Temperature- and field angular-dependent helical spin period characterized by magnetic dynamics in a chiral helimagnet MnNb$_3$S$_6$

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The chiral magnets with topological spin textures provide a rare platform to explore topology and magnetism for potential application implementation. Here, we study the magnetic dynamics of several spin configurations on the monoaxial chiral magnetic crystal MnNb$_3$S$_6$ via broadband ferromagnetic resonance (FMR) technique at cryogenic temperature. In the high-field forced ferromagnetic state (FFM) regime, the obtained frequency $f$ vs. resonance field $H_{\text{res}}$ dispersion curve follows the well-known Kittel formula for a single FFM, while in the low-field chiral magnetic soliton lattice (CSL) regime, the dependence of $H_{\text{res}}$ on magnetic field angle can be well-described by our modified Kittel formula including the mixture of a helical spin segment and the FFM phase. Furthermore, compared with the sophisticated Lorentz micrograph technique, the observed magnetic dynamics corresponding to different spin configurations allow us to obtain temperature- and field-dependent proportion of helical spin texture and helical spin period ratio $L(H)/L(0)$ via our modified Kittel formula. Our results demonstrated that field- and temperature-dependent nontrivial magnetic structures and corresponding distinct spin dynamics in chiral magnets can be an alternative and efficient approach to uncovering and controlling nontrivial topological magnetic dynamics.

chiral helimagnets, magnetic solitons, ferromagnetic resonance, phase diagram

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1 Introduction

Chiral helimagnets (CHM) possess nontrivial spin-textures with spiral or rotary alignment of spin moments, such as topological spin textures of magnetic skyrmions, which provide a platform to study the interesting topological physics and potential applications for spintronics [1-5]. MnNb$_3$S$_6$ and CrNb$_3$S$_6$ are typical chiral helimagnets with the same lattice structure [6,7], analogous electronic [8,9], and magnetic structures [10,11]. In the monoaxial chiral helimagnets [12], all spins are in the $ab$-plane and rotate at a definite angle...
arises from losing the inversion center in the magnetic atoms sublattice. The Heisenberg interaction (coefficient $J$) prefers all spins forming collinear arrangements (ferromagnetic or antiferromagnetic alignments). In contrast, the chiral DM interaction (coefficient $D$) favors the non-collinear alignment of spins and facilitates chiral magnetic orders [12]. Thus, their competition generates a chiral helimagnet with a fixed spin helix period $L(0)$ determined by the ratio of two interactions $L(0) = \tan^{-1}(D/J)$ [13,14].

However, under an external field $H$, the field-dependent Zeeman interaction will also compete with the above two magnetic interactions and can be used to achieve field-controllable spin textures [6,15,16]. Therefore, by tuning magnetic field or/and temperature, the chiral helimagnets can evolve from a CHM into a chiral magnetic soliton lattice (CSL) or forced ferromagnetic state (FFM) to achieve the minimal total energy in terms of the competition of several magnetic interactions [6,17,18]. Additionally, there exist several distinct nontrivial magnetic properties in these monoaxial hexagonal crystals [17-20]. As the schematics are shown in Figure 1(a), the external field $H$ tilts the spin direction of the spin soliton lattice, modulates the spin helix period $L(0)$ to $L(H)$ at $H < H_c$, and finally turns it into the FFM regime at $H > H_c$. The previous theoretical investigations of CHM [21,22] reported that the period of CSL can be described by the 1D chiral sine-Gordon model, which generally follows the formula $L(H_0)/L(0) = 4K(k)E(k)/\pi^2$ [23-26], where $K(k)$ and $E(k)$ are the elliptic integrals of the first and second kinds with modulus $k$ ($0 \leq k \leq 1$), respectively, and $H_0$ is the in-plane component of an external magnetic field. The elliptic modulus $k$ is given by $\sqrt{k/E(k)} = (H_{ab}/H)^{1/2}$ to minimize the CSL formation energy. The static and dynamic magnetic properties experiments confirm that the nontrivial spin configurations of these chiral helimagnets highly depend on the external magnetic field, dimensionality, and temperature [1,7,17,27-29]. Moreover, the Lorentz transmission electron microscopy also directly observed the temperature-dependent CSL state and its period in CrNb$_6$S$_6$ [15,14,30,31]. However, for MnNb$_3$S$_6$ helimagnet with the same lattice structure as CrNb$_3$S$_6$, the Lorentz transmission electron microscopy measurement failed to identify the spatial period of CSL because MnNb$_3$S$_6$ has a much lower magnetic order temperature $T_c \sim 45$ K and the weak field modulation of the helix period [32]. Therefore, a high sensitivity technique that can catch the spiral period information of MnNb$_3$S$_6$ and its evolution with the external magnetic field and temperature is urgently needed.

