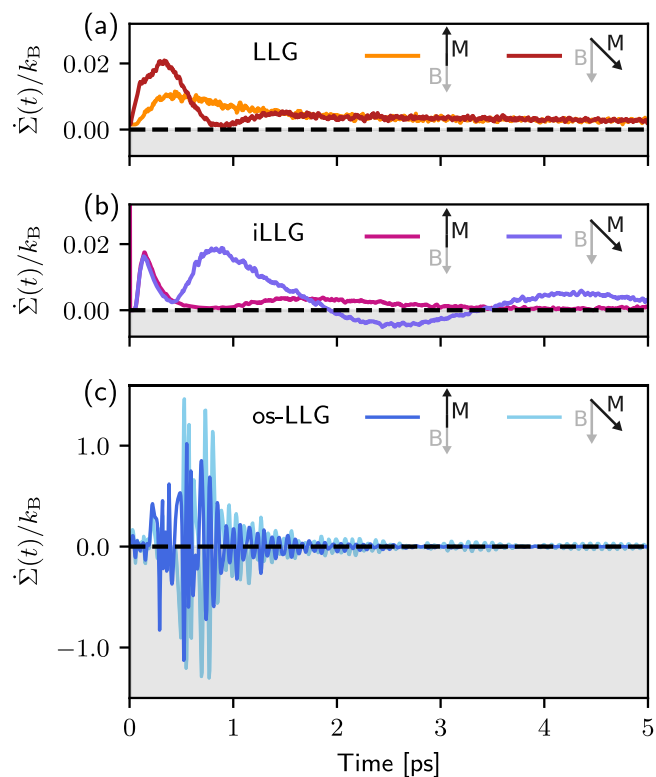


Fig. 3 | Entropy production rate of the LLG, iLLG, and os-LLG equation. **a** Entropy production rates of the LLG equation for parallel (orange) and canted (red) configurations stay positive for all times, while **b** for the iLLG equation, the canted (purple) configuration leads to temporarily negative entropy production rates. **c** The entropy production rates of the os-LLG equation oscillate quickly between positive and negative for both (parallel (dark blue) and canted (light blue)) configurations. The grey region highlights violations of contractivity. Traces shown are after taking a moving average over five data points that suppresses the fluctuations of the limited statistics (50,000 trajectories). The remaining parameters correspond to those in Fig. 2.

the first and second order expansion of Eq (5). Hence, we have a damping parameter of $\alpha = 0.15$ and an inertial parameter of $\tau = 0.8$ ps³⁹.

In the following, we will characterize the dynamics starting from two initial configurations, which capture two different geometries and field

strengths:

- parallel : $\mathbf{m}_0 = (0, 1.0, 0)$, $\mathbf{B}_{\text{eff}} = -0.35T \hat{\mathbf{y}}$;
- canted : $\mathbf{m}_0 = (\sqrt{0.5}, -\sqrt{0.5}, 0)$, $\mathbf{B}_{\text{eff}} = -1.5T \hat{\mathbf{y}}$.

The first configuration starts with anti-parallel alignment between $\mathbf{B}_{\text{eff}}$ and $\mathbf{m}_0$, while the canted configuration has a 45° angle between field and magnetization, as well as a stronger $\mathbf{B}_{\text{eff}}$. In all simulations, we choose a unitless temperature of $T_{\text{sim}} = 0.1$.

In order to be able to use the negative EPR to non-Markovianity link established by the theorem of ref. 49 (see Introduction), the initial conditions of the simulations need to be in the class of conditional steady states. We therefore selected initial conditions accordingly for the numerical results discussed below. A detailed discussion of how the initial conditions are obtained is given in Methods.

