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Yuriy Sizyuk, Natalia B. Perkins, and Peter Wölfle Phys. Rev. B **92**, 155131 — Published 19 October 2015 DOI: 10.1103/PhysRevB.92.155131

Lifting mean-field degeneracies in anisotropic spin systems

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In this work, we propose a method for calculating the free energy of anistropic classical spin systems. We use a Hubbard-Stratonovich transformation to express the partition function of a generic bilinear super-exchange Hamiltonian in terms of a functional integral over classical timeindependent fields. As an example we consider an anisotropic spin-exchange Hamiltonian on the cubic lattice as is found for compounds with strongly correlated electrons in multi-orbital bands and subject to strong spin-orbit interaction. We calculate the contribution of Gaussian spin fluctuations to the free energy. While the mean field solution of ordered states for such systems usually has full rotational symmetry, we show here that the fluctuations lead to a pinning of the spontaneous magnetization along some preferred direction of the lattice.

I. INTRODUCTION

Recent research activities on transition metal oxides suggest that the interplay of the strong spin-orbit coupling (SOC), crystal field (CF) interactions and electron correlations may lead to compass-like anisotropic interactions between magnetic degrees of freedom.¹ These anisotropic interactions have a generic form $J_{ij}^{\alpha}S_i^{\alpha}S_j^{\alpha}$ in which α depends on the direction of the particular link or bond and S denotes spin or pseudospin describing magnetic or orbital degrees of freedom.

The models in which compass-like anisotropies are dominating, or also the pure compass models, have been known for a long time. These models appear naturally in strongly correlated electron systems as minimal models to account for interactions between pseudospins describing orbital degrees of freedom.^{2–7} The compass-like anisotropies also arise as interactions between magnetic degrees of freedom in systems with strong SOC which might be realized in 4d and 5d transition metal oxides.⁸ However, in these systems, due to the extended nature of 4d and 5d orbitals, the compass interactions are always accompanied by the usual SU(2) symmetric Heisenbergtype exchange. These models are especially interesting because while the pure compass-like models are rare, the combined Heisenberg-compass models have been shown to be minimal models describing the magnetic properties of various materials. A review of the different realizations of compass models,^{2–21} their physical motivations, symmetries, unconventional orderings and excitations may be found in the recent paper by Nussinov and van den Brink.¹

One of the common features induced by compass-like anisotropies is frustration, arising from a competition of interactions along different directions and leading to the macroscopic degeneracy of the classical ground state and in addition to rich quantum behavior. In many cases, the pure compass models do not show conventional magnetic ordering because the degeneracy of the classical ground state is connected to discrete sliding symmetries of the model.^{4,9} Because these symmetries are intrinsic symmetries of the model, they can not be lifted by the order-by disorder mechanisms. Instead, the direct consequence of the existence of these symmetries is that the natural order parameters for pure compass models are nematic, which are invariant under discrete sliding symmetries.

The nematic order present in the compass model is fragile and is easily destroyed by the presence of the isotropic Heisenberg interaction which breaks some of the intrinsic symmetries of the model. In Heisenbergcompass models, some of the degeneracies become accidental. In these models, the true magnetic order might be selected by fluctuations via an order by disorder mechanism, removing accidental degeneracies and determining both the nature and the direction of the order parameter. Despite the simplicity of these models, the interplay of the Heisenberg and compass interaction leads to very rich phase diagrams even in the simplest case of the square lattice.¹⁵ For classical systems this mechanism requires finite temperatures, where entropic contributions of fluctuations to the free energy become effective.

In this work, we will be interested in studying the directional ordering transitions in the Heisenberg-compass model on the cubic lattice.¹⁶ From a historical perspective, the three-dimensional 90° compass model was the first model of this kind proposed by Kugel and Khomskii² in the context of the ordering of the t_{2q} orbitals in transition metal oxides with perovskite structure and then studied in more details by Khaliullin¹⁶ in application to $LaTiO_3$. The formal procedure which we will be using here is based on the derivation of the fluctuational part of the free energy by integrating out the Gaussian fluctuations, and determining which orientations of the vector order parameter correspond to the free energy minimum. To do so, we first express the partition function as a functional integral over classical fields. In this first paper we consider classical spins at finite temperature. Our starting point in evaluating this exact representation of the partition function is the mean-field solution, which usually does not reflect the anisotropic character of the interaction referring to the crystal lattice axes. As a next step, we evaluate the contribution of Gaussian fluctuations to the free energy of the mean field ordered state. The latter

carries the information embodied in the anisotropic spin interaction and therefore allows to define preferred directions of the spin order with respect to the lattice. We will not go beyond the simple evaluation of the contribution of fluctuations, e.g. by incorporating the fluctuation contribution self-consistently.

For simplicity, we choose the parameters of the model such that the ground state is ferromagnetic, i.e. we consider the Heisenberg interaction to be ferromagnetic and allow the compass interaction to be both ferromagnetic and antiferromagnetic. Our analysis shows that the profile of the fluctuation part of the free energy exhibits significant changes when the compass interactions become antiferromagnetic and exceed some critical value. For any ferromagnetic and weak antiferromagnetic compass interactions, the minima of the fluctuational part of the free energy are attained if the spontaneous magnetization vector points along one of the cubic axes. Once the antiferromagnetic compass interactions become strong, the minima of the free energy and thus the possible directions of the magnetization shift to one of the cubic body diagonals. Interestingly, this transition happens smoothly through an intermediate phase in which the locations of the minima slide along the unit sphere in a very peculiar way. As the compass interaction becomes more antiferromagnetic in the intermediate phase, the maxima and minima interchange in a symmetric manner. In order to do that, they continuously split and slide around each other. We found the intermediate phase to exist when the ratio of the compass to Heisenberg interactions is roughly -1.2 < K/J < -1.45. However, since the process is very smooth, it is difficult to determine the exact boundaries of the intermediate phase.

This paper is organized as follows. In section II we introduce the functional integral representation of the partition function for the spin systems with interactions described by the most general bilinear form of the superexchange Hamiltonian. The details of the method are outlined in the Appendix. In section III, we apply this framework to compute the angular dependence of the fluctuational part of the free energy for the ferromagnetic Heisenberg-compass model on the cubic lattice. Our results are presented and discussed in Section IV.

