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Magnetic nutation: Transient separation of magnetization from its angular momentum

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For nearly 90 years, precession and relaxation processes have been thought to dominate the magnetization dynamics. Only recently has it been considered that, on short time scales, an inertia-driven magnetization dynamics should become relevant, leading to additional nutation of the magnetization vector. Here, we trigger magnetic nutation via a sudden excitation of a thin $\mathrm{Ni}_{80}\mathrm{Fe}_{20}$ (Permalloy) film with an ultrashort optical pulse, that leads to an abrupt tilting of the effective field acting on the magnetic moments, separating the dynamics of the magnetization from that of its angular momentum. We investigate the resulting magnetization dynamics in the inertial regime experimentally by the time-resolved magneto-optical Kerr effect. We find a characteristic oscillation in the Kerr signal in the range ∼0.1 THz superimposed on the precessional oscillations with GHz frequencies. By comparison with atomistic spin dynamics simulations, we demonstrate that this observation cannot be explained by the well-known Landau-Lifshitz-Gilbert equation of motion but can be attributed to inertial contributions leading to nutation of the magnetization vector around its angular momentum. Hence, an optical and nonresonant excitation of inertial magnetization dynamics can trigger and control different magnetic processes, ranging from demagnetization via nutation to precession in a single device. These findings will have profound implications for the understanding of ultrafast spin dynamics and magnetization switching.

I. INTRODUCTION

The dynamics of magnetization in a magnetically ordered material is governed by precession [1]. Macroscopically, this precession leads to magnetic resonance phenomena that are used in a wide range of applications, from materials characterization to medical diagnostics. Microscopically, spin precession, or more precisely the precession of the spin's magnetic moment, explains the existence of spin waves and—in its quantized form—magnons as quasiparticles for the excitation of a magnetic ground state. Magnons are in the focus of contemporary spintronics [2,3] with applications in sensor devices, data processing, and energy efficient computing [4]. However, since spins are quantized angular momenta, there must be analogies to the mechanics of rotating bodies. If the rotation axis of a mechanical gyroscope is tilted away from the direction of the gravitational field by means of an external force, it will start to precess around the gravitational field, maintaining a constant angle with it. But there is a second type of dynamics: if one disturbs the precession such that the rotation axis of the gyroscope and its angular momentum are no longer aligned, an additional motion of the gyroscope around the angular momentum axis appears, known as the nutation [5,6].

In analogy to the magnetization dynamics, the axis of the gyroscope defines the magnetization. In principle, the orientation of the magnetization can deviate from the associated total angular momentum [7–11] (and in the following, we will not distinguish between spin and orbital contributions to the total angular momentum of the electronic system). For a gyroscope, the nutation dynamics can be triggered by a sudden torque pulse. In this work, we demonstrate that—in complete analogy to the mechanical case—we can trigger magnetic nutation via a sudden excitation of the magnetic system of a thin $\mathrm{Ni}_{80}\mathrm{Fe}_{20}$ (Permalloy or Py) film with an ultrashort laser pulse [Fig. 1(a)]. As in the mechanical case, a "sudden excitation" is defined by its time scale being substantially shorter than that of the resulting nutation, so the laser pulse must be much shorter than one period of nutation. This ultrashort pulse then leads to an abrupt tilting of the effective field acting on the spins' magnetic moments, which initiates a precession of the angular momentum [Fig. 1(b)]. Crucially, the magnetic inertia leads to a separation of the dynamics of the magnetization from that of the angular momentum of the spin system, initiating nutation. This effect is sketched in Fig. 1(c): In the absence of inertia, these dynamics would set in instantaneously with an abrupt change of the magnetic velocity, and magnetization and angular momentum would always be parallel. Considering inertia, the kinetic energy

