

# Dynamical Susceptibility and Elementary Excitations in Monolayer Ferroic Films Described by XZ Heisenberg Model

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The dynamical susceptibility and finite temperature collective excitations in monolayer spin film are calculated using the anisotropic exchange XZ-Heisenberg model for different spin magnitude and the functional integral method. Combining mean field and Gaussian approximation, it is shown that the internal transverse field induced by internal transverse exchange interaction, leads to a decrease (increase) of the spin wave energy in the temperature region below (above) the spin reorientation temperature. This temperature is of the same nature as the critical temperature of the transverse Ising model studied previously. Reduction of the external spin reorientation transverse field (the critical tuning parameter) by intrinsic transverse exchange is demonstrated. [doi:10.2320/matertrans.MD201714]

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## 1. Introduction

Ferroelectric (FE) nanostructures including ultrathin films (UTF) is one of most intriguing topics due to its potential applications in modern electronic nanoscale devices.<sup>1)</sup> A fundamental problem is that FE properties of nanostructures are not only declined with reducing size but also are strongly affected by other factors like the external pressure, atomic substitution or transverse field (TrF), which are generally called tuning parameters. Tuning parameters may be external or internal factors. The dependence of the phase diagram of FEs on the tuning parameter is given in the Scott *et al.* paper.<sup>2)</sup> The transverse Ising spin model (TIM) is the simplest microscopic model, which can be used for studying the influence of the tuning parameter on FE properties. Since the time De Gennes firstly used the transverse spin 1/2 Ising model to explain the ferroelectric phase of KDP<sup>3)</sup> up to now, the TIM has been applied successfully for solving important problems, such as: order-disorder phase transitions in FE materials, spin glass phenomenon, quantum phase transition,<sup>4)</sup> thin films.<sup>5-7)</sup> In the previous work,<sup>8)</sup> the role of the external TrF in damping of order-disorder phase transition temperature of UTF was investigated and offered an explanation of the thickness dependence of the Curie temperature of PbTiO<sub>3</sub> thin films. Reduction of the critical TrF with reducing the film thickness was also pointed out.

This paper is aimed to show that the intrinsic transverse exchange interactions between FE dipoles can further reduction of the external critical TrF. It is also shown that the TrF in the TIM for films, which has been studied for a long time,<sup>5-7)</sup> is corresponding to the spin reorientation (SR) problem of magnetic film (see Ref. 9). Without any long-range orders at finite temperature in spin systems described by two dimensional isotropic exchanges Heisenberg spin model,<sup>10)</sup> and with the TIM corresponding to an anisotropic non-diagonal Heisenberg exchange model, it is necessary to use the simplest anisotropic exchange model, such as XZ-Heisenberg (XZ-H) model. In this paper, the XZ-H model

with different spin-*S* values is used to calculate the dynamical susceptibility, elementary excitations of monolayer spin film (MLSF) at finite temperature. Unlike previous researches,<sup>5-8)</sup> we use functional integral method<sup>11)</sup> to calculate the Green functions (GFs) and dynamical susceptibilities with the Gaussian approximation. The transverse exchange interactions in the XZ-H model can be an origin of the decrease of magnitude of collective elementary excitations compared with that for the TIM.

The paper is organized as follows: Part 1 is for introduction. In Part 2, we introduce the model and calculation method. In Part 3, we obtain the dynamical susceptibility, the temperature- and field-dependence of collective excitation (spin wave), and discuss our results in detail. The conclusion is given in Part 4.

## 2. XZ Heisenberg Model and Green Functions

### 2.1 Hamiltonian of the XZ Heisenberg model for monolayer spin films

We consider a monolayer square spin film lattice<sup>8)</sup> containing *N* spins (*N* ≫ 1). A spin position is defined by **R<sub>j</sub>**—a two dimensional vector in the film plane and distance between *j*-th and *j'*-th spins is *R<sub>jj'</sub>* = |**R<sub>j</sub>** − **R<sub>j'</sub>**|. The Hamiltonian of the XZ anisotropic exchange Heisenberg model for spin system is given by

$$H = -h_0 \sum_j s_j^z - \Omega_0 \sum_j s_j^x - \frac{1}{2} \sum_{j,j'} [J_{jj'} s_j^z s_{j'}^z + L_{jj'} s_j^x s_{j'}^x]. \quad (1)$$

Here *h*<sub>0</sub>, *Ω*<sub>0</sub> are the external longitudinal and transverse fields given in energy unit (the effective dipole moment *μ* is included in the field *h*<sub>0</sub>). The *s<sub>j</sub><sup>z</sup>*, *s<sub>j</sub><sup>x</sup>* are components of the spin operator at site *j* in the crystallographic *xyz* frame, which has *z*-axis (*x*-axis) perpendicular (in-plane) to the square spin lattice. *J<sub>jj'</sub>* = *J*(*R<sub>jj'</sub>*), *L<sub>jj'</sub>* = *L*(*R<sub>jj'</sub>*) are exchange parameters between spin components along the *z* and *x* directions. We propose anisotropic exchange interactions *J<sub>jj'</sub>* ≠ *L<sub>jj'</sub>* and use the mean field approximation (MFA) as the zero-order

