

Atomistic spin dynamics with quantum colored noise

Fried-Conrad Weber^{1,2,*}, Felix Hartmann^{1,2}, Matias Bargheer^{1,2}, Janet Anders^{2,3}, and Richard F. L. Evans^{4,†}

¹*Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Wilhelm-Conrad-Röntgen Campus, BESSY II, 12489 Berlin, Germany*

²*Institute of Physics and Astronomy, University of Potsdam, Karl-Liebknecht-Str. 24–25, 14476 Potsdam, Germany*

³*Department of Physics and Astronomy, University of Exeter, Stocker Road, Exeter EX4 4QL, United Kingdom*

⁴*School of Physics, Engineering and Technology, University of York, York YO10 5DD, United Kingdom*



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The accurate prediction of temperature-dependent magnetization dynamics is a fundamental challenge in computational magnetism. While atomistic spin dynamics simulations have emerged as a powerful tool for studying magnetic phenomena, their classical nature leads to significant deviations from experimental observations, particularly at low temperatures. Here we present a comprehensive implementation of quantum-corrected atomistic spin dynamics into the VAMPIRE software package, based on the open-system Landau-Lifshitz-Gilbert equation with a quantum thermostat. Our implementation incorporates memory effects along with colored noise derived from quantum-mechanical considerations that improve the description of the equilibrium magnetization. We demonstrate excellent quantitative agreement with experimental magnetization curves for nickel and gadolinium across the full temperature range. Our results establish that incorporating quantum environmental effects and colored noise substantially enhances the predictive capabilities of atomistic spin dynamics simulations, providing a robust framework for modeling temperature-dependent magnetic phenomena in localized moment magnetic systems.

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I. INTRODUCTION

Atomistic spin models are foundational tools for investigating Heisenberg-like ferromagnets, offering detailed insights into magnetic properties arising from localized moments in lattice structures. These models, which describe magnetism through classical localized magnetic moments on a lattice coupled by exchange interactions, have successfully captured the properties of diverse magnetic systems—from binary alloys like FePt [1] and GdFe [2] to more complex materials such as Fe₃O₄ [3], IrMn [4], FeRh [5], YIG [6], and Nd₂Fe₁₄B [7,8]. Atomistic spin dynamics (ASD) simulations have proven invaluable for understanding ultrafast magnetization dynamics, enabling detailed studies of phenomena such as thermal fluctuation effects on Gilbert damping [9], subpicosecond laser-induced demagnetization in Ni [10], and thermally driven switching in GdFe [2]. Recent advancements have expanded the capabilities of ASD models through developments like Spin-Lattice Dynamics (SLD), which incorporates lattice vibrations [11–16]; Longitudinal-Spin Fluctuations (LSF), capturing Stoner-like effects in transition metals [17,18]; and high- and multitemperature frameworks describing energy flow between electronic, magnetic, and phononic subsystems following ultrafast laser

excitation [19,20]. These innovations have broadened the scope of ASD simulations to address increasingly complex magnetic phenomena, with LSF playing a particularly significant role for modeling itinerant magnetic materials [21].

However, ASD simulations face fundamental limitations due to their classical treatment of magnetic moments via the Landau-Lifshitz-Gilbert (LLG) equation with Gaussian white noise. This classical approach inadequately captures the temperature-dependent magnetization, especially near absolute zero, where quantum effects become significant. Recent advances have addressed these limitations through complementary approaches. Barker and Bauer [22] implemented a semiquantum thermostat that introduces correlated noise obeying quantum statistics without zero-point fluctuations, enabling accurate modeling of low-temperature magnetization in complex materials. Bergqvist and Bergman [23] developed a method incorporating the magnon density of states to apply quantum (Bose-Einstein) statistics in atomistic simulations, significantly improving the prediction of thermodynamic properties. Nussle *et al.* [24,25] developed a path-integral spin dynamics approach whereby one constructs an effective magnetic field to yield the correct quantum expectation value for classical ASD simulations. More recently, Berritta *et al.* [26] provided an analytical framework for temperature rescaling between classical and quantum regimes. Nevertheless, a fully microscopic model capable of accurately reproducing the temperature dependence of magnetization across all regimes has remained elusive.

In this paper we present an implementation of ASD using an open-system Landau-Lifshitz-Gilbert (os-LLG) equation derived from a microscopic open quantum system approach with colored noise [27]. To validate the method,

*Contact author: fried-conrad.weber@uni-potsdam.de

†Contact author: richard.evans@york.ac.uk

we compare the paramagnetic case (exchange constant $J_{ij} = 0$) with results from SPIDY [28], a software package designed for solving the os-LLG equation with colored noise for single and small clusters of spins. The integration of the os-LLG equation into VAMPIRE, a C++-based software package for ASD simulations [29], enables us to study the temperature-dependent magnetization of elementary room-temperature magnetic materials, including Ni, Fe, Co, and Gd. Our findings demonstrate that atomistic os-LLG simulations with a colored semiquantum (which we will call “quantum no-zero”) thermostat significantly improve the accuracy of temperature-dependent magnetization predictions for Ni and Gd. This paper aims to underscore the potential of os-LLG simulations in advancing the study of temperature-dependent magnetic phenomena while also highlighting their current limitations and areas for future development.

II. THEORY

Unlike the phenomenological LLG equation widely used in ASD simulations, the os-LLG equation emerges naturally from an open-quantum-system approach [27]. This fundamental derivation provides a more rigorous theoretical foundation for describing the dynamics of classical spin vectors interacting with a thermal bosonic bath. Historically, the temperature dependence of magnetization is described by Bloch’s law, $m(T) \sim m_0 T^{3/2}$, and attributed to the presence of quantized spin waves known as magnons [30]. As quantum quasiparticles with finite energy and spin-1 (due to the flip of a single spin from $+1/2\hbar \rightarrow -1/2\hbar$), these are assumed to be bosons and thus follow Bose-Einstein statistics. In textbooks and other works [31–33] this is extended to assume that the thermal bath itself is also bosonic, but a consideration of the origin of spin fluctuations suggests important contributions from the electron system, especially for ultrafast dynamics [34]. This suggests that for metallic magnets the relevant heat bath is likely fermionic, which may have important effects at short timescales and require modifications to the derivation of the quantum thermostat [27]. In this work we primarily consider the quasiequilibrium regime where our magnetic system is coupled to a bosonic heat bath reflecting lattice fluctuations.

The derived os-LLG equation for the i ’th spin $\mathbf{S}$ of Ref. [27] is

$$\dot{\mathbf{S}}_i(t) = \gamma \mathbf{S}_i(t) \times \left[\mathbf{B}_{\text{eff},i} + \mathbf{B}_{\text{th},i}(t) + \int_{t_0}^t dt' K(t-t') \mathbf{S}_i(t') \right]. \quad (1)$$

The first contribution to Eq. (1) is a time-independent effective field $\mathbf{B}_{\text{eff},i}$ that combines an applied field $\mathbf{B}_{\text{app}}$, an anisotropy field $\mathbf{B}_{\text{ani}}$, and an exchange field $\mathbf{B}_{\text{exc}}$, i.e., $\mathbf{B}_{\text{eff},i} = \mathbf{B}_{\text{app}} + \mathbf{B}_{\text{ani}} + \mathbf{B}_{\text{exc},i}$. The second term in Eq. (1) refers to a time-dependent stochastic thermal field $\mathbf{B}_{\text{th},i}(t)$, which arises from coupling each spin to a bosonic bath (e.g., phonons) [27,28]. The third and final contribution to Eq. (1) is a memory kernel term $K(t-t')$ that introduces non-Markovian dynamics by making the spin evolution dependent on its history. It contains the damping of the system and takes inertial corrections into account. The noise term and the memory kernel are related self-consistently by the fluctuation-dissipation theorem [27].

In general, the time correlations in the thermal noise are determined by setting the coupling between the spin system and the bath via a power spectrum

$$\tilde{P}(\omega, T) = \frac{1}{2} \int_{-\infty}^{\infty} dt' e^{i\omega t'} \langle B_{\text{th},i}^k(t) B_{\text{th},i}^l(t') \rangle_{\beta}, \quad (2)$$

where $\langle \bullet \rangle_{\beta}$ indicates averaging with respect to the thermal distribution. The indices k and l denote the different spatial direction of the noise field vector $\mathbf{B}_{\text{th},i}$, i.e., x , y , and z . In the following our power spectrum is of Lorentzian shape, i.e., $\tilde{P}^{\text{Lor}}(\omega, T) = I^{\text{Lor}}(\omega) \Theta(\omega, T)$, with

$$I^{\text{Lor}}(\omega) = \frac{\mathcal{A} \Gamma \hbar \omega}{(\omega_0^2 - \omega^2)^2 + \omega^2 \Gamma^2}, \quad (3)$$

where ω_0 represents the center frequency, Γ the width, and $\mathcal{A}$ the amplitude (coupling strength) of the Lorentzian spectral density $I^{\text{Lor}}(\omega)$. Here the Lorentzian form introduces a characteristic resonance frequency of the bath ω_0 that couples to the spin system but with a defined width Γ arising from phonon dispersion. The coupling strength $\mathcal{A}$ defines the strength of coupling between the bath and the spin system and is directly related to the Gilbert damping parameter α_G and the spin-orbit coupling. Choosing a Lorentzian spectral density introduces a spectral character to the effective power spectrum that promotes resonant thermal excitations at $\omega = \omega_0$ while depopulating the available excitations at higher frequencies $\omega > \omega_0$ (for a detailed discussion see Appendix A).

The temperature dependence of the power spectrum $\tilde{P}^{\text{Lor}}(\omega, T)$ is introduced by one of the following three distinct thermostats $\Theta(\omega, T)$:

$$\Theta_{\text{cl}}(\omega, T) = \frac{2k_B T}{\hbar \omega}, \quad (4)$$

$$\Theta_{\text{qu}}(\omega, T) = \coth \left(\frac{\hbar \omega}{2k_B T} \right), \quad (5)$$

$$\Theta_{\text{qnz}}(\omega, T) = \coth \left(\frac{\hbar \omega}{2k_B T} \right) - 1. \quad (6)$$

Equation (4) represents the classical thermostat [29], Eq. (5) the quantum thermostat [27], and Eq. (6) the quantum no-zero (QNZ) thermostat [22]. The quantum thermostat includes zero-point fluctuations at $T = 0$ K, while both the classical and QNZ thermostats ensure vanishing fluctuations at absolute zero. In the high-temperature limit $\hbar \omega \ll 2k_B T$ the quantum noise converges to the classical behavior.