We obtain the probability distribution of the magnetization $p(\mathbf{m}, t)$ by transformation of the normalized magnetization vector into spherical coordinates $\mathbf{m} = (r = |\mathbf{m}|, \varphi, \vartheta)$ with $\varphi \in [0, 2\pi)$ and $\vartheta \in [0, \pi]$, and $|\mathbf{m}| = 1$ remains unchanged by the dynamics. Next, we discretise the phase-space (φ, ϑ) by $\varphi/30$ and $\vartheta/15$ to obtain 450 bins (see Methods) and count at each time step t_n how often $\mathbf{m}$ lies within a certain bin. The total number of trajectories normalizes the counts to get a properly normalized probability distribution $p(\mathbf{m}, t)$, as shown in Fig. 1.

We start with a fixed initial magnetization (Fig. 1a) and let the system evolve until it reaches equilibrium. Regardless of the equation that governs the dynamics, Fig. 1b, c illustrate that the numerically obtained steady state (after 600 ps) is the Gibbs state, i.e., the expected magnetization configuration in thermal equilibrium (for further details see Methods). To calculate the relative entropy, we therefore use the Gibbs state $\pi_\beta(\mathbf{m})$, which is known analytically and whose numerically binned version we can obtain directly. The intermediate magnetization configurations (after 100 ps) for the LLG, iLLG and os-LLG equations are shown in Fig. 1d, e and f, respectively. They differ and therefore confirm that the three equations describe different dynamics. Knowing $p(\mathbf{m}, t)$ at each time step t_n , we calculate the relative entropy $D(p(\mathbf{m}, t) || \pi_\beta(\mathbf{m}))$ and the entropy production rate $\dot{S}(t)$ for all three simulated equations of motion.

The calculated relative entropy for the parallel configuration during the first 15 ps is shown in Fig. 2. As expected from the analytical discussion above, we observe very different behavior of the relative entropy during the first ≈ 15 ps of the three different magnetization equations. While the relative entropy for the LLG equation decreases monotonously, the relative entropy for the iLLG equation shows small variations, and the relative entropy for the os-LLG equations is clearly non-monotonous, where the high-frequency oscillations stem from the memory kernel in Eq. (5). All three cases become strictly monotonously declining for larger times, $t > 15$ ps (see Methods), indicating that the non-Markovian dynamics is present only on short, “ultrafast” timescales. As we show in the Methods, the overall magnitude of the numerically obtained relative entropy depends on the binning we choose. Therefore, what matters here are the relative differences between the three LLG-type dynamics Eqs. (3)–(5).

In Fig. 3, we compare the calculated entropy production rates $\dot{S}(t)$ for all three magnetization dynamics equations and both parameter configurations. Since the distribution $p(\mathbf{m}, t)$ is made up of a finite sample of $N_{\text{traj}} = 50,000$ trajectories, the numerically obtained entropy production rate shows visible fluctuations. To reduce the noise, we apply a moving time average on the entropy production rate over five data points. While some sampling noise remains, one can see that the entropy production rate of the LLG equation stays strictly positive for both configurations (staying above the black dashed line in Fig. 3a), while the entropy production rate of the iLLG temporarily becomes negative for the canted configuration, but stays strictly positive for the parallel one (see Fig. 3b). This is mainly attributed to the torque in the magnetic field that the magnetization vector initially experiences, which is larger for the canted configuration than the parallel one^{26,37}. For the os-LLG Eq. (5), the entropy production rate becomes temporarily negative in both configurations and changes sign frequently

