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Magnetostrictive hypersound generation by spiral magnets in the vicinity of magnetic field induced phase transition



Igor V. Bychkov^{a,b}, Dmitry A. Kuzmin^{a,b,*}, Alexander P. Kamantsev^c, Victor V. Koledov^c, Vladimir G. Shavrov^c

^a Chelyabinsk State University, 129 Br. Kashirinykh Str., Chelyabinsk 454001, Russian Federation

^b South Ural State University (National Research University), 76 Lenin Prospekt, Chelyabinsk 454080, Russian Federation

^c Kotelnikov Institute of Radio-engineering and Electronics of RAS, Mokhovaya Street 11-7, Moscow 125009, Russian Federation

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ABSTRACT

In present work we have investigated magnetostrictive ultrasound generation by spiral magnets in the vicinity of magnetic field induced phase transition from spiral to collinear state. We found that such magnets may generate transverse sound waves with the wavelength equal to the spiral period. We have examined two types of spiral magnetic structures: with inhomogeneous exchange and Dzyaloshinskii–Moriya interactions. Frequency of the waves from exchange-caused spiral magnetic structure may reach some THz, while in case of Dzyaloshinskii–Moriya interaction-caused spiral it may reach some GHz. These waves will be emitted like a sound pulses. Amplitude of the waves is strictly depends on the phase transition speed. Some aspects of microwaves to hypersound transformation by spiral magnets in the vicinity of phase transition have been investigated as well. Results of the work may be interesting for investigation of phase transition kinetics as well, as for various hypersound applications.

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1. Introduction

Nowadays, ideas on nanoscale heat control attract great researcher's attention. It is well known that heat transfer in solids occurs mainly by high-frequency phonons (with frequencies more than 0.1 THz), so, these ideas have lead to formation of such cutting-edge physics area like phononics [1–4]. Lower-frequency phonons (in frequency range from 100 MHz up to 0.1 THz), hypersound, cannot to propagate in solids on macroscopic scales due to large damping. For the same reason it is difficult to investigate these waves experimentally. Efforts to generate and detect such phonons are justified by the possibility of submicron scale and nanosecond time-scale investigation of materials properties. Inaccessible to the human ear sound vibrations with frequencies approximately from 20 kHz to 100 MHz, ultrasound, become the basis of a vast number of diagnostic and research technologies [5,6].

Due to great number of application, new ways of generating of ultra- and especially hyper- sound may be very useful. In present work we show the possibility of ultra- and hyper- sound generation by magnets with spiral spin structure during and in the

E-mail address: kuzminda@csu.ru (D.A. Kuzmin).

vicinity of magnetic field induced phase transition into collinear state.

Recently, electromagnetic waves radiation by Heusler"s alloy at structural phase transition has been experimentally observed [7]. Some possible mechanisms of this radiation have been proposed and theoretically investigated as well. In general, material"s energy changes to minimal value during phase transition. This change of energy may be released like electromagnetic waves, heat, sound etc. In magnetic materials spin, electromagnetic and acoustic waves are coupled. Excitation any of them leads to energy redistribution between all subsystems. One of manifestations of this effect is a well-known phenomenon of electromagnetic sound generation in magnetic materials (see, for example, review [8]). The features associated with electromagnetic-acoustic transformation became the basis for some methods of study and detection of phase transitions in magnetic materials [9-14]. In spiral magnets such hybridization of the oscillations has a number of features and may be controlled by external magnetic field [15,16]. In [17] authors experimentally observed and theoretically explained intensive electromagnetic-acoustic transformation and anomalies in transverse sound velocity in erbium single crystal at different phase transitions (including state with spiral magnetic structure). These features may be explained by resonant subsystems interaction in the vicinity of phase transition. Electromagnetic-sound transformation should take a place even far from resonances. It will be less efficiently, but may be more convenient for practical

^{*} Corresponding author at: Chelyabinsk State University, 129 Br. Kashirinykh Str., Chelyabinsk 454001, Russian Federation.

implementation. Features of such transformation are investigated in this paper. The possibility of sound generation by spiral magnets during phase transition has never been investigated. This issue is also covered in this paper.