While in this paper we solely consider a Lorentzian power spectrum for the noise [where $I(\omega)$ takes the form of Eq. (3)], it is also possible to consider the same equation of motion [Eq. (1)] and temperature dependencies [Eqs. (4)–(6)] with Ohmic noise (so called due to the white noise nature of thermal noise in an ideal resistor). There the spectral density of the bath is linear in frequency, i.e., $I^{\text{Ohm}}(\omega) \propto \alpha_G \omega$. In this case, Eq. (1) reduces to the standard LLG equation [27], and for a classical thermostat [Eq. (4)], the thermal fluctuations lose their time-correlated nature. Our approach allows for a number of different *flavors* of thermal bath within our open-system framework, summarized in Table I. Standard classical spin dynamics follows a classical thermostat with Ohmic (white) noise. The additional flexibility of this frameworks allows in-

TABLE I. Taxonomy of colored thermostats, including the Ohmic and Lorentzian bath spectral densities $I(\omega)$, each with classical, quantum, and quantum no-zero thermostats $\Theta(\omega, T)$.

$\tilde{P}(\omega, T)$	Acronym	$I(\omega)$	$\Theta(\omega, T)$
Classical noise	CO	Ohmic	T/ω
Quantum Ohmic	QO	Ohmic	$\coth(\omega/T)$
Quantum no-zero Ohmic	QNZO	Ohmic	$\coth(\omega/T) - 1$
Classical Lorentzian	CL	Lorentzian	T/ω
Quantum Lorentzian	QL	Lorentzian	$\coth(\omega/T)$
Quantum no-zero Lorentzian	QNZL	Lorentzian	$\coth(\omega/T) - 1$

dependent consideration of the effects of time correlation and quantum statistics, with and without zero-point fluctuations.

The Lorentzian-shaped spectrum $I^{\text{Lor}}(\omega)$ in Eq. (3) is used to approximate the physical bath, which is characterized by the phonon density of states [35]. The arising noise field $\mathbf{B}_{\text{th},i}$ is characterized by $\langle \mathbf{B}_{\text{th},i}(t) \rangle = 0$, and its temporal autocorrelation is given by $\langle B_{\text{th},i}^k(t) B_{\text{th},i}^k(t') \rangle \propto \delta_{ij} \delta_{kl} \chi(t - t', T)$, where $\chi(\cdot)$ depends on the chosen parameters of Eq. (3). In general, $\chi(t - t', T)$ is not δ correlated, leading to colored noise (as shown in Fig. 1).

To fix the three Lorentzian parameters ω_0 , Γ , and $\mathcal{A}$, we obtain ω_0 and Γ by fitting the Lorentzian spectral density $I^{\text{Lor}}(\omega)$ to the phonon density of states (DOS) [35], representing the long-time fluctuations of the heat bath. In this approximation we assume that the electronic system is always in its ground state and thermal fluctuations arise due to fluctuations of the lattice. The last free parameter $\mathcal{A}$ is fixed by comparing it to the (Gilbert) damping parameter of the material, via $\alpha_G = \mathcal{A}\Gamma/\omega_0^4$ [27]. More details of the fitting are given in Appendix B.

In summary, the two advantages of the os-LLG equation (1), compared to the standard LLG equation, are that it contains colored quantum thermostats and introduces non-Markovian effects in a thermodynamically consistent way. The Lorentzian power spectrum introduces a natural cut-off frequency at finite frequency, so that the characteristic system frequencies (given here by the phonons) are weighted and the noise is colored. Generally, colored noise with different thermostats and memory effects are considered to play a crucial role at short times on the microscale [36,37], but here we also consider the effects on the equilibrium magnetization.

III. NUMERICAL IMPLEMENTATION

To translate the theoretical advantages of a quantum thermostat with colored noise described by Eq. (1) into practical ASD simulations, we follow the numerical methods outlined in Refs. [27,28] and implement these into the VAMPIRE software package [29]. Below we outline our approach for generating colored noise and implementing non-Markovian spin dynamics into ASD as schematically illustrated in Fig. 1.

Before the noise is colored, white noise (with uncorrelated random fluctuations) is generated by drawing N random numbers ξ_i^k per spatial dimension k from a Gaussian probability distribution $p(\xi_i^k)$, with mean $\mu = 0$ and variance $\sigma = 1/\sqrt{dt}$, where dt is the integration step size [see Fig. 1(a)]. The white noise time signal $\xi_i^k(t)$ of spatial dimension k is Fourier transformed into frequency space such that $\tilde{\xi}_i^k(\omega)$ is obtained. The Fourier transformed noise is then scaled by multiplying by the square root of the Lorentzian power spectrum $\tilde{P}^{\text{Lor}}(\omega, T)$ [Eq. (3)], including the appropriate thermostat, see Table I to color the noise, shown schematically in Fig. 1(b). Finally the colored noise is inverse Fourier transformed, returning the noise to the time domain, expressed

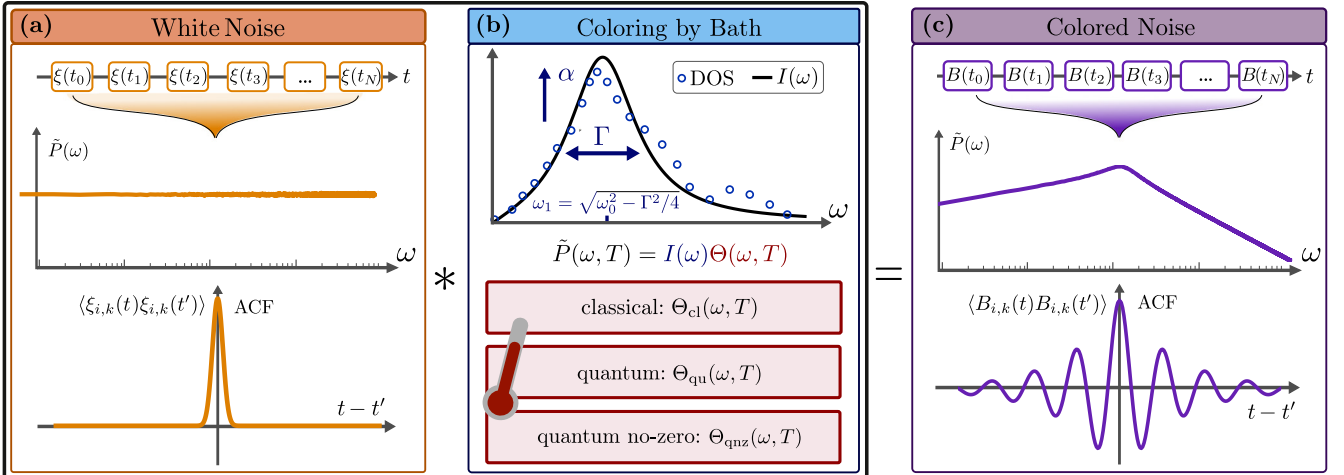


FIG. 1. Colored noise generation. In this figure we sketch schematically how the colored noise is generated (compare with Sec. III). (a) White noise is generated by drawing N random numbers ξ from a Gaussian probability distribution $p(\xi)$. The white noise is characterized by a flat power spectrum and a δ -shaped autocorrelation function (for better visibility approximated by a slim Gaussian distribution). (b) To color the noise the power spectrum needs to be fixed: Firstly, the magnetic environment is approximated by a Lorentzian spectral density $I^{\text{Lor}}(\omega)$ (black solid curve, middle panel). The parameters $(\mathcal{A}, \Gamma, \omega_0)$ are obtained by fitting $I^{\text{Lor}}(\omega)$ to the (phonon) density of states DOS (blue points). Secondly, one of three thermostats Θ (classical, quantum, quantum no-zero) must be selected. (c) Via the convolution in Eq. (7) the noise becomes colored, as the frequencies are weighted and the Lorentzian introduces a natural high-frequency cutoff. The colored noise $B_{\text{th},i}^k(t)$ has a shaped powers spectrum (2) and a finite autocorrelation function.

mathematically as the integral over frequency:

$$B_{\text{th},i}^k(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega t} \sqrt{\tilde{P}^{\text{Lor}}(\omega, T)} \tilde{\xi}_i^k(\omega). \quad (7)$$

The resulting noise is colored and possesses a finite autocorrelation in time that is characteristic of the chosen Lorentzian parameters, thermostat and temperature. Numerically we utilize the FFTW library to compute the Fourier transforms [38] efficiently in $N \log N$ time, where N is the number of elements. We minimize the memory usage by using two techniques discussed in Appendix C.

So far we have considered the generation of the thermal noise with the desired power spectrum and thermodynamics, but we have not considered exactly how to solve Eq. (1), including the memory kernel $K(t - t')$. Explicitly this would require an integration over all time to include the memory effects and the associated dissipation so that the fluctuation-dissipation theorem is satisfied. Previously it has been shown in Ref. [27] that under the assumption of Lorentzian bath spectrum in (2), Eq. (1) can be recast into a set of three coupled differential equations. Therefore, two auxiliary variables $\mathbf{V}$ and $\mathbf{W}$ [27,28] are needed to capture the non-Markovian nature of the os-LLG equation, given by

$$\dot{\mathbf{S}}_i(t) = \gamma \mathbf{S}_i(t) \times \left(\mathbf{B}_{\text{eff},i} + \frac{1}{\sqrt{S_0}} \mathbf{B}_{\text{th},i}(t) + \mathbf{V}_i(t) \right), \quad (8)$$

$$\dot{\mathbf{V}}_i(t) = \mathbf{W}_i(t), \quad (9)$$

$$\dot{\mathbf{W}}_i(t) = \mathcal{A} \mathbf{S}_i(t) - \omega_0^2 \mathbf{V}_i(t) - \Gamma \mathbf{W}_i(t), \quad (10)$$

where S_0 represents the length of the classical spin vector. Here the kernel $K(t - t')$ represents the dissipation of the energy to the bath and is constructed to obey the fluctuation-dissipation theorem [39] in the general case where the noise is colored and has a different thermostat [27]. Similar stochastic equations of motion for (single) spins were first discussed by Miyazaki and Seki [37,40] but with a different underlying system-bath coupling and therefore a different noise autocorrelation as in Eqs. (8)–(10). We also note that Suhl [41] developed a similar nonstochastic model that describes magnetization relaxation via magnetoelastic interaction into the lattice degrees of freedom. The exchange field $\mathbf{B}_{\text{exc},i}$ follows the vampire framework [29]. The thermal fields $\mathbf{B}_{\text{th},i}$ are precalculated for each atom and each spatial direction numerically by using the above method. Finally, Eqs. (8)–(10) are solved using a fourth-order Runge-Kutta (RK4) integrator, which computes four intermediate slopes (k_1 – k_4) to update the state:

$$y_{n+1} = y_n + \frac{dt}{6} (k_1 + 2k_2 + 2k_3 + k_4).$$

Because the exchange field depends sensitively on the instantaneous spin configuration, it is updated and recalculated at each intermediate RK4 step based on the spins' predicted positions. The precomputed thermal noise field is incorporated using the previously described time interpolation with decimation: for the first RK4 slope (k_1) we use the noise at time t_n , for the intermediate slopes (k_2 and k_3) we use the average of the noise at t_n and t_{n+1} , and for the final slope (k_4) we use the noise at time t_{n+1} . To validate our numerical implementation, we perform a consistency check in the paramagnetic regime

($J_{ij} = 0$) with the single spin implementation SPiDY [28]. The results are detailed in Appendix D and show excellent agreement between the two implementations.