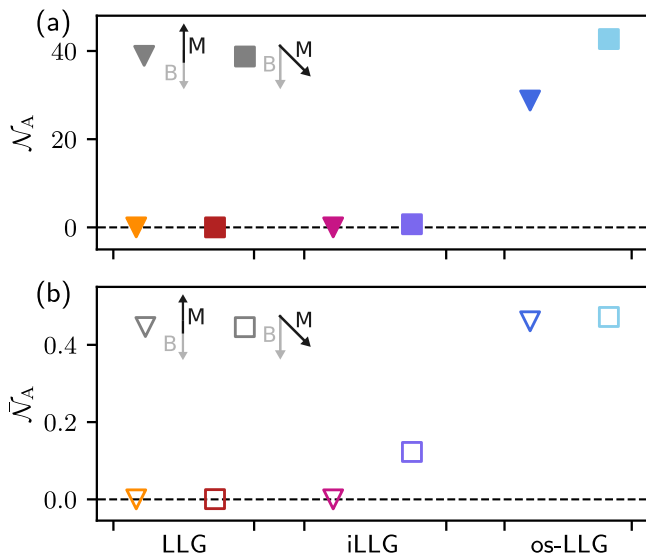


Fig. 4 | Magnitude of non-Markovianity. For the three different magnetization dynamics equations and the parallel (triangles) and canted (squares) configurations, we calculate (a) the non-Markovianity measures $\mathcal{N}_A$ Eq. (9) (filled symbols) (b) and $\bar{\mathcal{N}}_A$ Eq. (10) (empty symbols). For both measures, they are zero for both configurations in the LLG case (orange, red). In the iLLG case, they are zero for the parallel (pink) configuration but non-zero for the canted (purple) configuration. In the os-LLG case (dark blue, light blue), both non-Markovianity measures are non-zero for both configurations and significantly larger than in the iLLG case. The difference between the configurations is larger in the iLLG case, but smaller in the os-LLG case. The measures $\mathcal{N}_A$ and $\bar{\mathcal{N}}_A$ are evaluated over the first 12 ps of the dynamics for the trajectories shown in Fig. 3, to which the colors of the markers are matched accordingly.

(Fig. 3c). The magnitude of the entropy production rate is larger than that of the LLG and iLLG equations, which is due to the dynamics of the total system-bath setup.

To further quantify the differences in the entropy production rate, we employ the discrete versions of Eq. (9) and Eq. (10) to calculate the negative EPR window $\mathcal{N}_A$ and the relative EPR window $\bar{\mathcal{N}}_A$ from the entropy production rates presented in Fig. 3. The resulting values are plotted in Fig. 4. For the parallel configuration, we find that both EPR windows are zero for the LLG and iLLG equations, while it is non-zero for the os-LLG equation. For the canted configuration, only the LLG equation has a zero measure, while iLLG and os-LLG have finite values. The negative EPR window of the os-LLG equation has a value of $\mathcal{N}_A \sim 30$, which is one order of magnitude larger than for the iLLG equation $\mathcal{N}_A \sim 2$.

We remark that we have introduced a small threshold of $\epsilon = -0.001$, such that we only counted the regions in which $\dot{\Sigma}(t) < \epsilon$ holds. This is again to reduce fluctuations stemming from the finite number of simulated trajectories.

The relative EPR window $\bar{\mathcal{N}}_A$ additionally reveals that the non-Markovianity in the iLLG is significantly stronger for the canted configuration than for the parallel one. The results for $\bar{\mathcal{N}}_A$ also show that the non-Markovianity of the os-LLG does not differ as much between parallel and canted configurations as it does for the iLLG case. This underlines the non-Markovianity of the os-LLG beyond the inertial term, because the stronger excitation of the inertial term in the canted configuration does not lead to significant changes in the non-Markovianity.

The numerical results exemplify and complete the analytical results discussed above and in Supplementary Note 1 and Supplementary Note 2. If the magnetic system of interest is assumed to evolve via the standard LLG Eq. (3), it must show a strictly positive entropy production rate. In contrast, if the magnetic system of interest exhibits a negative entropy production rate, the system undergoes non-Markovian evolution, and memory effects must be accounted for, e.g., using the os-LLG equation.

Discussion

In summary, our manuscript uses the thermodynamic concept of negative entropy production rates to quantify non-Markovianity in magnetization dynamics on ultrashort timescales. We use three different equations of motion, i.e., LLG Eq. (3), iLLG Eq. (4), and os-LLG Eq. (5), to compare their non-Markovianity. Analytical arguments show that the LLG equation always has a positive entropy production rate, whereas the entropy production rates of the iLLG and os-LLG equations may temporarily become negative, depending on the initial conditions and magnetic field strengths.