II. REPRESENTATION OF THE PARTITION FUNCTION

We consider a system of identical classical spins \mathbf{S} on a lattice, interacting in an anisotropic fashion as indicated in the introduction, defined by the Hamiltonian

$$H = \frac{1}{2} \sum_{j,j'} \sum_{\alpha\alpha'} J_{j,j'}^{\alpha,\alpha'} S_j^{\alpha} S_{j'}^{\alpha'}$$
(1)

where j, j' label the lattice sites, $\alpha, \alpha' = x, y, z$ label the three components of the spin and $\mathbf{S}^2 = 1$. For the models with compass-like anisotropic and Heisenberg isotropic

We will be interested in the long-range ordered phases of the system. The mean field approximation of the order parameter usually leads to a highly degenerate manifold of states, e.g. a ferromagnetic state with spontaneous magnetization pointing in any direction. This degeneracy is lifted by the anisotropic components of the spin interaction, but only at the level of the fluctuation contribution to the free energy (action) S_{fl} . In the following we outline a method allowing to calculate S_{fl} , which is based on the Hubbard-Stratonovich transformation of the partition function for spin systems described by the generic Hamiltonian (1). We present details and discuss justifications for this method in the Appendix.

The partition function of the system is given by the integral over the Boltzmann weights of configurations

$$Z = \int [dS_j] \exp[-\beta \sum_{j\alpha,j'\alpha'} J_{jj'}^{\alpha\alpha'} S_j^{\alpha} S_{j'}^{\alpha'}] \delta(\mathbf{S}_j^2 - 1), \quad (2)$$

where $\beta = 1/k_B T$ is the inverse temperature, S_j^{α} are the components of the spin operator at site j.

It is useful to represent the Hamiltonian in the basis of the eigenfunctions $\chi_{n;j,\alpha}$ of the spin exchange matrix, defined by

$$\sum_{j',\alpha'} J_{jj'}^{\alpha\alpha'} \chi_{n;j',\alpha'} = \kappa_n \chi_{n;j,\alpha} \,.$$

For spins on a periodic lattice these eigenstates are labeled by wavevector \mathbf{q} (inside the first Brillouin zone) and index ν , characterizing three principle axes of the matrix \hat{J} . Thus, $|n\rangle = |\mathbf{q}, \nu\rangle$ and the normalized eigenfunctions take the form

$$\chi_{\mathbf{q},\nu;j,\alpha} = \frac{1}{\sqrt{N}} e^{i\mathbf{q}\cdot\mathbf{R}_j} u_{\nu,\alpha} \,,$$

where N is the number of lattice sites, the $u_{\nu,\alpha}$ are orthonormal real-valued eigenvectors, i.e. $\sum_{\alpha} u_{\nu,\alpha} u_{\nu',\alpha} = \delta_{\nu\nu'}$ and $\kappa_{\mathbf{q},\nu}$ are the eigenvalues of the spin exchange interaction matrix.

We now define the normal amplitudes of the spins as

$$S_{\mathbf{q},\nu} = \sum_{j,\alpha} \chi_{\mathbf{q},\nu;j,\alpha} S_j^{\alpha}$$

and express the Hamiltonian as

$$H = \sum_{\mathbf{q},\nu} \kappa_{\mathbf{q},\nu} S_{\mathbf{q},\nu}^* S_{\mathbf{q},\nu} , \qquad (3)$$

where $S^*_{\mathbf{q},\nu} = S_{-\mathbf{q},\nu}$. Commutation of classical spins allows us to employ a Hubbard-Stratonovich transformation in terms of classical fields $\varphi_{\mathbf{q},\nu}$ in order to represent

the interaction operator as a Zeeman energy operator of spins in a spatially varying magnetic field. As a result one finds the following representation of the partition function

$$Z = \int [d\varphi] \exp[-\beta \{ \sum_{\mathbf{q},\nu} |\kappa_{\mathbf{q},\nu}|^{-1} \varphi^*_{\mathbf{q},\nu} \varphi_{\mathbf{q},\nu} - \mathcal{S}_{loc}(\{\varphi^*_{\mathbf{q},\nu},\varphi_{\mathbf{q},\nu}\})$$
(4)

where the integration volume element is given by

$$[d\varphi] = \Pi_{\mathbf{q},\nu} \frac{i\beta d\varphi_{\mathbf{q},\nu}^* d\varphi_{\mathbf{q},\nu}}{2\pi |\kappa_{\mathbf{q},\nu}|} \,.$$

The contribution to the action in the case of classical spins is given by

$$S_{loc}(\{\phi_{\mathbf{q},\nu}^*,\phi_{\mathbf{q},\nu}\}) = \beta^{-1} \sum_j \ln[\sinh(2\beta\varphi_j)/2\beta\varphi_j] , \quad (5)$$

where $\varphi_j^2 = (\varphi_j^x)^2 + (\varphi_j^y)^2 + (\varphi_j^z)^2$, with $\varphi_j^\alpha \equiv \sum_{\mathbf{q},\nu} s(\kappa_{\mathbf{q},\nu}) \varphi_{\mathbf{q},\nu} \chi_{\mathbf{q},\nu;j,\alpha}^*$ and $s(\kappa_{\mathbf{q},\nu}) = 1$ for $\kappa_{\mathbf{q},\nu} < 0$ and $s(\kappa_{\mathbf{q},\nu}) = i$ for $\kappa_{\mathbf{q},\nu} > 0$. The Hubbard-Stratonovich identity used to derive the above functional integral is different for eigenmodes $\varphi_{\mathbf{q},\nu}$ with positive or negative eigenvalue $\kappa_{\mathbf{q},\nu}$, leading to the appearance of a complexvalued φ_j . The details of evaluating $\mathcal{S}_{loc}(\{\varphi_{\mathbf{q},\nu}^*,\varphi_{\mathbf{q},\nu}\})$ can be found in the Appendix.

III. APPLICATION TO THE CUBIC LATTICE

A. Isotropic Heisenberg interaction

In order to demonstrate how to perform the evaluation of the above representation of the partition function, we consider first the isotropic ferromagnetic Heisenberg model with nearest neighbor interactions on the cubic lattice. In this case, the Hamiltonian (1) reads

$$H = J \sum_{\langle j;j' \rangle} \sum_{\alpha} S_j^{\alpha} S_{j'}^{\alpha}, \qquad (6)$$

where the lattice summation is over nearest neighbors $\langle j, j' \rangle$ -bonds and J < 0. For the isotropic exchange interaction, the eigenvalues, $\kappa_{\mathbf{q},\nu} = J \sum_{\alpha} \cos q_{\alpha}$, are independent of ν , $\kappa_{\mathbf{q},\nu} = \kappa_{\mathbf{q}}$, and hence are degenerate.