IV. NUMERICAL RESULTS

To test our implementation in the multispin case, we assume a simple spin Hamiltonian, including nearest-neighbor exchange and coupling to an external magnetic field of the form [29]

$$\mathcal{H} = - \sum_{i<j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \mu_i \sum_i \mathbf{S}_i \cdot \mathbf{B}_{\text{app}}, \quad (11)$$

where J_{ij} is the exchange interaction between nearest neighbors, $\mathbf{S}_{i,j}$ represent local and neighboring atomic spin moment directions, respectively, μ_i is the local spin moment, and $\mathbf{B}_{\text{app}}$ is the applied field vector. The system of coupled spins described by the Hamiltonian is then time-integrated using the os-LLG at constant temperature to achieve a consistent thermodynamic state. We first consider a simulation of Ni, with a low magnetic moment of $\mu_i = 0.61 \mu_B$ and an intermediate case in terms of the strength of the quantum corrections. As described above in Sec. III, we characterize the bath by a Lorentzian spectral density $I^{\text{Lor}}(\omega)$. The width $\Gamma = 28.9$ rad THz and characteristic frequency $\omega_0 = 40.5$ rad THz are obtained by fitting the phonon DOS of Ni, see Fig. 2(a). Assuming that the Gilbert damping of Ni is $\mathcal{A} = 4655$ rad³ THz³, we obtain $\alpha_G = 0.05$. We perform a simple computational experiment, initializing all spins parallel to the x axis in a $B_z = 1.0$ T magnetic field and simulating the relaxation dynamics of the overall magnetization at different temperatures. Here we use a small system size of $(7 \text{ nm})^3$ with a face-centered cubic lattice containing around $N = 45\,000$ individual atomic spins. We use an exchange constant of $J_{ij} = 9.35 \times 10^{-22}$ J/link within the nearest-neighbor approximation. All parameters for the simulations are summarized in Table II, and the dependency of the thermostat and the exchange energy on the Curie temperature are detailed in Appendix E.

Previous works have identified a clear difference comparing classical and quantum thermostats in magnetic systems in the temperature dependence of the magnetic order parameter $m(T)$ given by $m(T) = |\sum_i \mu_i \mathbf{S}_i| / \sum_i \mu_i$, where μ_i is the moment length. At $T = 0$, without zero-point motion all spins are aligned and give a fully saturated state. The shape of the $m(T)$ curve is characteristic of the thermodynamic fluctuations of the spins with increasing temperature, expressed by an interpolation of Bloch (low-temperature) and Curie (high-temperature) behavior and given by

$$m(T) = \left[1 - \left(\frac{T}{T_c} \right)^{\alpha_m} \right]^{\beta_m}, \quad (12)$$

where $\beta_m \simeq 0.34$ is the usual magnetization critical exponent describing the shape of the curve near the Curie temperature [43,44], and α_m is an exponent describing the shape of the curve at low temperature. We note that the β_m exponent here better represents the data for the classical Heisenberg spin model than the usually known value of $\beta_m = 0.365$ [45,46]. This equation is a simplified form compared to that proposed by Kuz'min [47] but clearly separates classical and quantum

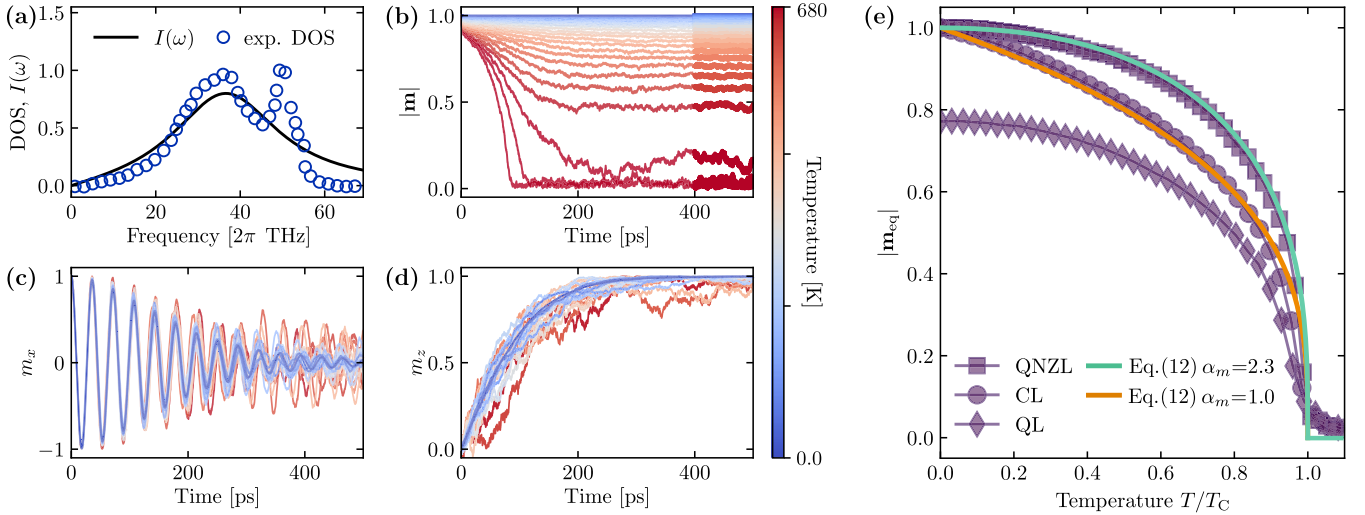


FIG. 2. Dynamics toward equilibrium and equilibrium magnetization with colored noise. (a) Fit of the measured phonon DOS for nickel (open blue circles) [42] with the Lorentzian spectral density $I^{\text{Lor}}(\omega)$ (black solid line) defining spin-bath coupling. Averaged dynamics of the (b) magnetization amplitude, (c) m_x component, and (d) m_z component, using QNZL noise for different temperatures [blue (0 K) to red (680 K) transition]. (e) Equilibrium magnetization for classical thermostat (circle), quantum thermostat (diamond), and quantum no-zero thermostat (squares). While the os-LLG simulation with the classical thermostat reproduces the classical ASD simulation results [10] (i.e., Eq. (12) with $\alpha_m = 1$, orange solid line), the os-LLG simulation with the QNZL noise matches Eq. (12) with $\alpha_m = 2.32$ fitted for nickel (green solid line). Simulations are for a $7 \times 7 \times 7$ nm block containing 45 000 spins.

behavior, allowing for cases where the system essentially follows the classical $m(T)$ curve [48]. The parameter α_m gives an indication of the *quantumness* of the system, with $\alpha_m = 1$ the classical limit and $\alpha_m > 1$ indicating quantum corrections, which are weak for Gd, strong for Fe, and intermediate for Co and Ni [10]. The os-LLG described in this paper for multispin systems naturally introduces quantum corrections to the spin fluctuations and we therefore use the temperature-dependent magnetization as a benchmark against experimental data to determine the accuracy of the parameters in our simulations.

The obtained averaged dynamics $|\mathbf{m}(t)|$, $m_x(t)$, and $m_z(t)$ for the quantum no-zero Lorentzian (QNZL) noise at different temperatures is shown in Figs. 2(b), 2(c), and 2(d), respectively. The system shows both precessional [Figs. 2(c) and 2(d)] and longitudinal [Fig. 2(b)] relaxation leading to a reduced magnetization length after around 200 ps, after which an average equilibrium value of the magnetization is calculated. We note that this deviation from experimental

measurements of ultrafast demagnetization likely occurs because we only include a phononic bath in our model; an electronic bath with higher frequency components would accelerate the equilibration timescale.

Starting with the classical thermostat and Lorentzian spectral density (CL noise), we find excellent agreement with the standard classical behavior described by Eq. (12) for $\alpha_m = 1$ (compare orange line and purple circles in Fig. 2), confirming that the equilibrium magnetization of the os-LLG equation successfully recovers the standard stochastic LLG with white noise in the classical limit.

Next, the os-LLG equation with QL noise (Table I) is simulated. This provides crucial insights into the low-temperature regime [see purple diamonds in Fig. 2(e)]. While it accurately captures the shape of the magnetization curve at low temperatures, a distinctive feature emerges at absolute zero. The presence of quantum zero-point fluctuations, $\Theta_{\text{qu}}(\omega, T = 0 \text{ K}) \neq 0$, creates a competition with the ferromagnetic ex-

TABLE II. Table of simulation parameters for the ferromagnetic elements Fe, Co, Ni and Gd [29]. The Lorentzian parameters are obtained by fitting the phonon density of states for Fe [49], Co [50], Ni [42], and Gd [51]. Simulations use the atomistic spin moment as the spin length and not multiples of $\hbar/2$. The exchange parameters for QNZL noise which reproduce the correct T_C vary from the exchange parameters for classical ASD [29]. For our simulations, we do not consider the structural phase transitions of Fe and Co.