Here, we perform the systematic ferromagnetic resonance experiment to investigate thoroughly the detailed dependence of magnetic dynamics corresponding to the nontrivial CSL in MnNb$_3$S$_6$ on the field magnitude, angle, and temperature. We find that chiral helimagnet MnNb$_3$S$_6$ exhibits a distinct field angular dependence of spin resonance in low-field nontrivial CSL from the uniform FMR in high-field FFM. Then, we propose a modified Kittel model considering partial helix spin textures, which can successfully describe the experimentally observed spin dynamics of the low-field nontrivial CSL at different temperatures. Moreover, the modified Kittel model also enables us to extract temperature- and field-dependent proportion of the helical spin texture and helical spin period ratio $L(H)/L(0)$, like the sophisticated Lorentz micrograph technique used in most chiral helimagnets. The demonstrated method can generally be used as an alternative and easy-access approach to explore interesting magnetic dynamics not just in MnNb$_3$S$_6$ and other topologically nontrivial chiral magnets.

Figure 1(b) and (c) show the phase diagram of spin textures in MnNb$_3$S$_6$ determined from the static field- and temperature-dependent proportion of the helical spin texture and helical spin period ratio $L(H)/L(0)$, like the sophisticated Lorentz micrograph technique used in most chiral helimagnets. The demonstrated method can generally be used as an alternative and easy-access approach to explore interesting magnetic dynamics not just in MnNb$_3$S$_6$ and other topologically nontrivial chiral magnets.

**Figure 1** (Color online) Several nontrivial spin configurations and their phase diagrams of MnNb$_3$S$_6$. (a) Schematic of spin configurations of several magnetic orders in monoaxial chiral helimagnets under the external magnetic field: CHM state at $H = 0$, CCP, TCSL, CSL states at $0 < H < H_c$, and FFM state at $H > H_c$, respectively. The orange and green arrows represent the $c$-axis of the MnNb$_3$S$_6$ crystal and the direction of the magnetic field $H$, respectively. Phase diagram of the specific magnetic orders in monoaxial chiral helimagnet MnNb$_3$S$_6$ crystal with $H // ab$-plane (b) and $H // c$ axis (c). The boundaries among CSL (blue region) and FFM states (green region) were determined by critical field (squares) obtained from the quasi-static magnetization hysteresis loops. The critical field data (solid circle) reported by others is also shown in the phase diagram [7]. $T_c$ represents the Curie temperature 45 K of MnNb$_3$S$_6$, determined from the $M$-$T$ curves. PM (orange region) represents paramagnetism.
temperature-dependent magnetic susceptibility results with \( H \) in the \( ab \)-plane and \( H \) parallel to the \( c \)-axis of the single-crystal sample, respectively (see Supporting Information for the magnetic susceptibility measurement results). Note that the critical fields obtained by the static magnetization loop have some deviations from previous reports \([11,33]\) due to different definition criteria and broad transition regions in \( M(H) \) curves. More specifically, the critical field of the phase diagram in Figure 1(b) is slightly higher than in our previous \( M(\dot{H}) \) curves. More specifically, the critical field of the phase at excitation frequency \( f \) varies from 5 to 20 GHz with 1 GHz steps, oblique field angle \( \theta \) increasing from 5 to 20 GHz with 1 GHz steps, oblique field angle \( \theta = 45^\circ \) and cryogenic temperature \( T = 4.5 \) K. Therefore, it is expected that the different dynamic properties corresponding to two distinct spin textures could be observed in our broadband FMR spectra.