A uniform ferromagnetic mean field solution is found by solving the saddle point equation

$$\frac{\partial}{\partial \varphi_{MF}} \mathcal{S} = -\frac{\partial}{\partial \varphi_{MF}} N[|\kappa_{\mathbf{q}=\mathbf{0}}|^{-1} (\varphi_{MF})^2 \qquad (7)$$
$$-\beta^{-1} \ln[\sinh(2\beta \varphi^{MF})/2\beta \varphi_{MF}]] = 0,$$

where we used $\varphi_{\mathbf{q}\nu}^{MF} = \sqrt{N}\varphi_{MF}\delta_{\mathbf{q},0}m_{0,\nu}, \ \varphi_j = \varphi_{MF}, m_{0,\nu}$ for the components of the unit vector along the magnetization in the reference frame defined by the principal axes of the interaction matrix (which are the cubic axes in this case), and N is the number of lattice sites. The

solution of the Eq.(7) gives us a non-linear equation for the mean-field parameter:

$$2|\kappa_{\mathbf{q}=\mathbf{0}}|^{-1}\varphi_{MF} - 2\coth(2\beta\varphi_{MF}) + \frac{1}{\beta\varphi_{MF}} = 0 \quad (8)$$

We solve this equation numerically at each temperature and get $\varphi_{MF}(T)$. Linearizing the equation (8) near the transition, we find the transition temperature $T_c = \beta_c^{-1} = 2|\kappa_{\mathbf{q}=\mathbf{0}}|/3$. We note in passing that a different length of the classical spin vector $|\mathbf{S}| = S_0$ may be simply scaled back to unit length by changing the temperature as $T' = S_0^2 T$. Choosing $S_0^2 = 3/4$ appropriate for quantum spin S = 1/2 we find the renormalized transition temperature $T'_c = |\kappa_{\mathbf{q}=\mathbf{0}}|/2$, which agrees with the quantum mean field transition temperature.

The fluctuation contribution is obtained by expanding the action in the fluctuation field $\delta \varphi_{\mathbf{q},\nu} = \varphi_{\mathbf{q},\nu} - \varphi_{\mathbf{q}\nu}^{MF}$ about the mean field solution to lowest order:

$$S = S_0 + S_{\rm fl}$$
(9)
$$S_0 = |\kappa_{\mathbf{q}=\mathbf{0}}|^{-1} \varphi_{MF}^2 - NT \ln[\sinh(2\beta\varphi_{MF})/2\beta\varphi_{MF}]$$

For Gaussian fluctuations, the fluctuation part of the free energy, or equivalently the action, $S_{\rm fl}$, is a bilinear function of $\delta \varphi_{\mathbf{q},\nu}$. It is given by

$$S_{\rm fl}\{\delta\varphi_{\mathbf{q},\nu}\} = \sum_{\mathbf{q};\nu,\nu'} A_{\mathbf{q},\nu\nu'}\delta\varphi_{\mathbf{q},\nu}^*\delta\varphi_{\mathbf{q},\nu'} \qquad (10)$$

where we defined matrices $A_{\mathbf{q},\nu\nu'}$ describing the weight of Gaussian fluctuations of wavevector \mathbf{q} and polarization ν as

$$A_{\mathbf{q}\nu\nu'} = |\kappa_{\mathbf{q}\nu}|^{-1} \delta_{\nu,\nu'}$$

$$-\frac{2}{3} [\beta_c (\delta_{\nu,\nu'} - m_{0,\nu} m_{0,\nu'}) + 3\beta r m_{0,\nu} m_{0,\nu'}] s(\kappa_{\mathbf{q}\nu}) s(\kappa_{\mathbf{q}\nu'})$$
(11)

Here, for shortness we introduced $r = 1/(2\beta\varphi_{MF})^2 - 1/\sinh^2(2\beta\varphi_{MF})$.

In the limit of small \mathbf{q} , it is instructive to separate the fluctuations into longitudinal (along \mathbf{m}_0) and transverse (perpendicular to \mathbf{m}_0) components, $\delta \varphi_{\mathbf{q}}^{l} = \mathbf{m}_0 \cdot \delta \varphi_{\mathbf{q}}$ and $\delta \varphi_{\mathbf{q}}^{tr} = \sum_{\mu=1,2} \mathbf{m}_{\mu} \varphi_{\mathbf{q},\mu}^{tr}$, respectively. We defined $\delta \varphi_{\mathbf{q},\mu}^{tr} = \mathbf{m}_{\mu} \cdot \delta \varphi_{\mathbf{q}}$, with $\mathbf{m}_1 = (\mathbf{m}_0 \times \mathbf{z})/|\sin\theta|$ and $\mathbf{m}_2 = \mathbf{m}_1 \times \mathbf{m}_0$, where $\cos \theta = \mathbf{m}_0 \cdot \mathbf{z}$. Despite the complex nature of fluctuational fields, their separation into transverse and longitudinal is possible in the limit of small \mathbf{q} , because the interaction eigenvalues $\kappa_{\mathbf{q}\nu} < 0$ and thus, $s(\kappa_{\mathbf{q}\nu}) = 1$ in this region of the BZ for any polarization component ν . Then, the longitudinal fluctuations contribute to the free energy as

$$S_{\rm fl,l} = \sum_{\boldsymbol{q}} [|\kappa_{\boldsymbol{q}}|^{-1} - 2\beta r] (\boldsymbol{m}_0 \cdot \delta \boldsymbol{\varphi}_{\boldsymbol{q}}^{\rm l}) (\mathbf{m}_0 \cdot \delta \boldsymbol{\varphi}_{-\boldsymbol{q}}^{\rm l}) \qquad (12)$$

The transverse fluctuations are gapless in agreement with Goldstone's theorem:

$$S_{\rm fl,tr} = \sum_{\boldsymbol{q},\kappa_{\mathbf{q},\nu}<0} [|\kappa_{\mathbf{q}}|^{-1} - \frac{2}{3}\beta_c] (\delta \boldsymbol{\varphi}_{\mathbf{q}}^{\rm tr} \cdot \delta \boldsymbol{\varphi}_{-\mathbf{q}}^{\rm tr})$$
(13)

since $\lim_{\mathbf{q}\to\mathbf{0}}[|\kappa_{\mathbf{q}}|^{-1} - \frac{2}{3}\beta_c] = 0$



FIG. 1: (Colors online) The magnitude of the action $S_{\rm fl}(\theta, \phi)$ defined by Eq. (18) is plotted on the surface of the unit sphere. The preferred directions of the magnetization, corresponding to the minima of the free energy, are shown by deep blue color. The energy scale is shown in units of J. (a) J = -1 and K = 0.75: the preferred directions of the magnetization are along the cubic axes. (b) J = -1 and K = 1.5: the preferred directions of the magnetization.

