Ultrafast spin dynamics near magnetization compensation point in the non-collinear state of rare-earth garnets

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Abstract

Magnetic state of ferrimagnets composed of several magnetic sublattices is described by a phase diagram owing a specific temperature point of magnetization compensation. Up to now spin dynamics in ferrimagnets has been investigated mostly in the collinear phase and most of the effects specific for the non-collinear phase near the compensation point were overlooked. Here we synthetized a dielectric ferrimagnetic with compensation point around room temperature to fill this gap. Peculiarities of quasi-ferromagnetic and -antiferromagnetic modes excited by ultrashort laser pulses in the non-collinear state are identified. The non-collinearity makes quasi-antiferromagnetic mode sensitive to the external magnetic field and brings frequencies of the two modes much closer together. The quasi-ferromagnetic mode frequency maximizes near the compensation point and vanishes towards the collinear phase. At the phase transition its amplitude angle significantly increases reaching 7°. This opens new opportunities for the applications of laser induced spin dynamics in ferrimagnets for magnonics.

Introduction

Ultrafast magnetic phenomena driven by femtosecond light pulses are emerging topics of modern material science. The growth in the number of studies in this field is stimulated by technological demands and fundamental puzzles ranging from ultrafast all-optical data storage\textsuperscript{1–5} to Boolean information processing technologies\textsuperscript{1–4} and quantum computing\textsuperscript{6} to spin waves propagation in antiferromagnets\textsuperscript{7} and magnon Bose-Einstein condensation\textsuperscript{6,8}.

Ferrimagnets are unique among ordered magnetic materials as they combine properties of both ferro- and antiferromagnets. Being composed of several antiferromagnetically coupled magnetic ions, these materials sustain low frequency quasi-ferromagnetic (q-FM) and high frequency quasi-antiferromagnetic (q-AFM), so-called “exchange” modes\textsuperscript{9} . The latter is not attainable by GHz conventional microwave stimuli and can be excited by femtosecond laser pulses due to either thermal or optomagnonic effects\textsuperscript{10}. Ultrafast spin dynamics along with ultrafast switching, domain wall motion and skyrmion formation were extensively studied in ferrimagnetic metals, such as GdFeCo\textsuperscript{11–16} and CoGd\textsuperscript{17–19}. In contrast to metals and alloys, insulating oxides are almost lossless at optical frequencies. It makes them perfect materials for essential applications such as all-optical non-thermal magnetic recording\textsuperscript{1,2} and control spin waves in all-dielectric nanostructures\textsuperscript{20}.

Chemical composition and structure of ferrimagnets stimulate exotic physical phenomena in which the role of exchange interaction is crucial. It leads to coexistence of angular momentum
and magnetization temperature compensation points \((T_A \text{ and } T_M)\), correspondingly. The latter one is characterized by zero net magnetization and can be verified via H-T phase diagrams. The theory of phase transitions in compensated ferrimagnets was previously extensively studied\(^{21–24}\). Below Curie temperature, sublattice magnetization in a ground state of a ferrimagnet can be converted from collinear to non-collinear, influencing spin dynamics. The collinear phase is accompanied with collinear alignment of both of the sublattices magnetizations and external magnetic field. The non-collinear phase is characterized by bevel of sublattices magnetizations combined with net magnetization slant to the external magnetic field.

Previous experiments on excitation of spin dynamics in rare-earth iron garnets (REIG) by femtosecond laser pulses were primarily concerned on collinear phase\(^9,25–28\). An all-optical excitation of the exchange resonance between rare-earth and transition metal sublattices far from \(T_M\) was studied in\(^9\) in an in-plane configuration of external magnetic field. The results were found to be in an agreement with the conventional Kaplan–Kittel theory\(^29\) which describes q-AFM mode far from the compensation point. In\(^28\) both an exchange and ferromagnetic resonances were investigated in a wide range of temperatures and an in-plane magnetic field. Despite \(T_M\) identification the features of spin dynamics near this point were not observed probably due to large applied external magnetic field (up to 100 kOe). As a result, the frequencies behavior was described by ferromagnetic resonance\(^30\) and Kaplan-Kittel theories. Consequently, only collinear magnetic phase was previously examined. The role of non-collinear phase on spin oscillations in the vicinity of magnetization compensation point has not yet been investigated. Nowadays, the room-temperature non-collinear phase, actively studied in various magnetic materials\(^{31–33}\), is a potential candidate for chiral spin textures based magnetic non-volatile memory and spintronic devices. Hence, the understanding of spin dynamics in this phase is of fundamental and practical demand.

In the current work we thoroughly investigated spin dynamics in the non-collinear phase of \(R_3Fe_5O_{12}\) rare-earth iron-garnet (RIG) in the vicinity of the magnetization compensation temperature point. We scrutinized magnetic phase transitions in the material via magneto-optical measurements and distinguished the temperature-dependent features of the spin modes by magneto-optical pump-probe technique. The experimental results are well described by the quasi-antiferromagnetic theoretical approach.

**Results**

Unique magnetic properties of RIG \((R_3Fe_5O_{12})\) are governed by its complex crystal structure. \(Fe^{3+}\) ions occupy sites in tetrahedral and octahedral sublattices of the cubic unit cell, the total magnetization of the \(Fe^{3+}\) ions in the tetrahedral positions is larger than in the octahedral ones. Uncompensated magnetic moment of \(Fe^{3+}\) ions gives rise to net magnetization \(M_{Fe}\) which is antiparallelly ordered to \(M_R\) of rear-earth element occupying the third (dodecahedral) sublattice. Due to huge exchange field between \(Fe^{3+}\) ions in tetrahedral and octahedral sublattices they can be treated as a single one with magnetization \(M_{Fe}\). As a result, the rare-earth element undergoes an exchange field produced by collective iron magnetization. The existence of net magnetic moment compensation point \((T_M)\) is mainly managed by strong temperature dependance of \(M_R\) while \(M_{Fe}\) is hardly affected\(^{22}\).

Here a 2.2 μm thick RIG film of composition \((Bi_{0.6}Gd_{2.4})(Fe_{4.28}Ga_{0.57}Ge_{0.15})O_{12}\) where rear-earth ions are represented by Gd is considered. The film is grown on (111) gadolinium gallium
garnet substrate and has a uniaxial magnetic anisotropy with the axis perpendicular to the film. We investigate the case when the external magnetic field is in-plane and therefore perpendicular to the anisotropy axis.

Ultrafast spin dynamics in the sample was launched by fs-laser pump pulses of circular polarization at 787 nm and observed by the Faraday rotation (FR) of the fs-laser probe pulses at 515 nm delayed with respect to the pump pulse in the configuration shown in Fig. 1a. We managed to observe tens GHz frequency spin oscillations for the temperatures in a close vicinity to the magnetization compensation point (Fig. 1b). The signals distinctly show two spin modes whose character is significantly modified when crossing $T_M$. To identify the modes and their physical origin we start from the static magnetic properties of the sample.