2. Theory and calculations

Let us consider spiral magnet crystal. For definiteness, we will assume that it has a hexagonal symmetry. It's properties may be described phenomenologically by the following free energy density

$$F = \frac{\alpha}{2} \left(\frac{\partial \mathbf{M}}{\partial x_i} \right)^2 + F_{in} + \frac{\beta_1}{2} M_z^2 + \frac{\beta_2}{2} M_z^4 - H M_z + b_{ijlm} M_i M_j u_{lm} + c_{ijlm} u_{ij} u_{lm,},$$
(1)

where α and β_i are exchange and anisotropy constants, b_{ijlm} and c_{ijlm} are the tensors of magnetostriction and elasticity, **M** is the crystal magnetization, *H* is the value of external magnetic field directed along *z*-axis (spiral axis), $u_{ij} = (\partial u_i/\partial x_j + \partial u_j/\partial x_i)/2$ are the components of the strain tensor, u_i are the components of displacement vector. Term F_{in} corresponds to inhomogeneous magnetic interaction. In case of exchange-caused spiral this is $F_{in} = \gamma (\partial^2 \mathbf{M}/\partial x_i^2)^2/2$, while in Dzyaloshinskii–Moriya interaction-caused case $F_{in} = \alpha_1 \mathbf{M} rot \mathbf{M}$, γ and α_1 are the constants of inhomogeneous exchange and Dzyaloshinskii–Moriya interaction, consequently. Ground state of such system corresponds to the minimum of free energy density. In case under consideration (i.e. when both anisotropy and external magnetic field distinguish *z*-axis) magnetization in ground sate will rotate in *xy*-plane. Magnetization may be expressed in the form

$$M_{0x} = M_0 \sin\theta \cos qz, \quad M_{0y} = M_0 \sin\theta \sin qz,$$

$$M_{0z} = M_0 \cos\theta,$$
 (2)

where M_0 is saturation magnetization, $q=2\pi/L$ is wavenumber of the spiral, *L* is spiral period, θ is angle between spiral axis and magnetization direction. When $\theta=\pi/2$, magnetic is in simple spiral phase, $\theta=0$ correspond to collinear ferromagnetic state. When $0 < \theta < \pi/2$ magnetic is in conical (ferromagnetic) spiral phase. Spiral angle θ depends on external magnetic field value and may be calculated by solving of equation

$$M_0 \cos\theta \left[\tilde{\beta}_1 + h_{me} + \left(\tilde{\beta}_2 - \frac{h_{me}}{M_0^2} \right) M_0^2 \cos^2\theta + \alpha q^2 + \tilde{\Delta} \right] + H = 0$$
(3)

where $\tilde{\beta}_1$ and $\tilde{\beta}_2$ are renormalized by magnetostriction anisotropy constants [15,16]. For exchange interaction-caused spiral structure we have $\gamma > 0$, $\alpha < 0$, $q = (-\alpha/2\gamma)^{1/2}$, $h_{me} = (b_{11} - b_{12})^2 M_0^2 / (c_{11} - c_{12})$, $\tilde{\Delta} = \gamma q^4$. In Dzyaloshinskii–Moriya interaction-caused case $\alpha_1 \neq 0$, $\alpha > 0$, $q = \alpha_1/\alpha$, $h_{me} = b^2 M_0^2 / 2\mu$, $\tilde{\Delta} = -2\alpha_1 q$. Magnetosriction is small enough and not affects the ground state configuration of magnetic subsystem.

Equilibrium deformations tensor has been calculated in [16]. It's components u_{xx}^{*} , u_{yy}^{*} , and u_{zz}^{*} are homogeneous and depend on the spiral angle θ , while the components u_{xz}^{*} , and u_{yz}^{*} are inhomogeneous.