	Fe	Co	Ni	Gd	Unit
Crystal structure	bcc	hcp	fcc	hcp	
Unit cell size a	2.866	2.507	3.524	3.636	Å
Atomic spin moment S_0	2.22	1.72	0.606	7.63	μ_B
Lorentzian amplitude $\mathcal{A}$	7812	4583	4655	246	$\text{rad}^3 \text{ THz}^3$
Lorentzian width Γ	31.5	27.1	28.9	10.0	rad THz
Lorentzian center ω_0	47.1	39.7	40.5	14.9	rad THz
Exchange energy J_{ij} (QNZL)	2.8×10^{-21}	2.6×10^{-21}	9.35×10^{-22}	5.35×10^{-22}	J/link
Curie temperature T_C	1043	1388	631	293	K

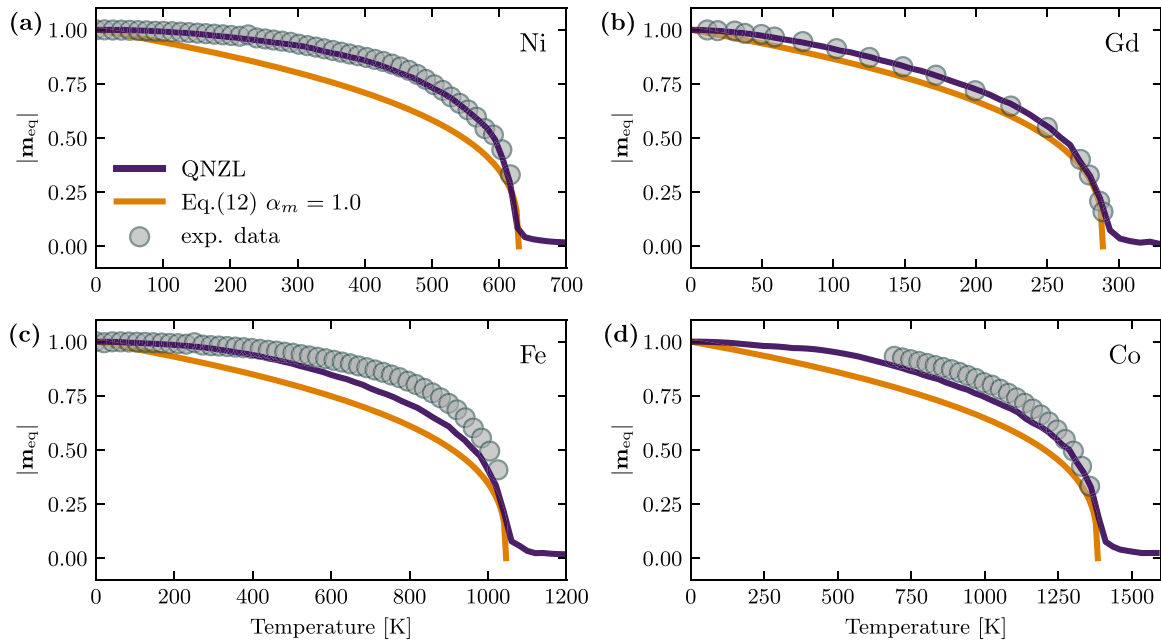


FIG. 3. Equilibrium magnetization for Ni, Gd, Fe, and Co. Comparison of simulated equilibrium magnetization using the os-LLG model with QNZL (purple solid line) and classical ASD simulations with classical white noise [29] (orange solid line). The gray circles represent experimental data for (a) nickel, (b) gadolinium, (c) iron [52], (d) and cobalt [53]. For nickel and gadolinium, the os-LLG model with QNZL noise shows excellent agreement with the measured temperature-dependent magnetization, while for cobalt and iron there is a difference between the simulated and experimental results. Simulations performed for approximately 45 000 spins with an effective field $B_z = 10$ mT. Further simulation parameters are detailed in Table II.

change coupling, resulting in a magnetization that remains below unity even at $T = 0$ K. Its origin is the fact that the thermodynamic fluctuations do not tend to zero in the quantum case but plateau to a low-temperature value that is temperature independent (a more detailed discussion of the zero-point fluctuations can be found in Appendix F).

We further consider a form of the noise with the same quantum thermostat but backing off the zero-point fluctuations (QNZL), which removes the residual noise as $T \rightarrow 0$, leading to a fully saturated state at $m(0) = 1$. This is similar to the form used by Barker and Bauer [6], where they used a quantum thermostat with an Ohmic power spectrum (QNZO). The QNZL thermostat successfully reproduces the temperature dependence of magnetization of Ni across the entire temperature range, capturing both quantum effects at low temperatures and classical behavior near and above the Curie temperature, see purple squares in Fig. 2(e). Our simulation results agree nicely with Eq. (12) fitted from the experimental data for Ni [green line in Fig. 2(e)].

For all thermostats we observe a slightly elevated magnetization in the paramagnetic phase near T_C , which we attribute to finite size effects in our simulations [see Fig. 2(e) for $T > T_C$]. This elevation is particularly noticeable given our relatively small sample size ($7 \times 7 \times 7$ nm), though it does not affect the overall validity of our results in the ferromagnetic phase.

Since the os-LLG successfully reproduces the temperature-dependent equilibrium magnetization for Ni, we next consider comparative simulations for the elemental ferromagnets. In Fig. 3 we compare the equilibrium magnetization of the os-LLG equation with QNZL noise (purple line) against

classical ASD (12) simulation results and experimental data (gray circles) for the four ferromagnetic materials: nickel (a), cobalt (b), iron (c), and gadolinium (d). Parameters for the different materials are given in Table II. Our simulations nicely recover the low-temperature magnetization of all four materials, with excellent agreement for nickel and gadolinium over the full temperature regime, showing a strong improvement compared to the usual classical white noise ASD simulations.

One important aspect of QNZ thermostats is that the shape (curvature) of the equilibrium magnetization curves is dependent on the length of the magnetic moment $S_0 := \mu_i$. This is different in standard ASD simulations with classical white noise (CO), where the shape of the curve is independent of the magnetic moment [27]. For example, comparing the QNZL magnetization curves of Ni and Gd in Figs. 3(a) and 3(b), respectively, we see that for a large magnetic moment (Gd) the equilibration curve tends toward a classical ASD curve (orange line). In contrast, for a short spin length S_0 (Ni), the QNZL magnetization curve shows a clear difference to the classical ASD curve. While the low-temperature behavior of $m(T)$ for Fe and Co is well reproduced by the QNZL noise, there are deviations at higher temperatures, Figs. 3(c) and 3(d). For cobalt this is mainly attributed to the phase transition from fcc to hcp cobalt at 700 K [50]. This transition is not taken into account in the simulations of Fig. 3(d) as the full temperature range is simulated by a fixed parameter set detailed in Table II. Next to that the deviation is likely caused by the larger moments of $\mu_{Co} = 1.72\mu_B$ and $\mu_{Fe} = 2.22\mu_B$, giving a closer correspondence to the classical $m(T)$ curve that is more apparent at higher temperatures [54].

V. DISCUSSION AND OUTLOOK

In this work we have implemented the os-LLG equation with colored quantum noise in the ASD software package VAMPIRE, enabling the investigation of large-scale, exchange-coupled spin systems. We have implemented three distinct thermostats with Lorentzian spectral densities characterizing quantum, quantum without zero-point motion, and classical behaviors. In the classical limit, we reproduce the usual classical behavior of standard atomistic spin models. While the quantum thermostat correctly captures the temperature scaling of magnetization, it introduces zero-point fluctuations, leading to nonunity magnetization at $T = 0$ K [27]. The QNZL noise, which omits these fluctuations, provides the most accurate description of the temperature-dependent equilibrium magnetization for the studied materials at low temperatures and is especially accurate for gadolinium and nickel over the full temperature range. Unlike previous approaches such as temperature rescaling [10], the quantum thermostat naturally introduces the quantum-mechanical nature of the heat bath from first principles, directly parameterized from material properties such as the phononic density of states.

In contrast to classical ASD simulations, in our os-LLG framework the spin length plays an important role, leading to a characteristic transition from quantum to classical temperature-dependent equilibrium magnetization for larger spin lengths. Assuming $S_0 = \mu_i$ within the QNZL thermostat, the large local moments of Fe and Co lead to more classical-like behavior than for small moments like Ni where $\mu_i \sim \hbar/2$, in contrast to experimental observations where the $m(T)$ for Co and Fe is more quantumlike [10]. This suggests that an appropriate value for S_0 for Fe and Co to reproduce experimental observations could be significantly lower than the full value of the local moment. Including longitudinal spin fluctuations [17] to reflect the itinerant character and Stoner excitations [55] in these materials could also bring the simulated $m(T)$ curves into closer agreement with experiment [18].

The choice of Lorentzian parameters has a modest effect on the shape of the $m(T)$ curve, which is mostly determined by the thermostat and spin length but can have important effects on the dynamic response. A nonflat bath spectral density departs from the paradigm of purely Markovian magnetization dynamics of atomistic spin models. The non-Markovianity arises by incorporating time-correlated fluctuations and a memory kernel in the equation of motion. This approach opens new avenues for experimental validation and for probing memory effects in magnetic systems. The ability to parametrize the bath directly from material properties—such as phonon densities of states—ensures a first-principles basis for both equilibrium and dynamic simulations.

In summary, our implementation of quantum colored noise in ASD significantly improves the modeling of equilibrium magnetization in local moment systems, thereby offering a more accurate description of their thermodynamic properties and establishing a foundation for future exploration of their dynamic behavior. The integration of fermionic (electron) baths alongside the current bosonic (phonon) approach represents a promising direction for capturing the complete spectrum of magnetic behavior across different materials and timescales. This can include effects such as ultrafast

demagnetization [56], nutation [57–59], and coherent interaction between phonons and the magnetization [60,61]. At present the extension of our method to fermionic baths is not straightforward and requires further research.

Further work could include modeling complex magnetic materials such as antiferromagnets or frustrated systems and including a dynamic thermostat capable of modeling nonequilibrium magnetization dynamics with a quantum thermostat and time-correlated noise.

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F.C.W. implemented the os-LLG into the VAMPIRE software package with assistance from R.F.L.E. The atomistic calculations were performed and the data analyzed by F.C.W., while F.H. drafted the manuscript and developed the framework for the model, based on earlier work by J.A. The project was supervised by J.A. and R.F.L.E. All authors contributed to the final manuscript and presentation of results.

The authors declare no competing interests.

DATA AVAILABILITY

The data that support the findings of this article are openly available [62], embargo periods may apply.