The magnetization state of a RIG film can be described in terms of the sublattice magnetizations $\mathbf{M}_{Fe}$ and $\mathbf{M}_R$ forming Néel vector $\mathbf{L} = \mathbf{M}_{Fe} - \mathbf{M}_R$ and magnetization vector $\mathbf{M} = \mathbf{M}_{Fe} + \mathbf{M}_R$ (see Fig. 1a). The angle $\theta = \frac{\theta_{Fe} - \theta_R}{2}$ characterizes the deviation of the Néel vector from the film plane, while the angle $\epsilon = \frac{\theta_{Fe} - \theta_R}{2}$ characterizes a bevel of the sublattice magnetization from a collinear antiparallel orientation. Here $\theta_{Fe}$ and $\theta_R$ are the angles between $\mathbf{M}_{Fe}$ and $\mathbf{M}_R$ and the film plane. As it can be shown by a rigorous theoretical considerations (see SI), $\mathbf{M}_{Fe,R}$ vectors always lie in the plane formed by the magnetic field and anisotropy axis.
Figure 1. (a) Configuration of the magnetic sublattices of the sample and time-resolved experiment. (b) Ultrafast spin dynamics for different temperatures below and above $T_M$. (c) The hysteresis loops for different temperatures of the sample and experimental configuration (inset). (d) Experimental (bright) and theoretical (blurred) phase diagram of different phases of the RIG film. Black curves represent phase transition boundary between non-collinear and collinear phases. (e) Temperature dependence of $\theta$ for the applied in-plane 4 kOe magnetic: black points – experimental data, red line – theoretical approximation. The arrows show sublattices magnetization vectors orientation with respect to external magnetic field for collinear (blue filling), non-collinear (green filling) and compensated (white line) states. (f)
Coercivity and Faraday rotation (FR) amplitude as a function of sample’s temperature. Blue filling shows the non-collinearity region.

Orientation of the Néel vector was experimentally studied by the measurements of the magneto-optical hysteresis loops for different temperatures (Fig.1c). The measurements were performed in the almost in-plane external magnetic field (the tilt angle is around 1 deg.) for the normal light incidence so that only the out-of-plane component of the gyration was detected. Since the gyration in RIG is mainly given by the contribution of Fe$^{3+}$ ions in octahedral and tetrahedral positions in the lattice, the observed Faraday polarization rotation corresponds to the behavior of $M_{Fe}$. At zero magnetic field $M_{Fe}$ is directed normally to the surface ($\theta_{Fe} = \pi/2$) due to the uniaxial magnetic anisotropy, while when an in-plane magnetic field is applied $M_{Fe}$ deflects from the normal. In this case $\theta_{Fe}$ can be found from: $\theta_{Fe}(H, T) = \arcsin \left( \frac{FR(H, T)}{FR(0, T)} \right)$, where $FR(H, T)$ and $FR(0, T)$ are angles of the Faraday rotation (FR) under applied magnetic field $H$ and zero magnetic field, correspondingly. Since the tilt between Fe and Gd sublattices given by angle $\epsilon$ is rather small (see SI Section II), $\theta_{Fe} \approx \theta$ and such measurement gives information about deflection of the Néel vector.

The ground state of the system is governed by the external magnetic field as well as by film magnetization and anisotropy, which are temperature dependent. The analysis of the system’s potential energy minimum (see SI sections I and II) reveals that the ground state is determined by the ratio of $m = M_{Fe} - M_{R}$ that changes rapidly with the temperature near the compensation point ($m=0$) and $m_{cr} = \chi_{\perp} H + 2K/H$, where $\chi_{\perp} = \frac{(M_{Fe}+M_{R})^2}{2\Lambda M_{Fe}M_{R}}$, $\Lambda$ is a constant that represents the exchange of Fe and rare-earth sublattices, $K = K_{Fe} + K_{R}$, $K_{Fe}$ and $K_{R}$ are anisotropy constants of each sublattice (numerical values are given in SI). Depending on the temperature and external magnetic field applied to the film, two magnetic phases of the considered easy-axis RIG film are possible: collinear and noncollinear. In the collinear phase existing for $|m| > m_{cr}$ vectors $M$ and $L$ are along $H$ and in-plane ($\theta = 0$), and the sublattice magnetic moments are antiparallel ($\epsilon = 0$). Non-collinear phase existing in the vicinity of the magnetization compensation point $|m| < m_{cr}$ provides the deflection of vectors $M$ and $L$ from $H$ and the sample plane ($\theta \neq 0$) which is accompanied with noncollinearity of the sublattices magnetic moments ($\epsilon \neq 0$, see SI Fig.S2). Thus, a second type phase transition from non-collinear to collinear phases occurs at $m = m_{cr}$. Phase diagram $\theta(T, H)$ and its crossection at $H = 4$ kOe is given in Fig. 1d,e, respectively.

Both experimental and theoretical results (Fig.1d) indicate that the temperature of the transition from the non-collinear to collinear states drops with the increase of external magnetic field. It is due to its dependence on the Zeeman energy, apart from the anisotropy and exchange energies. An experimental data obtained for $H=4$ kOe (Fig.1e) reveals a distinct jump of derivative $\partial \theta / \partial T$ for $\theta = at T_{cr} \sim 383$ K, indicating the second kind phase transition at $m = m_{cr}$. An additional transition point exists for $H=4$ kOe in the region below the compensation point at $T_{cr}' \sim 284$ K. Between these two critical values of temperature the ground state is non-collinear ($\theta \neq 0$). For the temperature interval $T_{cr}' < T < T_{cr}$ theory predicts bistability of the system due the degeneracy of $+\theta$ and $-\theta$ states (see SI, Section II). However, in our experiments a small out-of-plane magnetic field component exists which lifts this degeneracy and selects the direction of z-projection of $M = M_{Fe} + M_{R}$ parallel to this small out-of-plane field component. It results in a flip of $L$, which is nearly parallel to $M_{Fe}$, in a vicinity of the compensation point due to a change.
of the ratio of the almost oppositely directed $\mathbf{M}_{\text{Fe}}$ and $\mathbf{M}_{\text{R}}$ vectors while crossing the compensation point (Fig1c,e).

The sublattice magnetization and L vector flip when crossing $T_M \sim 336$ K is accompanied by the nonlinear coercivity field rise towards maximum at $T_M \sim 336$ K (blue symbols in Fig. 1f). The absolute value of the Faraday rotation decreases over the whole range of applied temperatures (red symbols in Fig. 1f) which is caused by decrease of $\mathbf{M}_{\text{Fe}}$.