To systematically explore the specific dynamics of chiral helimagnet \( \text{MnNb}_2\text{Se}_3 \), we measured the broadband FMR spectra carefully at several different temperatures \( T = 4.5, 10, 30, \) and 45 K. Figure 3(a)-(d) show the frequency-depending magnetic susceptibility results with \( H \) in the \( ab \)-plane and \( H \) parallel to the \( c \)-axis of the single-crystal sample, respectively. A 1 mm × 1 mm square-shaped single-crystal \( \text{MnNb}_2\text{Se}_3 \) with \(~10 \) μm thickness was fixed to the S-pole of the CPW by using the apiezon N-grease with high thermal conductivity and its \( c \) axis aligns along the \( z \)-axis (Figure 2(a)). All cryogenic-temperature FMR spectra data were collected using a homemade differential FMR measurement system combining the lock-in technique and a closed-cycle G-M refrigerator-based cryostat. Static magnetic field \( H \) can rotate in the \( y-z \) plane (Figure 2(a)) and be modulated with an amplitude of 1-2 Oe by a pair of secondary Helmholtz coils powered by an alternating current source with a low audio frequency of 129.99 Hz.

### 2 Experimental section

The differential ferromagnetic resonance (FMR) spectroscopy is based on a coplanar waveguide (CPW), illustrated in Figure 2(a). A 1 mm × 1 mm square-shaped single-crystal \( \text{MnNb}_2\text{Se}_3 \) with \(~10 \) μm thickness was fixed to the S-pole of the CPW by using the apiezon N-grease with high thermal conductivity and its \( c \) axis aligns along the \( z \)-axis (Figure 2(a)). All cryogenic-temperature FMR spectra data were collected using a homemade differential FMR measurement system combining the lock-in technique and a closed-cycle G-M refrigerator-based cryostat. Static magnetic field \( H \) can rotate in the \( y-z \) plane (Figure 2(a)) and be modulated with an amplitude of 1-2 Oe by a pair of secondary Helmholtz coils powered by an alternating current source with a low audio frequency of 129.99 Hz.

### 3 Results and discussion

#### 3.1 Spin resonance of the high-field FFM regime

Figure 2(b) shows the representative pseudocolor plot of normalized magnetic field-dependent FMR spectra obtained at excitation frequency \( f \) varying from 5 to 20 GHz with 1 GHz steps, oblique field angle \( \theta_{\text{hf}} = 45^\circ \) and cryogenic temperature \( T = 4.5 \) K. The inset of Figure 2(b) exhibits a representative differential FMR spectrum with \( f = 19 \) GHz, which can be well fitted using a differential Lorentzian function. The characteristic dynamic properties, e.g., the resonance field \( H_r \) and linewidth, can be extracted accurately from the fitting parameters of the experimental FMR spectra. As mentioned above, the external magnetic field can change the spin texture of \( \text{MnNb}_2\text{Se}_3 \). For instance, the low-field CSL with a nontrivial topological property will be driven into the trivial FM state by an in-plane magnetic field \( H_{\text{in}} \geq H_c \sim 0.51 \) kOe at \( T = 4.5 \) K. Therefore, it is expected that the different dynamic properties corresponding to two distinct spin textures could be observed in our broadband FMR spectra.

To systematically explore the specific dynamics of chiral helimagnet \( \text{MnNb}_2\text{Se}_3 \), we measured the broadband FMR spectra carefully at several different temperatures \( T = 4.5, 10, 30, \) and 45 K. Figure 3(a)-(d) show the frequency-depend
ependent resonance field $H_{\text{res}}$ extracted by fitting experimental FMR spectra with a differential Lorentzian function [34,35]. For high-field range $H > H_c \sim 0.51$ kOe, we found that the dispersion curves of $f$ vs. $H_{\text{res}}$ obtained at all four different temperatures can be well-fitted with the well-known Kittel formula as follows (see Supporting Information for specific derivation process):

$$f = \gamma \left( H_{\text{res}} \cos(\theta_M - \theta_0) + 4\pi M_s \cos(2\theta_M) \right) \times \left( H_{\text{res}} \sin(\theta_M) - 4\pi M_s \sin^2(\theta_M) \right),$$ (1)