The numerically calculated relative entropy of all three equations directly shows that they differ on short timescales (up to approximately 10 ps), while they converge in the long-time limit. Using two non-Markovianity measures Eqs. (9) and (10), which are based on the negativity of the entropy production rates, confirmed that the os-LLG Eq. (5) shows the largest magnitude of non-Markovianity. This evidences that by including a memory kernel in the os-LLG equation, the magnetization dynamics can become non-Markovian^{40,41,64}. Similarly, the iLLG Eq. (4) has a finite non-Markovianity measure under specific initial conditions. The quantitative results, therefore, agree with the analytical results and provide a platform for relative comparison of non-Markovianity in different dynamics. Since the calculations are based on experimental parameters, the quantification allows to compare independent experiments.

We note again that the details of the relative entropy and entropy production rate depend on the material parameters, the initial condition, the temperature and the statistics of the thermostat⁵⁵, and the different internal and external magnetic fields. However, whenever the magnetization dynamics is found to yield a negative entropy production rate (given the dynamics is undriven, detailed balance holds, and the fixed point of the dynamics is known), the correct theoretical framework cannot be the LLG equation.

While non-Markovianity in magnetic systems has been studied at least to some extent theoretically^{16,34,64–67}, only recently has this been connected to an experiment on ultrafast timescales⁴⁰. Further ultrafast experiments in (magnetic) solid-state materials may probe and detect non-Markovian evolution of magnetic or non-magnetic degrees of freedom in the future. Timescale separation is broken, for example, in experiments with strong magnon-phonon coupling⁶⁸ and (nonlinear) phonon-phonon interactions^{69–71}. One promising direction to experimentally access the entropy production is to use hot-electron bolometers⁷². Another is based on ultrafast thermodynamics, which links available measurements of magnetization or lattice dynamics directly to entropy production^{42,43}. This could reveal new physics as well as lead to new technological applications.

Future theoretical work is needed to address additive noise^{43,73} instead of multiplicative noise^{16,41,74}, extending the fluctuations by a longitudinal component, and to extend this classical analysis to the quantum regime^{49,75,76}.

Methods

Conditional steady states

For the theorem in ref. 49 to hold, we need to make sure that the initial conditions of the LLG, iLLG, and os-LLG equations are in the set of conditional steady states, which is defined as

$$\pi_{B|S} = \frac{e^{-\beta(H_B+V)}}{\int d\mathbf{x}_B e^{-\beta(H_B+V)}} = \frac{1}{\mathcal{Z}_B} e^{-\beta(H_{B|S}-H_S)}. \quad (11)$$

where the classical system-bath Hamiltonian is given by $H_{\text{tot}} = H_S + H_B + V_{\text{int}}$. For the os-LLG equation the system, bath, and interaction Hamiltonians are given as⁴¹

$$\begin{aligned} H_S &= -\omega_L \mathbf{m} \cdot \mathbf{B}_{\text{eff}}, \\ H_B &= \frac{1}{2} \int_0^\infty d\omega (\mathbf{p}_\omega^2 + \omega^2 \mathbf{x}_\omega^2), \\ V_{\text{int}} &= \mathbf{m} \cdot \int_0^\infty d\omega \sqrt{I(\omega)} \omega \mathbf{x}_\omega. \end{aligned}$$