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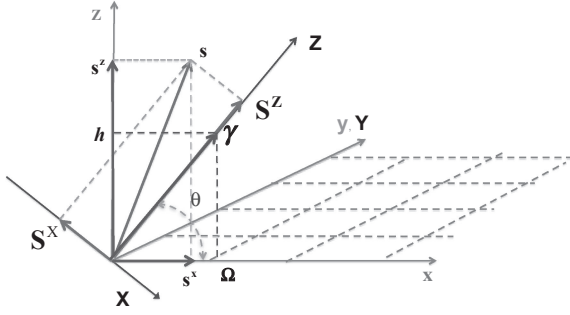


Fig. 1 Relative orientation between the crystallographic coordinates  $xyz$  and rotated  $XYZ$  systems. Incline angle of the spin direction above the film plane is denoted by  $\theta$ .

approximation with the Hamiltonian  $H_0$ . Under the influence of the total field  $\gamma$ , spins rotate and orient along the direction of this field in equilibrium state (see Fig. 1). Choosing a new coordinate  $XYZ$  system, which has  $Z$ -axis parallel to the direction of total field,  $\gamma$ , using the unitary transformation to transform the spin operator in the  $xyz$  system to the spin operator in the  $XYZ$  coordinate system (components of the spin operator in the  $XYZ$  system are denoted by capital letters)  $s_j^x = \frac{h}{\gamma} S_j^X + \frac{\Omega}{\gamma} S_j^Z$ ,  $s_j^z = -\frac{\Omega}{\gamma} S_j^X + \frac{h}{\gamma} S_j^Z$ , we rewrite Hamiltonian (1) in new form

$$H = H_0 + H_{\text{int}}. \quad (2)$$

MF and interacting parts of Hamiltonian are

$$H_0 = \frac{N}{2} \{J(0)m_z^2 + L(0)m_x^2\} - \gamma \sum_j S_j^Z, \quad (3)$$

$$H_{\text{int}} = -\frac{1}{2} \sum_{\alpha\alpha'} \sum_{j,j'} J_{jj'}^{\alpha\alpha'} \delta S_j^\alpha \delta S_{j'}^{\alpha'}. \quad (4)$$

The total field consists of longitudinal and transverse components

$$\begin{aligned} \gamma &= \sqrt{h^2 + \Omega^2}, \quad h = h_0 + J(0)m_z, \\ \Omega &= \Omega_0 + L(0)m_x, \end{aligned} \quad (5)$$

where  $m_z, m_x$  are statistical averages of spin projections per site  $\langle s_j^{z,x} \rangle = \text{Tr}(e^{-\beta H} s_j^{z,x}) = m_{z,x}$ .  $J(0), L(0)$  appeared in eq. (5) are the zero wave vector Fourier inversion of exchange parameters (see eq. (30)).  $H_{\text{int}}$  describes the interaction between the spin fluctuation components  $\delta S_j^\alpha$  ( $\alpha = X, Z$ ), and  $\delta S_j^Z = S_j^Z - \langle S_j^Z \rangle$ ,  $\delta S_j^X = S_j^X$ . Figure 1 shows relative orientation between the  $xyz$  and  $XYZ$  coordinates. Hamiltonian  $H_{\text{int}}$  has a quadratic form in the  $XYZ$  coordinate system.

Implementing Fourier transformation for the spin fluctuation operators, we obtain  $H_{\text{int}}$  in the wave vector representation

$$H_{\text{int}} = -\frac{1}{2} \sum_{\mathbf{k}, \alpha\alpha'} I^{\alpha\alpha'}(\mathbf{k}) \delta S^\alpha(\mathbf{k}) \delta S^{\alpha'}(-\mathbf{k}), \quad (6)$$

where anisotropic exchange interactions are represented by the following  $2 \times 2$  matrix

$$\begin{aligned} \hat{\mathbf{I}}(\mathbf{k}) &= \begin{pmatrix} I^{XX}(\mathbf{k}) & I^{XZ}(\mathbf{k}) \\ I^{ZX}(\mathbf{k}) & I^{ZZ}(\mathbf{k}) \end{pmatrix} \\ &= \frac{1}{\gamma^2} \begin{pmatrix} \Omega^2 J(\mathbf{k}) + h^2 L(\mathbf{k}) & \Omega h [L(\mathbf{k}) - J(\mathbf{k})] \\ \Omega h [L(\mathbf{k}) - J(\mathbf{k})] & h^2 J(\mathbf{k}) + \Omega^2 L(\mathbf{k}) \end{pmatrix}. \end{aligned} \quad (7)$$

$J(\mathbf{k}), L(\mathbf{k})$  are Fourier inversions of the exchange interactions given in eq. (1). The symmetric matrix  $\hat{\mathbf{I}}(\mathbf{k})$  means invariant not only under the sign change of the wave vector but also under the permutation of alpha index, i.e.  $I^{\alpha\alpha'}(\mathbf{k}) = I^{\alpha'\alpha}(\mathbf{k})$ . The free energy and the longitudinal, transverse static susceptibilities are given by ( $k_B T = \beta^{-1}$ )

$$F = -\beta^{-1} \ln \text{Tr}(e^{-\beta H}), \quad (8)$$

$$\chi_{st}^{zz} = \beta \mu^2 \frac{\partial^2 F}{\partial h_0^2}, \quad \chi_{st}^{xx} = \beta \mu^2 \frac{\partial^2 F}{\partial \Omega_0^2}. \quad (9)$$

The MFA solution for thermodynamic quantities of the MLSF is given in Ref. 8.