APPENDIX A: OHMIC VS LORENTZIAN POWER SPECTRUM

In Fig. 4 we compare the power spectra of an Ohmic (upper panels) and Lorentzian (lower panels) spectral density (Ni parameters, see Table II) with each of the three thermostats, i.e., classical, quantum, quantum-no-zero (3) (see also Table I). The Ohmic spectral density is given by $I^{\text{Ohm}}(\omega) \propto \alpha_G \omega$ and is commonly used in ASD simulations, e.g., [22,29]. Each power spectrum is plotted for four effective temperatures $\tilde{T}$ and shows how relevant the choice of the spectral density and the thermostat is.

It is helpful to discuss the relevant qualitative features of the choice of thermostat and spectral density on the power spectrum. For classical Ohmic noise (CO) in Fig. 4(a), the power spectrum is flat and simply increases in magnitude with increasing temperature. This means that there is no frequency bias to the excited fluctuations and therefore the noise has no spectral characteristics. This allows for high-frequency excitations to occur readily but also gives a classical scaling of the noise with temperature, leading to a classical temperature-dependent magnetization curve that is not accurate for most magnetic materials. The classical thermostat

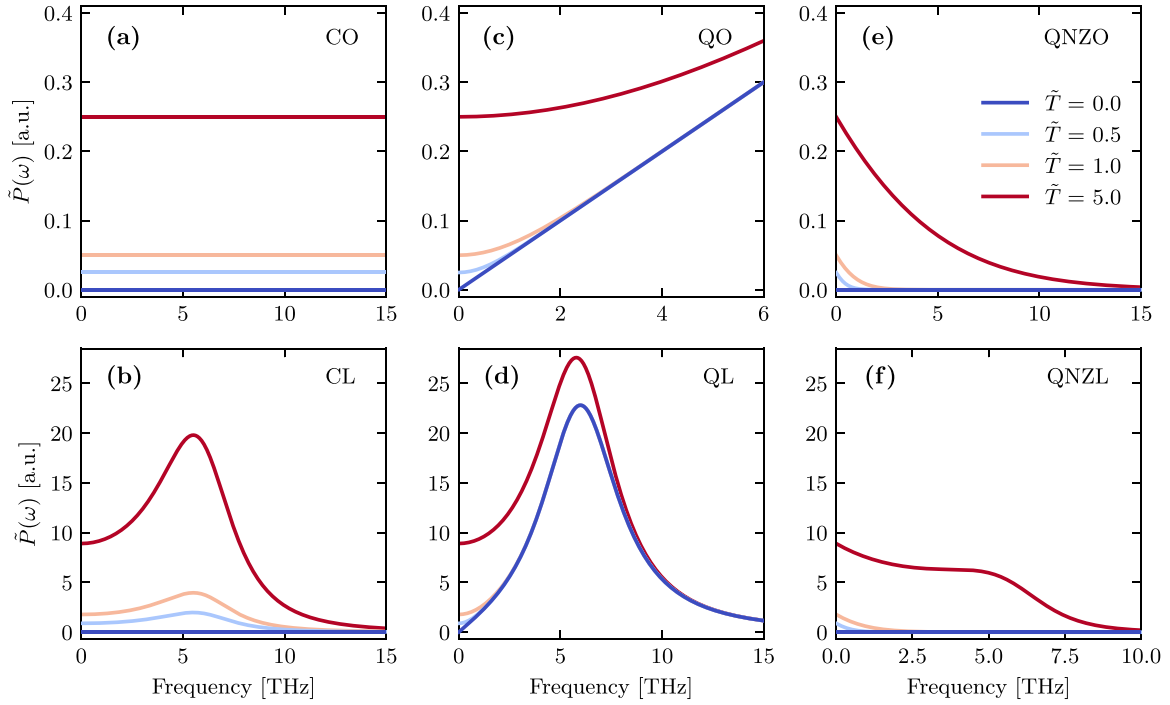


FIG. 4. Comparison of the Ohmic vs the Lorentzian power spectrum $\tilde{P}(\omega)$. We here compare the six different power spectra (cf. Table I). (a), (c), (e) Ohmic spectral density (CO, QO, QNZO) and (b), (d), (f) Lorentzian spectral density (CL, QL, QNZL) from left to right). This demonstrates that the choice of spectral density and of the thermostat is crucial. The effective (unit-free) temperature $\tilde{T}$ is given by $k_B T / \hbar \tilde{\omega}_L$. The Ohmic spectral density is given by $I^{\text{Ohm}}(\omega) \propto \alpha_G \omega$. The Lorentzian parameters for the Lorentzian power spectra are those for Ni (see Table II).

with Lorentzian (CL) noise in Fig. 4(b) introduces a spectral character to the effective power spectrum that promotes resonant thermal excitations at $\omega = \omega_0$ while depopulating the available excitations at higher frequencies $\omega > \omega_0$. Since ω_0 is at a relatively high frequency and significantly above the precession frequency $\tau_p = \gamma \alpha_G \sim 6 \times 10^{10}$ Hz, resonant effects only typically occur under ultrafast (subpicosecond) excitation. Correspondingly, the spin excitations relevant for the equilibrium magnetization are all below ω_0 and so the dominant effect is from the thermostat where, again, the power spectrum scales linearly with the temperature.

Introducing the quantum thermostat with Ohmic noise (QO) in Fig. 4(c) leads to a nonlinear scaling of the power spectrum amplitude with temperature, as well as a linear increase in the power spectrum as a function of frequency. This naturally leads to fluctuations at zero temperature due to remaining fluctuations of quantized phonon modes. The nonlinear scaling of the power spectrum with temperature is reminiscent of the qualitative effect of spin-temperature rescaling [10,26] and leads to a nonclassical temperature-dependent magnetization but within a quantum-mechanical framework. The use of the Lorentzian spectral density with quantum thermostat (QL) in Fig. 4(d) combines the spectral features described for the classical thermostat above, with the nonlinear scaling of the noise with temperature and zero-point motion.

It is possible to remove the zero-point motion effects of the quantum noise by using the thermostat in Eq. (6). Here the quantum no-zero thermalized power spectrum with Ohmic

noise (QNZO) in Fig. 4(e) decays rapidly in frequency, leading to a strong depopulation of high-frequency modes. This limits spin-wave excitations to low frequencies and can therefore significantly increase the Curie temperature, depending on the choice of S_0 . Finally, in the case of quantum no-zero with Lorentzian noise power spectrum in Fig. 4(f), some spectral character is introduced into the noise but only when the temperature is large enough that characteristic thermal excitation overlaps with ω_0 . At low temperatures the QNZO and QNZL should exhibit very similar behavior, while at high temperatures the Lorentzian nature of the noise allows for the high-frequency spin-wave modes characteristic of the phase transition and therefore does not increase the Curie temperature as much as the QNZO case. As before for the CL noise, with sufficient temperature the QNZL should exhibit spectral characteristics with non-Markovian behavior.

APPENDIX B: FITTING THE (PHONON) DOS DATA

In our implementation of the os-LLG equation, the Lorentzian parameters ($\mathcal{A}, \omega_0, \Gamma$) in units of rad THz are transformed to unitless variables via $\tilde{\mathcal{A}} = \mathcal{A} / \tilde{\omega}_L^3$, $\tilde{\omega}_0 = \omega_0 / \tilde{\omega}_L$, and $\tilde{\Gamma} = \Gamma / \tilde{\omega}_L$, and scaled via an *effective* Larmor frequency $\tilde{\omega}_L$, which is defined for a fixed field strength $|\mathbf{B}| = 1.0$ T. Thus, $\tilde{\omega}_L = |\gamma| \times 1 \text{ T} \approx 0.176 \text{ rad THz}$, with $|\gamma| \approx 1.76 \times 10^{11} \text{ rad/s/T}$ being the electron gyromagnetic ratio. The *effective* Larmor frequency $\tilde{\omega}_L$ is solely used to transfer the dimensionless simulation parameters into dimensioned parameters and vice versa [28].

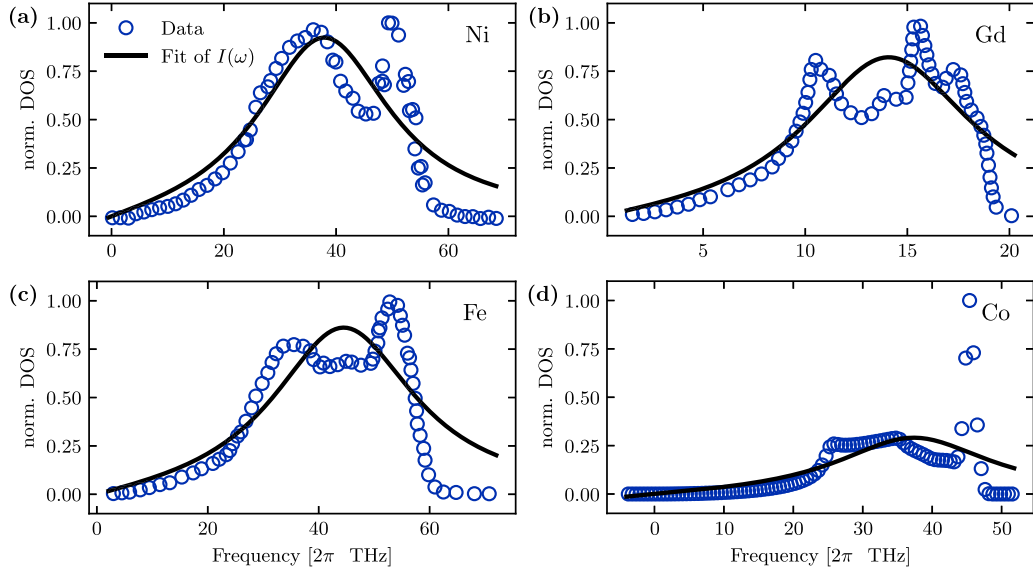


FIG. 5. Lorentzian fitting of the phonon DOS for four materials: Ni [42], Gd [51], Fe [49], and Co [50]. The fitted parameters are (a) Ni $\omega_0 = 40.5$ rad THz and $\Gamma = 28.9$ rad THz, (b) Gd $\omega_0 = 14.9$ rad THz and $\Gamma = 10.0$ rad THz, (c) Fe $\omega_0 = 47.1$ rad THz and $\Gamma = 31.5$ rad THz, and (d) Co $\omega_0 = 39.7$ rad THz and $\Gamma = 27.1$ rad THz. The parameters in units of rad THz are given in Table II.

In Fig. 5 we show the phonon DOS data (open blue circles) and the fitted Lorentzian (black solid line) for the four ferromagnetic materials (Ni [42], Gd [51], Fe [49], and Co [50]) discussed in this manuscript. The extracted Lorentzian parameters are given in Table II and in the caption of Fig. 5.