For investigation of sound waves generation we should to solve equation of motion for elastic medium

$$\rho \ddot{u}_i = \partial \sigma_{ij} / \partial x_j, \quad \sigma_{ij} = \partial F / \partial u_{ij}. \tag{4}$$

Let us consider only waves propagating along *z*-axis. Eq. (4) with (1) lead to the following wave equations

$$\begin{aligned} \ddot{u}_{\pm} &- v_t^2 \partial^2 u_{\pm} / \partial z^2 = 2\rho^{-1} b_{44} M_z \partial M_{\pm} / \partial z; \\ \ddot{u}_z &- v_l^2 \partial^2 u_z / \partial z^2 = 4\rho^{-1} b_{13} \big(M_x \partial M_x / \partial z + M_y \partial M_y / \partial z \big). \end{aligned}$$
(5)

In (5) we have introduced circular components $(u, M)_{+} = (u, M)_{+}$ $(M)_x \pm i(u, M)_y$, transversal and longitudinal sound velocities v_t $=(2c_{44}/\rho)^{1/2}$ and $v_l = (c_{33}/\rho)^{1/2}$, consequently. Right-hand sides of Eq. (5) mean the sound source functions, and in general may depend from both coordinate and time. Indeed, computation of time dependence of magnetization vector's components during phase transition (i.e. phase transition kinetics) is an individual problem. It may be modeled, for example, by time-dependent Ginzburg-Landau, Landau-Khalatnikov, or Landau-Lifshitz equations with different damping terms. We will consider only some simple for analysis ideal cases without solving of the phase transition kinetics problem. Our calculations are valid for processes when characteristic time of magnetization change is much larger than time of local quasi-equilibrium setting up in material. In real magnets local quasi-equilibrium distribution of magnetic moments is reached quickly due to strong exchange interaction [18].

For numerical estimations we will use following constants values [19]: $b_{ij} \sim 20 \text{ erg}/(\text{Oe} \times \text{cm}^4)$, $\rho \sim 10 \text{ g/cm}^3$, $v_t \sim 3 \times 10^5 \text{ cm/s}$, $v_{l} \sim 5 \times 10^{5}$ cm/s, $M_{0} \sim 500$ Oe. Period of the structure for Dzyaloshinskii-Moriya interaction-caused spiral magnets is usually much more that in case of exchange-caused spiral structures [20]. For example, $Fe_xCo_{1-x}Si$ alloys, which symmetry allows Dzyaloshinsky–Moria interaction, for x=0.3 in spiral state has a modulation period L=230 nm $(q\sim 3 \times 10^5 \text{ cm}^{-1})$ [21]. Other examples of the magnets with Dzyaloshinsky-Moria interaction-caused spiral structures are FeGe (L=70 nm, $q\sim8\times10^5$ cm⁻¹) [22] and MnSi (L=18 nm, $q\sim3\times10^6$ cm⁻¹) [23]. Different modulated states exist in erbium single crystal due to the competing exchange interaction. In conical state wavenumber of the structure is 5c²/21 (c²) $=2\pi/c$ is inverse lattice constant, c=0.56 nm is lattice constant) [24], i.e. $q \sim 3 \times 10^7 \text{ cm}^{-1}$. We will use $q \sim 10^5 \text{ cm}^{-1}$ and $q \sim 10^8$ cm⁻¹, consequently.

During phase transition, spin waves (or magnons) may also be excited. So, components of magnetization vector M_i should have the form $M_i(z,t)=M_{i0}(z,t)+m_i(z,t)$, where term $M_{i0}(z,t)$ means change of the ground state during the phase transition, while $m_i(z, t)$ correspond to excited spin waves. We will consider only sound waves excited by $M_{i0}(z,t)$ because usually, change of the ground state magnetization during the phase transition is much more than excited spin waves amplitude. In such a case it is easy to show, that only transverse sound will be excited. From (5) we will have

$$\ddot{u}_{\pm} - v_t^2 \partial^2 u_{\pm} / \partial z^2 = \pm i q \rho^{-1} b_{44} M_0^2 \sin(2\theta) \exp(\pm i q z).$$
(6)

Solution of (6) can be obtained by the Duhamel's principle [25]

$$u_{\pm}(z, t) = (2v_t)^{-1} \int_0^t \int_{z-v(t-s)}^{z+v(t-s)} f_{\pm}(\xi, s) d\xi ds, , \qquad (7)$$

where $f_+(z, t)$ is right-hand side of Eq. (6).