## 2.2 Functional integral representation for Green functions, Gaussian fluctuation approximation

The longitudinal and transverse susceptibilities of the MLSF are calculated via the free energy or spin-spin correlation functions

$$\chi^{zz} = \beta \mu^2 \sum_{j,j'} \langle \delta s_j^z \delta s_{j'}^z \rangle, \quad \chi^{xx} = \beta \mu^2 \sum_{j,j'} \langle \delta s_j^x \delta s_{j'}^x \rangle. \quad (10)$$

The spin-spin correlation functions appeared in eq. (10) can be calculated using imaginary time (Matsubara) Green function (GF) constructed from components of spin fluctuation operators  $\delta s_j^\alpha$  ( $\alpha = x, z$ ) in the spin lattice  $xyz$  coordinates within the Heisenberg representation, which is

$$\begin{aligned} G^{\alpha\alpha'}(\mathbf{R}_{jj'}, \tau_1 - \tau_2) &= \langle \hat{T} \delta \tilde{s}_j^\alpha(\tau_1) \delta \tilde{s}_{j'}^{\alpha'}(\tau_2) \rangle, \\ \delta \tilde{s}_j^\alpha(\tau) &= e^{\tau H} \delta s_j^\alpha e^{-\tau H}. \end{aligned} \quad (11)$$

Making Fourier transformation, we have

$$\begin{aligned} G^{\alpha\alpha'}(\mathbf{R}_{jj'}, \tau) &= \frac{1}{N} \sum_{\mathbf{k}, \omega} G^{\alpha\alpha'}(\mathbf{k}, \omega) e^{-i\mathbf{k}\mathbf{R} - i\omega\tau}, \\ \omega &= 2\pi n/\beta, \quad n \text{ is an integer number.} \end{aligned} \quad (12)$$

Denoting  $(\mathbf{k}, \omega)$  by a three-component wave vector  $\mathbf{q}$ , we obtain

$$\begin{aligned} G^{\alpha\alpha'}(\mathbf{q}) &= \langle \hat{T} \delta \tilde{s}^\alpha(\mathbf{q}) \delta \tilde{s}^{\alpha'}(-\mathbf{q}) \rangle \\ &= \frac{1}{\beta} \sum_{\mathbf{R}} \int_0^\beta d\tau G^{\alpha\alpha'}(\mathbf{R}, \tau) e^{+i\mathbf{k}\mathbf{R} + i\omega\tau}, \end{aligned} \quad (13)$$

$$\delta \tilde{s}^\alpha(\mathbf{q}) = \frac{1}{\beta \sqrt{N}} \sum_j \int_0^\beta \delta s_j^\alpha(\tau) e^{+i\mathbf{k}\mathbf{R}_j + i\omega\tau} d\tau,$$

$$\delta \tilde{s}_j^\alpha(\tau) = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}, \omega} \delta \tilde{s}_j^\alpha(\mathbf{k}, \omega) e^{-i\mathbf{k}\mathbf{R}_j - i\omega\tau}. \quad (14)$$

Fourier inversion of the dynamical susceptibility is then defined as

$$\chi^{\alpha\alpha'}(\mathbf{q}) = \beta \mu^2 G^{\alpha\alpha'}(\mathbf{q}). \quad (15)$$

The static susceptibilities in the long wavelength limit are readily derived from eq. (15)

$$\chi_{st}^{\alpha\alpha} = \beta \mu^2 G^{\alpha\alpha}(0). \quad (16)$$

Defining Fourier inversion of GFs in the  $XYZ$  coordinate frame and Heisenberg picture as

$$\begin{aligned}\Gamma^{\alpha\alpha'}(\mathbf{q}) &= \langle \hat{T} \delta \tilde{S}^\alpha(\mathbf{q}) \delta \tilde{S}^{\alpha'}(-\mathbf{q}) \rangle \\ &= \frac{\langle \hat{T} \delta S^\alpha(\mathbf{q}) \delta S^{\alpha'}(-\mathbf{q}) \sigma(\beta) \rangle_0}{\langle \sigma(\beta) \rangle_0},\end{aligned}\quad (17)$$

we can express GFs in the  $xyz$  coordinate  $G^{\alpha\alpha'}(\mathbf{q})$  via GFs in the  $XYZ$  coordinate as

$$\begin{aligned}G^{zz}(\mathbf{q}) &= \frac{\Omega^2}{\gamma^2} \Gamma^{XX}(\mathbf{q}) - \frac{h\Omega}{\gamma^2} [\Gamma^{ZX}(\mathbf{q}) + \Gamma^{XZ}(\mathbf{q})] \\ &\quad + \frac{h^2}{\gamma^2} \Gamma^{ZZ}(\mathbf{q}),\end{aligned}\quad (18)$$

$$\begin{aligned}G^{xx}(\mathbf{q}) &= \frac{h^2}{\gamma^2} \Gamma^{XX}(\mathbf{q}) + \frac{h\Omega}{\gamma^2} [\Gamma^{ZX}(\mathbf{q}) + \Gamma^{XZ}(\mathbf{q})] \\ &\quad + \frac{\Omega^2}{\gamma^2} \Gamma^{ZZ}(\mathbf{q}).\end{aligned}\quad (19)$$