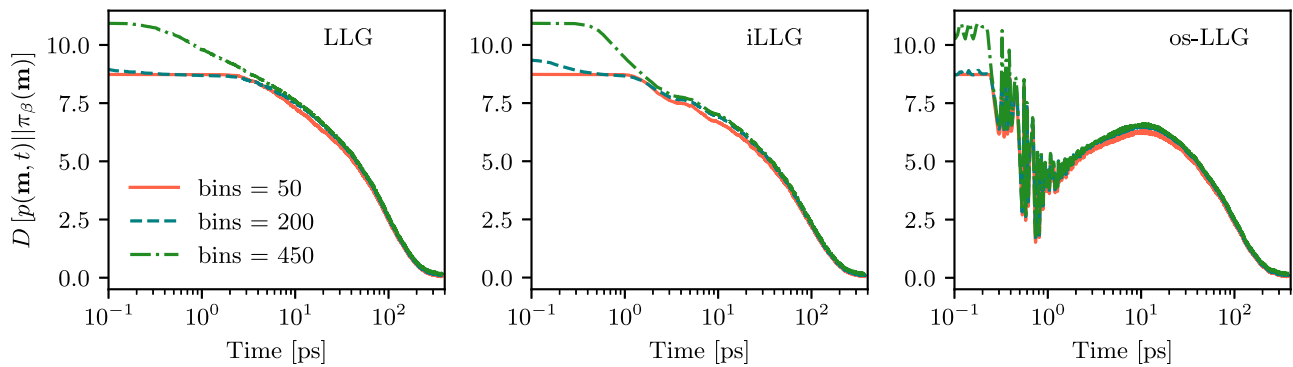


Fig. 5 | Relative entropy for different levels of coarse-graining. Left panel: The relative entropy over time until the system has arrived at equilibrium (i.e., $D(t) = 0$ and $\dot{D}(t) = 0$). The system evolves via the standard LLG equation, but the resolution of the coarse-graining is increased from 50 (orange solid line) to 200 (blue dashed

line) to 450 (green dash-dotted line) bins. Center panel and right panel: The same as in the left panel, but the system evolves via iLLG and os-LLG equations, respectively. The initial condition and the magnetic field are fixed by the parallel parameter configuration. The relative entropy is calculated for $N_{\text{trac}} = 2000$ trajectories.

After completing the square, the conditional steady state is given by

$$\pi_{\text{B|S}} = \frac{1}{Z_{\text{B}}} \exp \left(-\beta \frac{1}{2} \int_0^\infty d\omega \left(\mathbf{p}_\omega^2 + \omega^2 \left(\mathbf{X}_\omega + \frac{\sqrt{I(\omega)} \mathbf{m}}{\omega^{3/2}} \right)^2 - \frac{I(\omega) \mathbf{m}^2}{2\omega} \right) \right), \quad (12)$$

where in equilibrium the bath oscillators are displaced by $\mathbf{X}_{\text{eq}} = -\sqrt{I(\omega)} \omega^{-3/2} \mathbf{m}$, due to the interaction with the magnetization vector $\mathbf{m}$. The last term can be absorbed into the bath partition function Z_{B} . This is translated into the equations of motions (Eqs. (S41)–(S42)), which we solve numerically, by noticing that $\mathbf{v}$ is an effective bath position, defined as

$$\mathbf{v} = \int_0^\infty d\omega \sqrt{I(\omega)} \mathbf{X}_\omega. \quad (13)$$

For a bath in equilibrium we have $\mathbf{X}_\omega \rightarrow \mathbf{X}_{\text{eq}} = \sqrt{I(\omega)} \omega^{-3/2} \mathbf{m}$, such that

$$\mathbf{v}_{\text{eq}} = \mathbf{m} \int_0^\infty d\omega \frac{I(\omega)}{\omega} = \frac{\mathcal{A} \mathbf{m}}{\omega_0^2}, \quad (14)$$

for the Lorentzian spectral density introduced in the main text. Therefore, we find that the initial conditions in Eqs. (S41)–(S42) have to be chosen as $\mathbf{v}_0 = \mathcal{A} \mathbf{m} / \omega_0^2$ and $\mathbf{w}_0 = (0, 0, 0)$. Alternatively, one can deduce this by requiring that $\dot{\mathbf{v}} = \dot{\mathbf{w}} = 0$ in the conditional steady state, which leads to the same initial conditions.