First of all let us consider infinitely fast phase transition, corresponding to the spiral suppression. Despite an evident nonreality of such a process, this situation corresponds to the maximum change of sound source function and, thus, to maximum amplitude of the emitted sound. We will consider such process only for estimation of fundamentally maximal value of amplitude of emitted sound. This process may be modeled by step-like time dependence of spiral angle θ . From (6) one can see, that source function is equal to zero for both simple spiral phase ($\theta = \pi/2$) and collinear ferromagnetic state ($\theta = 0$), and has a maximum at $\theta = \pi/4$. So, if we will have infinitely fast phase transition from simple spiral phase to collinear ferromagnetic one, sound waves will not be emitted. Maximum sound waves generation will take a place at

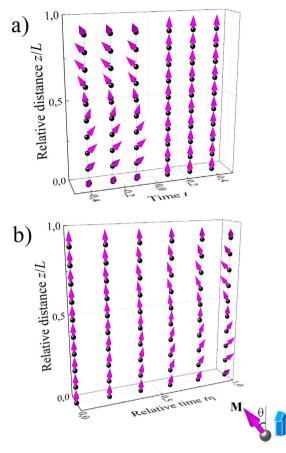


Fig. 1. (single-column) Model processes of phase transitions in spiral magnets: (a) show an infinitely fast transition from cone state with $\theta = \pi/4$ to collinear ferromagnetic state; (b) show the process when $sin[2\theta(t)] = \eta t$, $t < \eta^{-1}$, i.e. linear time dependence of sound source function.

phase transition from $\theta = \pi/4$ to collinear ferromagnetic state. Mathematically, it may be expressed as $\sin[2\theta(t)] = \Theta(t)$, where $\Theta(t)$ is Heaviside step function (see Fig. 1a). In such case integral (7) may be easily calculated:

$$u_{\pm}(z, t) = \pm i b_{44} M_0^2 \exp[\pm i q z] [1 - \cos(q v_t t)] / \rho q v_t^2.$$
(8)

This corresponds to the inhomogeneous distribution of displacements with oscillations. The oscillations frequency is $\omega = qv_t$. For Dzyaloshinskii–Moriya interaction-caused spiral magnets linear frequency $f = \omega/(2\pi) \sim 5$ GHz, and amplitude is $u_{\pm} \sim 10^{-5}$ cm. In exchange-caused case $f \sim 5$ THz, $u_{\pm} \sim 10^{-8}$ cm. Both cases correspond to hypersound oscillations.

Let us consider now, how the phase transition speed will affect on sound generation. For these purposes we will consider model process, when $\sin[2\theta(t)] = \eta t$, $t < \eta^{-1}$ (i.e. linear time-dependence of source function, see Fig. 1b). Calculations show, that the following sound oscillations will be excited:

$$u_{\pm}(z, t) = \pm \eta \frac{ib_{44}M_0^2}{\rho q^2 v_t^3} \Big[qv_t t - \sin(qv_t t) \Big] \exp[\pm iqz], \quad t < \eta^{-1}.$$
(9)