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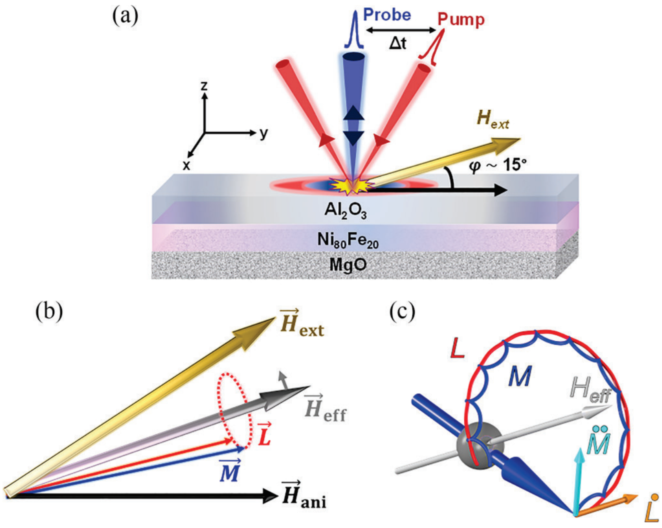

FIG. 1. (a) Schematic of the pump-probe experiment. (b) The pump pulse leads to a sudden reduction of the anisotropy field $\vec{H}_{\text{ani}}$ which, in turn, leads to a sudden tilting of the effective field $\vec{H}_{\text{eff}}$ so that the equilibrium direction for the magnetization vector $\vec{M}$ changes within some hundred fs. Consequently, the angular momentum $\vec{L}$ starts to precess. (c) Magnetic inertia causes the dynamics of the magnetization to be separated from the angular momentum dynamics. As the angular momentum starts to precess around the effective field, the magnetization accelerates toward the effective field direction, initiating nutation dynamics on top of precession.

cannot increase instantaneously. Instead, as we will show later on, the magnetization starts to accelerate towards the effective field direction giving rise to nutation. The additional nutation is clearly a consequence of the sudden change of the effective field direction.

According to theoretical predictions [7–19], nutation could add a rich variety of effects to the magnetization dynamics, including nutational resonances [8,19], nutational spin waves [20–22], a shift of the known precessional resonances [19,23], switching processes driven via a resonant excitation of nutation [24,25], and even nutational auto-oscillations [26] appear to be possible. However, all these studies are purely theoretical and—despite its fundamental importance—the experimental investigation of nutation effects is still in its infancy. Only recently, the resonant excitation of inertial spin dynamics has been experimentally observed, where THz pump-optical probe measurements were used to trigger and detect the nutation resonance [27,28]. However, our results go beyond the mere demonstration of the existence of magnetic nutation, as previously shown in [27,28], but show that the separation of the angular momentum from its magnetic moment is an inherent phenomenon occurring during the relaxation of a nonequilibrium excitation of a magnetic system using ultrashort optical pulses. This "separation" induced by nutation means that the angular momentum and magnetic moment vectors are no longer parallel to each other. This leads to a qualitatively different behavior of the dynamics of magnetization ($\vec{M}$) and angular momentum ($\vec{L}$)—a nutating and precessing $\vec{M}$, but only a precessing and non-nutating $\vec{L}$. Moreover, we provide a detailed theoretical understanding of the observed nutation dynamics. Thus, our study allows us to control the integration of different processes of magnetization dynamics, ranging from ultrafast demagnetization via nutation to precession in a single experiment.