B. Fluctuations due to anisotropic compass interactions

Next, in addition to the isotropic Heisenberg term, let us take into consideration an anisotropic compass interaction, K. The constraint that the ferromagnetic mean field solution remains stable is satisfied for all negative (ferromagnetic) values of K and for positive values K < 3|J|.

In the presence of the anisotropic compass interaction, the model (1) reads

$$H = \sum_{j;j'} \sum_{\alpha} J^{\alpha}_{jj'} S^{\alpha}_j S^{\alpha}_{j'}$$
(14)

where the exchange interaction is given by

$$J_{jj'}^{\alpha} = \frac{1}{2} \delta_{j',j+\tau} [J + K \delta_{\alpha,|\tau|}]$$
(15)

The index $\tau = \pm x, \pm y, \pm z$ labels nearest neighbor sites, where $|\tau|$ denotes a direction in spin space. The eigenvalues of the operator $J^{\alpha \alpha'}_{jj'}$ defined in the previous section are given by

$$\kappa_{\mathbf{q}\nu} = \sum_{\alpha} (J + K\delta_{\alpha,\nu}) \cos q_{\alpha} \tag{16}$$

The eigenvectors \mathbf{u}_{ν} are again along the three cubic axes, such that the components are $u_{\nu\alpha} = \delta_{\nu\alpha}$. This time the three eigenvalues for given \mathbf{q} are not degenerate (except in the limit $\mathbf{q} \to 0$) and the fluctuation contribution to the free energy will therefore depend on the orientation of the spontaneous magnetization. We may again use the representation of the partition function Z as a functional integral over the Fourier components $\varphi_{\mathbf{q}\nu}$ of the auxilliary field.

Provided J < 0 and K < 3|J| the mean field solution φ_{MF} is given as before by solving the transcendental equation (8) numerically. The fluctuation contribution to the free energy is obtained by expanding the action in the fluctuation field about the mean field solution to lowest order. We get

$$Z = C \exp(-\beta S_0) \int [d\delta \varphi] \exp(-\beta S_{\rm fl} \{\delta \varphi_{\mathbf{q},\nu}\}), \quad (17)$$

where the fluctuation part of the action is given by Eq.(10) and Eq.(11) In the following, we show that by comparison to the isotropic model, Eq. (17) manifestly breaks rotational invariance, which results in a selection of preferred directions of the order parameter which minimize the free energy.

The 3 × 3-matrix $A_{\mathbf{q},\nu\nu'}$ may be diagonalized and has eigenvalues $\lambda_{\gamma,\boldsymbol{q}}$ and eigenvectors $\boldsymbol{v}_{\gamma,\boldsymbol{q}}$, $\gamma = 0, 1, 2$. This allows us to express $\sum_{\nu\nu'} A_{\mathbf{q},\nu\nu'} \delta\varphi_{\mathbf{q},\nu}^* \delta\varphi_{\mathbf{q},\nu'} = \sum_{\gamma} \lambda_{\gamma,\boldsymbol{q}} \delta\varphi_{\boldsymbol{q},\gamma} \delta\varphi_{-\boldsymbol{q},\gamma}$, where $\delta\varphi_{\boldsymbol{q},\gamma} = \boldsymbol{v}_{\gamma,\boldsymbol{q}} \cdot \delta\varphi_{\boldsymbol{q}}$. The integration over the fluctuation amplitudes may now be performed and gives

$$S_{\rm fl} = \beta^{-1} \frac{1}{2} \sum_{\mathbf{q},\rho} \{ \ln |\lambda_{0,\mathbf{q}}\lambda_{1,\mathbf{q}}\lambda_{2,\mathbf{q}}| \}$$
(18)

where we chose $s(\kappa_{\mathbf{q}\nu}) = \pm i$ for $\kappa_{\mathbf{q}\nu}$ following the procedure described at the end of the appendix. Alternatively, we may use that $|\lambda_{0,q}\lambda_{1,q}\lambda_{2,q}| = |\det\{A_{\mathbf{q},\nu\nu'}\}|$, saving the trouble of having to determine the eigenstates of $A_{\mathbf{q},\nu\nu'}$.

Let us now derive the explicit expression for the fluctuation contribution for an arbitrary orientation of $\mathbf{m}_0 = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$. Inserting this into the definition of $A_{\mathbf{q},\nu\nu'}$ given by Eq.(11), we find its elements to be

$$\begin{aligned} A_{q,00} &= |\kappa_{q,x}|^{-1} - \frac{2}{3}s(\kappa_{q,x})s(\kappa_{q,x})(\beta_{c}(1 - s_{\theta}^{2}c_{\phi}^{2}) + 3\beta r s_{\theta}^{2}c_{\phi}^{2}) \\ A_{q,01} &= -\frac{2}{3}s(\kappa_{q,x})s(\kappa_{q,y})(3\beta r - \beta_{c})c_{\phi}s_{\phi}s_{\theta}^{2} \\ A_{q,10} &= A_{q,01} \\ A_{q,02} &= -\frac{2}{3}s(\kappa_{q,x})s(\kappa_{q,z})(3\beta r - \beta_{c})c_{\phi}c_{\theta}s_{\theta} \\ A_{q,20} &= A_{q,02} \\ A_{q,11} &= |\kappa_{q,y}|^{-1} - \frac{2}{3}s(\kappa_{q,y})s(\kappa_{q,y})(\beta_{c}(1 - s_{\theta}^{2}s_{\phi}^{2}) + 3\beta r s_{\theta}^{2}s_{\phi}^{2}) \\ A_{q,12} &= -\frac{2}{3}s(\kappa_{q,y})s(\kappa_{q,z})(3\beta r - \beta_{c})s_{\phi}c_{\theta}s_{\theta} \\ A_{q,21} &= A_{q,12} \\ A_{q,22} &= |\kappa_{q,z}|^{-1} - \frac{2}{3}s(\kappa_{q,z})s(\kappa_{q,z})(\beta_{c}s_{\theta}^{2} + 3\beta r c_{\theta}^{2}), \end{aligned}$$
(19)



FIG. 2: (Colors online) The magnitude of the action $S_{\rm fl}(\theta, \phi)$ defined by Eq.(18) computed in the intermediate phase. The energy scale is shown in units of J. (a) J = -1 and K = 1.25. The free energy shows the early stages of the splitting of each maximum along one of the cubic diagonals into three maxima and the splitting of the each minimum along one of the cubic directions into four minima. (b) J = -1 and K = 1.3. The full splitting of each maximum into three and each minimum into four minima. The maxima are moving towards cubic face diagonals and the minima are moving towards cubic body diagonals. (c) J = -1 and K = 1.4. The minima of the free energy reach cubic diagonal directions. The maxima of the free energy along [1,1,0], [1,0,1], [0,1,1] are splitting into two and going towards cubic directions.



