

High-frequency susceptibility tensor in the inertial regime: Prediction for subterahertz magnonics in ferromagnets

Jonas Wiemeler^{✉,*}, Michael Farle[✉], and Anna S. Semisalova^{✉,†}*Faculty of Physics and CENIDE, University of Duisburg-Essen, 47057 Duisburg, Germany*

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Inertial spin dynamics alters the resonant spin wave excitation resulting in the modification of the resonance field and relaxation in microwave spectroscopy of ferromagnets described by the conventional Landau-Lifshitz-Gilbert (LLG) equation. Here we present a complete theoretical framework which quantitatively predicts the dynamic response of the magnetization for ferromagnetic systems exhibiting inertia. We compute the tensor of the high-frequency magnetic susceptibility analytically using the linearization of the inertial LLG equation, with fewer approximations than in the inertial Smit-Beljers approach. Our solution yields the precession (ferromagnetic resonance) and nutation resonance frequency, relaxation and intensity for an arbitrary free energy density, allowing straightforward quantitative analysis of inertial effects in magnetic resonances. Using the typical parameters of permalloy as an example we calculate the impact of materials parameters including Gilbert damping and inertial relaxation time on the conventional magnetic resonant response as well as the amplitude, linewidth, and frequency of the typically subterahertz nutation resonance.

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I. INTRODUCTION

High-frequency spin dynamics at subterahertz frequencies became a focus of recent theoretical and experimental studies as it potentially contributes to energy efficient spin-based information processing, based on the control of spin-current generation and spin-to-charge current conversion at terahertz frequency [1].

For application of the precession at subterahertz frequencies mostly antiferromagnetic materials have been considered as most promising [2,3]. The reason is that the classical Landau-Lifshitz-Gilbert (LLG) equation yields ultrahigh-frequency (beyond 100 GHz) dynamic response of ferromagnets only for high magnetic fields of dozens of Tesla or high magnetocrystalline anisotropy [4,5]. The concept of magnetic inertia challenges the understanding of the magnetic susceptibility of magnetic materials in the subterahertz range and introduces a nutation resonance [6–15]. In recent experimental research the excitation of ultrahigh-frequency spin dynamics due to nutation in ferromagnetic thin films has been observed in permalloy $\text{Ni}_{81}\text{Fe}_{19}$, $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ and cobalt [16,17]. Recent studies also demonstrated the possibility of nonresonant excitation of spin nutation using ultrashort laser pulses [18]. The ability to excite the magnetization nutation coupled to its precession [19] significantly broadens the

potential range of applications of “conventional” ferromagnets in THz spin dynamics. Aside from our phenomenological detailed analysis presented below, several fundamental questions on the atomistic origin of the inertia in itinerant ferromagnets have been discussed [20–23]. In general, it is assumed that spin-orbit effects determine the separation and position of the conventional precession (i.e., ferromagnetic resonance, FMR) and nutation resonances as well as the relaxation of the nutation resonance superimposed on the conventional GHz-range precession. Until now, several novel concepts such as ultrafast nutational magnetization switching [24–26], spin pumping [27] and spin torque [28,29] in the presence of inertia, effects of confinement and emergence of low-frequency nutation in nanoparticles [30,31] have been reported.

Taking into account inertia effects one finds a decrease of the conventional GHz resonance in comparison to the one without inertia (classical LLG). Consequently the analysis of microwave absorption and dispersion relation by using just the LLG equation must have lead to small errors in the determination of values for relaxation and effective anisotropy parameters. Fortunately the inertia-induced changes of precession resonance characteristics are very small at low frequency and become noticeable above approximately 100 GHz excitation [19].

Furthermore, the inertia of magnetization is predicted to result in the excitation of nutation spin waves [32–37]. In thin isotropic ferromagnetic films, for example, nutation surface spin waves have been shown theoretically to propagate at much higher frequencies than Damon-Eshbach spin waves and have negative group velocity [33]. Similarly, the appearance of high-frequency exchange-mediated nutation spin waves and its inverse direction to precession waves in case of Dzyaloshinskii-Moriya interaction (or due to other nonreciprocity effects) was reported in [36].