II. RESULTS AND DISCUSSION

A. Experimental results

Spin dynamics triggered by sudden excitation of a thin $Ni_{80}Fe_{20}$ film (thickness of 5 and 2.8 nm) with an ultrashort optical pulse was observed using a time-resolved magneto-optical Kerr effect (TR-MOKE) setup based on a two-color, noncollinear pump-probe technique [see Fig 1(a) and the Supplemental Material [29]]. The samples are epitaxially grown on double-sided polished MgO substrates by molecular beam epitaxy (MBE) technique at room temperature and are capped with a 3-nm Al_2O_3 layer. After the femtosecond (fs) pump pulse, the magnetization of the system is partially or completely lost within hundreds of fs, which is known as ultrafast demagnetization [30–32]. This is generally followed by a fast recovery of magnetization within sub-ps to a few ps and a slower recovery within hundreds of ps, known as fast and slow remagnetization respectively. The slower recovery is accompanied by a precession of the magnetization [32–35]. On the shorter time scales of this slow recovery period (<300 ps), we have observed that the usual precessional dynamics is enriched by an additional oscillation with higher frequency. To quantify the frequencies of these oscillations, we have recorded TR-MOKE data up to 300 ps with very high time step resolution (as shown in Figs. 2(a) and 2(b) for 5- and 2.8-nm Py samples, respectively measured at $\mu_0 H_{\text{ext}} = 113\,\text{mT}$ and $F = 4.6\ \text{mJ cm}^{-2}$) and subtracted the ferromagnetic resonance (FMR) background with the frequency obtained from the fast-Fourier transform (FFT) of the precessional oscillations [32]. The precessional data up to longer time delays are provided in the Supplemental Material [29]. The zoomed views of the background subtracted TR data for a small time window showing nutational oscillations are depicted in Figs. 2(c) and 2(d). The TR-MOKE data up to 300 ps at $\mu_0 H_{\text{ext}} = 96\,\text{mT}$ and $F = 4.6\ \text{mJ cm}^{-2}$ are provided in the Supplemental Material [29]. The FFT power spectra obtained from the FMR background subtracted entire TR data trace (up to 300 ps) for the two samples, giving a clear mode at ~0.1 THz in both cases, are plotted in Figs. 2(e) and 2(f), respectively. Next, we have scanned the time-resolved traces (with an interval of 100 ps), and performed the FFT in each successive time window. The resulting frequency vs time profiles for the samples measured at different magnetic fields and pump fluences are summarized in Fig. 3. In all cases, a distinct mode is observed at ~0.1 THz, which we assign to the nutation. Further details of the data analysis are discussed in the Supplemental Material [29]. The nutation frequency shows a negligible dependence on the applied magnetic field and pump fluence. The TR sum signals are provided in the Supplemental Material [29], where we do not see any mode at ~0.1 THz. This is a strong criterion for interpreting our 0.1 THz oscillation as purely magnetically driven. An alternative interpretation for such high-frequency dynamics could be exchange-dominated perpendicular standing

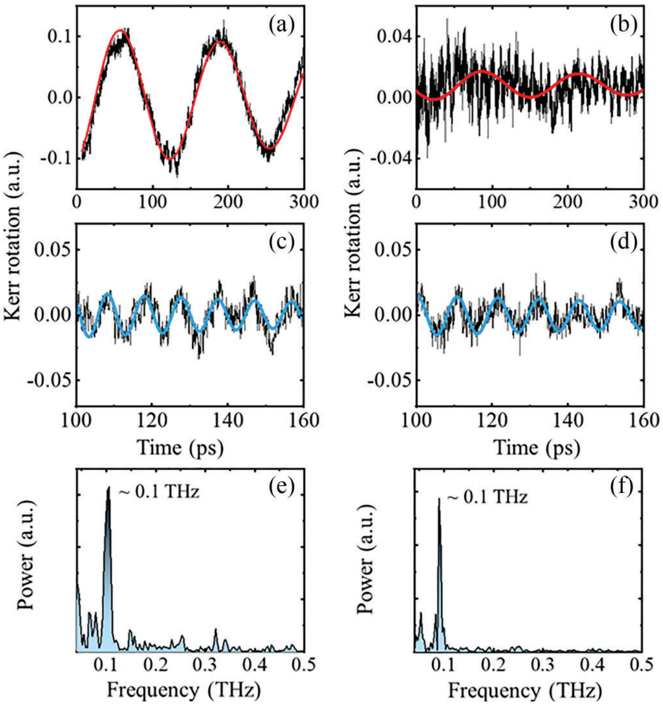

FIG. 2. Time-resolved Kerr rotation data measured up to 300 ps for (a) 5-nm and (b) 2.8-nm Py samples at $\mu_0 H_{\text{ext}} = 113\,\text{mT}$ and $F = 4.6\ \text{mJ cm}^{-2}$. The black lines are the experimental data and the red lines represent the FMR background. Zoomed views of the background subtracted TR data of (c) 5-nm and (d) 2.8-nm Py samples for small time window (100–160 ps) showing nutational oscillations. The black lines are the experimental data and the blue lines are the sinusoidal fit to the data points. The FFT power spectra from the background subtracted entire TR data (up to 300 ps) of (e) 5-nm and (f) 2.8-nm Py sample showing a strong peak at ~0.1 THz. The x axis is plotted from 30 GHz to 0.5 THz.