APPENDIX C: GENERATING COLORED NOISE

We utilize the FFTW library to compute the colored noise in the frequency domain efficiently in $N \log N$ time, where N is the number of elements. Since computing the colored noise requires storing the entire noise array in memory, we employ two optimizations to reduce the memory usage. Without using these optimizations, the memory required to store the noise for a system of 10 000 atoms over a simulation time of 1 ns with a time step of 0.1 fs would be approximately 2.4 TB (8 byte $\times$ $3 \times 10\,000 \times 10^7$; double precision floating point, three components of noise per atom, 10 000 atoms, 10^7 time steps), not including the memory required for the FFTW library itself, making this approach unfeasible for most computers. The first optimization is to generate the noise on a coarser time grid

than the simulation time step. The colored noise varies on timescales of 1–100 fs, so we can generate the noise with a time step $dt_{\text{coarse}} = D \times dt_{\text{sim}}$, where D is a decimation factor determined by the characteristic frequencies of the noise. We select D such that dt_{coarse} satisfies the Nyquist criterion for the highest significant frequency component in the power spectrum, typically $D \approx 10$ –100 for our Lorentzian parameters. The noise values at intermediate simulation time steps are then obtained through linear interpolation. The plot in Fig. 6(a) shows the output for noise generated on a 0.1-fs fine time grid (purple line) and compares it with noise generated with the described method on a coarse time grid (open circles). To preserve the correct statistical properties, the initial white noise is scaled $\sqrt{dt_{\text{sim}}/dt_{\text{coarse}}}$, which ensures that the standard deviation of the interpolated noise and the autocorrelation function matches that of noise generated directly on the fine time grid. This approach reduces both memory requirements and computational costs by a factor of D , making it feasible to generate colored noise for large-scale simulations, while maintaining the essential statistical properties that influence the system dynamics.

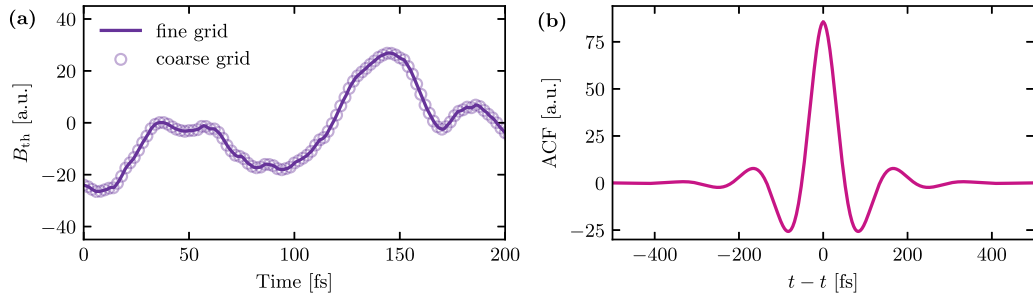


FIG. 6. Noise trace and corresponding autocorrelation function for Ni parameters at $T = 100$ K for QL. (a) The noise trace generated on a fine 0.1-fs time grid (lines) compared to noise generated on a coarse 2-fs time grid (dots). (b) The autocorrelation function of the noise for the Ni parameters. The autocorrelation decays within hundreds of femtoseconds.

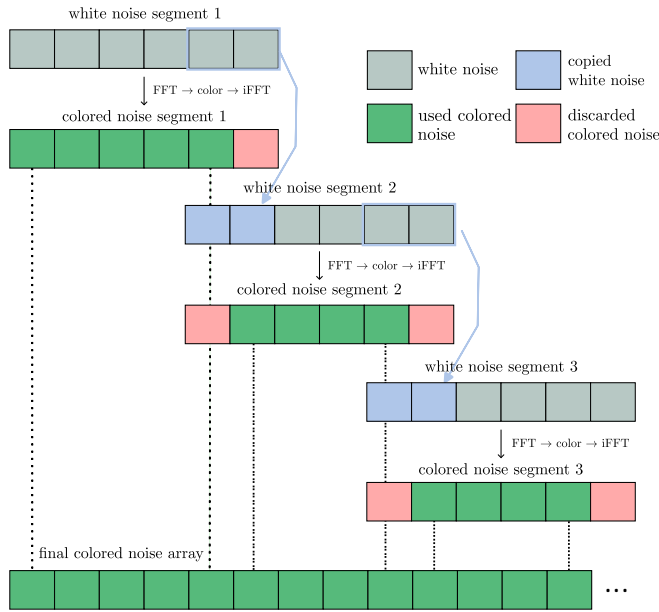


FIG. 7. Sketch of noise segmentwise noise generation. First a white noise segment is generated and transformed into frequency domain where the noise is colored and then inversely transformed into time domain. From the first colored noise segment, the last part is discarded and the others used for simulation until all the noise is used up. Then the last two subsections of the white noise used in step 1 are copied into the first subsections of the second white noise window. Then again, the colored noise is generated but now the first and last segment are discarded. The same procedure is then repeated until the simulation ends.

The second optimization is to generate the noise in smaller time segments rather than the entire simulation duration at once. We divide the total simulation time into n segments, each of length $t_{\text{seg}} = t_{\text{sim}}/n$. We generate the colored noise for each segment independently; however, we ensure the continuity of the noise at the segment boundaries by overlapping the segments. The method we employ is sketched in Fig. 7. We generate a white noise segment of a certain length which is divisible by six. For the first segment we do the FFT of the white noise, into the frequency domain, apply the coloring function, and then do the inverse FFT to obtain the colored noise in the time domain. We then only keep the first 5/6 (green boxes) of the generated noise and discard the last sixth (red box). For the next segment we populate the first 2/6 of this segment with the last 2/6 of the previous white noise segment (blue arrow) and fill the rest with new white noise. We then repeat the FFT, coloring, and inverse FFT process for this new white noise array, but we only keep 4/6 of the generated colored noise, discarding the first 1/6 and last 1/6. This process is repeated for all segments until the entire simulation duration is covered. This approach ensures that the colored noise is continuous across segment boundaries while also retaining the statistical properties of the noise. Important to consider is that the segment length t_{seg} must be significantly larger than the noise correlation time to avoid artifacts in the noise statistics. Figure 6(b) shows the autocorrelation function for the Ni parameters used in this paper at $T = 100$ K. The autocorrelation decays on timescales of hundreds of femtosec-

onds, allowing to cut the memory cost for long-timescale simulations by a factor of 100. By combining these two optimizations, we can efficiently generate colored noise for large-scale atomistic spin dynamics simulations without exceeding memory limitations.

APPENDIX D: COMPARING THE PARAMAGNETIC DYNAMICS

To validate our implementation of the os-LLG equation with colored noise in vampire, we first examined the paramagnetic case ($J = 0$) through comparison with SPiDY [28].

Figures 8(a) and 8(b) demonstrate excellent agreement between the dynamics of SPiDY (black line, open circles) and our os-LLG vampire implementation (red line, open triangles). In addition, Fig. 8(c) shows perfect agreement between the paramagnetic steady state of the two approaches. The equilibrium magnetization was determined by evolving the system under a fixed quantum thermostat [see Eq. (5)] until the magnetization amplitude $m(t) = |\langle \mathbf{m}(t) \rangle|$ reached steady state. For each temperature the final magnetization amplitude was computed by averaging over a time window in the steady-state regime.

To compare the accuracy in more detail, we ran paramagnetic simulations with a classical thermostat (4) with $T = 0$ K. The system of coupled differential equations (8)–(10) becomes deterministic, and the accuracy is not falsified by a limited number of stochastic realizations. We find that the absolute error (AE) between the two approaches is $|\text{AE}| < 6 \times 10^{-6}$. Therefore we can use our implementation of SPiDY into VAMPIRE to study magnetic materials beyond paramagnets.

APPENDIX E: CURIE TEMPERATURE FOR THE DIFFERENT THERMOSTATS

In Fig. 9 we demonstrate that the choice of thermostat has a strong influence on the numerically observed Curie temperature T_C of the os-LLG equation (1) framework. We find, for the Lorentzian parameters of Ni and a given exchange energy J_{ij} , that the QNZL thermostat consistently predicts a larger T_C than the CL and QL thermostats. This is directly linked to the discussion of the different power spectra in Appendix A. In contrast, in classical ASD simulations with VAMPIRE, using white noise, an exchange energy of $J_{ij} = 6.0 \times 10^{-21}$ J/link has been reported to reproduce the correct T_C [29]. It follows that the thermostat quite drastically changes the observed Curie temperature. Future work is needed to gain a detailed understanding of all its implications.

APPENDIX F: NONUNITY MAGNETIZATION FOR QL NOISE

QL noise naturally leads to zero-point fluctuations, which significantly reduces the magnetization at low temperatures, rendering our QL noise simulations unphysical, compare with Fig. 2(e). This, however, could not be a problem of the theory but rather the model to simulate the equilibrium magnetization as the spins in ASD are treated