The thermodynamic average given in eq. (17) is  $\langle \dots \rangle_0 = \text{Tr}(e^{-\beta H_0} \dots) / \text{Tr}(e^{-\beta H_0})$ .  $\sigma(\beta)$  is the scattering matrix, which can be presented in the functional integral of auxiliary fields (see Ref. 11 and references therein) as

$$\begin{aligned}\sigma(\beta) &= \int D\psi \exp \left\{ -\frac{1}{2\beta} \sum_{\mathbf{q}\alpha\alpha'} I_{\alpha\alpha'}^{-1}(\mathbf{k}) \psi_\alpha(\mathbf{q}) \psi_{\alpha'}(-\mathbf{q}) \right\} \\ &\quad \times \hat{T} \exp \left\{ \sum_{\mathbf{q}\alpha} \psi_\alpha(\mathbf{q}) \delta S^\alpha(\mathbf{q}) \right\}.\end{aligned}\quad (20)$$

$I_{\alpha\alpha'}^{-1}(\mathbf{k})$  is matrix element of the inverse matrix  $\hat{\mathbf{I}}^{-1}(\mathbf{k})$ .

A measure of the functional integration is given by

$$\begin{aligned}\int D\psi \dots &= \prod_\alpha \int_{-\infty}^{+\infty} \frac{d\psi_\alpha(0)}{\sqrt{2\pi \det[\hat{\beta}\hat{\mathbf{I}}(0)]}} \prod'_{\mathbf{q}\neq 0} \int_{-\infty}^{+\infty} \frac{d\psi_\alpha^c(\mathbf{q})}{\sqrt{\pi \det[\hat{\beta}\hat{\mathbf{I}}(\mathbf{k})]}} \\ &\quad \times \int_{-\infty}^{+\infty} \frac{d\psi_\alpha^s(\mathbf{q})}{\sqrt{\pi \det[\hat{\beta}\hat{\mathbf{I}}(\mathbf{k})]}} \dots\end{aligned}\quad (21)$$

$\psi_\alpha^c(\mathbf{q})$  and  $\psi_\alpha^s(\mathbf{q})$  are correspondingly the real and imaginary parts of complex field variable  $\psi_\alpha(\mathbf{q})$ . The prime in the eq. (21) implies that  $\mathbf{q}$  varies in the half-space. The free energy of the MLSF is represented in the functional integral form of the field variables as

$$F = F_0 - \frac{1}{\beta} \ln \int D\psi \exp(-\Phi[\psi]),\quad (22)$$

where the functional  $\Phi[\psi]$  has the following form

$$\begin{aligned}\Phi[\psi] &= \frac{1}{2\beta} \sum_{\mathbf{q}\alpha\alpha'} I_{\alpha\alpha'}^{-1}(\mathbf{k}) \psi_\alpha(\mathbf{q}) \psi_{\alpha'}(-\mathbf{q}) \\ &\quad - \ln \left\langle \hat{T} \exp \left\{ \sum_{\mathbf{q}\alpha} \psi_\alpha(\mathbf{q}) \delta S^\alpha(\mathbf{q}) \right\} \right\rangle_0.\end{aligned}\quad (23)$$

It is ready to obtain the Green functions  $\Gamma^{\alpha\alpha'}(\mathbf{q})$  with the functional average. We have

$$\begin{aligned}\Gamma^{\alpha\alpha'}(\mathbf{q}) &= -[\beta\hat{\mathbf{I}}(\mathbf{k})]_{\alpha\alpha'}^{-1} \\ &\quad + \sum_{\alpha_1\alpha_2} [\beta\hat{\mathbf{I}}(\mathbf{k})]_{\alpha\alpha_1}^{-1} [\beta\hat{\mathbf{I}}(\mathbf{k})]_{\alpha_1\alpha_2}^{-1} \overline{\psi_{\alpha_1}(\mathbf{q}) \psi_{\alpha_2}(-\mathbf{q})},\end{aligned}\quad (24)$$

where the upper bar stands for the functional average

$$\overline{f[\psi]} = \frac{\int D\psi \exp\{-\Phi[\psi]\} f[\psi]}{\int D\psi \exp\{-\Phi[\psi]\}}.\quad (25)$$

In Gaussian approximation, the functional  $\Phi[\psi]$  has quadratic form

$$\Phi_G = \frac{1}{2} \sum_{\mathbf{q}\alpha\alpha'} A_{\alpha\alpha'}^{-1}(\mathbf{q}) \psi_\alpha(\mathbf{q}) \psi_{\alpha'}(-\mathbf{q}).\quad (26)$$

