Quantum Dynamics of Multiferroic Helimagnets: a Schwinger-Boson Approach

Hosho Katsura,1 Shigeki Onoda,2 Jung Hoon Han,3 and Naoto Nagaosa1,4

1Department of Applied Physics, The University of Tokyo,
7-3-1, Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
2Condensed Matter Theory Laboratory, RIKEN, Wako, Saitama 351-0198, Japan
3BK21 Physics Research Division, Department of Physics,
Sungkyunkwan University, Suwon 440-746, Korea
4Cross Correlated Materials Research Group, Frontier Research System,
Riken, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

We study the quantum dynamics/ fluctuation of the cycloidal helical magnet in terms of the Schwinger boson approach. In sharp contrast to the classical fluctuation, the quantum fluctuation is collinear in nature which gives rise to the collinear spin density wave state slightly above the helical cycloidal state as the temperature is lowered. Physical properties such as the reduced elliptic ratio of the spiral, the neutron scattering and infrared absorption spectra are discussed from this viewpoint with the possible relevance to the quasi-one dimensional LiCu2O2 and LiCuVO4.

PACS numbers: 71.70.Ej, 75.30.Kz, 75.80.+q, 77.80.-e

Frustration, competition between interactions, in magnets has been an intriguing issue in the field of classical/quantum magnetism over several decades. In the usual case, even with the competing exchange interactions $J_{ij}$’s, their Fourier transform $J(q)$ has the maximum at some wavevector $q = Q$, and the classical ground state becomes the helimagnet [1]. This is because of the constraint of the fixed length of the classical spin, i.e., $|S_i| = $fixed. In strongly frustrated quantum magnets, on the other hand, the long-range order is possibly destroyed and novel ground states without magnetic order may be realized. Many possibilities such as chiral spin liquid [2], spin-nematic [3] and spin- Peierls/valence-bond-crystal [4] states are theoretically proposed. Another possibility is a magnetically ordered state realized by the order-by-disorder mechanism when the corresponding classical system has continuously degenerate ground states [5].

Recently a renewed interest has been focused on the cycloidal helimagnets from the viewpoint of multiferroics, which exhibit both magnetic and ferroelectric properties [6,7]. These materials shed some new light on the frustrated magnets since the electric polarization is closely related to the vector spin chirality $S_i \times S_j$ [8,9,10,11,12], which has been the subject of intensive interests. Namely, it was found that the electric polarization $(P)$ produced by the neighboring spins $(S_i$ and $S_j$) can be written as

$$P = \alpha e_{ij} \times (S_i \times S_j),$$

where $e_{ij}$ denotes the unit vector connecting the sites $i$ and $j$. This relation has a physical interpretation in terms of spin current induced between noncollinear spins due to frustration [8].

Magnetic materials with the finite vector spin chirality include wide range of systems such as three dimensional (3D) magnets RMnO$_3$ $(R =$ Gd, Tb, Dy) with spin $S = 2$ [13,14,15,16], the kagome staircase compound Ni$_3$V$_2$O$_8$ with $S = 1$ [17], $S = 1/2$ quantum spin chains LiCu$_2$O$_2$ [18,19], LiCuVO$_4$ [20] and the quasi-one-dimensional (1D) molecular helimagnet with $S = 7/2$ [21]. Depending on the temperature, dimensionality, and the magnitude of the spin $S$, the role of the classical/quantum spin fluctuations differs and the theoretical studies on these fluctuations are needed for the consistent interpretation of the phase diagram and the physical properties of these systems. Especially, the low dimensionality enhances both thermal and quantum fluctuations leading to the breakdown of the conventional (classical + spin wave) picture for helimagnets. The possible chiral spin states without the magnetic long range ordering have been proposed theoretically for classical [22,23,24] and quantum [25,26,27] spin systems. However, the systematic study of the quantum fluctuation in the helimagnets including the finite temperature effect is rare, which is addressed in this paper and will be complementary to the works mentioned above.

In this paper, we study the quantum/thermal fluctuation in the helimagnet in terms of the Schwinger Boson (SB) approach. The advantage of the SB method is that it can describe the length of the ordered moment as a soft variable. Namely, in the constraint on the Schwinger boson number at each site,

$$\sum_\sigma b_{j\sigma}^\dagger b_{j\sigma} = 2S,$$

it can be decomposed into the condensed (classical) part and the fluctuating part. Therefore, the degrees of classical/quantum fluctuation and the ordered moment can be described in a unified fashion in this method [28]. In the SB language, the paramagnetic to collinear transition is described by the density wave instability of bosons, while the collinear to helical one corresponds to the Bose-Einstein condensation (BEC) of SB.