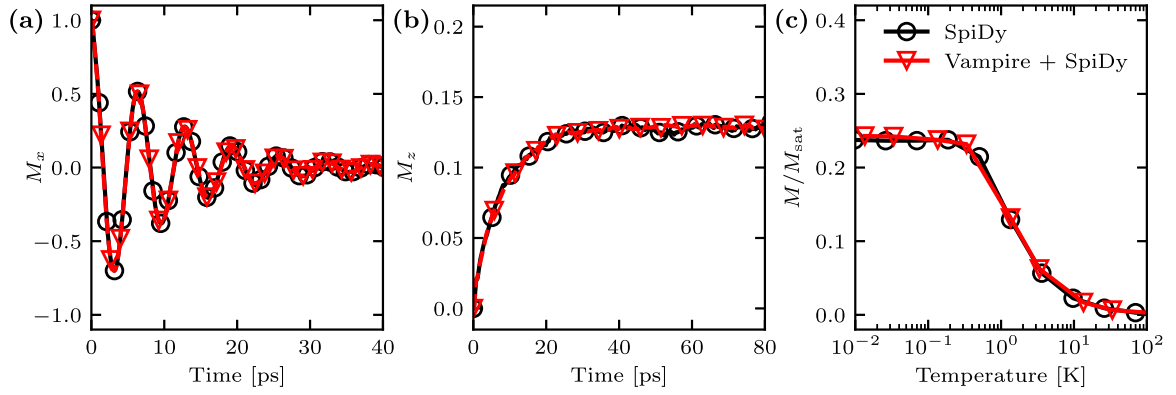


FIG. 8. Paramagnetic magnetization dynamics. Here we compare the paramagnetic spin dynamics calculated from SpiDy [28] (black line, open circles) and the implementation of the os-LLG equation (1) into vampire (red line, open triangles) while employing a quantum thermostat $\tilde{\lambda}_{\text{qu}}$ (5). (a), (b) We find excellent agreement between the two approaches, here exemplarily shown for the M_x and M_z components of the magnetization. (c) This leads to an excellent agreement between the steady states of the two implementations over the complete temperature range. The Lorentzian simulation parameters are taken from Set 1 of Ref. [27], i.e., $\tilde{A} = 10.0 \tilde{\omega}_L^3$, $\tilde{\omega}_0 = 7.0 \tilde{\omega}_L$, and $\tilde{\Gamma} = 5.0 \tilde{\omega}_L$, with $\tilde{\omega}_L = 0.176 \text{ rad THz}$. Further, the spin length of the classical spin vector is $S_0 = \hbar/2$, the external field is $\mathbf{H}_{\text{ext}} = (0, 0, 1) \text{ T}$. For (a) and (b) the single spin temperature is $T = 1.35 \text{ K}$ and the dynamics is averaged over 50 000, respectively, over 13 500 spin trajectories for panel (c). The spin is initially aligned along the positive x direction, i.e., $\mathbf{S}_0 = (1.0, 0.0, 0.0)$.

classically, which overestimated the effect of zero-point fluctuations.

Additionally, zero-point fluctuations of atomic spin moments raises an interesting and important question with regard to the electronic structure of a magnetic material in the presence of a phononic heat bath. In standard electronic structure calculations such as density functional theory

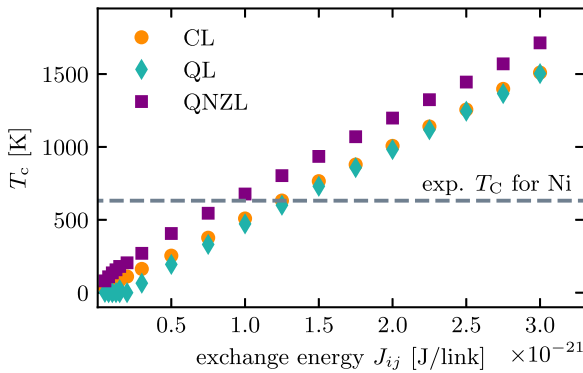


FIG. 9. Curie temperature of the three different thermostats for varying exchange energies. We vary the exchange energy J_{ij} and determine the corresponding Curie temperature T_C . The classical (orange circle), quantum (green diamonds), and quantum-no-zero (purple squares) thermostats show the same linear trend for $J_{ij} > 0.3 \times 10^{-21} \text{ J/link}$. However, close attention should be paid to the slopes of the three thermostats. The classical and quantum thermostat have a similar slope, whereas the slope of the quantum-no-zero noise is steeper. This implies that the prediction of the Curie temperature and the exchange energy is strongly related to choice of the thermostat. We choose the same Lorentzian parameters as for Ni, compare to Table II. The experimentally measured Curie temperature for Ni ($T_C = 631.2 \text{ K}$) is indicated by the gray dashed line. In classical ASD simulations with vampire an exchange energy of $J_{ij} = 6.0 \times 10^{-21} \text{ J/link}$ reproduces the correct T_C [29].

(DFT), a time-independent solution is assumed where local fluctuations of nuclear (atomic) positions are averaged out, the electron density is then minimized, and the local spin moment is calculated, typically with good estimations of the local moment value in comparison with experimental data. In some respects the quantum spin fluctuations are included within the electronic structure, projecting the net magnetic moment onto a classical vector which is the relevant observable quantity. On the other hand, small zero-point fluctuations of the lattice will naturally induce spin fluctuations due to small changes in the electronic structure. Within the framework of the os-LLG, the strength of these quantum fluctuations is determined by the parameter S_0 , which for correspondence with the classical case is assumed the same as the local atomic spin moment, $S_0 := \mu_i$. This assumes that the strength of the fluctuations in

TABLE III. List of symbols used in the paper and their meanings (excluding standard and redundant definitions).

Symbol	Description
$K(t - t')$	Memory kernel capturing non-Markovian effects
$\tilde{P}(\omega, T)$	Power spectrum of the thermal noise
$I(\omega)$	Spectral density of the bath (Ohmic or Lorentzian)
$\Theta(\omega, T)$	Thermostat determining temperature dependence of noise
ω_0	Resonance (center) frequency of the Lorentzian bath
Γ	Width of the Lorentzian spectral peak (broadening)
$\mathcal{A}$	Coupling strength between spin system and bath (spin-orbit coupling)
$\tilde{\omega}_L$	Effective Larmor frequency: $\tilde{\omega}_L = \gamma \cdot 1 \text{ T}$
$\mathbf{V}_i, \mathbf{W}_i$	Auxiliary variables for memory effects in numerical integration

the case of QL noise is based on the small and finite length of the spin moment μ_i rather than some auxiliary quantity such as the mass of the atom (that determines the strength of the zero-point motion). In addition, quantum fluctuations of the electronic bath itself are not considered here nor in DFT calculations of electronic structure and may also add additional sources of noise. It is possible to separate the values of S_0 (which governs the strength of the fluctuations) and μ_i (which governs the strength of coupling to an external magnetic field) through an $s-d$ model that explicitly considers

local and itinerant magnetic moments, but that is somewhat beyond the scope of the present article, as it requires a more detailed understanding of the physical nature of the thermal bath.

APPENDIX G: SYMBOLS

For a comprehensive list of symbols used in this article see Table III.

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- [1] O. N. Mryasov, U. Nowak, K. Y. Guslienko, and R. W. Chantrell, Temperature-dependent magnetic properties of FePt: Effective spin Hamiltonian model, *Europhys. Lett.* **69**, 805 (2005).
 - [2] T. A. Ostler, R. F. L. Evans, R. W. Chantrell, U. Atxitia, O. Chubykalo-Fesenko, I. Radu, R. Abrudan, F. Radu, A. Tsukamoto, A. Itoh, A. Kirilyuk, T. Rasing, and A. Kimel, Crystallographically amorphous ferrimagnetic alloys: Comparing a localized atomistic spin model with experiments, *Phys. Rev. B* **84**, 024407 (2011).
 - [3] Z. Nedelkoski, D. Kepaptsoglou, L. Lari, T. Wen, R. A. Booth, S. D. Oberdick, P. L. Galindo, Q. M. Ramasse, R. F. L. Evans, S. Majetich, and V. K. Lazarov, Origin of reduced magnetization and domain formation in small magnetite nanoparticles, *Sci. Rep.* **7**, 45997 (2017).
 - [4] S. Jenkins, R. W. Chantrell, and R. F. L. Evans, Atomistic simulations of the magnetic properties of $\text{Ir}_x\text{Mn}_{1-x}$ alloys, *Phys. Rev. Mater.* **5**, 034406 (2021).
 - [5] J. Barker and R. W. Chantrell, Higher-order exchange interactions leading to metamagnetism in FeRh, *Phys. Rev. B* **92**, 094402 (2015).
 - [6] J. Barker and G. E. W. Bauer, Thermal spin dynamics of yttrium iron garnet, *Phys. Rev. Lett.* **117**, 217201 (2016).
 - [7] Y. Toga, M. Matsumoto, S. Miyashita, H. Akai, S. Doi, T. Miyake, and A. Sakuma, Monte Carlo analysis for finite-temperature magnetism of $\text{Nd}_2\text{Fe}_{14}\text{B}$ permanent magnet, *Phys. Rev. B* **94**, 174433 (2016).
 - [8] S. Miyashita, M. Nishino, Y. Toga, T. Hinokihara, T. Miyake, S. Hirotsawa, and A. Sakuma, Perspectives of stochastic micro-magnetism of $\text{Nd}_2\text{Fe}_{14}\text{B}$ and computation of thermally activated reversal process, *Scr. Mater.* **154**, 259 (2018).
 - [9] S. Sampana Pai, J. Chureemart, R. W. Chantrell, R. Chepulskey, S. Wang, D. Apalkov, R. F. L. Evans, and P. Chureemart, Temperature and thickness dependence of statistical fluctuations of the Gilbert damping in Co-Fe-B/MgO bilayers, *Phys. Rev. Appl.* **11**, 044001 (2019).
 - [10] R. F. L. Evans, U. Atxitia, and R. W. Chantrell, Quantitative simulation of temperature-dependent magnetization dynamics and equilibrium properties of elemental ferromagnets, *Phys. Rev. B* **91**, 144425 (2015).
 - [11] M. Strungaru, M. O. A. Ellis, S. Ruta, O. Chubykalo-Fesenko, R. F. L. Evans, and R. W. Chantrell, Spin-lattice dynamics model with angular momentum transfer for canonical and microcanonical ensembles, *Phys. Rev. B* **103**, 024429 (2021).
 - [12] P.-W. Ma, C. H. Woo, and S. L. Dudarev, Large-scale simulation of the spin-lattice dynamics in ferromagnetic iron, *Phys. Rev. B* **78**, 024434 (2008).
 - [13] J. Tranchida, S. Plimpton, P. Thibaudau, and A. Thompson, Massively parallel symplectic algorithm for coupled magnetic spin dynamics and molecular dynamics, *J. Comput. Phys.* **372**, 406 (2018).
 - [14] G. P. Müller, M. Hoffmann, C. Dißelkamp, D. Schürhoff, S. Mavros, M. Sallermann, N. S. Kiselev, H. Jónsson, and S. Blügel, Spirit: Multifunctional framework for atomistic spin simulations, *Phys. Rev. B* **99**, 224414 (2019).
 - [15] J. Hellsvik, D. Thonig, K. Modin, D. Iuşan, A. Bergman, O. Eriksson, L. Bergqvist, and A. Delin, General method for atomistic spin-lattice dynamics with first-principles accuracy, *Phys. Rev. B* **99**, 104302 (2019).
 - [16] H. Lange, S. Mankovsky, S. Polesya, M. Weißenhofer, U. Nowak, and H. Ebert, Calculating spin-lattice interactions in ferro- and antiferromagnets: The role of symmetry, dimension, and frustration, *Phys. Rev. B* **107**, 115176 (2023).
 - [17] P.-W. Ma and S. L. Dudarev, Longitudinal magnetic fluctuations in Langevin spin dynamics, *Phys. Rev. B* **86**, 054416 (2012).
 - [18] M. O. A. Ellis, M. Galante, and S. Sanvito, Role of longitudinal fluctuations in L_{10}FePt , *Phys. Rev. B* **100**, 214434 (2019).
 - [19] D. Zahn, F. Jakobs, Y. W. Windsor, H. Seiler, T. Vasileiadis, T. A. Butcher, Y. Qi, D. Engel, U. Atxitia, J. Vorberger, and R. Ernstorfer, Lattice dynamics and ultrafast energy flow between electrons, spins, and phonons in a 3D ferromagnet, *Phys. Rev. Res.* **3**, 023032 (2021).
 - [20] M. Pankratova, I. P. Miranda, D. Thonig, M. Pereiro, E. Sjöqvist, A. Delin, O. Eriksson, and A. Bergman, Heat-conserving three-temperature model for ultrafast demagnetization in nickel, *Phys. Rev. B* **106**, 174407 (2022).
 - [21] E. K. Delczeg-Czirjak, O. Eriksson, and A. Ruban, The effect of longitudinal spin-fluctuations on high temperature properties of $\text{Co}_3\text{Mn}_2\text{Ge}$, *Scr. Mater.* **226**, 115213 (2023).
 - [22] J. Barker and G. E. W. Bauer, Semiquantum thermodynamics of complex ferrimagnets, *Phys. Rev. B* **100**, 140401(R) (2019).
 - [23] L. Bergqvist and A. Bergman, Realistic finite temperature simulations of magnetic systems using quantum statistics, *Phys. Rev. Mater.* **2**, 013802 (2018).
 - [24] T. Nussle, S. Nicolis, and J. Barker, Numerical simulations of a spin dynamics model based on a path integral approach, *Phys. Rev. Res.* **5**, 043075 (2023).
 - [25] T. Nussle, S. Nicolis, I. Sofos, and J. Barker, Path-integral spin dynamics with exchange and external field, *Phys. Rev. B* **112**, 054404 (2025).
 - [26] M. Berritta, S. Scali, F. Cerisola, and J. Anders, Accounting for quantum effects in atomistic spin dynamics, *Phys. Rev. B* **109**, 174441 (2024).