Here  $\hat{\mathbf{A}}^{-1}(\mathbf{q})$  is an inverse matrix of the symmetric matrix  $\hat{\mathbf{A}}(\mathbf{q})$  with its elements

$$A_{\alpha\alpha'}^{-1}(\mathbf{q}) = [\beta\hat{\mathbf{I}}(\mathbf{k})]_{\alpha\alpha'}^{-1} - \delta_{\alpha\alpha'} M_\alpha(\omega),\quad (27)$$

$$\begin{aligned}\hat{\mathbf{M}}(\omega) &= \begin{pmatrix} M_X(\omega) & 0 \\ 0 & M_Z(\omega) \end{pmatrix} \\ &= \begin{pmatrix} b_s(\beta\gamma)/\beta(\gamma - i\omega) & 0 \\ 0 & b'_s(\beta\gamma)\delta_{\omega,0} \end{pmatrix}.\end{aligned}\quad (28)$$

Taking functional integration in eq. (24), one gets

$$\hat{\Gamma} = \hat{\mathbf{M}}(\omega) [\hat{\mathbf{I}} - \beta\hat{\mathbf{I}}(\mathbf{k})\hat{\mathbf{M}}(\omega)]^{-1}.\quad (29)$$

Equation (29) is the solution in the matrix form for the GFs in the  $XYZ$  coordinate system. GFs and susceptibilities of the MLSF in the crystallographic  $xyz$  coordinate system are derived straightforwardly from eqs. (15)–(29).

### 3. Susceptibilities, Elementary Excitations and Discussion

In this part, we apply above theory for calculation of the susceptibilities and elementary excitations in the MLSF taking into account only the exchange interactions between nearest neighbor (NN) spins. Exchange integrals in the NN approximation and other parameters have explicit forms as

$$\begin{aligned}J(\mathbf{k}) &= 2J[\cos(k_x a) + \cos(k_y a)], \\ L(\mathbf{k}) &= 2L[\cos(k_x a) + \cos(k_y a)].\end{aligned}\quad (30)$$

Using eqs. (17)–(30), we obtain the longitudinal and transverse GFs for the MLSF

$$\begin{aligned}\begin{cases} G^{zz}(\mathbf{q}) \\ G^{xx}(\mathbf{q}) \end{cases} &= \begin{cases} \frac{\Omega^2}{\gamma^2} M_X(\omega) + \frac{h^2}{\gamma^2} M_Z(\omega) - \beta L(\mathbf{k}) M_X(\omega) M_Z(\omega) \\ \frac{h^2}{\gamma^2} M_X(\omega) + \frac{\Omega^2}{\gamma^2} M_Z(\omega) - \beta J(\mathbf{k}) M_X(\omega) M_Z(\omega) \end{cases} \\ &\quad \times \frac{1}{1 - \beta [I^{XX}(\mathbf{k}) M_X(\omega) + I^{ZZ}(\mathbf{k}) M_Z(\omega)] + \beta^2 J(\mathbf{k}) L(\mathbf{k}) M_X(\omega) M_Z(\omega)}.\end{aligned}\quad (31)$$

We note that, when  $L(\mathbf{k}) = 0$ , the upper formula in eq. (31) simplifies to the similar result for the longitudinal Green function of the 3D-TIM case obtained in Ref. 12 except for the wave vector  $\mathbf{k}$  defined in the 2D reciprocal space. Using eq. (31), we obtain the expressions for longitudinal and transverse dynamical susceptibilities, which are

$$\left\{ \begin{array}{l} \frac{\chi^{zz}(\mathbf{q})}{\mu^2} \\ \frac{\chi^{xx}(\mathbf{q})}{\mu^2} \end{array} \right\} = \left\{ \begin{array}{l} \frac{\Omega^2}{\gamma^2} b_s(\beta\gamma) + \beta b'_s(\beta\gamma) \left[ \frac{h^2}{\gamma} - L(\mathbf{k}) b_s(\beta\gamma) \right] \delta_{\omega,0} \\ \frac{h^2}{\gamma^2} b_s(\beta\gamma) + \beta b'_s(\beta\gamma) \left[ \frac{\Omega^2}{\gamma} - J(\mathbf{k}) b_s(\beta\gamma) \right] \delta_{\omega,0} \end{array} \right\} \times \frac{1}{\gamma - \frac{[J(\mathbf{k})\Omega^2 + L(\mathbf{k})h^2] b_s(\beta\gamma)}{\gamma^2} - \beta b'_s(\beta\gamma) \left\{ J(\mathbf{k}) \frac{h^2}{\gamma} + L(\mathbf{k}) \left[ \frac{\Omega^2}{\gamma} - J(\mathbf{k}) b_s(\beta\gamma) \right] \right\} \delta_{\omega,0} - i\omega}, \quad (32)$$

where  $b'_s(x)$  is the derivative of the Brillouin function  $b_s(x)$ . The static longitudinal and transverse susceptibilities for the MLSF are defined at zero frequency and at the long wavelength limit ( $\mathbf{q} = 0$  or  $\mathbf{k} = 0$ ,  $\omega = 0$ ) of the eq. (32).