*Contact author: jonas.wiemeler@uni-due.de†Contact author: anna.semisalova@uni-due.de

The smaller frequency of precession spin waves in both backward volume and Damon-Eshbach geometry [33] is similar to the shift of the ferromagnetic resonance frequency due to inertia [19]. Other studies showed that the inertia effects can also be seen in magnetic nanodiscs with a vortex structure. The gyrotropic motion of the vortex core excited by microwaves is overlayed with an inertia-caused rotating spin wave mode of opposite handedness [38]. These results are expected to stimulate further research of inertia-driven effects in the high-frequency range which might contribute to comprehensively investigated high-frequency magnon dynamics for applications in information processing [39,40].

The dynamics of magnetization $\mathbf{M}$ in the inertial regime is described by the inertial Landau-Lifshitz-Gilbert (iLLG) equation proposed in the works of J.-E. Wegrowe [6–8]:

$$\frac{d\mathbf{M}}{dt} = -|\gamma|\mu_0\mathbf{M} \times \mathbf{H}_{\text{eff}} + \frac{\alpha}{M_S}\mathbf{M} \times \frac{d\mathbf{M}}{dt} + \frac{\eta}{M_S}\mathbf{M} \times \frac{d^2\mathbf{M}}{dt^2}, \quad (1)$$

where γ is the gyromagnetic ratio, μ_0 is the vacuum magnetic permeability, M_S is the saturation magnetization, $\mathbf{H}_{\text{eff}}$ is an effective magnetic field which includes all anisotropy, exchange, and applied magnetic fields, α is the dimensionless Gilbert damping constant, and η is the inertial relaxation time. Earlier studies have focused mainly on the dependence of the nutation resonance frequency on η [19,41]. Up to now a general solution of the iLLG for arbitrary free energy density landscapes has been missing. Our novel framework for computational inertia predictions presents a complete analytical solution of the iLLG equation for the high-frequency AC susceptibility tensor allowing to determine the resonant microwave absorption for an arbitrary free energy density, which takes all materials parameters into quantitative account.

Our findings suggest a route for experimental detection of nutation resonance using high-frequency/high-field ferromagnetic resonance in ferromagnets and guide the prediction for suitable materials for experimental studies with respect to the relative intensities of conventional and nutational resonance.

II. RESONANCE AND HIGH-FREQUENCY SUSCEPTIBILITY TENSOR WITHIN INERTIAL REGIME

We perform the analysis for the uniform magnetization precession mode, which is the standard macrospin approximation inherent in the analysis of ferromagnetic resonance. The susceptibility tensor is calculated in accordance with the Ansatz proposed in [42] for a uniform precession. The inertial LLG equation (1) together with internal magnetic fields $\mu_0\mathbf{H}_{\text{eff}}$ (e.g., anisotropy fields are derived from the free energy density) is treated as a hypersurface $\mathbf{L}(\mathbf{m}, \mathbf{b})$:

$$\mathbf{L}(\mathbf{m}, \mathbf{b}) := -|\gamma|\mu_0\mathbf{M} \times \mathbf{H}_{\text{eff}} + \frac{\alpha}{M_S}\mathbf{M} \times \frac{d\mathbf{M}}{dt} + \frac{\eta}{M_S}\mathbf{M} \times \frac{d^2\mathbf{M}}{dt^2} - \frac{d\mathbf{M}}{dt} \stackrel{!}{=} 0 \quad \forall t, \quad (2)$$

where $\mathbf{m}(t)$ and $\mathbf{b}(t)$ are microwave-induced magnetization and the AC microwave field. $\mathbf{m}, \mathbf{b}$ are assumed as sufficiently small to treat the resonance problem in the linear regime

of microwave excitation which is a common experimental condition. The hypersurface $\mathbf{L}(\mathbf{m}, \mathbf{b})$ can be approximated in first-order Taylor expansion around the dynamic component of the magnetization $\mathbf{m} \rightarrow 0$ and $\mathbf{b} \rightarrow 0$ yielding

$$\mathbf{L}(\mathbf{m}, \mathbf{b}) \approx \mathbf{L}(0, 0) + \underline{\underline{J}}_{\mathbf{m}, \mathbf{b}} \cdot (m_x, m_y, m_z, b_x, b_y, b_z)^T, \quad (3)$$

with $\underline{\underline{J}}_{\mathbf{m}, \mathbf{b}}$ being the Jacobian matrix for $\mathbf{L}$ in $\mathbf{m}$ and $\mathbf{b}$. Equation (3) can be further split into magnetization $\underline{\underline{J}}_{\mathbf{m}}$ and field $\underline{\underline{J}}_{\mathbf{b}}$ part as well as solved for $\underline{\underline{\chi}}$, given the definition of the susceptibility $\mathbf{m} = \underline{\underline{\chi}} \cdot \mathbf{b}$:

$$\underline{\underline{\chi}}_{\text{hf}} = -[(\underline{\underline{J}}_{\mathbf{m}})^{-1} \cdot \underline{\underline{J}}_{\mathbf{b}}]. \quad (4)$$