spin-wave (PSSW) modes across the film thickness. However, we can exclude these modes to be relevant here, since theoretically predicted PSSW modes in Py films of similar thickness appear at much higher frequencies. The calculated PSSW mode frequencies are ~1 THz and ~0.5 THz for 2.8- and 5-nm-thick Py films, respectively. Additionally, the PSSW modes exhibit an inverse relationship with sample thickness, which is not observed here.

B. Inertial Landau-Lifshitz-Gilbert equation

For nearly 90 years, magnetizations dynamics has been described by the Landau-Lifshitz (LL) equation, which—with minor modifications by Gilbert—is known as the Landau-Lifshitz-Gilbert (LLG) equation [1,36],

$$\frac{\mathrm{d}\vec{M}}{\mathrm{d}t} = -\gamma\vec{M} \times \left[\mu_0\vec{H}_{\text{eff}} - \frac{\alpha}{\gamma M_{\text{S}}}\frac{\mathrm{d}\vec{M}}{\mathrm{d}t}\right], \tag{1}$$

where γ is the gyromagnetic ratio, M_{S} is the saturation magnetization, α is the Gilbert damping constant, and $\vec{H}_{\text{eff}}$ is the effective field which includes contributions not only from an external magnetic field, but also from exchange interactions and anisotropies. The first term on the right-hand side of Eq. (1) accounts for the precession of magnetization vector ($\vec{M}$) around $\vec{H}_{\text{eff}}$. The second term with a first-order time derivative of $\vec{M}$ is the Gilbert damping term [36], which occurs due to the transfer of energy and angular momentum of $\vec{M}$ to the environmental degrees of freedom and leads to a relaxation of $\vec{M}$ towards the direction of $\vec{H}_{\text{eff}}$. However, in deriving the LLG equation, the assumption is made that, analogous to the rigid body, only one component of the diagonal inertial tensor in the body-fixed system is finite [36]. Considering an inertial tensor of the form $I = \text{diag}(I_1, I_1, I_3)$ leads to an additional second-order time derivative term in the equation of motion that plays the role of inertia for the magnetization.

The resulting inertial Landau-Lifshitz-Gilbert (ILLG) equation reads [8,37]

$$\frac{\mathrm{d}\vec{M}}{\mathrm{d}t} = -\gamma\vec{M} \times \left[\mu_0\vec{H}_{\text{eff}} - \frac{\alpha}{\gamma M_{\text{S}}}\frac{\mathrm{d}\vec{M}}{\mathrm{d}t} - \frac{\eta}{\gamma M_{\text{S}}}\frac{\mathrm{d}^2\vec{M}}{\mathrm{d}t^2}\right]. \tag{2}$$

The second order derivative term on the right-hand side of Eq. (2) gives rise to inertia and can lead to an additional oscillatory motion superimposed on top of the usual precession dynamics, known as nutation. The nutation parameter η linearly depends on I_1 and is thus directly linked to the new structure of the inertial tensor.

The time scale of the nutation is defined by η which is expected to be in the range of ps or even less, leading to oscillations of much higher frequency (sub-THz range) than those associated with spin precession (GHz range). Note that the definition of η in Eq. (2) is slightly different from former work [8]. Here it is a time scale that is independent of the damping constant α. Different types of derivations of the inertial term include phenomenological arguments [9] as well as first principles calculations [11] and a relativistic approach based on the Dirac equation [15,18]. The determination of the value of the parameter η is, however, an open question, as different studies indicate values ranging from a few femtoseconds to hundreds of picoseconds [8,10,12,27,37]. Assuming a certain value for the parameter η, however, the angular frequencies for precession and nutation (for dynamics close to equilibrium) in a magnetic field $\mu_0 H_{\text{ext}}$ can be approximated from the following formula [25,37]:

$$\omega_{\text{nu}} = -\frac{\sqrt{1 + 4\gamma\eta\mu_0 H_{\text{ext}}} + 1}{2\eta} \approx -\frac{1}{\eta}$$
$$\omega_{\text{p}} = \frac{\sqrt{1 + 4\gamma\eta\mu_0 H_{\text{ext}}} - 1}{2\eta} \approx \gamma\mu_0 H_{\text{ext}}. \tag{3}$$