FIG. 3: (Colors online) The finite temperature phase diagram of the model (14) restricted to K < 3|J|. The red dashed line shows the borders of the intermediate phase. The phases on the left and on the right from the intermediate phase have the magnetizations pointing along cubic axes and cubic body diagonals, respectively. Both the anisotropic interaction K and the temperature T are measured in the units of J.

where, to shorten notations, we denote $\sin \theta(\phi) \equiv s_{\theta(\phi)}$ and $\cos \theta(\phi) \equiv c_{\theta(\phi)}$. The interactions are defined as $\kappa_{q,x}^{-1} = 1/((J+K)\cos q_x + J\cos q_y + J\cos q_z), \ \kappa_{q,y}^{-1} = 1/((J+K)\cos q_y + J\cos q_x + J\cos q_z)$ and $\kappa_{q,z}^{-1} = 1/((J+K)\cos q_z + J\cos q_x + J\cos q_y)$. We see that the matrix $A_{\alpha\alpha'}$ has a rather complex structure as a function of qand angles θ and ϕ . This gives rise to a complex behavior of the eigenvalues $\lambda_{0,q}, \ \lambda_{1,q}$ and $\lambda_{2,q}$.

IV. RESULTS AND DISCUSSIONS

We now present the results we obtained for $S_{\rm fl}(\theta, \phi)$ by performing numerical integration in Eq.(18). The angular dependencies of $S_{\rm fl}(\theta, \phi)$ for various values of Kare presented in Fig.1 and Fig.2, where the magnitude of $S_{\rm fl}(\theta, \phi)$ as a function of orientation of the spontaneous magnetization is shown as a color-coded plot on the unit sphere. The calculations for all the plots in Fig.1 and Fig.2 are performed at temperature $\beta = \beta_c + 1$ and assuming J = -1. We see that $S_{\rm fl}(\theta, \phi)$ has a non-trivial dependence on the direction of the order parameter defined by angles θ and ϕ which is modified when we change the parameters of the model. This peculiar angular dependence of $S_{\rm fl}(\theta, \phi)$ is inherited from non-trivial angular dependencies of $\lambda_{0,q}$, $\lambda_{1,q}$ and $\lambda_{2,q}$.

In Fig.1 (a), we present the profile of $S_{\rm fl}(\theta, \phi)$ computed for K = 0.75. We can see, that $S_{\rm fl}(\theta, \phi)$ is minimized when the magnetization is directed along one of the cubic axes. We note that the cubic directions are also selected for other values of ferromagnetic compass interactions (K < 0) and up to the limit of the pure ferromagnetic compass model. In Fig.1 (b), we increased the compass interaction to be equal to K = 1.5. We see that the minima of $S_{\rm fl}(\theta, \phi)$ are achieved when the magnetization is directed along one of the [1,1,1] axes, i.e. along the cubic diagonals, indicating that a rotation of the order parameter takes place as a function of K.

Let us understand how the transition between these two ferromagnetic phases with cubic easy axes and easy axes along cubic diagonals takes place. In Fig. 3, we presented the finite temperature phase diagram of the model (14). The ferromagnetic order is stable for K < 3|J|. The mean field transition temperature is shown by a blue line. At small K, the magnetization points along cubic axes. At large K, the magnetization points along cubic diagonals. These two phases are separated by a phase in which the magnetization points along some intermediate direction. The borders of the intermediate phase are shown by red dashed lines. The characteristic profiles of the fluctuation free energy in the intermediate phase are shown in Fig. 2(a)-(c). Fig.2(a) shows $S_{\rm fl}(\theta, \phi)$ computed for K = 1.25. The free energy shows the early stage of the splitting of each maximum along one of the cubic diagonals into three maxima and the splitting of each minimum along one of the cubic directions into four minima. For example, the maximum along [1,1,1] direction continuously splits into three maxima which slide towards the cubic face diagonals [1,1,0], [1,0,1], and [0,1,1]. At the same time the minima at cubic directions slide toward cubic diagonals. For example, the minimum at the [1,0,0] direction splits into four minima, which slide to replace the maxima along [1,1,1], [1,-1,1], [1,1,-1], [1,-1,-1]directions. At Fig.2(b) we slightly increased the value of the compass interaction and set it equal to K = 1.3. Here we see the full splitting of each maximum into three and each minimum into four minima. The maxima are moved towards cubic face diagonals and the minima are moved towards cubic diagonals. In Fig. 2(c) we set K = 1.4, which corresponds to the final stage of the deformation of the free energy profile before it has the structure shown in Fig.1(b). Here we see that the maxima of the free energy along [1,1,0], [1,0,1], [0,1,1] are splitting into two and are going towards the cubic axes directions but do not yet reach it. So in this way the maxima and minima slide around each other to replace each other as we change the parameters.

V. CONCLUSION

The magnetic properties of heavy transition metal oxides such as iridates and others are emerging as a new fascinating field offering opportunities to realize strongly frustrated quantum spin systems in the laboratory. In these systems the combination of multi-band electronic structure and strong Coulomb and Hund's couplings with strong spin-orbit interaction can give rise to extremely anisotropic spin exchange interactions of the compass type. Mean field solutions of these models are often untouched by the anisotropies of the model and show the full isotropy of pure Heisenberg models, in contrast with experimental observation. In this paper we addressed the question how the system selects special preferred directions of the mean field order parameter vector. We restricted ourselves to the case of a ferromagnetic order parameter, but the analogous question exists for antiferromagnetic or more complicated ordered structures. We find that the high degeneracy of the ferromagnetic mean field solution is lifted by the free energy contribution from thermal fluctuations. We calculated the fluctuation contribution for a Heisenberg-compass model of classical spins on a three dimensional cubic lattice with nearest neighbor interactions - an isotropic Heisenberg coupling J < 0 (which we take as the energy unit), and a compass coupling K. The ferromagnetic state is found if K < 3|J|. Rather than exploring the full phase diagram, we focused on one typical temperature $T = T_c/(1+T_c)$ where T_c is the mean field transition temperature and show a qualitative phase diagram in Fig.3. For values of K < 1.2 the system is found to choose preferred directions of the spontaneous magnetization along one of the cubic axes. For 1.5|J| < K < 3|J| the preferred directions are found to be along the space diagonals. The two domains are separated by a region in which the minima and maxima of the free energy split into four and three, respectively, and perform an interesting dance around each other. In this intermediate phase one thus has not just six equilibrium orientations, but twenty four. Exactly how these transitions happen, in particular as a function of temperature, will be the subject of future work. The thermodynamic properties of these intermediate phases at elevated temperature, when thermally activated transitions between different orientations of finite domains may occur is another field to be explored. In the temperature regime considered here we expect the classical approximation to be valid. A generalization to quantum spin systems of the approach presented here is in preparation.

