Nutation Wave as a Platform for Ultrafast Spin Dynamics in Ferromagnets

I. Makhfudz,1, a) E. Olive,1 and S. Nicolis2
1) GREMAN, UMR 7347, Université de Tours-CNRS, INSA Centre Val de Loire, Parc de Grandmont, 37200 Tours, France
2) Institut Denis Poisson, Université de Tours, Université d’Orléans, CNRS (UMR7013), Parc de Grandmont, F-37200, Tours, France

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The remarkable experimental discovery of femtosecond scale laser pulse-driven demagnetization process in ferromagnetic nickel in mid 90s spurred a flurry of experimental and theoretical research activities in ultrafast magnetization dynamics. Standard theoretical description of magnetization dynamics based on Landau-Lifshitz-Gilbert equation is justified only for slow enough spin phenomena while those occurring at very short time scales are much less understood. The past two decades have observed a remarkable theoretical development predicting the emergence of dynamical inertia in magnetization dynamics at very short time scale, focusing mostly on the dynamics of a single spin, leading to the prediction of a new type of spin motion called nutation. We advance this theoretical progress by considering inertial effect on the dynamics of a system of interacting spins. We demonstrate the occurrence of a new type of collective mode referred to as nutation wave, shown to have massive relativistic dispersion relation with characteristic speed and frequency well exceeding those of the more familiar spin wave. These excellent properties make nutation wave a prospective candidate to be a platform for optically-driven ultrafast spintronic devices.

Spin dynamics plays a crucial role in technological applications of magnetism and is one of the mainstays in the emerging field of spintronics1. As the demand for efficient and high-speed electronic devices is becoming more urgent than ever, the drive towards fast or even ultrafast spin dynamics has been an object of intense pursuit by the research community. The existing magnetic switching technology is limited in speed for real applications in magnetic data storage devices5.

While magnetization dynamics is normally described by the well known Landau-Lifshitz-Gilbert equation6,7, recent theoretical studies have found that a term, corresponding to the inertia of a spin or single domain of uniformly magnetized ferromagnet, appears at short time scales8-10,11,12,13,14 and is also distinct from the inertia of topological defects or spin textures, such as that of a Skyrmion bubble15. A useful mechanical analogy is a spinning top: This inertia term originates from the transverse components of the inertia tensor I_{xy(y)}8-9. It gives rise to a so far-neglected type of spin motion; the nutation of the spin, which is a cycloidal motion with a high characteristic frequency, which is of particular relevance in ultrafast spin dynamics. A recent experiment has reported the observation of such high-frequency nutation dynamics16.

In this contribution to SPIE Optics+Photonics 2021 Conference, we point out that a type of collective excitation appears in spin chains, in the presence of such a one-site inertia term, when the spins interact through a spin-exchange interaction. This excitation takes the form of a wave of nutation, be it referred to as the “nutation wave”. The corresponding single-particle excitation, called the “nutation”, here, is found to have a gap at zero wave vector and the dispersion relation of a relativistic particle, that we identify with the Higgs mode arising from the coupling between the scalar bosons, that describe the magnetization profile and an emergent gauge boson, that describes the phase of the profile, along the chain. Experiments that could probe these properties are proposed.

We can describe the appearance of the nutation wave as follows. Consider the single–site inertial Landau-Lifshitz-Gilbert (ILLG) equation6,8,9,10,11:

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \left[ \mathbf{H}_{\text{eff}} - \eta \left( \frac{d\mathbf{M}}{dt} + \frac{\tau}{N} \frac{d^2\mathbf{M}}{dt^2} \right) \right]$$

(1)

in terms of the magnetization vector \(\mathbf{M}\), where \(\gamma\) is the gyromagnetic constant, \(\eta\) is the Gilbert damping and \(\tau\) is the spin relaxation time. The equation (1) has a microscopic origin as a torque equation and is applicable even to an individual spin (or magnetic moment). The second time derivative is an inertia term that has been shown to give rise to nutation, that describes oscillatory motion on top of the damped precessional motion produced by the first two terms on the right hand side6,8,9,10,11.