Similar expressions, which reproduce the first term of the expansions, were also derived in Ref. [13]. Note the different sign of ω_{nu} and ω_p, which indicates that the sense of rotation of nutation is opposite to that of precession. In our experiments, we observed a nutation frequency of ~0.1THz, so that for our samples, $\eta = 1/\omega_{\text{nu}}$ comes out to be ~1.6 ps, which is about five times larger than the corresponding value reported in Ref. [27]. The differences in the nutation frequencies are intriguing and need to be discussed. We observe the nutation frequency in the same order of magnitude as previously observed [27], and in the range predicted by theory [38,39]. In our work, a different excitation scheme may have caused some modifications in the system properties, affecting η. We performed measurements after strong ultrafast demagnetization

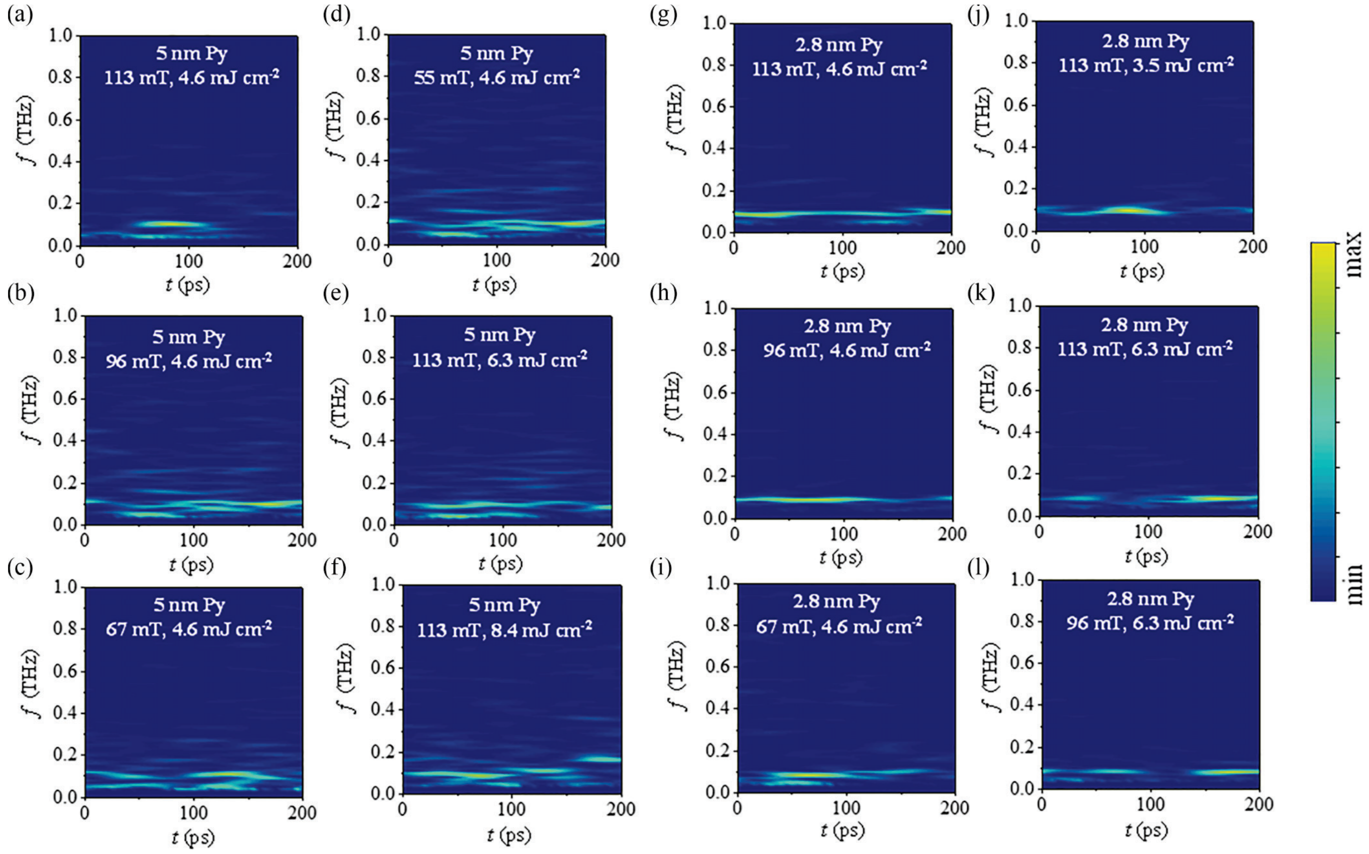

FIG. 3. Frequency vs time plots obtained from the FFT of the background subtracted experimental time-resolved Kerr rotation signals for the 5-nm and 2.8-nm Py samples at different magnetic fields and pump fluences showing a clear mode at ∼0.1 THz in each case. The sample thickness and values of $\mu_0 H_{\text{ext}}$, F are shown in the respective plots.

by femtosecond optical pulse (∼1.5 eV), which resulted in the generation of hot electrons above the Fermi level. In contrast, Ref. [27] used much lower photon energy and pulse intensity of the driving THz field (∼1 meV), resulting in the absence of hot electrons and subsequent quenching of the magnetization on picosecond time scales. Therefore, we can conclude that Ref. [27] measured nutation in a close to equilibrium regime, while our case involved a regime far from equilibrium, which could affect η and the nutation frequency. A more detailed discussion can be found in the Supplemental Material [29] (see also Refs. [37–40] therein). Further investigation of this time-dependent behavior is needed for more insight into the dynamics.

C. Theoretical analysis