Ultrafast spin dynamics was studied in a wide temperature range which allowed us to observe non-collinear states below and above compensation point ($T_m=336$ K), as well as the dynamics at the phase transition into the collinear state ($T_c=383$ K at $H = 4$ kOe) and after it (Fig.2a). Figure 2a depicts the time-resolved Faraday polarization rotation (TRFR) induced by circularly polarized light at fixed 4 kOe field and different temperatures of the sample. Two kinds of oscillations: high frequency ones at the time scale of 50 ps (Fig. 2a, inset) and low frequency ones at the time scale of 400 ps are clearly observed. Fourier spectra of these signals (Fig.2b and inset) confirm the presence of the low- and high- frequency modes significantly dependent on the film temperature. Notably, the high-frequency component in the TRFR signal was not observed for the temperatures above $T_M$. Data on the magnitude and frequency of these modes was extracted from the Fourier spectra and compared with the theoretical values (Fig.3 a,b).

![Image](https://via.placeholder.com/150)

**Figure 2.** (a) Ultrafast magnetization dynamics represented by the probe Faraday transients in the range of 350 ps for different temperatures from 303 K (below $T_M$) to 393 K (above $T_M$). Higher and lower frequency modes are seen. Inset – the probe Faraday transients for the range of 50 ps demonstrating the higher frequency mode in detail. (b) Fourier transform spectrum of the observed Faraday transients demonstrating q-FM mode and q-AFM mode (inset) for different temperatures. Blue lines show a shift of the q-FM mode frequency with the temperature rise.

Theoretical analysis of the spin dynamics in the current ferrimagnetic system is performed based on the Euler-Lagrange equations of motion (SI, Section I). In the case of non-collinear phase ($\epsilon \neq 0$ or $|m| < m_c$) assuming gyromagnetic ratios of Fe and Gd ions, $\gamma_{\text{Fe}}$ and $\gamma_{\text{Gd}}$, the same: $\gamma_{\text{Fe}} = \gamma_{\text{Gd}} = \gamma$, the mode frequencies are given by:

$$f_{q=\text{AFM,q=FM}} = \frac{1}{2\sqrt{2\pi}} \gamma H (A + B + \zeta \pm \sqrt{(A + B)^2 + 2\zeta B})^{\frac{1}{2}}, \quad (1)$$
where \( A = 1 - \frac{|m| \cos \theta}{\chi H} \), \( B = \left( 2 \cos \theta - \frac{|m|}{\chi H} \right)^2 \), \( \zeta = \frac{4K}{\chi H^2} \). In Eq. (1) the sign “+" corresponds to the higher frequency, q-AFM, while the sign “-” corresponds to the lower frequency, q-FM (inset in Fig. 3a). Noteworthy, the term “quasi” is typically used for canted AFMs, such as FeBO3 owing uncompensated magnetic moment\(^37\) which is similar to the ferrimagnetic IGs. Expressions analogous to Eq. (1) can be derived for the collinear phase (see SI). The frequencies found from Eq. (1) (solid curves in Fig. 3a) are nicely consistent with the experimental findings (dots in Fig. 3a). It allows to use Eq.(1) to broaden the view of modes frequencies behavior and calculate frequencies of q-FM and q-AFM modes for different values of the external magnetic field and temperature (Fig. 3 b-d). In Fig. 3 b,c the frequencies of both modes are shown by color plot versus H and T, while Fig. 3d represents dependence of \( f_{q-AFM, q-FM} \) on \( H \) at a fixed temperature below the compensation point. A boundary between the collinear and non-collinear phases are shown in Fig. 3 b,c by dashed white line.

**Figure 3.** (a) Experimentally found frequencies of the q-FM (blue dots) and q-AFM (brown dots) modes compared to theoretical curves calculated from Eq.(1). (b) Amplitude of the magnetization deflection angle from the ground state versus temperature for the q-FM (blue dots) and q-AFM modes (brown dots). All measurements were done at the external magnetic field \( H = 4 \) kOe. (c-d) Oscillation frequencies of the q-AFM (b) and q-FM modes (c) as a function of external magnetic field and temperature calculated using Eq. 1. White dashed lines at (b) and (c) indicate the boundary of phase transition between non-collinear and collinear phases. (d) Slices of (b) and (c) at T=313 K together with experimental points.
In the non-collinear state the features of the q-AFM and q-FM modes significantly differ from those far from $T_M$. As for the q-AFM mode, in the collinear phase far from $T_M$ it has a character of the Kaplan-Kittel exchange mode whose frequency is unaffected by external magnetic field and primarily influenced by the exchange magnetic field$^{9,25,28}$. In the non-collinear state, however, the situation dramatically changes: the quasi-AFM mode frequency becomes magnetic field-dependent (Fig.3b) which gives an important tool for its control unavailable for the collinear states far from $T_M$.

The behavior of the q-FM mode is nearly the opposite. Usually, in the collinear state far from $T_M$, the frequency of the ferromagnetic mode significantly depends on the applied magnetic field (for example, see ref. $^{28}$). Fig.3a demonstrates that in the non-collinear state near $T_M$ the q-FM mode exhibits very weak magnetic field dependence and becomes completely insensitive to the external magnetic field at $T_M$ (Fig. 3a,e). A peculiar non-monotonous dependence of $f_{q-FM}$ on the magnetic field is observed in the non-collinear phase near the transition to the collinear one: it decreases with the increase of the magnetic field (Fig.3a) which is in a high contrast with what happens above the transition in the collinear state and in the ferromagnetic materials. Moreover, at the transition point the theory predicts that the frequency of this mode would tend to zero (Fig. 3a,c).

It should be also noted that at the temperatures far from $T_M$, the frequencies of the q-FM and q-AFM modes usually have several orders difference. For example, frequencies of 3 GHz and 410 GHz were observed for the q-FM and q-AFM modes for the films without compensation point at similar experimental conditions$^{25}$. Our study shows (Fig.3a) that near the compensation point $f_{q-AFM}$ and $f_{q-FM}$ become close to each other: $f_{q-AFM}$ decreases and $f_{q-FM}$ increases for the temperature increase to $T_M$ so that both modes have extremums at this temperature. There is a small frequency gap at $T_M$: $\Delta\omega = \gamma \left( \frac{2K}{X_{\perp}} H^2 - \frac{2K}{X_{\perp}} \right)$, which can be controlled by external magnetic field.