For a more detailed derivation of $\underline{\underline{\chi}}$ see [42]. The magnetic anisotropies and other magnetic fields, which interact with the magnetization, are taken into account by the effective magnetic field $\mu_0\mathbf{H}_{\text{eff}}$. This can be derived from the Helmholtz free energy density F in spherical coordinates. For simplicity we focused on a free energy density consisting of the Zeeman term and the shape anisotropy in the thin film limit [43], assuming magnetocrystalline anisotropy K_{ani} as negligible, but any arbitrary number of energy terms could be added if needed:

$$F(\theta_H, \theta_M, \phi_H, \phi_M) = -\mu_0\mathbf{M} \cdot \mathbf{H} + \mu_0 \frac{M_S^2}{2} \cos^2 \theta. \quad (5)$$

F must be minimized numerically with regards to the applied field $\mu_0\mathbf{H}$ (given in spherical coordinates by angles θ_H and ϕ_H), to find the equilibrium position θ_M and ϕ_M of the magnetization. By this approach, it is possible to take into account the dragging effect and nonaligned modes in the FMR calculation.

The complete transverse high-frequency susceptibility $\chi_{\perp}$ dataset has been calculated for the case of a thin film with g -factor $g = 2.09$, saturation magnetization $\mu_0 M_S = 1$ T, Gilbert precession resonance damping $\alpha \in [0.001, 0.1]$, inertial relaxation time $\eta \in [0.01, 1]$ ps and external applied magnetic field $\mu_0 H \in [1, 9]$ T. Resonance field, amplitude and linewidth have been evaluated for both precession and nutation absorption resonance peaks. For easier comparison to experiments, in which the derivative of the absorption is recorded, we used the peak-to-peak linewidth $\Delta\omega^{pp}/2\pi = \frac{\Delta\omega^{\text{FWHM}}}{2\pi\sqrt{3}}$ instead of full width at half maximum (FWHM).

III. RESULTS AND DISCUSSION

A. High-frequency susceptibility and nutation resonance

Figure 1 shows the dimensionless imaginary part of the dynamic transverse susceptibility $\chi_{\perp}(\omega, M, \mu_0 H, g, \alpha, \eta)$ of thin ferromagnetic film as a function of frequency for different inertial relaxation times η calculated according to Eq. (4). We chose $\mu_0 M_S = 1$ T, $g = 2.09$ ($\gamma/2\pi = 29.25$ GHz/T), $\alpha = 0.005$ and an in-plane applied magnetic field $\mu_0 H = 1$ T. The two resonances corresponding to the precession (FMR) at low frequency (left) and the nutation at frequency up to 1 THz can be clearly resolved. The inset shows the magnified frequency range of the precession, highlighting the shift of the frequency towards lower values with increasing η . The bottom plot in Fig. 1 shows exemplary spectra at $\eta = 0.18$ ps