For a deeper analysis of the dynamics of magnetization and angular momentum in the inertial regime we perform atomistic spin dynamics simulations based on the classical Heisenberg model and the ILLG equation (details are provided in the Supplemental Material [29]). To be able to compare with our experiments, we use a permalloy model from Ref. [41], where the atomic magnetic moments are located on an fcc lattice with 20% iron atoms and 80% nickel atoms. We use the nutation parameter of $\eta = 1.6$ ps obtained from our experiments above and a magnetic field $\vec{B} = (0,\ 110,\ 30)^{\mathrm{T}}$ mT with an out-of-plane component as in the experiments. Then we solve the stochastic ILLG equation on an atomistic level as described in the Supplemental Material [29] and calculate the time-dependent magnetization curve $M_z(t)$ shown in Fig. 4(a). After a thermal excitation with a heat pulse, the system shows the precessional dynamics around the effective field with a period in the range of hundred picoseconds. Due to the nutation, a second oscillation on the single picosecond time scale arises. This can be seen in the inset of Fig. 4(a), showing that the nutational mode has a period in the range of some picoseconds. The Fourier transform in Fig. 4(b) shows that the nutation frequency is ∼0.1 THz. With these parameters, the nutation frequency is very similar to the experimentally determined one from Fig. 3 and also matches the analytical expectation from Eq. (3). Note, however, that here the actual frequency and not the angular frequency is considered.

Furthermore, our simulations allow us to investigate the behavior of the angular momentum $\vec{L}$ of the system, which is—due to the inertia—no longer parallel to the magnetization [37]. The relation between the magnetization $\vec{M}$ and the corresponding angular momentum $\vec{L}$ (per volume) is given by [37]

$$\vec{L} = \frac{1}{\gamma}\vec{M} - \frac{\eta}{\gamma M_{\mathrm{S}}}\vec{M} \times \frac{\mathrm{d}}{\mathrm{d}t}\vec{M}. \tag{4}$$

This equation also follows from the new structure of the inertial tensor with a finite I_1 and describes a separation of