Let’s now consider spin dynamics at around second kind phase transition between the non-collinear and collinear phases, where the frequency curve derivative $\frac{\partial f}{\partial T}$ breaks (at around $T_{cr} = 383 \, K, H = 4 \, kOe$). At the boundary between the non-collinear and collinear phases, an intriguing feature appears. The q-FM mode’s amplitude increases pronouncedly and reaches a maximum at $T_{cr}$ (Fig. 3e). At this, its frequency drops down to 7 GHz (Fig. 3a), which indicates a soft mode character$^{24}$. This phenomenon is explained by a significant increase of the magnetic susceptibility which takes place at the magnetic phase transition of the second kind. Similar growth of the spin amplitude was previously observed for phase transitions in metallic GdFeCo$^{22}$. Nevertheless, we should emphasize that here we demonstrate optical excitation of the soft mode in the ferrimagnetic dielectric at room temperature which is quite important for advanced ultrafast spin control. The excitation efficiency of spin dynamics at the soft mode is enhanced by more than 3 times if compared to the collinear phase near the transition temperature (for instance, at 393 K) and 4 times far from the transition temperature (for instance, at 413 K).

Discussions

To conclude, we investigated ultrafast spin dynamics in the non-collinear magnetic phase of rare-earth iron garnet near compensation temperature in this study. We demonstrated several peculiarities of spin dynamics in a non-collinear state which are in a high contrast with the
dynamics of the exchange and ferromagnetic modes in a collinear state far from the compensation points. We have shown that when temperature approaches the compensation point the frequency of q-AFM and q-FM modes behave oppositely: the former decreases, while the latter one grows. The situation changes after crossing the compensation point for higher temperatures. We also discovered that transition from non-collinear state to collinear one is accompanied with transformation of q-FM mode to the soft one which is accompanied by a huge increase of the excitation efficiency and q-FM mode amplitude. The magnetization deflection amplitude of the soft mode is more than 4 times larger than for the collinear state (at 413 K) and up to 10 times higher than for the non-collinear phase (at 343 K). As the deflection angle of the soft mode was found to reach ~7°, it can be potentially interesting for nonlinear magnonic phenomena such as Bose-Einstein condensation\textsuperscript{38,39} and superfluidity.

We also found that, in contrast to the collinear phase where the q-AFM mode is field-independent and q-FM mode strongly depends on field, in the noncolinear phase the behavior becomes upside-down: q-AFM mode turns to a field-dependent character, while the q-FM mode gets almost field independent which is in agreement with theoretical predictions based on q-AFM approximation of Euler-Lagrange equations of motion.\textsuperscript{27,28} The approach described in current study is universal and can be applied for RIG with various rare-earth ions. The described methodology allows temperature control of magnetization states of RIG for magnonic and spintronic devices.

**Methods**

**Sample fabrication**

The Bi3+ ions have a large ionic radius, therefore, for the synthesis of BiIG films the (Ca)\textsubscript{3}(GaZrMg)\textsubscript{5}O\textsubscript{12} substrate with the large lattice parameter \(a_s = 1.2494 \text{ nm}\) was chosen. The film (BiGd)\textsubscript{3}(FeAlGa)\textsubscript{5}O\textsubscript{12} was grown by liquid phase epitaxy method from an overcooled solution-melt on a horizontally fixed \(111\) substrate at the isothermal conditions. The Bi\textsubscript{2}O\textsubscript{3}–B\textsubscript{2}O\textsubscript{3}–PbO oxides were used as a solvent. Sample growth temperature \(T_g\) was 742.5 C.

A mismatch between the crystal lattice of the substrate and the magnetic film, \(\Delta a = -0.007 \text{ Å}\) was determined by X-ray diffraction method. The film thickness \(h=2.2 \text{ µm}\) were found from optical transmittance spectra. A magnetopolarimeter was used to measure the magnitude of the Faraday effect wavelength of 515 nm 4.54 deg/µm. The compensation temperature \(T_M=336\text{K}\) was determined by the sign change of the Faraday effect.

**Static and time-resolved magneto-optical measurements**

For static magneto-optical (MO) measurements of hysteresis loops the sample was placed into in-plane external magnetic field of electromagnet (AMT&C Troitsk). The beam with \(\sim250 \text{ fs}\) pulse duration was produced from a tunable optical parametric amplifier (Avesta PARUS), which was pumped by a 1 kHz high-energy Yb regenerative amplifier (Avesta TETA). Linearly polarized (E vector directed along external magnetic field) normally incident 515 nm light pulses were utilized for polarization rotation measurements. The light was modulated by optical chopper (Thorlabs) at 500 Hz. Passed through the sample light pulses were detected using balanced photodetector (Newport Nirvana 2007). Electrical signal from photodetector was detected using lock-in detector (Zurich instruments MFLI).
For time-resolved magneto-optical measurements the sample was placed into constant in-plane external magnetic field of electromagnet (AMT&C Troitsk). 787 nm circularly polarized pulses pumped the sample at ~10° of polar angle. Linearly polarized along external magnetic field probe pulses of 515 nm hit the sample at normal incidence. The pump and probe pulses were focused on the sample to a 100 μm and 40 μm spots correspondingly. The pump and probe beams fluence were 30 mJ/cm² and 0.3 mJ/cm² correspondingly. Temporal overlap between pump and probe pulses was varied by using a 600 mm motorized translation stage (Thorlabs) with a retroreflector in the control beam optical path. The pump pulses were modulated by optical chopper (Thorlabs) at 500 Hz. Passed through the sample probe pulses were detected using balanced photodetector (Newport Nirvana 2007). Electrical signal from photodetector was detected using lock-in detector (Zurich instruments MFLI). The sample was heated by Peltier element electrically controlled with current stabilization. Temperature of the sample was controlled by thermistor.

**Theoretical description**

The RIG ferrimagnetic film was theoretically analyzed using a two-sublattice model. Quasi-antiferromagnetic approximation was used to describe the magnetization behavior near the compensation point, which means the two sublattice magnetizations were considered nearly antiparallel expect for a small bevel angle. To obtain the static ground state of the ferrimagnet, we calculated minima of the potential energy using Lagrange and Hamiltonian functions. Dynamics of the system was analyzed using Euler-Lagrange equations of motion. The presented approach can be used for a wide class of the ferrimagnetic materials, so we put the thorough and detailed description of this analysis to Supplementary, see Sections I-III.