One can see that sound oscillations with the same frequency are excited. Amplitude of exited oscillations linearly depends on the "phase transition speed" η . We may calculate, that $u_{\pm} \sim 10^{-21}$ $\times \eta$ cm and $u_{\pm} \sim 10^{-27} \times \eta$ cm for Dzyaloshinskii–Moriya and exchange interaction-caused spiral magnets, respectively. For example, ultrafast magnetization reversal processes have a time of magnetization switching about 10^{-12} s, or $\eta \sim 10^{12}$ s⁻¹. So, we will have amplitudes $u_{\pm} \sim 10^{-9}$ cm Dzyaloshinskii–Moriya caused spiral magnets, while in case of exchange-caused spiral magnets the amplitude will be very small. Note, that we have considered an infinitely long crystal. This has lead to the exciting of standing sound waves. For finite crystal depending on acoustic characteristics of surroundings, one may to expect two typical scenarios. In case of perfectly acoustically conjugated surroundings and spiral magnet (i.e. when acoustic characteristics of both mediums at frequencies of emitted sound are equal) two sound pulses with characteristic time length of about $\tau \sim d/v_t$, where *d* is the sample size will be emitted from both surfaces of the sample. For sample size $d \sim 1$ mm we will have time length $\tau \sim 10^{-6}$ s. Generally, in case of arbitrary surroundings exited waves will be partially reflected from the sample boundaries and emitted into surroundings. The details of this process are depend on boundary conditions on displacements and should be considered separately.

Let us consider now the features of microwave generation of sound by spiral magnets. In such a case we cannot neglect influence of spin waves generations, because it will be the main mechanism of sound generation. Change of the ground state may be used only for tuning of some properties of emitted sound waves. So, wave Eq. (5) may be transformed in linear approximation into the followings:

$$\begin{aligned} \partial^{2} u_{\pm} / \partial z^{2} &- v_{t}^{-2} \ddot{u}_{\pm} = -b_{44} c_{44}^{-1} (M_{z0} \partial m_{\pm} / \partial z + m_{z} \partial M_{\pm 0} / \partial z); \\ \partial^{2} u_{z} / \partial z^{2} &- v_{l}^{-2} \ddot{u}_{z} = -2b_{13} c_{33}^{-1} (M_{x0} \partial m_{x} / \partial z + m_{x} \partial M_{x0} / \partial z + M_{y0} \partial m_{y} / \partial z \\ &+ m_{y} \partial M_{y0} / \partial z). \end{aligned}$$

One can see that now both longitudinal and transversal sound waves may be excited. Transversal magnetization oscillations may excite both longitudinal and transverse sounds, while longitudinal magnetization oscillations will affect only on transverse sound generation. At phase transition into collinear ferromagnetic state longitudinal sound will disappear, and transversal sound will be excited only by transversal magnetization oscillations. Following to [5] we will assume that magnetization oscillations at ferromagnetic resonance are homogeneous, i.e. $\partial m_i(z, t)/\partial z \approx m_{i0} \left[\delta(z) - \delta(z - d) \right] \exp(-i\omega t),$ $m_i(z, t) = m_{i0}\Theta(z)\Theta(d-z)\exp(-i\omega t)$, where $\delta(z)$ is Dirac's deltafunction, d is sample size. Wave Eq. (10) may be solved with using of Green's functions. Green's function for transverse and longitudinal waves $G_t(z, z_0)$, respectively, consist of two terms. The first one is $(i/2 k_{tl})\exp(ik_{tl}|z-z_0|)$ corresponding to the wave excited at z_0 and propagating in +z direction, while the second one is $(i/2 k_{tl})\exp(ik_{tl})$ $z+z_0$) corresponding to the wave exited at the same point but propagating in opposite direction, here $k_{t,l} = \omega / v_{t,l}$. Therefore complete Green's function has the form

$$G_{t,l}(z, z_0) = ik_{t,l}^{-1} \exp(-ik_{t,l}z) \cos(k_{t,l}z_0)$$
(11)

Calculations show that amplitudes of the excited waves are

$$\begin{aligned} u_{\pm} &= -\frac{ib_{44}M_0}{k_t c_{44}} \left\{ m_{\pm 0} \cos \theta \Big[1 - \cos(k_t d) \Big] \right. \\ &\pm iq \frac{m_{20} \sin \theta}{q^2 - k_t^2} \Big[k_t \exp(\pm iq d) \sin(k_t d) \mp iq \big(1 - \exp(\pm iq d) \cos(k_t d) \big) \Big] \Big\}; \\ u_z &= -\frac{4ib_{13}M_0 \sin \theta}{k_l c_{33}} \left\{ m_{x0} \frac{k_l}{q^2 - k_l^2} \Big[q \sin q d \sin k_l d - k_l \cos q d \cos k_l d \Big] \right. \\ &- m_{y0} \sin q d \Big[\cos k_l d - \frac{q \sin k_l d}{q + k_l} \Big] \Big\}. \end{aligned}$$
(12)