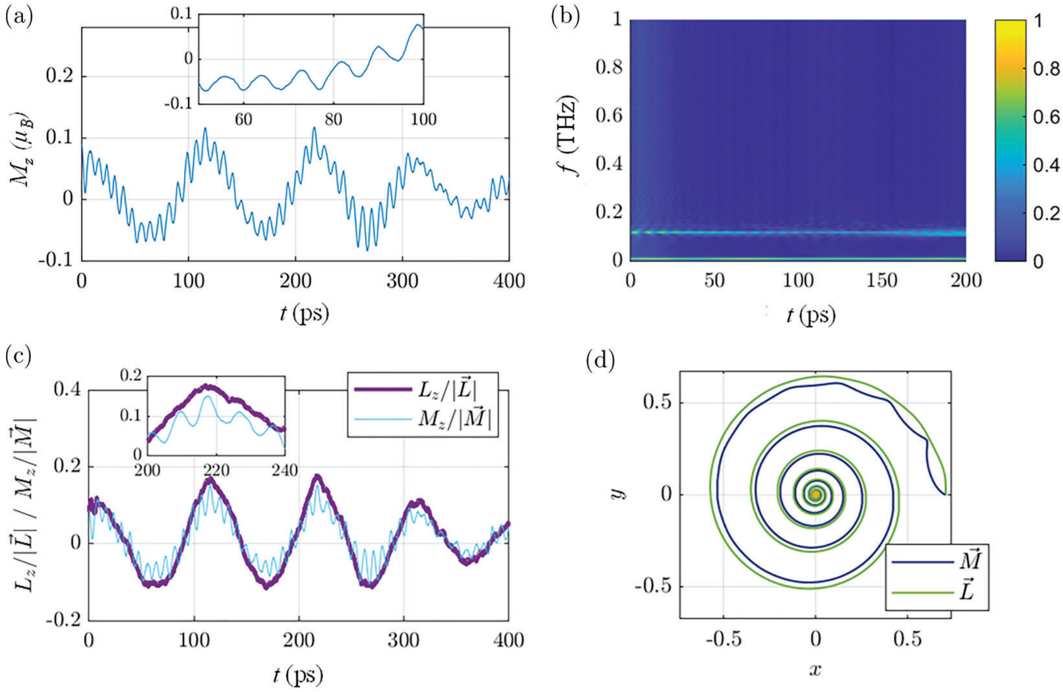

FIG. 4. (a) Time-dependent magnetization curve of the simulated Py model after an excitation with a rectangular ultrashort heat pulse. The magnetization shows both precession and nutation. (b) Frequency vs time plot of the spectral power density $P_z f^2$, unveiling the nutation frequency of ~0.1 THz and the precession frequency of ~0.01 THz. The spectral power density is multiplied by f^2 to reduce the thermal background noise. (c) Temporal dependence of the reduced magnetization and angular momentum of the out-of-plane component, showing that the magnetization nutates around the angular momentum. (d) Sketch of the trajectories of magnetization and angular momentum following a sudden tilting of the effective field, which is then perpendicular to the image plane.

the directions of the magnetization vector and the angular momentum vector. In the case of vanishing nutation (i.e., $\eta = 0$), these two vectors are aligned. They are separated for a finite value of η and a time-dependent magnetization. The ratio between the magnetization and the part of the angular momentum pointing in the magnetization direction (the projection of $\vec{L}$ on $\vec{M}$) is still given by the gyromagnetic ratio.

The temporal behavior of the system's angular momentum is shown in Fig. 4(c). On long time scales the angular momentum and the magnetization show the same precessional dynamics in the frequency range of single GHz, which is identical to the case of vanishing nutation. We point out that the nutation mode is only reflected in the magnetization, since only the magnetic moments nutate around their associated angular momentum. On the contrary, the angular momentum just precesses around the external field. As our simulations demonstrate, the nutation leads to a separation of the magnetic moment from its angular momentum, not only on the atomistic scale but also on a macroscopic level, splitting up $\vec{L}$ and $\vec{M}$ for a system of interacting magnetic moments. We experimentally measure the dynamics of the magnetization ($\vec{M}$), but not the angular momentum ($\vec{L}$), because the magneto-optical (MO) effects are given by the (transient) response of the spin-polarized electronic band structure. Therefore, a direct experimental verification of Eq. (4) is not possible. The sudden emergence of the nutation, as triggered by the laser pulse, can be understood when bringing the ILLG equation into an explicit form [37] that resembles the Newton's equation of motion:

$$\eta \frac{\mathrm{d}^2}{\mathrm{d}t^2}\vec{M} = -\frac{\gamma}{M_\mathrm{S}}\vec{M} \times (\vec{M} \times \mu_0 \vec{H}_\mathrm{eff}) - \alpha \frac{\mathrm{d}}{\mathrm{d}t}\vec{M} - \frac{1}{M_\mathrm{S}}\vec{M} \times \frac{\mathrm{d}}{\mathrm{d}t}\vec{M} - \eta \frac{1}{M_\mathrm{S}^2}\vec{M}\left(\frac{\mathrm{d}}{\mathrm{d}t}\vec{M}\right)^2. \tag{5}$$

Initially we consider a resting magnetization $\vec{M}$ parallel to the effective field $\vec{H}_\mathrm{eff}$. A sudden tilting (here via a fs laser pulse) of the effective field away from the magnetization direction starts the dynamics of both magnetization and angular momentum. As the initial magnetization rests, $\frac{\mathrm{d}}{\mathrm{d}t}\vec{M} = 0$ applies and only the first term on the right side of Eq. (5) is nonzero. Thus, the magnetization accelerates towards the effective field, perpendicular to the precession direction. After this initial acceleration, other terms have to be considered, leading to the magnetization dynamics as sketched schematically in Fig. 4(d). For the angular momentum, however, the initial acceleration of $\vec{M}$ leads to an immediate velocity $\frac{\mathrm{d}}{\mathrm{d}t}\vec{L} = -\frac{\eta}{\gamma M_\mathrm{S}}\vec{M} \times \frac{\mathrm{d}^2}{\mathrm{d}t^2}\vec{M} = -\vec{M} \times \mu_0 \vec{H}_\mathrm{eff}$ according to Eqs. (4) and (5), pointing in the precession direction. So, while the magnetization responds inert and accelerates due to the nutation term in the ILLG equation, the angular momentum starts to precess instantaneously, which makes it inertia free in contrast to the magnetization. Consequently, the magnetization does not follow the angular momentum instantaneously after the excitation. An important indicator of the dynamics of nutation is that it decays more rapidly than precession.

III. CONCLUSION

In conclusion, using the TR-MOKE experiments and atomistic spin dynamics simulations, we show that in thin $Ni_{80}Fe_{20}$ films coherent magnetic nutation can arise from a strong nonequilibrium of the spin system after ultrafast optical excitation leading to the transient separation of magnetization from its angular momentum. These findings cannot be explained by the ubiquitous Landau-Lifshitz-Gilbert equation but agree with an extension of this equation of motion that contains a second-order time derivative describing inertia. The magnetic nutation is in the high-frequency regime (sub-THz range) and superimposed on the precession dynamics (GHz range). Our results demonstrate that the magnetic nutation and the corresponding separation of the angular momentum from its magnetic moment is an intrinsic phenomenon occurring during the relaxation of an ultrafast nonequilibrium excitation of a magnetic system and does not only rely on its resonant excitation [27,28]. Hence, an optical excitation of inertial spin dynamics can enable and control the integration of different magnetic processes, ranging from demagnetization via nutation to precession in one device. This will have significant consequences in the field of ultrafast spintronics. The discovery of Einstein and de Haas [42] has connected magnetic moment with angular momentum, and, hence, linked electrodynamics with mechanics. Our findings show that nutation can separate effects that rely on magnetization dynamics from those that rest on properties of the associated angular momentum. While most effects in spintronics rely on magnetic or magneto-optic effects, the recently discovered ultrafast transfer of spin angular momentum into the lattice [43] rests on (spin plus mechanical) angular momentum conservation. Similarly, the recently established research into chiral phonons [44,45] can connect mechanical with magnetic degrees of freedom [46,47]. The separation of these quantities—magnetic moment and angular momentum—on ultrashort time scales will have profound implications for the understanding of ultrafast spin related physics.

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