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Supplementary information

Ultrafast spin dynamics near magnetization compensation point in the non-collinear state of rare-earth garnets

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Section I. Determination of the magnetic characteristics of the sample

The comparison of theory with experimental data becomes valuable only when one uses the information of the magnetic values of the sample, needed for calculations, such as the saturation magnetization $M_s$, uniaxial anisotropy constant $K$, magnetic susceptibility $\chi_\perp$, and some others. For the proposed study, the all above three quantities are important, and it is necessary to know the value of the saturation magnetization depending on the temperature $M_s(T)$ in a range in the vicinity of the compensation temperature point. Although one can easy find temperature curve $M_s$ of iron-garnet [Si1, Si2], it may vary depending on the composition of the material. For this reason we calculate a temperature magnetization curve using simulation molecular field theory (Fig. S1). The curve corresponds to the composition (Bi\textsubscript{0.6}Gd\textsubscript{2.4})(Fe\textsubscript{4.28}Ga\textsubscript{0.57}Ge\textsubscript{0.15})O\textsubscript{12}, which is used in our experimental studies, is presented in Figure S1. As it can be seen, the compensation temperature is 336 K, which is in full agreement with our experimental results. At the same time, in the vicinity of the compensation point, the magnetization lies in the range of 0–5 emu/cm\textsuperscript{3}, which is too in agreement with the experimental data from the literature for the samples of other composition [Si1, Si2]. The dependence in Fig. S1 allows one to match the theoretical and experimental curves due to connection between the temperature $T$ (which is recorded in the experimental study) and the magnetization $M$ (which the theory operates on).

Next, in order to determine the uniaxial anisotropy constant $K$, the transverse ferromagnetic resonance measurements at 292 K were used (Fig 2S). This temperature point corresponds $M_s$=8 emu/cm\textsuperscript{3}. According the well-known expression for the transverse ferromagnetic resonance ($2\pi f/\Gamma = H_n + 2K/M_s - 4\pi M_s$) taking into account the resonance frequency $f = 9430$ MHz and external field (please, see Fig. S2) $H_n$=1095 Oe (\(\Gamma=1.33\times10^7\) s\textsuperscript{-1} Oe\textsuperscript{-1}) one can obtain $K$ as approximately 14000 erg/cm\textsuperscript{3}. This value was subsequently used in all theoretical calculations.

For the magnetic susceptibility $\chi_\perp$ value, we used the quantity $3.9 \cdot 10^4$, which was estimated from other experimental data.
Section II. The Lagrangian of a two-sublattice system in the quasi-antiferromagnetic approximation

The ferrimagnetic rare earth iron garnet (like the one in our studies) can be represented theoretically as the two-sublattice system. One of the sublattices is represented by Fe ions, and the other one - by rare-earth Gd ions which are coupled by the exchange interaction. The external magnetic fields applied to the system can also be taken into account. Here we use the assumption that the applied external field is much smaller than the Hilbert exchange field.

Lagrangian method can be applied to describe the magnetization vectors of sublattices for obtaining the precession equations (oscillation equations or) of the antiferromagnetic vector.

Two-sublattice system can be characterized by $M_R$ and $M_{Fe}$ magnetization vectors corresponding to the rare earth and iron sublattices, respectively, and having $M_R$ and $M_{Fe}$ values and $\theta_R$, $\varphi_R$, and $\theta_{Fe}$, $\varphi_{Fe}$, angles in the spherical coordinate system (see Fig.1 in the main text).
Using the well-known relation for the kinetic energy in micromagnetism [Si3-Si5], the Lagrange function $\mathcal{L}$ of the system can be written as

$$
\mathcal{L} = -\frac{M_{Fe}}{\gamma_{Fe}} \sin \theta_{Fe} \frac{\partial \varphi_{Fe}}{\partial t} - \frac{M_{R}}{\gamma_{R}} \sin \theta_{R} \frac{\partial \varphi_{R}}{\partial t} - \Phi,
$$

(1)

where $\gamma_{R}$ and $\gamma_{Fe}$ are the gyromagnetic ratios, $\Phi$ is the potential energy, and $t$ is the time. Lagrangian Eq. (1) is a four-dimensional function of 3 spatial coordinates and time, therefore a general analytical solution cannot be obtained in many cases. For the two-sublattice system, so-called quasi-antiferromagnetic approximation considering nearly anti-parallel alignment of the sublattices can be used to simplify the description [Si3-Si5].

Therefore, antiferromagnetic vector $\mathbf{L} = \mathbf{M}_{Fe} - \mathbf{M}_{R}$ can be introduced and characterized by the angle set $\theta$ and $\varphi$, and:

$$
\begin{align*}
\theta_{Fe} &= \theta + \varepsilon, \quad \theta_{R} = \varepsilon - \theta, \\
\varphi_{Fe} &= \varphi + \beta, \quad \varphi_{R} = \pi + \varphi - \beta
\end{align*}
$$

(2)

where the parameters $\varepsilon \ll 1$ and $\beta \ll 1$ characterize the noncollinearity of the magnetization vectors of the sublattices. If $\varepsilon = \beta = 0$, according to Eq. (2) the $\theta_{Fe} = -\theta_{R}$ and $\varphi_{Fe} = \pi + \varphi_{Fe}$, and that means the sublattices magnetization vectors are collinear and antiparallel. Therefore, for sublattice magnetization vectors one can obtain the following coordinates in Cartesian system:

$$
\begin{align*}
\mathbf{M}_{Fe} = M_{Fe} \begin{pmatrix}
(\cos \theta - \varepsilon \sin \theta)(\cos \varphi - \beta \sin \varphi) \\
(\cos \theta - \varepsilon \sin \theta)(\sin \varphi + \beta \cos \varphi) \\
(\sin \theta + \varepsilon \cos \theta)
\end{pmatrix}
\\
\mathbf{M}_{R} = M_{R} \begin{pmatrix}
(\cos \theta + \varepsilon \sin \theta)(-\cos \varphi - \beta \sin \varphi) \\
(\cos \theta + \varepsilon \sin \theta)(\beta \cos \varphi - \sin \varphi) \\
(\varepsilon \cos \theta - \sin \theta)
\end{pmatrix}
\end{align*}
$$

(3)

By substituting the expressions for the sublattice magnetization vectors Eq. (3) into Eq. (1), neglecting the terms $\varepsilon \cdot \dot{\beta}$, and taking into account the property of Lagrangian function to be invariant under the addition of the full derivative of an arbitrary function of coordinates and time, one can obtain the Lagrangian of the two-sublattice system as a function of the angles $\theta$ and $\varphi$, parameters $\varepsilon$ and $\beta$, and their time derivatives:

$$
\mathcal{L} = -\frac{m}{\gamma} \dot{\varphi} \sin \theta - \frac{\mathcal{M}}{\gamma} (\dot{\varphi} - \beta \dot{\theta}) \cos \theta - \Phi,
$$

(4)

Where $\mathcal{M}$ is the sum of the magnetization moduli of the sublattices, $\mathcal{M} = M_{Fe} + M_{R}$, the $\gamma = \gamma_{R} = \gamma_{Fe}$ is the gyromagnetic ratio and the $m = M_{Fe} - M_{R}$ is the difference between the magnetization moduli of the sublattices. In the experimental measurements, we considered $m$ to be $M_{s}$ (see Section I). Lagrangian function determined by Eq. (4) depends on the potential energy $\Phi$, which is determined by the material properties (the type of exchange interaction and magnetic anisotropy, shape and size that determine demagnetization energy, etc.) and the configuration of the applied magnetic field.
Figure S1. A ferromagnetic film with uniaxial magnetic anisotropy in the external magnetic field $H$: the easy axis is denoted by the unit vector $n$, $L$ is the aniferromagnetic vector. The $\theta$ angle is measured from the projection of the vector $L$ onto the XOY plane.