Acknowledgements. We thank Michel Gingras, George Jackeli, Yoshi Kamiya, Alberto Hinojosa-Alvarado and Ioannis Rousochatzakis for useful discussions. N.P. and Y.S. acknowledge the support from NSF Grants DMR-1005932 and DMR-1511768. P.W. thanks the Department of Physics at the University of Wisconsin-Madison for hospitality during several stays as a visiting professor. P.W. also acknowledges partial support by an ICAM senior fellowship and through the DFG research unit "Quantum phase transitions". N.P. acknowledges the hospitality of KITP and partial support by the National Science Foundation under Grant No. NSF PHY11-25915.

Appendix A: Hubbard-Stratonovich transformation of the partition function for spin systems

1. General formulation

The Hubbard-Stratonovich (H-S) transformation is based on the mathematical identity

$$\exp[-ax^{2}] = \frac{1}{\sqrt{\pi|a|}} \int dy \exp[-\frac{y^{2}}{|a|} + 2s(a)xy)] \quad (A1)$$

where we defined

$$s(a) = \begin{cases} 1, & \text{if } a < 0\\ i, & \text{if } a > 0 \end{cases}$$
(A2)

For a > 0 we may as well use s(a) = -i. We will later make use of this ambiguity when we evaluate the *y*-integrals approximately, which may lead to imaginaryvalued contributions.

In the above H-S-transformation x may be a number or an operator. In the case it is an operator, we use the eigenfunctions $|n\rangle$ of \hat{x} defined by

$$\widehat{x}|n\rangle = x_n|n\rangle$$

to prove that

$$\exp[-a\widehat{x}^{2}]|n\rangle = \exp[-ax_{n}^{2}]|n\rangle$$
(A3)
$$= \frac{1}{\sqrt{\pi|a|}} \int dy \exp[-\frac{y^{2}}{|a|} + 2s(a)x_{n}y)]|n\rangle.$$
$$= \frac{1}{\sqrt{\pi|a|}} \int dy \exp[-\frac{y^{2}}{|a|} + 2s(a)\widehat{x}y)]|n\rangle$$

This identity also works for complex (nonhermitian) x and y:

$$\exp\left[-a\widehat{x}^{\dagger}\widehat{x}\right] = \frac{i}{2\pi|a|} \int dy^{*}dy \exp\left[-\frac{y^{*}y}{|a|} + s(a)(\widehat{x}^{\dagger}y + h.c.)\right]$$

We now turn to the case of the partition function of a spin system with generic interaction Hamiltonian (1). In order to use the mathematical identities we need to represent the Hamiltonian (1) in terms of normal coordinates. To this end we define the normalized eigenstates of the exchange interaction operator

$$\sum_{j',\alpha'} J_{jj'}^{\alpha\alpha'} \chi_{n;j',\alpha'} = \kappa_n \chi_{n;j,\alpha}$$
(A4)

in terms of which we have

$$J_{jj'}^{\alpha\alpha'} = \sum_{n} \kappa_n \chi_{n;j,\alpha}^* \chi_{n;j',\alpha'}, \qquad (A5)$$

where the $\chi_{n;(j',\alpha')}$ are a complete and orthonormal set of eigenfunctions and thus obey

$$\sum_{j,\alpha} \chi_{n;j,\alpha}^* \chi_{n';j,\alpha} = \delta_{n,n'}$$

$$\sum_{n} \chi_{n;j,\alpha}^* \chi_{n;j',\alpha'} = \delta_{j,j'} \delta_{\alpha,\alpha'}$$
(A6)

For spins on a periodic lattice, the eigenstates $|n\rangle = |\mathbf{q}, \nu\rangle$ are labeled by wavevector \mathbf{q} and spin component ν , and the eigenfunctions take the form

$$\chi_{\mathbf{q},\nu;j,\alpha} = \frac{1}{\sqrt{N}} e^{i\mathbf{q}\cdot\mathbf{R}_j} u^{\alpha}_{\mathbf{q}\nu} \tag{A7}$$

where $u^{\alpha}_{\mathbf{q}\nu}$ are normalized real valued eigenvectors, i.e. $\sum_{\alpha} u^{\alpha}_{\mathbf{q}\nu} u^{\alpha}_{\mathbf{q}\nu} = 1$, and $\kappa_{\mathbf{q},\nu}$ are the eigenvalues of the spin exchange operator.

We now define the normal amplitudes of the spin operators as

$$S_{\mathbf{q},\nu} = \sum_{j,\alpha} \chi_{\mathbf{q},\nu;j,\alpha} S_j^{\alpha} \tag{A8}$$

and express the Hamiltonian (1) as

$$H = \sum_{\mathbf{q},\nu} \kappa_{\mathbf{q},\nu} S^*_{\mathbf{q},\nu} S_{\mathbf{q},\nu}$$
(A9)

where $S_{\mathbf{q},\nu}^* = S_{-\mathbf{q},\nu}$.