- [27] J. Anders, C. R. J. Sait, and S. A. R. Horsley, Quantum Brownian motion for magnets, *New J. Phys.* **24**, 033020 (2022).
- [28] S. Scali, S. Horsley, J. Anders, and F. Cerisola, SpiDy.jl: Open-source Julia package for the study of non-Markovian stochastic dynamics, *J. Open Source Software* **9**, 6263 (2024).
- [29] R. F. L. Evans, W. J. Fan, P. Chureemart, T. A. Ostler, M. O. A. Ellis, and R. W. Chantrell, Atomistic spin model simulations of magnetic nanomaterials, *J. Phys.: Condens. Matter* **26**, 103202 (2014).
- [30] F. Bloch, Zur Theorie des Ferromagnetismus, *Z. Phys.* **61**, 206 (1930).
- [31] K. Sun, M. Kang, H. Nuomin, G. Schwartz, D. N. Beratan, K. R. Brown, and J. Kim, Quantum simulation of spin-boson models with structured bath, *Nat. Commun.* **16**, 4042 (2025).
- [32] L. Chen, Y. Yan, M. F. Gelin, and Z. Lü, Dynamics of the spin-boson model: The effect of bath initial conditions, *J. Chem. Phys.* **158**, 104109 (2023).
- [33] C. R. Hogg, F. Cerisola, J. D. Cresser, S. A. R. Horsley, and J. Anders, Enhanced entanglement in multi-bath spin-boson models, *Quantum* **8**, 1357 (2024).
- [34] A. Born, R. Decker, R. Büchner, R. Haverkamp, K. Ruotsalainen, K. Bauer, A. Pietzsch, and A. Föhlisch, Thresholding of the Elliott-Yafet spin-flip scattering in multi-sublattice magnets by the respective exchange energies, *Sci. Rep.* **11**, 1883 (2021).
- [35] S. Nemati, C. Henkel, and J. Anders, Coupling function from bath density of states, *Europhys. Lett.* **139**, 36002 (2022).
- [36] Y. V. Pershin and M. Di Ventra, Memory effects in complex materials and nanoscale systems, *Adv. Phys.* **60**, 145 (2011).
- [37] U. Atxitia, O. Chubykalo-Fesenko, R. W. Chantrell, U. Nowak, and A. Rebei, Ultrafast spin dynamics: The effect of colored noise, *Phys. Rev. Lett.* **102**, 057203 (2009).
- [38] M. Frigo and S. Johnson, The design and implementation of FFTW3, *Proc. IEEE* **93**, 216 (2005).
- [39] H. B. Callen and T. A. Welton, Irreversibility and generalized noise, *Phys. Rev.* **83**, 34 (1951).
- [40] K. Miyazaki and K. Seki, Brownian motion of spins revisited, *J. Chem. Phys.* **108**, 7052 (1998).
- [41] H. Suhl, Theory of the magnetic damping constant, *IEEE Trans. Magn.* **34**, 1834 (1998).
- [42] M. Kresch, O. Delaire, R. Stevens, J. Lin, and B. Fultz, Neutron scattering measurements of phonons in nickel at elevated temperatures, *Phys. Rev. B* **75**, 104301 (2007).
- [43] A. S. Arrott and B. Heinrich, Application of magnetization measurements in iron to high temperature thermometry, *J. Appl. Phys.* **52**, 2113 (1981).
- [44] A. Arrott, Equations of state along the road to Arrott's last plot, *J. Magn. Magn. Mater.* **322**, 1047 (2010).
- [45] S. Milošević and H. E. Stanley, Equation of state near the critical point, II. Comparison with experiment and possible universality with respect to lattice structure and spin quantum number, *Phys. Rev. B* **6**, 1002 (1972).
- [46] M. Campostrini, M. Hasenbusch, A. Pelissetto, P. Rossi, and E. Vicari, Critical exponents and equation of state of the three-dimensional Heisenberg universality class, *Phys. Rev. B* **65**, 144520 (2002).
- [47] M. D. Kuz'min, Shape of temperature dependence of spontaneous magnetization of ferromagnets: Quantitative analysis, *Phys. Rev. Lett.* **94**, 107204 (2005).
- [48] H. Sato, P. Chureemart, F. Matsukura, R. W. Chantrell, H. Ohno, and R. F. L. Evans, Temperature-dependent properties of CoFeB/MgO thin films: Experiments versus simulations, *Phys. Rev. B* **98**, 214428 (2018).
- [49] E. Alp, W. Sturhahn, T. Toellner, J. Zhao, M. Hu, and D. Brown, Vibrational dynamics studies by nuclear resonant inelastic x-ray scattering, *Hyperfine Interact.* **144**, 3 (2002).
- [50] R. Lizárraga, F. Pan, L. Bergqvist, E. Holmström, Z. Gercsi, and L. Vitos, First principles theory of the hcp-fcc phase transition in cobalt, *Sci. Rep.* **7**, 3778 (2007).
- [51] Q. Li, H. Ehteshami, K. Munro, M. Marqués, M. I. McMahon, S. G. MacLeod, and G. J. Ackland, Nonexistence of the s - f volume-collapse transition in solid gadolinium at pressure, *Phys. Rev. B* **104**, 144108 (2021).
- [52] J. Crangle, G. M. Goodman, and W. Sucksmith, The magnetization of pure iron and nickel, *Proc. R. Soc. London A* **321**, 477 (1971).
- [53] H. E. Nigh, S. Legvold, and F. H. Spedding, Magnetization and electrical resistivity of gadolinium single crystals, *Phys. Rev.* **132**, 1092 (1963).
- [54] F. Cerisola, M. Berritta, S. Scali, S. A. R. Horsley, J. D. Cresser, and J. Anders, Quantum-classical correspondence in spin-boson equilibrium states at arbitrary coupling, *New J. Phys.* **26**, 053032 (2024).
- [55] S. Streib, R. Cardias, M. Pereiro, A. Bergman, E. Sjöqvist, C. Barreteau, A. Delin, O. Eriksson, and D. Thonig, Adiabatic spin dynamics and effective exchange interactions from constrained tight-binding electronic structure theory: Beyond the Heisenberg regime, *Phys. Rev. B* **105**, 224408 (2022).
- [56] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Ultrafast spin dynamics in ferromagnetic nickel, *Phys. Rev. Lett.* **76**, 4250 (1996).
- [57] R. Mondal, L. Rozsa, M. Farle, P. M. Oppeneer, U. Nowak, and M. Cherkasskii, Inertial effects in ultrafast spin dynamics, *J. Magn. Magn. Mater.* **579**, 170830 (2023).
- [58] K. Neeraj, N. Awari, S. Kovalev, D. Polley, N. Zhou Hagström, S. S. P. K. Arekapudi, A. Semisalova, K. Lenz, B. Green, J.-C. Deinert, I. Ilyakov, M. Chen, M. Bawatna, V. Scalera, M. d'Aquino, C. Serpico, O. Hellwig, J.-E. Wegrowe, M. Gensch, and S. Bonetti, Inertial spin dynamics in ferromagnets, *Nat. Phys.* **17**, 245 (2021).
- [59] A. De, J. Schlegel, A. Lentfert, L. Scheuer, B. Stadtmüller, P. Pirro, G. von Freymann, U. Nowak, and M. Aeschlimann, Magnetic nutation: Transient separation of magnetization from its angular momentum, *Phys. Rev. B* **111**, 014432 (2025).
- [60] V. Unikandanunni, R. Medapalli, M. Asa, E. Albisetti, D. Petti, R. Bertacco, E. E. Fullerton, and S. Bonetti, Inertial spin dynamics in epitaxial cobalt films, *Phys. Rev. Lett.* **129**, 237201 (2022).
- [61] F. Hartmann, V. Unikandanunni, M. Bargheer, E. E. Fullerton, S. Bonetti, and J. Anders, Intrinsic non-Markovian magnetization dynamics, *arXiv:2512.07378*.
- [62] F.-C. Weber, F. Hartmann, J. Anders, R. F. L. Evans, and M. Bargheer, Atomistic spin dynamics with quantum colored noise, Zenodo (2026), doi:10.5281/zenodo.18391021.