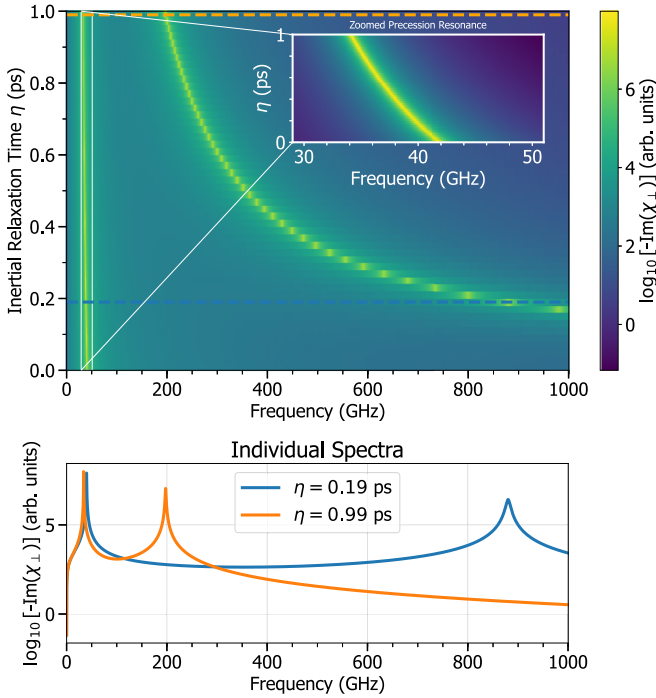


FIG. 1. Color plot of the imaginary part of the transverse susceptibility of a thin film ($N_{\perp} = 1$, $N_{\parallel} = 0$, magnetocrystalline anisotropy $K_{\text{ani}} = 0$, saturation magnetization $\mu_0 M_S = 1$ T, $g = 2.09$ ($\gamma/2\pi = 29.25$ GHz/T), Gilbert damping $\alpha = 0.005$ and external magnetic DC field $\mu_0 H = 1$ T). The $\log_{10}$ amplitude of $\text{Im}(\chi_{\perp})$ is given by the color according to the color bar to the right of the plot. The precession mode is visible as the low frequency signal below 50 GHz and shifts to slightly lower frequencies with increasing η , see inset for a detailed view. The nutation mode shifts from ≈ 200 GHz to 1 THz between $\eta = 1$ ps and $\eta = 0.18$ ps. One recognizes that for large η the conventional (FMR) resonance is repelled by the “approaching” nutation resonance as is expected for two coupled oscillators as a function of increasing coupling strength [44,45]. The inset illustrates this repulsion by magnifying the area of precession resonance. The bottom plot shows exemplary spectra selected from the colormap. The corresponding spectra at $\eta = 0.18$ ps and $\eta = 0.99$ ps are visible in the colormap as horizontal lines with matching color.

and $\eta = 0.99$ ps. In the following we analyze the resonance position, the linewidth, and the amplitude computed from the resonance spectra $\chi_{\perp}(\omega/2\pi)$. Within our model we find that the two resonances are coupled, i.e., their linewidth reduces and amplitude increases when the nutation resonance frequency becomes sufficiently low with higher η values and approaches the precession resonance frequency. This is the expected level repulsion for coupled oscillators with increasing coupling strength [44,45].

B. Effect of Gilbert damping and inertial relaxation time on magnetic resonances

Now, we describe how the nutation resonance parameters are influenced by material parameters such as the Gilbert damping α and the inertial relaxation time η . Magnetocrystalline anisotropy is considered as negligible [9,17] as for example is the case of permalloy or amorphous CoFeB thin

films. While our approach can be applied to any shape of a ferromagnetic sample (any shape anisotropy with its demagnetization factors can be included), here we focus on thin films, demagnetization factors $N_{\perp} = 1$ and $N_{\parallel} = 0$ as the most relevant candidates for applications in ultrafast spin dynamics, similarly to our earlier work [19]. One may note, however, that in the magnetic monolayer limit of ultrathin films deviations from the classical homogeneous demagnetization factors have to be taken into account [46,47].

Figure 2(a) shows the decrease of the nutation resonance frequency $\omega_N/2\pi$ of a thin film (magnetic field is applied in plane) as a function of the inertial relaxation time η . It can approximately be described as inversely proportional to η with an offset to higher frequency due to the magnetostatic energy (shape anisotropy) and Zeeman energy [19,41]: $\omega_N \approx \frac{1}{\eta} + |\gamma|(\frac{\mu_0 M_S}{2} + \mu_0 H)$ for low η values. Taking into account available data on inertial relaxation time η , one can see that the applied magnetic field typically used in experiments as well as the shape anisotropy, i.e., the saturation magnetization, only weakly affects the nutation resonance frequency in subterahertz to THz range. Figure 2(a) shows the nutation resonance frequency for a fixed Gilbert damping $\alpha = 0.005$ as a function of η . The variation of the nutation frequency for $\alpha \in [0.001, 0.1]$ is negligible, as will be discussed below. The applied magnetic field of few T has a sizable effect and leads to an increase of the nutation frequency as illustrated by curves for 1, 4, and 9 T, where the shift of the nutation resonance to higher frequency corresponds to the increase of precession frequency of the aligned FMR mode. The different behavior of nutation resonance in the nonaligned configuration of magnetization was discussed earlier in [19].