where $\gamma = 2.9$ kOe/MHz is the gyromagnetic ratio, $4\pi M_s = 4\pi M_s + H_k$ is the effective magnetization, $M_s$ is the saturation magnetization determined from static magnetization measurements, $H_k$ is the effective anisotropy field, the out-of-plane angle of the external field $\theta_M = 30^\circ$ and magnetization $\theta_0$. For monoaxial chiral helimagnet MnNb$_2$S$_4$ with easy-plane anisotropy, the magnetocrystalline anisotropy constants $K_{u1}$, $K_{u2}$ are defined as $H_k = -(2K_{u1} + 2K_{u2})/(\mu_0 M_s)$, where $K_{u2}$ can be neglected here because it is a fourth-order small item. Thus, $K_{u1}$ can be calculated by $K_{u1} = -(\mu_0 M_s H_k)/2$. The Kittel formula (eq. (1)) can well fit the high-field dispersion relation, indicating that all spins have a uniform precession under the high-field range, consistent with discussed magnetic field-forced FM state at $H > H_c$ in the $H$-$T$ phase diagram above (Figure 1(b)). Note that, for $T = 45$ K, only $f$ vs. $H_{\text{res}}$ data in the high field range was used to be fitted because it exhibits a significant deviation at the low field range due to the strong spin fluctuation near its critical magnetic order temperature $T_c = 45$ K.

Furthermore, we can obtain temperature-dependent effective magnetization $M_{\text{eff}}$, out-of-plane angle of magnetization $\theta_M$, and magnetocrystalline anisotropy constant $K_{u1}$, which were together with the saturation magnetization $M_s$ measured by SQUID magnetometer shown in Figure 3(c) and (f). Analogous to $M_s$, $K_{u1}$ exhibits a monotonic decrease with increasing temperature and rapidly reduces to near zero while temperature approaches $T_c = 45$ K. Moreover, the temperature-dependent equilibrium position of magnetization $\theta_M$ shows that the magnetic moment is more accessible to follow external magnetic field $H$ due to the decrease of $H_k$ and demagnetized field with increasing temperature.

In addition to the discussed $f$ vs. $H_{\text{res}}$ dispersion relation above, the linewidth $\Delta H$, characterized by using the full width at half maximum (FWHM), can be used to analyze the Gilbert damping constant. FWHM is determined by fitting experimental FMR spectra with a differential Lorentzian function [34,35]. Figure 4(a)-(f) show the dependence of FWHM on the excitation frequency $f$ at different temperatures $T = 4.5, 10, 20, 30, 40,$ and $45$ K with $H$ in the $ab$-plane ($\theta_0 = 0^\circ$). For the high-field FFM regime ($H > H_c$), the relation of linewidth $\Delta H$ vs. $f$ can be well-fitted with the following formula $\Delta H = \Delta H_0 + \alpha \Delta f/\gamma$, where $\Delta H_0$ is the inhomogeneous linewidth broadening constant, $\alpha$ is the Gilbert damping factor. One can easily see that the linewidth obviously deviates from the linear fitting in the low-field range, which is caused by the emerging CSL phase in the low field, well consistent with the discussed $f$ vs. $H_{\text{res}}$ dispersion relation in Figure 3. The temperature dependence of the Gilbert damping constants $\alpha$ corresponding to high-field FFM regime (inset of Figure 4(f)) shows a gradual enhancement with increasing temperature at the low-temperature range far below $T_c = 45$ K, and then suddenly reaches 0.11 at 40 K from 0.05 at 30 K when the temperature approaches to $T_c$. The significant broadening of the linewidth near Curie temperature $T_c$ is related to the thermal effect-induced strong spin fluctuation.