a)Electronic mail: Author to whom correspondence should be addressed: imakhfudz@gmail.com
Existing studies have so far mostly assumed perfectly uniform magnetization, subject to uniform external field $\mathbf{H}_{\text{eff}} = H$. In realistic situations, there would be a spatial variation in the magnetization $\mathbf{M}$ even in single domain system. We focus here on the consequences of such a spatial variation. In the presence of exchange interaction, the effective field is given by $\mathbf{H}_{\text{eff}} = \mathbf{H} + J_{ij} \partial^2 \mathbf{M}/\partial x_i \partial x_j$ in the ILLG model for which $d_{ij} = J \delta_{ij}, i, j = x$, now becomes

$$\frac{d \mathbf{M}}{dt} = \gamma \mathbf{M} \times \left( \mathbf{H}_{\text{eff}} - \eta \left( \frac{d \mathbf{M}}{dt} + \tau \left( \frac{d^2 \mathbf{M}}{dt^2} - \frac{v_{\text{iner}}^2}{\eta} \frac{\partial^2 \mathbf{M}}{\partial x^2} \right) \right) \right)$$

where

$$v_{\text{iner}}^2 = \frac{J}{\eta} \tau$$

Eq. (2) is a nonlinear wave equation. We shall study its solutions for the case of a single ferromagnetic domain with small spatial non-uniformities, i.e. no domain walls or solitons.

In order to describe small fluctuations or excitations, it is necessary to define an appropriate background and study the linear excitations about it. To this end we shall linearize eq. (2), by writing its solution in the form

$$\mathbf{M}(x,t) = \mathbf{M}_0 + \tilde{\mathbf{M}}(x,t)$$

where $\mathbf{M}_0$ is a spatially uniform magnetization. Furthermore, we rewrite eq.(2) in space-time translation invariant form that give the following equation

$$D_\tau \tilde{\mathbf{M}} = \gamma \left[ \mathbf{M}_0 \times \left( \tilde{\mathbf{H}} - \eta \left( D_\tau + \tau D_{x=x=+} D_{x=-} \right) \tilde{\mathbf{M}} \right) - \tilde{\mathbf{H}} \times \tilde{\mathbf{M}} \right]$$

where the space-time translation operator is given by $D_\tau = \partial_\tau - \nu \partial_\sigma$ (with $\nu = \pm \tilde{v}_g, \tilde{v}_{x=\pm}$). The velocity $\tilde{v}_g$ is the characteristic velocity of a travelling wave solution of the eq.(5) while $\tilde{v}_{x=\pm} = s v_{\text{iner}} \pm v_g$ is another velocity that reflects the relativistic part of the equation of motion. Eq. (5) contains a part that mimics the one in the standard wave equation. A wave $\psi(\pm ct)$, traveling with speed $v_{x=\pm} = sc$.

In order to verify that such a propagating wave solution exists, beyond the linear approximation, we solved eq. (2) numerically. Fig. 1. obtained from solving the equation around

![FIG. 1. The time evolution of the spins in terms of the spherical angles: $\phi(x,t)$ (a) and b)) and $\theta(x,t)$ (c) and d)) (in radians) e) The spherical vector illustration of the time evolution of a spin at fixed $x = 0$ displaying a nutation f) The snapshots of the spin chain at $t_0 = 0$ and $t_1 = 2$ showing a propagating nutation wave (blue arrow shows the wave propagation direction along $x$). The spherical coordinate frame for $\mathbf{M}$ uses the polar angle $\theta$ and azimuthal angle $\phi$ as per standard conventions. One unit of distance (time) in the figures corresponds to $a = 2(\tau = 10^{-14}s)$. FIG. 2. The profile of the magnetization vector with nutation around the equatorial plane.]

A static background as shown in Fig. 2. clearly shows the occurrence of the nutation wave in terms of the spherical angles $\theta(x,t)$ and $\phi(x,t)$. The existence of the nutation wave is made possible by the exchange field that "mediates" the propagation of this wave.