The influence of η on the nutation resonance amplitude A_N and the linewidth $\Delta\omega_N^{pp}/2\pi$ is shown in Figs. 2(b) and 2(c). The variations in A_N and $\Delta\omega_N^{pp}/2\pi$ with η are inversely related as they both are determined by the integral intensity of the absorption line which remains constant. The larger inertial relaxation time leads to an increase of nutation amplitude and to the decrease of the nutation resonance linewidth, thus favoring its experimental detection. In contrast, the nutation resonance in ferromagnetic materials with high Gilbert damping is expected to be broad and of low amplitude.

The ferromagnetic resonance frequency $\omega_p/2\pi$, amplitude A_p and linewidth $\Delta\omega_p^{pp}/2\pi$, [Figs. 2(d)–2(f)], exhibit similar trends with respect to inertial relaxation time, namely, a red shift of the precession and similar changes of $\Delta\omega_p^{pp}/2\pi$ and A_p . The latter is easy to follow by observing a reduction of the nutation and precession resonance frequencies with higher inertial relaxation time η [Figs. 2(a) and 2(d)]. When the nutation frequency reaches the GHz-range, where typically FMR frequencies belong to, the precession frequency experiences the stronger red shift and repulsion from the nutation (Fig. 1, inset), clearly demonstrating a coupling between these two modes. Overall, the effect of η parameter on the FMR absorption typically measured at frequencies below 100 GHz is only moderate [Figs. 2(d)–2(f)]—larger η leads to a reduction of the resonance frequency $\omega_p/2\pi$ and the linewidth, and to an insignificant increase of the amplitude for relatively low magnetic field such as 1 T. For example, for a considered case of an in-plane applied magnetic field of 1 T and $\alpha = 0.005$ we