Figure 4 (Color online) Temperature- and field-dependence of the FMR linewidth. The FMR linewidth FWHM vs. $f$ experimental data (symbols) obtained at in-plane field $H (\theta_0 = 0^\circ)$, $T = 4.5$ K (a), 10 K (b), 20 K (c), 30 K (d), 40 K (e), and 45 K (f). The linear lines are the fitting results of the FWHM data with $\Delta H = \Delta H_0 + \alpha \Delta f/\gamma$. The dashed lines are the extension of the linear fitting as guides to the eye. The regions with green and blue backgrounds represent the high-field FFM regime and low-field CSL regime, respectively. Inset in (f) shows dependence of the Gilbert damping constant $\alpha$ on temperature determined by the linear fittings of the data in (a)-(f).
3.2 Spin resonance of the low-field CSL regime

Unlike the high-field FFM regime, the low-field CSL regime includes two magnetic structures, helical spin texture and FM phase. Compared with a single FM state, the mixture of helical spin segment and FM part in the CSL regime is expected to exhibit distinct magnetic dynamics due to the change of various magnetic interaction energies of the whole system. As discussed in Figure 3(a) and (d), the experimentally obtained dispersion results deviate significantly from the Kittel formula eq. (1) at the low-field range. Analogous to the unsaturated magnetic domain system [36], we derived a modified Kittel formula (eq. (2)) for this mixture of spin textures by reconsidering the total magnetic interaction energy of the system via setting the proportions of the helical spin segment and FM phase as $q$ and $p = 1 - q$, respectively (see Supporting Information for specific derivation process).

The modified FMR Kittel formula is given as follows:

$$f = \gamma \times \left[ (qH_\text{res}\sin\theta_M\sin\theta_H + pH_\text{res}\cos(\theta_M - \theta_H) + 4\pi M_\text{eff}\cos(2\theta_M)) \right] \left[ (qH_\text{res}\sin\theta_M\sin\theta_H + pH_\text{res}\cos(\theta_M - \theta_H) - 4\pi M_\text{eff}\sin^2\theta_M) \right]^{-1},$$

(2)

where $1/q = L(H_c)/L(0)$ can be proved strictly. Setting $q = 0$ in the modified Kittel model can be returned to the standard Kittel formula (eq. (1)) for the pure ferromagnetic state. As mentioned above, the proportion of helical spin texture $q$ depends significantly on in-plane field component $H_\text{in}$ [13,14]. Therefore, it is difficult to get a reliable fitting result about the exponentially obtained $f_{\text{FMR}}$ vs. $H_\text{res}$ dispersion relation in the low-field CSL regime. Because the spin helix period $L(H)$ (proportional to $1/q$) shows a significant in-plane magnetic field dependent divergence around critical field $H_c$ [13,14].

To further investigate spin dynamics in the low-field nontrivial CSL regime, we adopted out-of-plane angular-dependent FMR spectra. Because the critical field $H_c$ from CSL transferring to FFM is expected to be higher at large $\theta_H$ due to the demagnetization field and strong easy-plane magnetic anisotropy. More specifically, we quantitatively calculate the in-plane component of the resonance field obtained in the out-of-plane angular-dependent FMR spectra and find it only changes by 2.6% at $f = 6$ GHz, $T = 5$ K (see details in Supporting Information), avoiding the in-plane field-induced significant modulation of $q$. Figure 5 shows the dependence of resonance field $H_\text{res}$ on out-of-plane angle $\theta_H$ from $0^\circ$ to $90^\circ$ with $T = 5$ K at different resonant frequencies. Figure 5(a)-(h) show that the experimental angular-dependent $H_\text{res}$ results can be well fitted by the modified Kittel formula eq. (2) (see the detailed fitting process in Supporting Information). The non-zero helical spin proportion $q$ under low excitation frequency (less 8 GHz) indicates the existence of the CSL state in the studied oblique field range with a low in-plane component field $H_\text{in} < H_c$, consistent with the discussion of $f$ vs. $H_\text{res}$ curves at the in-plane field above. Figure 5(i) and (j) show the field dependence of the obtained fitting parameter $q$ and the helical spin period ratio $L(H_\text{res})/L(0)$. Helical spin proportion $q$ gradually decreases with increasing field and reaches zero corresponding to the disappearance of helical spin texture when the resonant field is above its critical field $H_c \sim 5.1$ kOe at $T = 5$ K, similar to the previously reported field-dependent spin helix period $L(H)$ of CSL state in chiral helimagnet CrNb$_2$S$_6$ by using the Lorentz micrograph technique [13,14].