We seek to apply the above mathematical identities (A1)-(A3) to each normal component separately. This requires the normal components of the spin operators to commute with each other, which is certainly true for the classical spins. Then using the Hubbard-Stratonovich transformation one may express the Boltzmann weight operator of each normal mode in terms of normal field amplitudes $\varphi_{\mathbf{q},\nu}$ as

$$\begin{split} \exp[-\beta\kappa_{\mathbf{q},\nu}S^{*}_{\mathbf{q},\nu}S_{\mathbf{q},\nu}] &= (A10)\\ \frac{\imath}{2\pi\beta|\kappa_{\mathbf{q},\nu}|} \int \int d\varphi^{*}_{\mathbf{q},\nu}d\varphi_{\mathbf{q},\nu}\\ \exp[-|\beta\kappa_{\mathbf{q},\nu}|^{-1}\varphi^{*}_{\mathbf{q},\nu}\varphi_{\mathbf{q},\nu} + s(\kappa_{\mathbf{q},\nu})(S^{*}_{\mathbf{q},\nu}\varphi_{\mathbf{q},\nu} + h.c.)] &= \\ \frac{\imath\beta}{2\pi|\kappa_{\mathbf{q},\nu}|} \int \int d\varphi^{*}_{\mathbf{q},\nu}d\varphi_{\mathbf{q},\nu}\\ \exp[-\beta\{|\kappa_{\mathbf{q},\nu}|^{-1}\varphi^{*}_{\mathbf{q},\nu}\varphi_{\mathbf{q},\nu} + s(\kappa_{\mathbf{q},\nu})(S^{*}_{\mathbf{q},\nu}\varphi_{\mathbf{q},\nu} + h.c.)\}] \end{split}$$

The complete Boltzmann weight operator may be expressed, again using the commutability of the normal mode operators, as

$$\exp[-\beta \sum_{\mathbf{q},\nu} \kappa_{\mathbf{q},\nu} S^*_{\mathbf{q},\nu} S_{\mathbf{q},\nu}] = \int [d\varphi]$$
(A11)

$$\exp\left[-\beta \sum_{\mathbf{q},\nu} \{|\kappa_{\mathbf{q},\nu}|^{-1} \varphi_{\mathbf{q},\nu}^* \varphi_{\mathbf{q},\nu} + s(\kappa_{\mathbf{q},\nu}) (S_{\mathbf{q},\nu}^* \varphi_{\mathbf{q},\nu} + h.c.)\}\right]$$

where $\varphi^*_{{\bf q},\nu}=\varphi_{-{\bf q},\nu}$. The integration volume element is given by

$$[d\varphi] = \Pi_{\mathbf{q},\nu} \frac{i\beta d\varphi^*_{\mathbf{q},\nu} d\varphi_{\mathbf{q},\nu}}{2\pi |\kappa_{\mathbf{q},\nu}|}$$

Next we find that the partition function of an interacting classical spin system on an infinite periodic lattice may be expressed as

$$Z =$$
(A12)

$$= C \int [d\varphi] \exp[-\beta \sum_{\mathbf{q},\nu} |\kappa_{\mathbf{q},\nu}|^{-1} \varphi_{\mathbf{q},\nu}^* \varphi_{\mathbf{q},\nu} - S_{loc}(\{\varphi_{\mathbf{q},\nu}\})],$$

where C is a constant. The contribution $S_{loc}(\{\varphi_{\mathbf{q},\nu}\})$ to the action is given by

$$\mathcal{S}_{loc}(\{\varphi_{\mathbf{q},\nu}\}) = \frac{1}{\beta} \sum_{j} \ln W_j \tag{A13}$$

and W_j is computed by taking into account the constraint of the unit length of classical spins, $\mathbf{S}_j^2 = 1$, and integrating over all directions of spin at each lattice site

$$W_{j} = \int \frac{dS_{j}d\Omega_{j}}{2\pi} \exp[2\beta \sum_{\alpha} \varphi_{j}^{\alpha} S_{j}^{\alpha}] \delta(\mathbf{S}_{j}^{2} - 1)$$
$$= \int \frac{d\Omega_{j}}{4\pi} \exp[2\beta \sum_{\alpha} \varphi_{j}^{\alpha} S_{j}^{\alpha}]$$
$$= \frac{\sinh 2\beta |\varphi_{j}|}{2\beta |\varphi_{j}|}.$$
 (A14)

This gives

$$\mathcal{S}_{loc}(\{\varphi_{\mathbf{q},\nu}\}) = \frac{1}{\beta} \sum_{j} \ln[\frac{\sinh 2\beta |\varphi_j|}{2\beta |\varphi_j|}].$$
(A15)

Here we defined the complex-valued three-component field φ_i^{α} at each lattice site j as

$$\varphi_{j}^{\alpha} = \sum_{\mathbf{q},\nu} s(\kappa_{\mathbf{q},\nu}) \mathcal{R}e\{\varphi_{\mathbf{q},\nu}^{*}\chi_{\mathbf{q},\nu;j,\alpha}\}$$
(A16)
$$= \sum_{\mathbf{q},\nu} s(\kappa_{\mathbf{q},\nu})\varphi_{\mathbf{q},\nu}\chi_{\mathbf{q},\nu;j,\alpha}^{*}$$
$$= \varphi_{R,j}^{\alpha} + i\varphi_{L,j}^{\alpha}.$$

Observing that $\kappa_{\mathbf{q},\nu} = \kappa_{-\mathbf{q},\nu}$, we get

$$\varphi_{R,j}^{\alpha} = \operatorname{Re}\{\varphi_{j}^{\alpha}\} = \sum_{\mathbf{q},\nu,\kappa_{\mathbf{q},\nu}<0} \varphi_{\mathbf{q},\nu}\chi_{\mathbf{q},\nu;j,\alpha}^{*} \qquad (A17)$$
$$\varphi_{I,j}^{\alpha} = \operatorname{Im}\{\varphi_{j}^{\alpha}\} = \sum_{\mathbf{q},\nu,\kappa_{\mathbf{q},\nu}>0} \varphi_{\mathbf{q},\nu}\chi_{\mathbf{q},\nu;j,\alpha}^{*}.$$

The field amplitude is determined by

$$\varphi_j = \sqrt{(\varphi_{R,j} + i \varphi_{I,j})^2},$$
 (A18)

where $\boldsymbol{\varphi}_{R,j} = \left(\varphi_{R,j}^x, \varphi_{R,j}^y, \varphi_{R,j}^z\right)$ and $\boldsymbol{\varphi}_{I,j} = \left(\varphi_{I,j}^x, \varphi_{I,j}^y, \varphi_{I,j}^z\right)$.