Elementary excitation (or spin wave) energies in the free MLSF are obtained from the poles of the longitudinal and transverse GFs or those of the dynamical susceptibilities by the process of analytic continuation, replacing  $i\omega \rightarrow \omega$ . We note that the denominators of the transverse and longitudinal GFs are identical, thus they have the same poles. The dispersion relation of elementary excitation energies at finite temperature  $T$  is given by

$$\omega(\mathbf{k}) = \gamma - \frac{b_s(\beta\gamma)}{\gamma^2} [J(\mathbf{k})\Omega^2 + L(\mathbf{k})h^2]. \quad (33)$$

The total field value  $\gamma$  in the formula (33), total polarization  $m$  per site and its components are found by solving the MFA equations<sup>8)</sup>

$$\begin{aligned} m_z &= \frac{h}{\gamma} b_s(\beta\gamma), & m_x &= \frac{\Omega}{\gamma} b_s(\beta\gamma), \\ m &= \sqrt{m_z^2 + m_x^2} = b_s(\beta\gamma). \end{aligned} \quad (34)$$

We also examine the case when the external longitudinal field is absent ( $h_0 = 0$ ) for simplicity. Here and after, all energy quantities are measured in unite of the NN exchange strength  $J$ , lengths are measured in unit of the spin lattice constant  $a$ . Dimensionless temperature is  $\tau = k_B T / J$  and the others are marked by upper bar, for example  $\Omega_0 / J = \bar{\Omega}_0$ . We note that, there are two kinds of phase transitions in the present problem: the spin reorientation transition driven by the transverse field and the order-disorder phase transition resulted from competition between total field and temperature. The critical temperature in the TIM widely used in literature up to now<sup>4-8)</sup> is in fact the spin reorientation (SR) temperature  $\tau_R(\bar{\Omega}_0)$ , and defined from condition  $m_z(\tau_R, \bar{\Omega}_0) = 0$  or

$$\frac{\bar{\Omega}_0}{1 - \bar{L}} = 4b_s\left(\frac{\bar{\Omega}_0}{\tau_R(1 - \bar{L})}\right). \quad (35)$$

The order-disorder phase transition temperature  $\tau_C$  should be defined from condition  $m(\tau_C, \bar{\Omega}_0 = 0) = 0$ . The SR temperature depends on the TrF and equals to the order-disorder phase transition temperature, if the transverse field is zero  $\tau_R(\bar{\Omega}_0 = 0) = \tau_C$ . Figure 2 shows the MFA results for the temperature-dependence of the total polarization  $m$ , its components and incline angle  $\theta = \arctg(m_z/m_x)$ . For

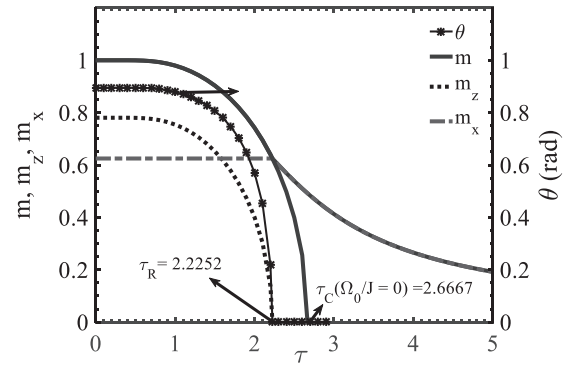


Fig. 2 Dependences of the total polarization  $m$  and its components  $m_z, m_x$ , incline angle  $\theta$  on temperature. Here  $S = 1$ ,  $h_0 = 0$ ,  $\bar{L} = 0.6$  for all curves. Curves  $m_z, m_x$  are plotted for  $\bar{\Omega}_0 = 1.0$ . The  $m$  curve is drawn for  $\bar{\Omega}_0 = 0$ . Values of the spin reorientation temperature  $\tau_R$  and order-disorder phase transition temperature  $\tau_C$  are shown.

$\bar{\Omega}_0 \neq 0$ , total polarization  $m$  tends to zero when temperature increases and  $\tau_C$  is defined approximately by the fastest slope of tangent lines of the  $m(\tau, \bar{\Omega}_0)$  curve. The specific TrF, that makes SR temperature be zero, is the critical transverse field  $\bar{\Omega}_{0C}$  (or the critical tuning parameter in general case).  $\bar{\Omega}_{0C}$  depends on the film thickness.<sup>8)</sup>