We choose the coordinate system in the following way. XY plane coincides with the film plane; Z-axis is collinear to the film normal. Uniform stationary external magnetic field $H$ is applied along the X axis ($H=\{H,0,0\}$, here and below we assume that $H>0$). The easy axis of the uniaxial effective magnetic anisotropy $n=\{0,0,1\}$ is directed along the film normal away from the substrate (the Z axis). The demagnetization energy for the thin film is proportional to $2\pi((M_{Fe}+M_{R}),n)^2$ and is much smaller than Zeeman and anisotropy energy in the vicinity of the compensation point, therefore it could be neglected. Thus, the potential energy of such system can be written as:

$$
\Phi = -\left( (M_{Fe} + M_{R})H + \Lambda M_{Fe} M_{R} - K_{Fe} \frac{(M_{Fe} n)^2}{M_{Fe}^2} - K_{R} \frac{(M_{R} n)^2}{M_{R}^2} \right)
$$

In Eq. (5) $K_{Fe}>0$ and $K_{R}>0$ are the magnetic anisotropy constants of each sublattice, $\Lambda>0$ is the intersublattice exchange constant and the “+” sign of the second term represents the antiferromagnetic character of the exchange interaction between the sublattices. Using Eq. (3) and taking into account $H$ and $n$ directions, one can write (5) as:

$$
\Phi = -mH \cos \theta \cos \varphi + \mathcal{H} \beta \cos \theta \sin \varphi + \mathcal{H} \varepsilon \sin \theta \cos \varphi + \frac{\delta}{2} (\varepsilon^2 + \beta^2 \cos^2 \theta - 1) - K \sin^2 \theta
$$

In Eq. (6) $\delta = 2\Lambda M_{Fe} M_{R}$, $K$ is the effective magnetic anisotropy constant, $K=K_{Fe}+K_{R}$. Considering $\varepsilon$ and $\beta$ as $O(1/\Lambda)$ the terms proportional to higher than $1/\Lambda$ powers can be neglected to obtain Eq. (6). Thus terms proportional to the $\varepsilon^2 \beta^2$ were neglected in case of calculation of the exchange term in (6), the terms proportional to the $\varepsilon \beta$ were neglected in case of calculation of the Zeeman term, and all terms proportional to the $\varepsilon$ or $\beta$ in case of anisotropy energy term.
Next, substituting the relation for the potential energy (6) into the Lagrange function (4) and considering \((\varepsilon, \beta)\) as a generalized coordinates set, with respect to them one can calculate the Lagrange equations:

\[
\begin{align*}
\frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{\varepsilon}} - \frac{\partial \mathcal{L}}{\partial \varepsilon} &= 0, \\
\frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{\beta}} - \frac{\partial \mathcal{L}}{\partial \beta} &= 0
\end{align*}
\]

which gives the relation between two sets of coordinates, \((\varepsilon, \beta)\) and \((\phi, \theta)\):

\[
\varepsilon = -\frac{\mathcal{H}}{\gamma} \cos \theta \cos \phi - \frac{\mathcal{H} H}{\gamma} \sin \theta \cos \phi
\]

\[
\beta \cos \theta = \frac{\mathcal{H}}{\gamma} \left( \dot{\theta} - H \sin \phi \right)
\]

Substituting Eq. (7) into Eq. (4) with the potential energy Eq. (5), one can exclude the noncollinearity parameters \(\varepsilon\) and \(\beta\) and reduce the Lagrangian of the two-sublattice system to a function of only two angles \(\phi\) and \(\theta\) of the antiferromagnetic vector \(L\) and their time derivatives:

\[
\mathcal{L}_{\text{eff}} = -\frac{\chi_\perp}{2} \left[ \left( \frac{\phi}{\gamma} \cos \theta + H \sin \theta \cos \phi \right)^2 + \left( H \sin \phi - \frac{\dot{\theta}}{\gamma} \right)^2 \right] - \phi m \sin \theta + m H \cos \theta \cos \phi + K \sin^2 \theta
\]

where \(\chi_\perp = \frac{\mathcal{H}^2}{\gamma}\). Lagrangian in form Eq. (8), in contrast to Eq. (4), is the function of two variables, which greatly simplifies the system analysis.

**Section III. The potential energy and equilibrium states of the system**

The equilibrium states of the two-sublattice system can be found as the minimum of the effective potential energy obtained using the known relationship between the Lagrange and the Hamiltonian functions for the stationary case \(\phi = 0\) and \(\dot{\theta} = 0\):

\[
U_{\text{eff}} = -\frac{\chi_\perp}{2} H^2 \sin^2 \theta \cos^2 \phi - \frac{\chi_\perp}{2} H^2 \sin^2 \phi - m H \cos \theta \cos \phi - K \sin^2 \theta
\]

Eq. (9) shows that antiferromagnetic vector \(L\) in the equilibrium state characterized by \(\theta_0, \phi_0\) angles always lie in the XZ plane \((\phi_0 = 0\) for \(m > 0\) or \(\phi_0 = \pi\) for \(m < 0\)). This can be explained using the first term in Eq. (5) that provide minima if the scalar product \((M, H) > 0\) of the sum magnetization \(M = M_{Fe} + M_R\) and external magnetic field \(H\) is positive. On the other hand, \(L\) and \(M\) vectors are nearly parallel for \(m > 0\) and nearly antiparallel for \(m < 0\).

The values of \(\theta_0\) corresponding to the equilibrium state depend on the system parameters, as shown in Table 1.