We now derive the dispersion relation of the nutation wave
by expanding the magnetization vector \( \mathbf{M} \) in eq.(2) using
\[
ed \det \left[ i \omega \delta_{ij} - (i \gamma \eta \omega + i \gamma \eta \tau \omega^2) - \gamma \omega \tau \nu_{\text{iner}} k^2 \right] = 0
\] (7)
where "det" represents the determinant of the \( 3 \times 3 \) matrix in its argument and \( I, J, K = x, y, z \) label the components of the vector \( \mathbf{M} \). \( \sum_k M_{0,k} = M_z^2 \) while \( \nu_{\text{iner}} \) is the totally antisymmetric tensor. This equation is further subject to the constraint \( |\mathbf{M}|^2 = M_z^2 \) which gives
\[
\mathbf{M}_0 \cdot \delta \mathbf{M} = 0
\] (8)
to linear order in \( \delta \mathbf{M} \) which simply means that the fluctuation of the magnetization \( \delta \mathbf{M} \) is normal to the the static background \( \mathbf{M}_0 \), as illustrated in Fig. 2. The constraint reduces the effective matrix into \( 2 \times 2 \) matrix and results in the following polynomial equation for \( \omega \)
\[
\omega^2 = (\omega_k - \alpha \omega (i + \omega \tau))^2
\] (9)
where
\[
\omega_k = \gamma JM_z k^2
\] (10)
The solutions of eq.(9) consist of two pairs of solutions; the two pairs differ only by opposite directions of propagation. We will only give the equations for the right-moving solutions, whose (complex-valued) dispersion relations are given by
\[
\omega_{\pm}(k) = \left( \pm 1 + i \alpha \right) \sqrt{4 \alpha \tau \omega_k + (\pm 1 + i \alpha)^2}
\] (11)
We will first focus on the real part of the complex energy dispersions eqn. (11).
The first solution \( \omega_+(k) \) turns out to give a non-relativistic (quadratic) dispersion relation associated with a precession mode
\[
\epsilon_{\text{sw}}(k) = \text{Re}[\omega_+(k)] = \gamma JM_z k^2
\] (12)
at small wave vector \( k \), which corresponds to the standard form of a spin wave dispersion\(^{20} \). The important point is that this dispersion is gapless; i.e. \( \epsilon_{\text{sw}}(k = 0) = 0 \).
The second solution \( \omega_-(k) \) turns out to give a massive dispersion relation. Its real part is given by
\[
\epsilon_{\text{sw}}(k) = \text{Re}[\omega_-(k)] \simeq \frac{1}{2 \alpha \tau} + \sqrt{\nu_{\text{iner}}^2 k^2 + \frac{1}{4 \alpha^2 \tau^2}}
\] (13)
where \( \alpha = \gamma M_z \eta \) with a gap \( m = 1/(\alpha \tau) \). We interpret this second solution as a propagation of the nutation; that is, the nutation wave that turns out to have a massive relativistic dispersion, having single-particle excitations (the "nutations") with mass \( m \).
Writing the full complex wave dispersion relations as
\[
\omega_{\pm}(k) = \omega_{\pm}(k) - i \alpha \omega_k
\] (14)
where \( \omega_{\pm}(k) = \epsilon_{\text{sw}(nw)}(k) \), the imaginary part \( \omega_k \) for spin wave is found to be
\[
\omega_k = \frac{1}{\tau} - \alpha \omega_k = \alpha m - K k^2
\] (15)
which implies that the zero wave vector nutation wave actually decays faster and as the wave vector increases (or wave length decreases), the nutation wave becomes even more robust (slowly-decaying).
More precisely, the robustness of the nutation wave can be described by the ratio between the imaginary and real parts
\[
r(k) = \frac{\omega_k}{\omega_{\pm}(k)} = \frac{\alpha m - K k^2}{m/\tau + \sqrt{\nu_{\text{iner}}^2 k^2 + (\frac{m}{\tau})^2}}
\] (17)
Physically, \( 1/r(k) \) gives the number of periods that a nutation wave with wave vector \( k \) covers as it propagates and decays with time. Defining \( k = (m/(2 \nu_{\text{iner}})) \kappa = k_0 \kappa \), where \( \kappa \) is a dimensionless wave vector parameter and \( k_0 = m/(2 \nu_{\text{iner}}) \) is the crossover wave vector from quadratic to linear dispersion, we have
\[
r(\kappa) = 2 \alpha \frac{1 - \kappa^2}{1 + \sqrt{\kappa^2 + 1}}
\] (18)
Since \( \alpha \lesssim 0.1 \) in real materials and the dispersion is practically linear once \( \kappa \gtrsim 1 \) as described by the group velocity
\[
v_g(k) = \frac{\partial \omega_k(k)}{\partial k} = v_{\text{iner}} \frac{\kappa}{\sqrt{\kappa^2 + 1}}
\] (19)
that already gives \( v_g \simeq v_{\text{iner}} \) for \( \kappa \gtrsim 1 \), we conclude that \( r \ll 1 \) indeed, indicating that we have a high speed nutation wave that will be robust against decay within time scale for observation \( \tau_d = 2 \pi/\omega_{\pm}(k_0) \gg \tau, 2 \pi/\omega_k(k_0) \) and over a length
scale $l_d \sim v_{iner} \tau_d$. Indeed for realistic materials parameters that we will exemplify at the end of this section, we find that

$$l_d = \frac{8 \pi v_{iner} \tau}{3} \gg a$$  \hspace{1cm} (20)$$

where $a$ is the lattice constant of typical spin chain materials.