In presence of nonzero TrF, temperature dependences of the collective excitation energy in different temperature regions (lower and higher  $\tau_R$ ) are given below i/ for  $\tau \leq \tau_R$

$$\begin{aligned} \bar{\omega}(\mathbf{k}) &= \bar{\gamma}' \left\{ 1 - \frac{1}{2} (\cos k_x + \cos k_y) \left[ \frac{\bar{\Omega}_0^2}{\bar{\gamma}'^2 (1 - \bar{L})} + \bar{L} \right] \right\}, \\ \bar{\gamma}' &= 4b_s\left(\frac{\bar{\gamma}'}{\tau}\right), \end{aligned} \quad (36)$$

ii/ for  $\tau = \tau_R$

$$\bar{\omega}(\mathbf{k})|_{\tau_R} = \frac{\bar{\Omega}_0}{1 - \bar{L}} \left\{ 1 - \frac{1}{2} (\cos k_x + \cos k_y) \right\}, \quad (37)$$

iii/ for  $\tau_R \leq \tau$

$$\begin{aligned} \bar{\omega}(\mathbf{k}) &= \bar{\gamma}' \left\{ 1 - \frac{1}{2\bar{L}} (\cos k_x + \cos k_y) \left( 1 - \frac{\bar{\Omega}_0}{\bar{\gamma}'} \right) \right\}, \\ \bar{\gamma}' &= 4\bar{L}b_s\left(\frac{\bar{\gamma}'}{\tau}\right) + \bar{\Omega}_0. \end{aligned} \quad (38)$$

Figure 3(a) shows the comparison between the energy of the elementary excitations of the XZ-H model ( $\bar{L} \neq 0$ ) and the TIM ( $\bar{L} = 0$ ) when  $k_x = k_y = 0.5$ . We can see that the internal transverse exchange  $\bar{L}$  in the XZ-H model leads to a

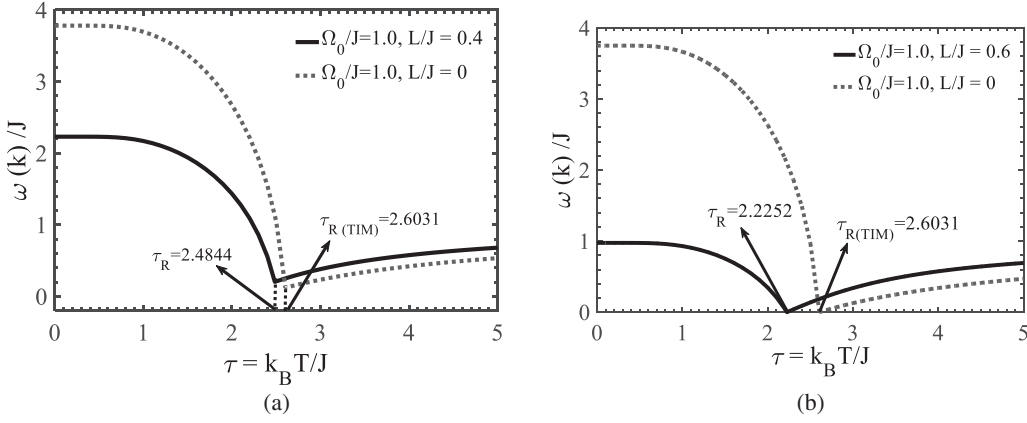


Fig. 3 Dependence of the intensity of the spin wave of the MLSF on temperature:  $k_x = k_y = 0.5$  (a/),  $k_x = k_y = 0$  (b/). The values of the spin and longitudinal field are  $S = 1$ ,  $h_0 = 0$  and the others are given in insets.

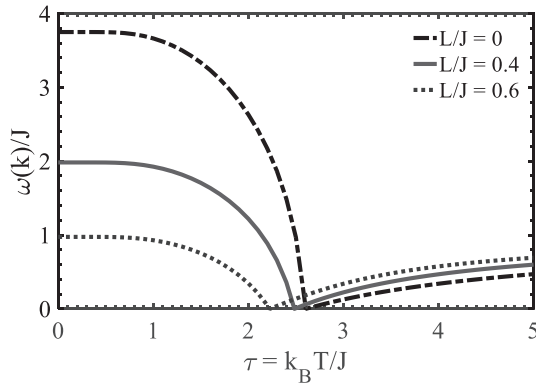


Fig. 4 Influence of the internal transverse exchange  $\bar{L}$  on the SR temperature  $\tau_R$ . Values of the parameters are:  $\bar{\Omega}_0 = 1$ ,  $h_0 = 0$ ,  $S = 1$ ,  $k_x = k_y = 0$ .

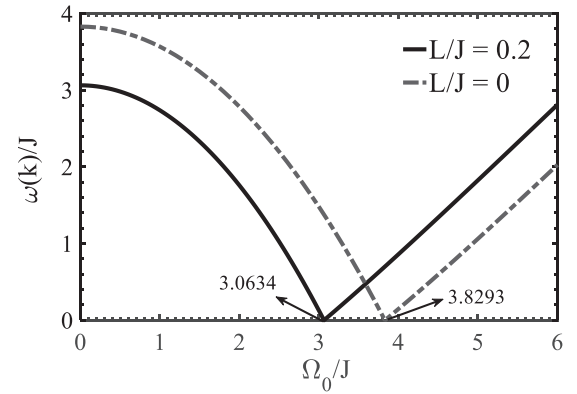


Fig. 5 Dependence of the long wave length spin wave energy ( $\mathbf{k} = 0$ ) on the transverse field. Solid (dashed) curve corresponds to the XZ Heisenberg model (TIM). Values of parameters are  $\tau = 1.2$ ,  $S = 1$ ,  $h_0 = 0$ . Magnitude of the SR field ( $\bar{\Omega}_{0R}$ ) for the XZ-H and TIM models is shown.

decrease of the SR phase transition temperature compared with that of the TIM. For  $\bar{L} < 1$ , the energy of the elementary excitation of the XZ-H model is quite smaller (a little larger) than that of the TIM when  $\tau < \tau_R$  ( $\tau > \tau_R$ ). All the curves in the Fig. 3(a) are similar to the temperature dependence of the soft mode curves in  $\text{PbTiO}_3$  (see Ref. 13). At the SR temperature, the long wave length excitation energy is zero (Fig. 3(b)).