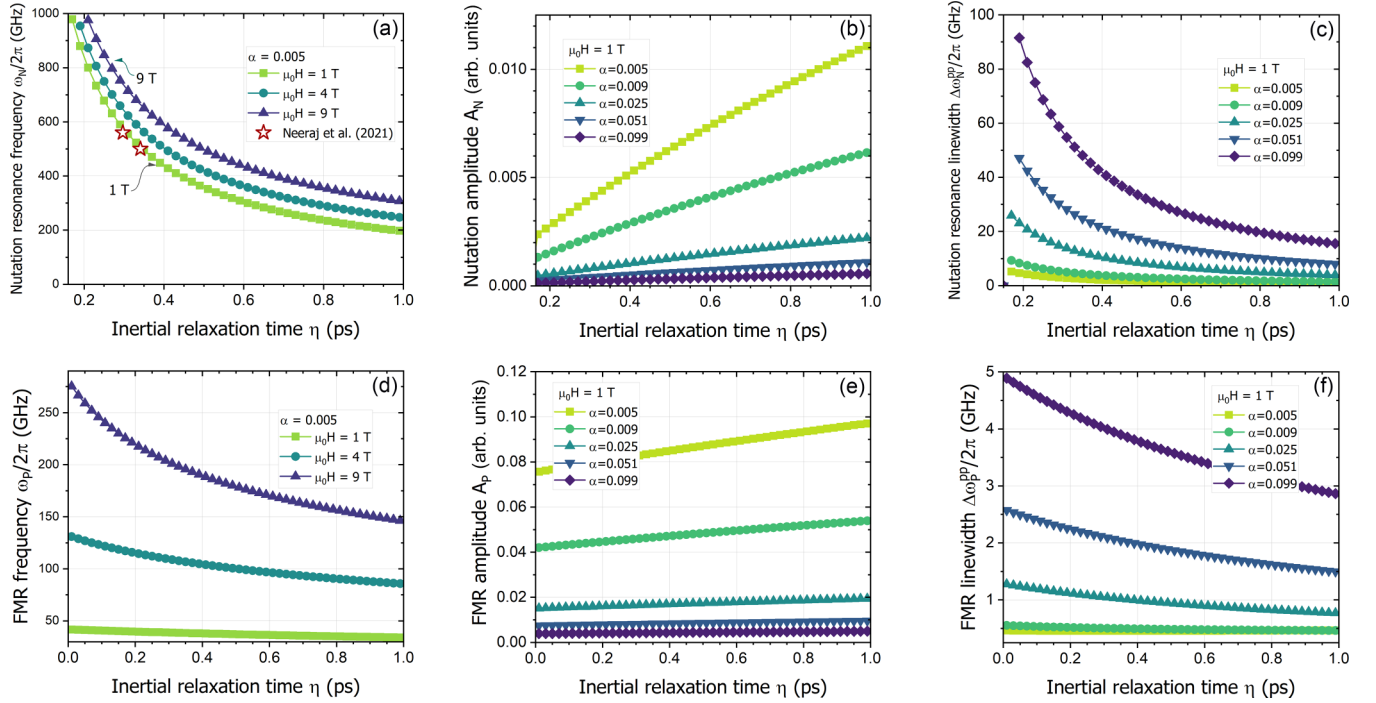


FIG. 2. The nutation (top) and precession (bottom) resonance parameters as a function of inertial relaxation time η for different Gilbert damping α according to Eq. (4): resonance frequency of nutation (a) (literature data from [17] for permalloy and CoFeB thin films measured at 0.35 T are indicated by stars for comparison) and precession (FMR) resonance (d), amplitude of nutation (b) and precession (e) resonance, the linewidth of nutation (c) and precession (f) resonance. The calculation parameters are $\mu_0 M_S = 1$ T, $g = 2.09$ ($\gamma/2\pi = 29.25$ GHz/T), magnetic field $\mu_0 H$ is applied in plane of a thin homogeneously magnetized film.