Effective model—we study quasi-1D and two-dimensional (2D) Heisenberg models with the exchange
interactions shown in Fig. 1 where $J_1$ is ferromagnetic, while $J_2$ are antiferromagnetic, leading to the frustration. The interchain/interplane interaction $J_{\perp}$ is assumed to be sufficiently weak, and will be treated by the mean field theory. The spin-$S$ operators are represented by $S^\alpha = b^\dagger_\alpha (\sigma^\alpha_{\sigma})/2 b_{\sigma}$, where $\sigma^\alpha$ ($\alpha = x, y, z$) are the Pauli matrices and the repeated indices are summed over. First, we assume that the resonating-valence-bond (RVB) correlation is dominant and neglect the other mean-field decoupling. This assumption is valid for the low-dimensional multiferroics such as LiCuVO$_4$ [29], LiCu$_2$O$_2$ [18] and Ni$_3$V$_2$O$_8$ [17]. The mean-field Hamiltonians of the quasi-1D model is given by

$$H^{\text{MF}}_{1D} = \sum_{k\sigma}(\lambda - 2\eta_1 \cos kx) b^\dagger_{k\sigma} b_{k\sigma} + \sum_{k} 2[\eta_2 \sin(2kx) + \eta_{1\perp}(\sin ky + \sin kz)] b^\dagger_{k\uparrow} b_{k\downarrow} + h.c. + 2N(\eta_1^2/J_1 + \eta_{1\perp}^2/J_2 + 2\eta_2^2/J_2 - S\lambda),$$

where $N$ is the total number of sites and $b_{k\sigma}$ is the Fourier transform defined by $b_{k\sigma} = \sum_{\nu} e^{-i k \nu R} b_{\nu \sigma} / \sqrt{N}$. In $H^{\text{MF}}_{1D}$, $\lambda$ denotes the chemical potential for the bosons and the order parameters $\eta_1$, $\eta_2$ and $\eta_{1\perp}$ are $J_{1}(b^\dagger_{i\sigma} b_{i+\perp,\sigma})/2$, $J_{2}(b^\dagger_{i\sigma} c_{i\sigma} b_{i+2\perp,\sigma})/2(1)$ and $J_{1\perp}(b^\dagger_{i\sigma} c_{i\sigma} b_{i+2\perp,\sigma})/2(1)$ ($\hat{e} = \hat{y}, \hat{z}$), respectively, with $\epsilon_{\perp} = -\epsilon_{\parallel} = 1$. RVB order parameters are assumed to be real and spatially uniform. In a parallel way, we can derive the quasi-2D mean-field Hamiltonian $H^{\text{MF}}_{2D}$. The Hamiltonian $H^{\text{MF}}_{2D}$ can be diagonalized by the Bogoliubov transformation as $H^{\text{MF}}_{2D} = \sum_{k\sigma} \omega(k)(\gamma_{k\sigma}^\dagger \gamma_{k\sigma} + 1/2) - 2N(\lambda + 1/2) + \text{const.}$, with the dispersion relation $\omega(k)^2 = (\lambda - 2\eta_1 \cos kx)^2 - (2\eta_2 \sin(2kx) + 2\eta_{1\perp}(\sin ky + \sin kz))^2$. The transformation between $\gamma_{k\sigma}$ and $b_{k\sigma}$ is given by

$$\begin{pmatrix} b_{k\uparrow} \\ b_{k\downarrow}^{\dagger} \end{pmatrix} = \begin{pmatrix} \cosh \theta_k & \sinh \theta_k \\ \sinh \theta_k & \cosh \theta_k \end{pmatrix} \begin{pmatrix} \gamma_{k\uparrow}^\dagger \\ \gamma_{k\downarrow} \end{pmatrix},$$

with $\tanh 2\eta_2 = -(2\eta_2 \sin kx + 2\eta_{1\perp}(\sin ky + \sin kz))/\lambda$. The chemical potential $\lambda$ is determined by the condition (2). $\eta$’s are obtained by minimizing the mean-field free energy $F^{\text{MF}}$. Figure 2 shows the numerically obtained $\eta_1$, $\eta_2$ and the gap $\Delta(T) = \omega(Q/2)$ of the 1D spin-1/2 model as a function of $J_1/J_2$. The transition temperature of $\eta_2$ is analytically given by $T_{\text{RVB}} = J_2(S + 1/2)/\ln(1 + 1/S)$. We have also numerically studied the 2D model at finite temperature and obtained similar results. From $\eta$’s, we can estimate the minima of the dispersion $\omega(k)$ as $\pm Q/2 = \pm (Q/2, \pi/2, \pi/2)$. $Q$ is determined to satisfy $(\lambda - 2\eta_1 \cos Q^2)\eta_1 \sin Q^2 = 4(\eta_2 \sin Q + 2\eta_{1\perp})\eta_2 \cos Q$.