One can see that amplitudes of excited sound have a complex dependence from the sample size, spiral wavenumber and wavenumber of excited sound wave. So, for example, when $qd = \pi n$, where *n* is an integer number (this means that the sample have an integer value of half spiral periods), longitudinal sound may be excited only by *x*-polarized microwaves. Amplitudes (12) depend

on the spiral angle, which may be controlled by external magnetic field (see Eq. (3)). In case of $\sin\theta = 0$, i.e. when $H > H_{cr} = M_0 \Big[\tilde{\beta}_1 + \tilde{\beta}_2 M_0^2 + \alpha q^2 + \tilde{\Delta} \Big]$, only transverse sound will be excited. When H=0 (i.e. $\cos\theta=0$) longitudinal sound may be excited by transverse microwave, while transverse sound may be excited by longitudinal microwaves. So, external magnetic field affects the polarization and amplitude of emitted sound. For microwaves wavelength of excited wave is usually much more that the spiral period, so, condition $k_{t,l} \ll q$ is satisfied. For such approximation we will have

$$u_{\pm} = -\frac{ib_{44}M_0}{k_t c_{44}} \left\{ m_{\pm 0} \cos \theta \left[1 - \cos(k_t d) \right] \mp i m_{z0} \sin \theta \left[1 - \exp(\pm i q d) \right] \right\};$$

$$u_z = -\frac{4ib_{13}M_0 \sin \theta}{k_l c_{33}} m_{y0} \sin q d.$$
(13)

Eq. (13) shows that longitudinal sound is mainly excited by *y*-polarized microwave and may be excited only in spiral state $(\sin\theta\neq 0)$. This sound will disappear at phase transition into collinear ferromagnetic state. Transversal sound may be excited by transverse microwaves in simple or conical ferromagnetic spiral states. Excitation of this sound will disappear at $\cos\theta=0$.

Transverse sound may be excited by longitudinal microwave as well when magnetic is in conical ferromagnetic spiral state.

Full potential energy density of excited oscillations $W = c_{33}(u_{zz})^2$ $(2+2c_{44}](u_{xz})^2+(u_{yz})^2]$. Fragments of frequency and size dependencies of this energy density calculated from exact solutions (12) at some frequency and size range for different polarizations of exciting microwave are shown on Fig. 2. One can see that these dependencies have an oscillating behavior. These oscillations correspond to interference of the sound waves propagating from opposite surfaces of the sample. The other reason for oscillations is periodical source function. Eq. (12) have a resonances corresponding to conditions $k_{t,l} = q$. At microwave frequencies these resonances may have a place only for magnets with Dzyaloshinskii-Moriya interaction-caused spiral structure $(q \sim 10^5 \text{ cm}^{-1}, \omega_{res} \sim$ $qv_{t,l} \sim 10^{10}$ rad/s). For exchange spiral magnets resonant frequencies are of terahertz range ($q \sim 10^8$ cm⁻¹, $\omega_{res} \sim qv_{tl} \sim 10^{13}$ rad/s). At such frequencies magnetization oscillations may be inhomogeneous and formulas (12) cannot be used. Calculations show also that frequency range near resonance may be distinguished by behavior relative to the spiral angle θ (external magnetic field H_0). Far enough from resonance, acoustic energy excited by x- or y- polarized microwaves