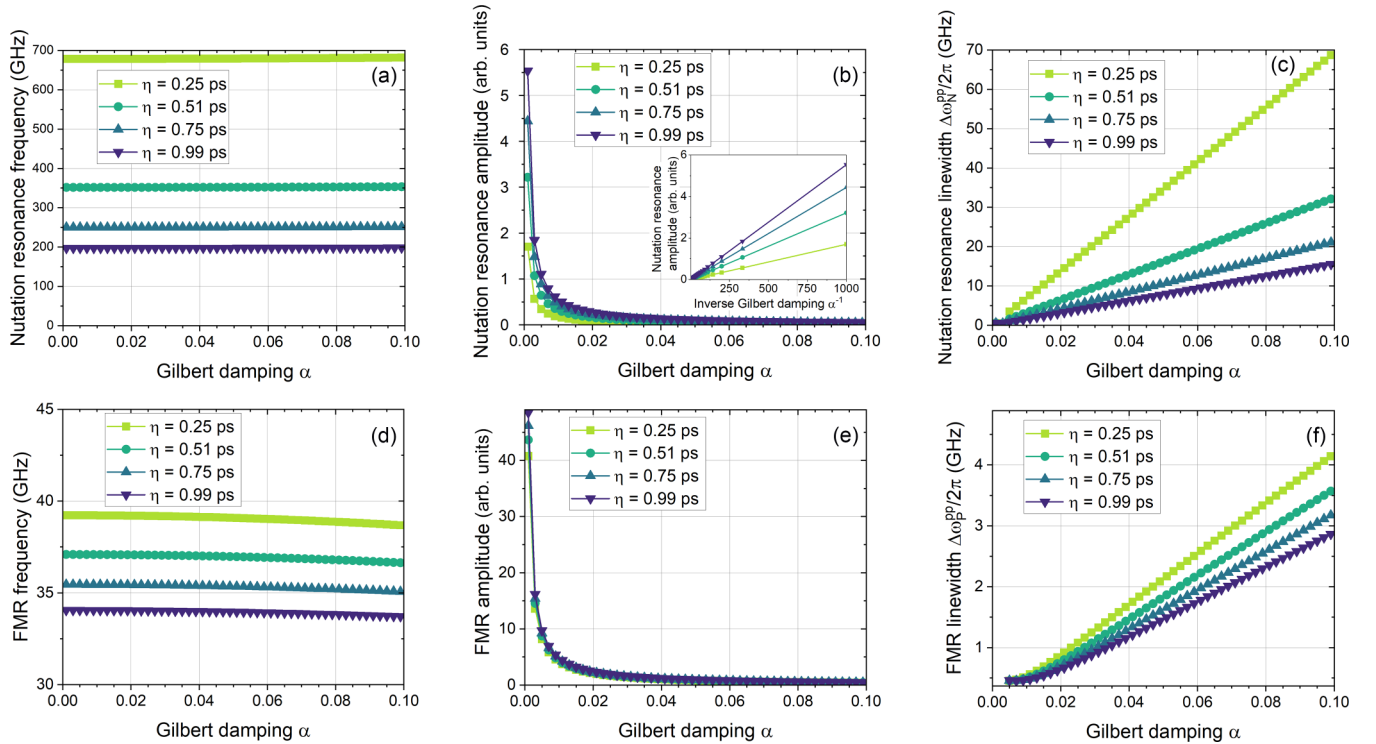


FIG. 3. The nutation (top) and precession (bottom) resonance parameters as a function of Gilbert damping parameter α for different inertial relaxation time η for a field $\mu_0 H = 1$ T applied in plane. The values are calculated from the high-frequency susceptibility $\chi_\perp$ according to Eq. (4): resonance frequency of nutation (a) and precession (FMR) resonance (d), amplitude of nutation (b) and precession (e) resonance, the linewidth of nutation (c), and precession (f) resonance. Inset in (b): amplitude of the nutation resonance A_N plotted vs. inverse Gilbert damping constant $1/\alpha$. The calculation parameters for a thin film are $\mu_0 M_S = 1$ T, $g = 2.09$ ($\gamma/2\pi = 29.25$ GHz/T).

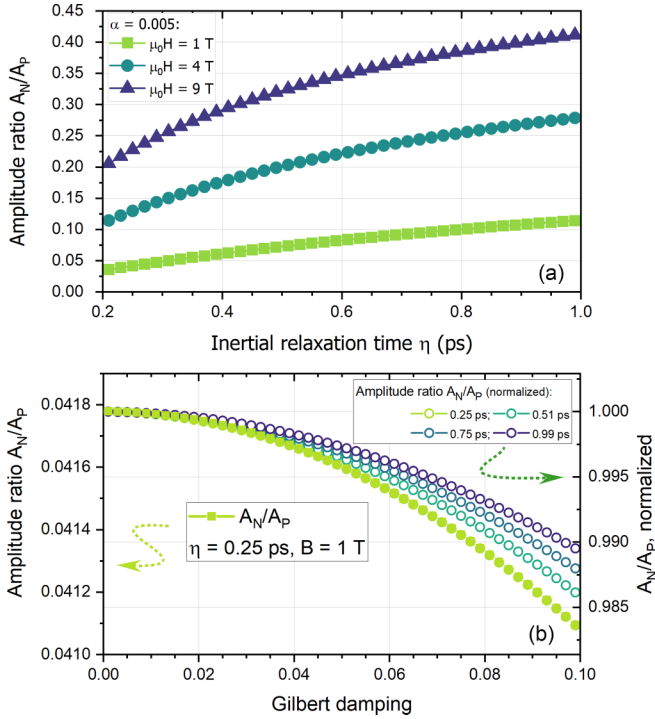


FIG. 4. The effect of inertial relaxation time η (a) and Gilbert damping α (b) on the ratio of nutation and precession amplitudes. Right axis (b) shows the relative (normalized) change of the amplitude ratio variation for different inertial relaxation time η . The calculation parameters for a thin film are $\mu_0M_S = 1$ T, $g = 2.09$ ($\gamma/2\pi = 29.25$ GHz/T), magnetic field μ_0H is applied in plane.

find a shift of $\omega_P/2\pi$ from 41.7 GHz to 34.1 GHz (18%) when η is increased from 0.01 ps to 1 ps, and this shift is less pronounced for smaller fields commonly used for measurements of microwave absorption and FMR. This explains why inertial effects have not been noticed in the context of FMR and spin wave physics until recently. We suggest that despite the small variation of $\omega_P/2\pi$ with η , magnetization inertia effects could be detected by measuring the frequency dependence of the precession ferromagnetic resonance position in strong magnetic fields and analyzing its deviation from the results of the LLG equation [19].