The nutation wave dispersion relation eq.(13) describes a relativistic massive particle, which thus defines a completely different excitation from that of a spin wave, which is gapless and non-relativistic at small wave vectors, as illustrated in Fig. 3.

![Dispersion Relations (Frequency versus Wave Vector k)](image)

FIG. 3. An illustrative profile of the dispersion relations of the nutation wave $\varepsilon_{nw}(k)$ (upper curve) and of spin wave $\varepsilon_{sw}(k)$ (lower curve) where the units are $m = 10^{14}$ Hz for the frequency and $k_0 = 0.714A^{-1}$ for the wave vector $k$. The dashed straight lines are guides to the eye on the linear-in-$k$ part of the nutation wave dispersion at large $k$.

We now compare the characteristic frequencies of the two waves. At the crossover wave vector $k_0$, the nutation wave frequency is found to be $\varepsilon_{nw}(k_0) = (1 + \sqrt{2})m/2$. On the other hand, we find that for the spin wave $\varepsilon_{sw}(k_0) = m/4$. This indicates that the nutation wave always has a higher frequency than spin wave when the former enters the linear part of its dispersion.

Our results can be directly verified in one-dimensional or quasi-one-dimensional magnetic materials such as CuCl$_2$ − TMSO, C$_4$H$_8$SO(TMSO), C$_2$H$_6$SO(DMSO) and (C$_2$H$_{11}$NH$_3$)$_2$CuCl$_4$. Considering C$_4$H$_8$SO(TMSO) and C$_2$H$_6$SO(DMSO), the best fit gave exchange constants $J_{exch}/k_B = 39K$ and 45K (where $k_B$ is the Boltzmann constant) and lattice spacing $a \approx 2A$. As an illustration, using $J = J_{exch}/(M_s a)$, $\gamma = 1.75 \times 10^{11}$ sA/kg, $\tau = 10^{-14}$ s, $M_s = 10^6$ A/m and $a = \gamma M_s = 0.10$, we obtain the nutation wave resonance frequency at $k = k_0$ to be $\varepsilon_{nw}(k_0) \approx 1.207 \times 10^{15}$ Hz and characteristic velocity $v_{iner} = \sqrt{\gamma M_s / (\pi \alpha)} \approx 22 \times 10^4$ m/s which is nearly one order of magnitude larger than the characteristic group velocity of a spin wave $v_{sw} = \partial \omega_{sw}(k)/\partial k|_{k=sw} \approx 3 \times 10^3$ m/s corresponding to the typical value known in literature (e.g. 24–25), with $k_{sw} \approx 0.14k_0$. This estimate thus validates our conclusion regarding the primacy of the nutation wave in terms of its characteristic speed and frequency. Even more realistic smaller values of $\alpha$ still give larger nutation wave velocity $v_{iner}$ than spin wave $v_{sw}$. Using $\alpha = 0.01, \tau = 10^{-12}$ s for example, we obtain $\varepsilon_{nw}(k_0) \approx 1.207 \times 10^{14}$, $v_{iner} \approx 7 \times 10^4$ m/s as illustrated in Fig. 3., and $k_0 = 0.7144A^{-1}, l_d = 586A, r(k_0) = 0.006$, indicating a robust wave.

The nutation wave would appear as a peak, along with a gap, in the structure factor $S(\omega, k)$, as measured in inelastic neutron scattering experiments where the peak is given by $\omega = \varepsilon_{nw}(k)$. From a practical point of view, the nutation wave may have significant consequences in the field of spintronics. In particular, despite being smaller in amplitude, compared to precession spin wave\textsuperscript{11}, the nutation wave has much higher characteristic frequencies than the latter. Furthermore, the robustness of the nutation wave at short wavelengths is perfect for spintronic applications, such as in realizing much faster magnetic switching processes\textsuperscript{26}.

We have demonstrated the emergence of new collective mode using essentially classical approach to magnetism in terms of classical spin vector driven by external fields. In realistic situations, these fields may take the form of laser pulse carrying alternating magnetic field of microwave or optical frequency. It is desirable to study the pertinent spin-photon coupling which would be responsible for the excitation of the collective mode quantum mechanically. This may require rather sophisticated analytical techniques beyond the scope of the present work, which may also uncover novel phenomena not described by our classical approach. In the end, the final goal of such research direction would be practical applications of inertial effect in realizing ultrafast spin dynamics to be implemented in high speed spintronics devices.

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