**Table S1.** Equilibrium states of a two-sublattice magnetic system in the external field
| System parameters | Stationary value of angle $\theta$ | Stationary value of angle $\phi$ | Scheme of the equilibrium state |
|--------------------|-----------------------------|-----------------------------|-------------------------------|
| I                  | $mH \geq \chi_\perp H^2 + 2K$ | $\theta_0 = 0$               | $\phi_0 = 0$                 |
| II                 | $0 < mH < \chi_\perp H^2 + 2K$ | $-\pi/2 < \theta_0 < \pi/2$, $\cos \theta_0 = \frac{mH}{\chi_\perp H^2 + 2K}$ | $\phi_0$ Not defined          |
| III                | $m = 0$                      | $\theta_0 = \pm \pi/2$      | $\phi_0$ Not defined          |
| IV                 | $0 < |m| H < \chi_\perp H^2 + 2K$, $m < 0$ | $-\pi/2 < \theta_0 < \pi/2$, $\cos \theta_0 = \frac{-mH}{\chi_\perp H^2 + 2K}$ | $\phi_0 = \pi$               |
Cases (I) and (V), so-called collinear phase, corresponding to $|m|H \geq \chi_{\perp}H^2 + 2K$, with $\theta_0=0$ describe antiferromagnetic vector $L$ collinear to $H$. This is similar to the ferromagnetic state, as the sum magnetization $M=M_{Fe}+M_{R}$ is parallel to $H$, while $L$ and $H$ parallel or antiparallel alignment is determined by the mutual orientation of $L$ and $M$ vectors that depends on the ratio between $M_{Fe}$ and $M_{R}$.

Cases (II) and (IV), so-called non-collinear phase, are realized for rather small values of $m$ less than critical one $|m| < m_c = \chi_{\perp}H + 2K/H$. The direction of the antiferromagnetic vector in the equilibrium state is determined as:

$$\cos \theta_0 = \frac{|m|H}{\chi_{\perp}H^2 + 2K}$$  \hspace{1cm} (10)

It is important, that Eq. (10) has two solutions, so it means that there are two equilibrium states with $+\theta_0$ and $-\theta_0$. Moreover, Eq.(9) shows that these states are degenerate as they have the same potential energy $U_{eff}$. Therefore, for the same temperature that determines $m$, and the same external magnetic field there are two equivalent equilibrium positions described by $+\theta_0$ and $-\theta_0$ angles.

According to Eq. (10) for $m=m_c \ \theta_0=0$, which means that this is the second-order phase transition from the collinear to non-collinear state at this point.

In the third case (III) describing the magnetization compensation point $m=0$, the antiferromagnetic vector $L$ in the equilibrium state is directed strictly along the easy axis of the effective anisotropy $\mathbf{n}$, and perpendicular to the external field $H$, so $\theta_0$ equals $\pm \pi/2$.

Figure S2 shows how these cases appear in the experimentally studied $(Bi_{0.6}Gd_{2.4})(Fe_{4.28}Ga_{0.57}Ge_{0.15})O_{12}$ film with the parameters, discussed in Section I. The transition between the collinear and non-collinear phases determined as $|m| = \chi_{\perp}H + 2K/H$ is shown by the black line.
Figure S2. $\theta(T, H)$ diagrams showing (I)-(V) cases. (a) $\theta(T, H)$ 3D plot showing the degeneracy and non-collinearity in the vicinity of the magnetization compensation point. (b), (c) false-color plots showing $\theta(T, H)$ dependencies for $\theta \geq 0$ and $\theta \leq 0$ solution branches. $m=m_c$ condition is shown by the black line.

The cases (II), (III) and (IV) are accompanied by the physical phenomenon of static non-collinearity, which has a dual nature. On the one hand, the antiferromagnetic vector $L$ is non-collinear to the external magnetic field vector $H$, as described above, and, on the other hand, the magnetization vectors of the $M_R$ and $M_{Fe}$ of the sublattices are also non-collinear to each other. Using the Eq. (2), Eq. (7), Eq. (10), the $\Delta$ is the angle between $M_{Fe}$ and $-M_R$, $\Delta = |\theta_R + \theta_{Fe}|$, is defined as:

$$\Delta = 2|x| = \frac{2xH}{\delta} \sin \theta_0$$

(11)

The $\Delta$ angle reaches its maximum $\Delta=2xH/\delta$ at the magnetization compensation point $m=0$. With the increase of $m$, $\Delta$ decreases reaching $\Delta=0$ at $|m|=m_c$. For $|m|>m_c$ where $\theta_0 = 0$ sublattice magnetizations are collinear to each other, and the case can be called a «collinear phase». At the same time, both in the collinear and non-collinear states $M_R$ and $M_{Fe}$ lie in the XZ plane and $\beta = 0$. 
The $\Delta$ angle (in radians) vs temperature, according to Eq. (11).

We have considered the configuration where the external magnetic field $H$ is applied strictly in-plane so that $\theta > 0$ and $\theta < 0$ states have the same energy according to Eq.(9). However, practically it is very complicated to apply the magnetic field strictly in-plane with a good precision, so that small out-of-plane magnetic field component always exists. The rigorous theoretical analysis of such a configuration is rather complicated and is out of the scope of this manuscript. However, it is obvious that the presence of $H_z$ component lifts the degeneracy between the $\theta > 0$ and $\theta < 0$ states with $+M_z$ and $-M_z$ projections. According to Eq. (5), the state with $M_zH_z > 0$ has lower energy than with $M_zH_z < 0$. Thus, for the configurations where $H_z$ is not zero, the state with $M_z$ component parallel to this small out-of-plane field is preferable. As $L=M_R-M_{Fe}$ is nearly parallel to $M_R$, $L_z$ and $M_z$ have the same signs for $m>0$ and the opposite signs for $m<0$. This results in the $L$ flip in a vicinity of the compensation point due to a change of the ratio of the almost oppositely directed $M_{Fe}$ and $M_R$ vectors and $m$ sign change while crossing the compensation point.

**Figure S3.** The $\Delta$ angle (in radians) vs temperature, according to Eq. (11)

**Figure S4.** $\theta(H, T)$ diagram in the presence of the small out-of-plane magnetic field.

**Section IV. Spin dynamics and mode frequencies**
In order to calculate the Lagrange equations for the angles $\theta$ and $\phi$ of the antiferromagnetic vector $\mathbf{L}$,

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\text{eff}}}{\partial \dot{\theta}} \right) - \frac{\partial \mathcal{L}_{\text{eff}}}{\partial \theta} + \frac{\partial \mathcal{A}}{\partial \theta} = 0$$

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\text{eff}}}{\partial \dot{\phi}} \right) - \frac{\partial \mathcal{L}_{\text{eff}}}{\partial \phi} + \frac{\partial \mathcal{A}}{\partial \phi} = 0$$

it is necessary to know the form of the Rayleigh function of the micromagnetic system. For the two-sublattice case, it can be written as follows [Si3-Si5]:

$$\mathcal{A} = \frac{\alpha}{2} \left( \frac{M_r (\dot{\theta}^2 + \cos^2 \theta_{r} \dot{\phi}^2)}{\gamma_r} + \frac{M_{Fe} (\dot{\theta}^2 + \cos^2 \theta_{Fe} \dot{\phi}^2)}{\gamma_{Fe}} \right)$$  