The opposite trend is observed when comparing materials with different Gilbert damping α (Fig. 3). While the nutation resonance frequency varies negligibly with α [Fig. 3(a)], materials with lower Gilbert damping show a larger nutation amplitude [Fig. 3(b)] and narrower resonance line [Fig. 3(c)] which is dominated by the lower dissipation rate. The analogous dependencies are also observed in the precession resonance mode of the iLLG equation—however with a factor of 6 to 18 lower frequency, an order of magnitude larger amplitude and much smaller resonance linewidth [Figs. 3(d)–3(f)].

The linear increase of the linewidths $\Delta\omega_P^{pp}/2\pi$ and $\Delta\omega_N^{pp}/2\pi$ with α is accompanied by an inversely proportional decrease of the corresponding amplitudes A_P and A_N , which favors the detection of nutation resonance in ferromagnets with low Gilbert damping such as Py [48], Fe₆₀Al₄₀ [49] Fe,

Co, Ni epitaxial magnetic thin films [50,51], CoFe [52,53], and Yttrium Iron Garnets (YIG) [54,55].

At the same time we note that the linear relation between the precession resonance linewidth $\Delta\omega_P^{pp}/2\pi$ and damping constant α is modified by an increase of the inertial relaxation time η as can be seen in Fig. 3(f) (darker lines), indicating the reduced enhancement of the linewidth $\Delta\omega_P^{pp}/2\pi$ and precession relaxation time with α due to a coupling with the nutation mode at larger η . The physical microscopic origin of the complex interplay between damping α and the inertial relaxation time, η , in ferromagnetic solids has yet to be identified. However, theoretical and experimental reports suggest that, similarly to Gilbert damping, the inertial relaxation time may be related to spin-orbit coupling [16,21]. Here we use the phenomenological iLLG approach to predict the most promising material parameters for experimental detection of the inertial effects in high-frequency spin dynamics. Since ultrahigh-frequency FMR techniques are rapidly developing, the search for suitable materials for measurements is expected to be most fruitful for ferromagnets with a large inertial relaxation time η . These materials, although η has not been understood from first principles yet, are expected to exhibit nutation in lower subterahertz frequency range.

The ratio of resonance amplitudes A_N/A_P [Figs. 4(a), 4(b)] is another important parameter defining the possibility of the experimental detection of the nutation resonance since its amplitude is at least one order of magnitude smaller than the amplitude of the FMR absorption line [Figs. 3(b), 3(e)] (see also [7]). Fig. 4 shows the effect of the inertial relaxation time η (top) and the Gilbert damping α (bottom) on the ratio A_N/A_P . The variation of the amplitudes ratio with α is negligible in our case study, decreasing by 1%–2% for the largest value of Gilbert damping. The relative change of the amplitudes ratio [right axis in Fig. 4(b)] insignificantly varies with η showing a small reduction for higher inertial relaxation time. Interestingly, at the same time the ratio A_N/A_P strongly depends on the strength of the applied field [Fig. 4(a)], highlighting further the effect of narrowing both resonance lines when the nutation frequency approaches the typical FMR frequency range [see also Figs. 2(c), 2(f)]. Finally, the relative amplitude of the nutation with respect to precession is boosted significantly by the increase in η [Fig. 4(a)] which further confirms that the materials with higher inertial relaxation time might be the best candidates for investigating the novel phenomena of inertial spin dynamics.

IV. CONCLUSIONS

In summary, we provide a theoretical framework for the calculation of the complete high-frequency magnetic susceptibility and magnetic resonance modes by the inertial Landau-Lifshitz-Gilbert equation, with the expansion of the “Unified description of collective magnetic excitations” [42] to the inertial regime, which can be used to fit experimental resonance data with fewer assumptions than required by the standard Smit-Beljers approach. Any magnetic free energy density can be selected as a starting point for the inherent dynamic susceptibility calculation. Using the example of a thin film, we provide the detailed dependence of resonance

modes and their amplitudes, linewidths, and frequencies on the Gilbert damping α and inertial relaxation time η . We show that the linewidth of both precession and nutation resonances reduces, and the amplitude increases when the nutation resonance frequency becomes sufficiently low and approaches the precession frequency for higher η values. The relative amplitude of the nutation resonance line with respect to precession was shown to increase with η as well as with the applied magnetic field. This work provides a guideline to identify materials which allow the detection of inertial effects in ferromagnets using time or frequency domain experiments.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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