Figure 4 shows reduction tendency of the SR temperature with increase of the internal transverse exchange magnitude  $\bar{L}$ .

We are now interested in the behavior of the elementary excitation at given temperature below Curie temperature  $\tau_C$ , with various TrFs. If we measure the amplitude of the spin wave in ferroic thin film starting from temperature  $\tau \leq \tau_R(0) = \tau_C$  we observe a decrease of the spin wave magnitude with increase of TrF until  $\bar{\Omega}_0 = \bar{\Omega}_{0R}$ .  $\bar{\Omega}_{0R}$  is called the SR field and defined by the condition  $m_z(\tau, \bar{\Omega}_{0R}) = 0$ , or

$$\frac{\bar{\Omega}_{0R}}{1 - \bar{L}} = 4b_s \left( \frac{\bar{\Omega}_{0R}}{(1 - \bar{L})\tau} \right). \quad (39)$$

Obviously, the SR temperature of the MLSF decreases from value  $\tau_R(0) = \tau_C$  to the value  $\tau_R(\bar{\Omega}_{0R}) = \tau$ . Below and above the SR field, the dispersion law of the elementary excitation of the XZ-H model is given by

i/ for  $\bar{\Omega}_0 \leq \bar{\Omega}_{0R}(\tau)$

$$\bar{\omega}(\mathbf{k}) = \bar{\gamma} \left\{ 1 - \frac{1}{2} (\cos k_x + \cos k_y) \left[ \frac{\bar{\Omega}_0^2}{\bar{\gamma}^2 (1 - \bar{L})} + \bar{L} \right] \right\},$$

$$\bar{\gamma} = 4b_s \left( \frac{\bar{\gamma}}{\tau} \right), \quad (40)$$

ii/ for  $\bar{\Omega}_0 = \bar{\Omega}_{0R}(\tau)$

$$\bar{\omega}(\mathbf{k}) = \frac{\bar{\Omega}_{0R}}{1 - \bar{L}} \left[ 1 - \frac{1}{2} (\cos k_x + \cos k_y) \right], \quad (41)$$

iii/ for  $\bar{\Omega}_0 \geq \bar{\Omega}_{0R}(\tau)$

$$\bar{\omega}(\mathbf{k}) = \bar{\gamma}' - 2(\cos k_x + \cos k_y) b_s \left( \frac{\bar{\gamma}'}{\tau} \right),$$

$$\bar{\gamma}' = 4\bar{L} b_s \left( \frac{\bar{\gamma}'}{\tau} \right) + \bar{\Omega}_0. \quad (42)$$

Figure 5 exhibits variation of the intensity of the spin wave energy in the MLSF with an increase of the transverse field  $\bar{\Omega}_0$  at temperature  $\tau = 1.2$ .

In the region  $\bar{\Omega}_0 > \bar{\Omega}_{0R}$ , the spin excitation is called tunneling mode in the bulk TIM model.<sup>14)</sup> According to us, it is better to call it the same spin wave mode. In this TrF region, spin vectors of the MLSF are directed in the film plane. In the Fig. 5, the spin reorientation field  $\bar{\Omega}_{0R}$  for the XZ Heisenberg (TIM) model is 3.0634 (3.8293), respectively.



We see that, the internal transverse exchange essentially reduces the SR transverse field.

The SR in the MLSF studied in the present work with anisotropic the XZ-Heisenberg model, and the field induced SR in monolayer film with isotropic Heisenberg exchange interactions and single ion anisotropy,<sup>9)</sup> have the same origin—the transverse field or the tuning parameter. In other competing interaction cases, role of tuning parameter can be a ratio of competing antiferromagnetic to ferromagnetic interaction in frustrated ferromagnetic films<sup>11)</sup> or this ratio in the theory for the first order magnetization process in polycrystalline perovskites.<sup>15)</sup> The SR point of view developed in this work may be applied to investigate the excitation spectra with the tuning parameter switched on. The SR concept can be applied to explain the “critical temperature-transverse magnetic field” or the “critical temperature-tuning parameter” phase diagrams of transverse Ising magnets given in Ref. 16.

#### 4. Conclusions

Dynamical susceptibility and elementary excitations in ferroic (magnetic or ferroelectric) single layer spin film was calculated using the anisotropic exchange XZ Heisenberg model in transverse and longitudinal fields. It is shown that the transverse internal exchange interaction reduces (enhances) the intensity of the collective excitation below (above) spin reorientation temperature. The “spin reorientation temperature-transverse field” phase diagram in the XZ Heisenberg model and the TIM is “critical temperature-tuning parameter” diagram. The external spin reorientation field is decreased by intrinsic transverse exchange interaction.

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