In other words, we need to hold the magnetization fixed at $\mathbf{m}_0$ until the bath has equilibrated (adapted) itself to this new magnetization direction. At finite temperature, $T_{\text{sim}} > 0$, the initial conditions are drawn from a Gaussian distribution around $\mathbf{v}_0$ and $\mathbf{w}_0$ with variance $\sigma_v = T_{\text{sim}} / \omega_0^2$ and $\sigma_w = T_{\text{sim}}$, respectively.

For the inertial LLG equation, the initial spin velocity $\mathbf{v}_0$ needs to be fixed, which can not be explicitly written in the open-system framework as above. By keeping the initial magnetization $\mathbf{m}_0$ fixed, one finds that the conditional steady state is given by a Gaussian distribution of the tangent plane at $\mathbf{m}_0$,

$$\pi_{\mathbf{v}|\mathbf{m}_0} \propto \exp(-\beta \alpha \mathbf{v}^2). \quad (15)$$

Therefore, the initial spin velocity $\mathbf{v}_0$ is given by a Gaussian in the tangent plane at $\mathbf{m}_0$ with variance $\sigma_v = T_{\text{sim}} / (\alpha \tau)$.

For the LLG equation, no additional degrees of freedom are needed to simulate the dynamics; hence, $\mathbf{m}_0$ is enough to initialize the system. In other words, for the LLG and inertial LLG equations, the bath equilibrates instantaneously. With the choice of these initial conditions, we satisfy the requirement of being in the set of conditional steady states from ref. 49, and

we can directly deduce that negative entropy production rates indicate non-Markovian dynamics.

Mean force Gibbs state

Knowing the steady state of the system is important because it is the fixed point of its dynamics and therefore necessary for the calculation of the relative entropy⁴⁹. Generally speaking, in strong-coupling thermodynamics^{77–79}, the steady state is not always the Gibbs state $\pi_\beta = e^{-\beta E_a} / Z$, with β and $Z = \sum_a e^{-\beta E_a}$ being the inverse temperature and the partition function, respectively, and mean force corrections to the Gibbs state are relevant⁸⁰. However, for a classical three-dimensional *isotropic* spin-boson system, i.e., the underlying open system model for the os-LLG Eq. (5), the steady state is always given by the Gibbs state π_β . This holds for any coupling strength⁸¹. Therefore, we do not need to further specify the coupling strength here (see Fig. (1)).

Influence of coarse-graining on the relative entropy

Here, the influence of the coarse-graining on the numerically calculated relative entropy is discussed. The relative entropy $D[p(\mathbf{m}, t) || \pi_\beta(\mathbf{m})]$ of the LLG, iLLG, and os-LLG equations for a coarse-graining of 50 (orange solid line), 200 (blue dashed line), and 450 (green dash-dotted line) bins (see Fig. 5) is compared. The finer the graining, the larger the initial relative entropy. Further, finer coarse-graining is helpful to resolve the inertial motion of the magnetization; however, at the same time, it increases the statistical noise (middle and right panel in Fig. 5).

Data availability

The numerically generated and analyzed datasets of the current study are available in the Zenodo Repository, <https://doi.org/10.5281/zenodo.21353289>⁸².

Code availability

The code developed for this study is openly available in Zenodo, <https://doi.org/10.5281/zenodo.21353289>⁸². The SPIDY software is available online⁶².

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Author contributions

F.H. and F.T. discussed and developed the initial idea. F.H. did the analytical and numerical calculations of the entropy production rates. F.T. developed code to numerically solve the iLLG equation. F.T. (lead) and F.H. (supportive) wrote a first draft of the manuscript. All authors finalized the paper. R.M.G. and J.A. supervised the project.

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Competing interests

The authors declare no competing interests.

Additional information

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