We now derive the contribution of Gaussian fluctuations to the free energy for the ferromagnetic mean field state which we denote as φ_{MF} . To this end, we expand $S_{loc}(\{\varphi_{\mathbf{q},\nu}\})$ (A15) in terms of the fluctuation amplitudes and separate the mean field and fluctuational contributions. First, we expand the field amplitude φ_j to bilinear order in the fluctuation amplitudes:

$$\varphi_{j} = \varphi_{MF} + \delta\varphi_{j}, \qquad (A19)$$

$$\delta\varphi_{j} = \frac{1}{2\varphi_{MF}} [2\varphi_{MF} \cdot (\delta\varphi_{R,j} + i\delta\varphi_{I,j}) + \delta\varphi_{R,j}^{2} - \delta\varphi_{I,j}^{2}] - \frac{1}{2\varphi_{MF}^{3}} [\varphi_{MF} \cdot (\delta\varphi_{R,j} + i\delta\varphi_{I,j})]^{2}.$$

Using the Eqs. (A17), we now obtain the expressions for $\delta \varphi_j$ and $\delta \varphi_j^2$ in terms of $\varphi_{\mathbf{q},\nu}^*, \varphi_{\mathbf{q},\nu}$, keeping quadratic

(Gaussian) terms only:

$$\sum_{j} \delta\varphi_{j} = \frac{1}{2\varphi_{MF}} \sum_{\mathbf{q},\nu,\nu'} \delta_{\nu,\nu'} s(\kappa_{\mathbf{q}\nu}) s(\kappa_{\mathbf{q}\nu'}) \varphi_{\mathbf{q},\nu}^{*} \varphi_{\mathbf{q},\nu'}$$
$$-\frac{1}{2\varphi_{MF}} \sum_{j} \delta\varphi_{j}^{2} \qquad (A20)$$
$$\sum_{j} \delta\varphi_{j}^{2} = \sum_{\mathbf{q},\nu,\nu'} s(\kappa_{\mathbf{q}\nu}) s(\kappa_{\mathbf{q}\nu'}) m_{0,\nu} \varphi_{\mathbf{q},\nu}^{*} \varphi_{\mathbf{q},\nu'} m_{0,\nu'}$$

Next, we expand Eq. (A15) step by step as

$$\sinh 2\beta |\varphi_j| = \sinh(2\beta(\varphi_{MF} + \delta\varphi_j))$$
$$= \sinh(2\beta\varphi_{MF})[1 + 2(\beta\delta\varphi_j)^2] + \cosh(2\beta\varphi_{MF})2\beta\delta\varphi_j$$

and further

$$\begin{split} &\ln[\sinh(2\beta\varphi_j)/2\beta\varphi_j] \\ &= \ln\sinh(2\beta(\varphi_{MF} + \delta\varphi_j)) - \ln(2\beta(\varphi_{MF} + \delta\varphi_j)) \\ &= \ln[\sinh(2\beta\varphi_{MF})/(2\beta\varphi_{MF})] \\ &+ [2\beta\varphi_{MF}\coth(2\beta\varphi_{MF}) - 1] \frac{\delta\varphi_j}{\varphi_{MF}} \\ &+ \frac{1}{2} [-\frac{(2\beta\varphi_{MF})^2}{\sinh^2(2\beta\varphi_{MF})} + 1] (\frac{\delta\varphi_j}{\varphi_{MF}})^2. \end{split}$$

The fluctuation part of the free energy is then given by

$$S_{fl} = -\beta^{-1}\delta \sum_{j} \ln[\sinh(2\beta\varphi_j)/2\beta\varphi_j] =$$

$$= -\frac{4}{3}\beta_c\varphi_{MF} \sum_{j}\delta\varphi_j \qquad (A21)$$

$$-\frac{1}{2\beta\varphi_{MF}^2} [1 - \frac{(2\beta\varphi_{MF})^2}{\sinh^2(2\beta\varphi_{MF})}] \sum_{j}\delta\varphi_j^2,$$

where we have used that $2\beta\varphi_{MF} \coth(2\beta\varphi_{MF}) - 1 = \frac{4}{3}\beta_c\beta\varphi_{MF}^2$. Substituting the expressions for $\delta\varphi_j, \delta\varphi_j^2$ and defining $r = 1/(2\beta\varphi_{MF})^2 - 1/\sinh^2(2\beta\varphi_{MF})$, we get

$$\mathcal{S}_{\mathrm{fl}}\{\delta\varphi_{\mathbf{q},\nu}\} = \sum_{\mathbf{q};\nu,\nu'} A_{\mathbf{q},\nu\nu'}\delta\varphi_{\mathbf{q},\nu}^*\delta\varphi_{\mathbf{q},\nu'} \qquad (A22)$$

where we defined matrices $A_{{\bf q},\nu\nu'}$ describing the weight of Gaussian fluctuations of wavevector ${\bf q}$ and polarization ν as

$$A_{\mathbf{q}\nu\nu'} = |\kappa_{\mathbf{q}\nu}|^{-1} \delta_{\nu,\nu'}$$

$$-\frac{2}{3} [\beta_c(\delta_{\nu,\nu'} - m_{0,\nu}m_{0,\nu'}) + 3\beta r m_{0,\nu}m_{0,\nu'}] s(\kappa_{\mathbf{q}\nu}) s(\kappa_{\mathbf{q}\nu'})$$

The fluctuation matrix $A_{\mathbf{q}\nu\nu'}$ will in general be nonhermitian, and its eigenvalues will be complex. We now use that $A_{\mathbf{q}\nu\nu'}$ is an even function of \mathbf{q} and divide \mathbf{q} -space into $q_x > 0$ ($M_>$) and $q_x < 0$ ($M_<$). For \mathbf{q} -modes $\phi_{\mathbf{q},\nu}$ with $\mathbf{q} \in M_>$ we choose $s(\kappa_{\mathbf{q}\nu}) = +i$, whereas for $\mathbf{q} \in M_<$ we choose $s(\kappa_{\mathbf{q}\nu}) = -i$, where $\kappa_{\mathbf{q}\nu} > 0$ in both cases. Then we have $A_{-\mathbf{q}\nu\nu'} = A^*_{\mathbf{q}\nu\nu'}$ and as a result of the functional integration we will get

$$Z = Z_{MF} \int [d\phi] \exp\left[-\beta \sum_{\mathbf{q},\nu,\nu'} A_{\mathbf{q},\nu\nu'} \varphi_{\mathbf{q},\nu}^* \varphi_{\mathbf{q},\nu'}\right]$$

$$= Z_{MF} \exp\left[-\frac{1}{2} \sum_{\mathbf{q}\in M_{>}} \ln(\det(A_{\mathbf{q},\nu\nu'}) \det(A_{\mathbf{q},\nu\nu'}^*))\right]$$

$$= Z_{MF} \exp\left[-\frac{1}{2} \sum_{\mathbf{q}} \ln|\det(A_{\mathbf{q},\nu\nu'})|\right]$$
(A24)

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