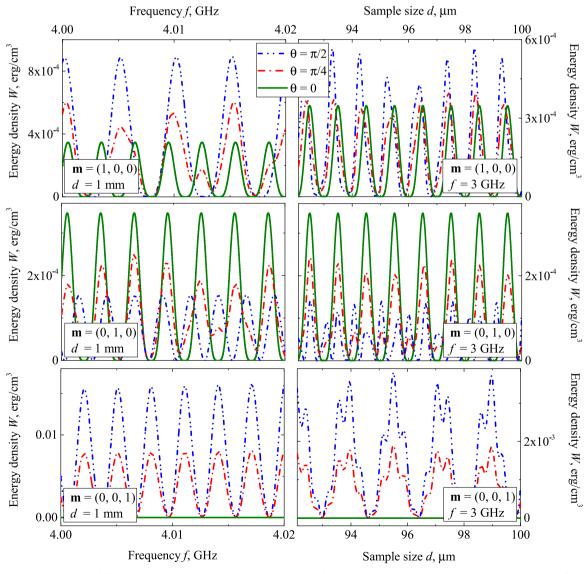


Fig. 2. (double-column) Fragments of frequency and size dependencies of energy density of the sound emitted by spiral magnet placed in microwave field with different polarization. Spiral wavenumber correspond to Dzyaloshinskii–Moriya interaction-caused spiral structure $q = 10^5$ cm⁻¹.

decreases with increasing of spiral angle, while near resonance it increases.

3. Results and discussion

We investigated the possibility of sound generation by spiral magnets during and in the vicinity of magnetic field induced phase transitions. We showed that magnets with Dzyaloshinskii–Moriya interaction-caused spiral structure may emit low frequency (about some GHz) hypersonic pulses during fast phase transition from conical spiral phase to collinear state, while exchange-caused spiral magnets may generate THz pulses, corresponding heat phonons excitation. Amplitude of the emitted pulses is linearly depends on "phase transition speed". This dependence may be used for investigation the details of phase transition kinetics.

We investigated also electromagnetic microwave-to-sound transformation by spiral magnets and found that it may be efficiently controlled by external magnetic field. Microwave-to-sound transformation may be controlled by microwave polarization (or sample orientation) as well. Such transformation may be used for controllable hypersonic generation. For investigation of the details of slow phase transitions in spiral magnets dependence of emitted sound energy on the spiral angle may be used.

In present work we used continual approach. It is correct far from the boundaries of the first Brillouin zone. For processes of electromagnetic microwave-to-sound transformation this condition is well satisfied (wavelength of the generated sound $\lambda_{lt} = 2\pi v$ $L_{l,t}/\omega \sim 10^{-5}$ cm is much more then lattice constant). The same situation holds for the sound generation at phase transition in spiral magnets with Dzyaloshinsky-Moria interaction (wavelength equal to the spiral period, which reaches hundreds of nanometers). In magnetic materials with exchange interaction-caused spiral structure modulation period is usually about tens of lattice constants, i.e. the condition is satisfied worse. This can lead to some difference between the calculated amplitudes and frequencies. It is known that near the Brillouin zone boundaries the speed of sound is reduced. Estimation can be carried out from the model of elastic chain consisting of one atomic species. Dispersion low of acoustic oscillations in such model has a form $\omega = \nu \sin(ka)/a$, where a is lattice constant, v is speed of the sound wave near centre of the first Brillouin zone. So, for magnets with inhomogeneous exchange interaction, for $k = q = 2\pi/L$, $L \approx 10a$, one may estimate phase speed of the sound $v^*(q) = \omega/k|_{k=q} \approx v \times \sin(2\pi/10)/(2\pi/10) \approx 0.94 v$. Estimations show, that taking into account the discreteness of the medium in the first approximation leads to decrease in the frequency of emitted sound on about 6% and to increase in the amplitude on about 10%, which does not change the order of magnitude in estimations we made above. The model used to describe sound generation is correct when sample size is much larger than the spiral period, i.e. for magnets with Dzyaloshinsky-Moria interaction $d > 10 \,\mu\text{m}$, while for magnets with inhomogeneous exchange interaction d > 10 nm. Thus, spiral magnets can be used as micro- and nano- hypersonic wave's generators.

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