To further investigate the temperature effect on the helical spin period of the low-field CSL regime, we also adopted out-of-plane angular-dependent FMR spectra at different temperatures below $T_c$. Figure 6 shows the dependence of $H_\text{res}$ on out-of-plane angle $\theta_H$ from $0^\circ$ to $90^\circ$ with $f = 6$ GHz.

**Figure 5** (Color online) Out-of-plane angular dependence of FMR spectra at 5 K. The angular dependence of resonance field $H_\text{res}$ at $T = 5$ K, $f_\text{res} = 4$ GHz (a), 6 GHz (b), 7 GHz (c), 8 GHz (d), 10 GHz (e), 12 GHz (f), 15 GHz (g), and 17 GHz (h), respectively. The solid red lines are the results of fitting with the modified Kittel formula eq. (2) described in the main text. Field dependence of the fitting parameter $q$ (i) and the helical spin period ratio $L(H_\text{res})/L(0)$ (j), respectively. The error bar of $q$ is defined in Supporting Information.
Similarly, the obtained angular-dependent $H_{\text{res}}$ data can also be well fitted with the modified Kittel formula eq. (2) shown as solid red fitting curves in Figure 6(a)-(f). Figure 6(g) and (h) show the temperature dependence of the obtained fitting parameter $q$ and $L(H_{\text{in}})/L(0)$. $L(H_{\text{in}})/L(0)$ gradually increases with increasing temperature and reaches infinity when the resonant field is above its critical field $H_c$ at $T \geq 40$ K, also consistent with the previous reports [13,14].

3.3 Phase diagram determined by spin dynamics

In addition to the phase diagram consisting of the FFM and CSL state, as shown in Figure 1(b), determined by the static magnetization characteristics, the dynamic analysis can also provide us with a detailed phase diagram of the CSL state. We quantitatively estimate the proportion of the helical spin texture $q$ (or helical period $L(H)$) from the angular-dependent dispersion relation of spin dynamics. We measure a series of out-of-plane angular-dependent FMR spectra with $f = 4, 7, 8, 10, 12, 15, 17$ GHz at different temperatures $T = 5, 10, 20, 30, 40, 45$ K (see the details in Supporting Information). After the analysis of dispersion relations as discussed above, we obtain the contour plot in terms of the component of the helical spin texture $q$ (equal to $L(0)/L(H)$) in the plane of temperature and in-plane field (Figure 7(a)), being overall consistent with the two-dimension phase diagram (Figure 1(b)) determined by static magnetic susceptibility measurements. In the low-field CSL regime, the spin helix period $L(H)$ gradually increases with increasing the applied external in-plane magnetic field because an in-plane field can help to enhance the FM segment in CSL due to the Zeeman effect. Figure 7(b) shows that the helical spin proportion $q$ vs. normalized in-plane field $H/H_c$ curves obtained at different temperatures collapse into a single field dependence curve.

Our results are consistent with the field dependence of $L(H)$ obtained by the Lorentz micrograph technique [13,14,31], confirming that analysis of out-of-plane field angular dependence of spin resonance using the modified Kittel formula can be regarded as another valid approach to probe the topological spin texture period in chiral magnets.

4 Conclusion

In summary, several specific spin textures and their distinct dynamics of the chiral helimagnet MnNb$_3$S$_6$ have been characterized detailly by field- and temperature-dependent static magnetization and broadband differential FMR spectroscopy. The high-field FFM follows the standard Kittel dispersion relation of a single domain FM state. In contrast, the low-field nontrivial CSL prefers the modified Kittel formula including the partial helix spin texture. Furthermore, like the sophisticated Lorentz micrograph technique, the modified Kittel model proposed in this work as an alternative and easy access approach enables us to extract the temperature- and field-dependent helical spin period ratio $L(H)/L(0)$ quantitatively from the angular-dependent FMR dispersion relations.
persion relation obtained at different temperatures. Our results find that the specific angular-dependent magnetic dy-
namics of nontrivial magnetic states proved in our work
provide a vital clue to exploring interesting magnetic dy-
namics in other topologically nontrivial chiral magnets.

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Supporting Information

The supporting information is available online at http://phys.scichina.com
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