(12)

where $\alpha$ is the Hilbert damping parameter. Substituting Eq. (2) into Eq. (12), and neglecting the terms containing $\varepsilon$ and $\beta$ in powers above 1st, one can calculate the derivatives of $\mathcal{A}$ with respect to generalized velocities $\dot{\phi}$ and $\dot{\theta}$. Lagrange equations for angles $\phi$ and $\theta$ can be obtained using Eq.(8) as:

$$-\frac{\mathcal{M}^2}{\delta^2} \dot{\phi} \cos \phi - \frac{\mathcal{M}^2}{\delta} \frac{H}{\gamma} \dot{\phi} \sin \theta \cos \theta - H^2 \sin \theta \cos \theta \cos^2 \phi +$$

$$-H \cos \phi \cos 2\theta \frac{\dot{\phi}}{\gamma} \cos \theta - m \sin \theta \cos \phi + K \sin 2\theta = \frac{\alpha \mathcal{M}}{\gamma} \dot{\theta}$$

(13)

$$\frac{\mathcal{M}^2}{\delta} \left( -\frac{\phi}{\gamma} \dot{\theta} \sin 2\theta + H \dot{\phi} \cos 2\theta \cos \phi - H \dot{\phi} \sin \theta \cos \theta \sin \phi + \frac{\phi}{\gamma} \cos^2 \theta \right) - \frac{m}{\gamma} \dot{\theta} \cos \theta +$$

$$+ \frac{\mathcal{M}^2}{\delta} \frac{\phi}{\gamma} \sin \theta \cos \theta \sin \phi + H^2 \sin^2 \theta \cos \phi \sin \phi \right) + \frac{\mathcal{M}^2}{\delta} \left( \frac{\dot{\theta}}{\gamma} - H \sin \phi \right) H \cos \phi$$

(14)

The further analysis of the behavior of the angles of the antiferromagnetic vector and the calculation of the frequencies of the corresponding oscillations require the linearization of equations (13)-(14) near the equilibrium states considered in the previous section. Substituting $\phi_0 = \phi_0 + \phi_l$ and $\theta_0 = \theta_0 + \theta_l$, where $\phi_0$ and $\theta_0$ correspond to the equilibrium angular coordinates of the vector $\mathbf{L}$, $\phi_l << 1$ and $\theta_l << 1$ are small deviations, so that higher than the first orders of these values are neglected, one can obtain the following equations. For the collinear phase ($\theta_0 = 0$):

$$\dot{\phi}_l + \frac{\alpha \mathcal{M}}{\chi} \gamma \dot{\phi}_l + \left( \frac{m}{\chi} \gamma^2 - \gamma^2 H^2 - \frac{2K\gamma^2}{\chi} \right) \dot{\phi}_l \pm \left( 2\gamma H - \frac{m\gamma}{\chi} \right) \dot{\phi}_l = 0$$

(15)

$$\dot{\phi}_l + \frac{\alpha \mathcal{M}}{\chi} \gamma \dot{\phi}_l + \left( \frac{m}{\chi} \gamma^2 - \gamma^2 H^2 \right) \phi_l \pm \left( 2\gamma H - \frac{m\gamma}{\chi} \right) \dot{\phi}_l = 0$$
For the non-collinear \((\theta_0 \neq 0)\) phase:

\[
\dot{\phi}_i + \frac{\alpha \gamma \gamma}{\chi} \dot{\theta}_i + \left( \gamma^2 \mathcal{H}^2 + \frac{2K^2}{\chi} \right) \left( 1 - \cos^2 \theta_0 \right) \phi_i \mp \left( 2\gamma H \cos^2 \theta_0 - \frac{m \gamma}{\chi \gamma} \cos \theta_0 \right) \dot{\phi}_i = 0
\]

\[
\dot{\phi}_i \cos^2 \theta_0 + \frac{\alpha \gamma \gamma}{\chi} \phi_i \cos^2 \theta_0 + \cos^2 \theta_0 \left( \frac{2K^2}{\chi} \right) \phi_i \pm \left( 2\gamma H \cos^2 \theta_0 - \frac{m \gamma}{\chi \gamma} \cos \theta_0 \right) \dot{\phi}_i = 0
\]

(16)

The first set of signs in each equation corresponds to the case \(m > 0 \theta_0 > 1\) or \(m < 0 \theta_0 < 1\), while the second one to the other cases.

Eq. (15) and (16) have harmonic solutions \(\theta_i = \theta_i \exp(i\omega t)\) and \(\phi_i = \phi_i \exp(i\omega t)\) with the frequencies \(\omega\) determined as the zeros of Eq. (15),(16) determinant:

\[
\omega_{qAM,qFM} = \gamma H \left( A' + B' - \frac{\zeta}{4} \pm \sqrt{\left( \frac{\zeta}{4} - B' \right)^2 + 2A'B'} \right)^{\frac{1}{2}}
\]

(17)

for \(|m| \geq \chi H + 2K/H\); and

\[
\omega_{qAM,qFM} = \frac{\gamma H}{\sqrt{2}} \left( A + B \pm \sqrt{(A + B)^2 + 2B} \right)^{\frac{1}{2}}
\]

(18)

for \(0 \leq |m| < \chi H + 2K/H\).

In (17) and (18) \(\zeta = \frac{4K}{\chi H^2}\), \(A = 1 - \frac{|m| \cos \theta_0}{\chi H}\), \(B = \left( 2 \cos \theta_0 - \frac{|m|}{\chi H} \right)^2\), \(A' = \frac{|m|}{\chi H} - 1\), \(B' = \frac{1}{2} \left( 2 - \frac{|m|}{\chi H} \right)^2\). The choice of the sign above the root corresponds to the choice of the higher \((\omega_1)\) or lower \((\omega_2)\) frequency branch, the quasi-ferromagnetic and quasi-antiferromagnetic modes. The frequency relations are invariant to sign inversion of \(m\). Notice that in case of significant difference of the gyromagnetic ratios of the sublattices, e.g. \(\gamma_R \neq \gamma_F\), the frequencies of quasi-FM and quasi-AFM modes differ below and above compensation point \(\omega_1(m) \neq \omega_1(-m)\), while the equilibrium states \(\theta_0(m) = \theta_0(-m)\) are the same. Detailed consideration of these cases is out of the scope of the present study.

Eq. (18) remains valid for the magnetization compensation point \(m = 0\) and \(\cos \theta_0 = 0\):

\[
\omega_{1,2} = \gamma \sqrt{\frac{2K}{\chi} + \frac{H^2}{2}} \pm \frac{H^2}{2}
\]

(19)

and can be obtained directly from the linearized Lagrange equations for the other coordinate system choice with \(Y\) axis directed along the easy axis of the effective anisotropy \(n\) which helps to avoid uncertainty of \(\varphi\) for \(m = 0\), and to avoid consequent zeroing of the precession equation, unlike Eq. (16).
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