To describe the low-energy physics of the model, it is useful to construct an effective continuum model. First, we suppose that $\eta$’s are non-zero. Next we expand the dispersion around the minima up to quadratic order in $k \pm Q/2$. The effective dispersion relations of $\gamma$-particles are those of massive relativistic bosons and explicitly given by $\Omega(k) = \sqrt{\Delta(T)^2 + c_{\parallel}^2 |k_{\parallel}|^2 + c_{\perp}^2 |k_{\perp}|^2}$, where $k_{\parallel}$ is the vector along (within) the chain (plane) while $k_{\perp}$ is that perpendicular to the chain (plane). The spin wave velocities $c_{\parallel}$ and $c_{\perp}$ can be written in terms of $\eta$’s, in principle. Now the effective Hamiltonian of our system is

$$H^{\text{eff}} = \sum_{k\sigma} \sum_{\alpha = \pm} \Omega(k)(\gamma_{k\sigma}^\dagger \gamma_{k\alpha} + 1/2),$$

where $\alpha = + (-)$ indicates that the momentum is around $+Q/2 (-Q/2)$. When the gap $\Delta(T) = \omega(Q/2)$ vanishes,
Collinear phase— To study the instability toward the magnetic ordering, we consider the mean-field decoupling of the interchain/interplane interaction corresponding to the density wave instability of the SB and treat the resulting one/two-dimensional problem \[ \mathcal{H}_{1D/2D} + \mathcal{H}_{\text{int}} \]

where \( z \) is the coordination number along interchain/interplane direction and \( S_{Q} \) is expressed by them as \( \langle a \times b \rangle = 0 \) and \( \langle a \times b \rangle \neq 0 \), respectively [24]. The interaction between \( \gamma \)-bosons, when translated from that between \( b \)-bosons by Eq. (4), is enhanced near the bottom of the dispersion, inversely leading to the density wave instability before the occurrence of BEC. From the rotational symmetry in spin space, we can set \( a^2 = b^2 = 0 \) without loss of generality. By introducing \( s = (a^2 - ia^2) + i(b^2 - ib^2) \) and \( t = (a^2 - ia^2) - i(b^2 - ib^2) \), we can rewrite \( \mathcal{H}_{\text{int}} \) as \( \mathcal{H}_{\text{int}} \approx \left( zJ_\perp / 2 \sum_{k=0}^{\infty} \left| |s^2 - (b^2 - Q^2 + 2b \cdot k + Q^2 + \text{h.c.}) + (zJ_\perp / 2) \sum_{k=0}^{\infty} \left| \left| 2t^2 - (b^2 \cdot k + Q^2 + 2b + \text{h.c.}) \right| \right. \right) \).

The summations over \( k \) are restricted to around 0 since our continuum model is valid only in the low-energy region. The free-energy density corresponding to the Hamiltonian \( \mathcal{H}_{1D/2D} + \mathcal{H}_{\text{int}} \) can be written in a decoupled form: \( f(x^2) \propto f(y^2), \) where \( x = zJ_\perp |s| \) and \( y = zJ_\perp |t| \). Since the helical order is related to \( x \) and \( y \) through \( \langle a \times b \rangle \propto x^2 - y^2 \), we conclude that the collinear phase appears if \( f(x^2) \) has a global minimum at \( x^2 = 0 \). In the quasi-1D case, \( f(x^2) - f(0) \) can be expanded in terms of \( x^2 = A x^2 + B x^3 \) with

\[
A = \frac{1}{\Delta(T)} \left( \frac{\Delta(T)}{2zJ_\perp} - \frac{S + 1/2}{888(T)} \right), \\
B = \frac{1}{\Delta(T)^3} \left( S + 1 \right) \frac{\delta(T)^3}{128} \frac{(1 - 2\delta(T)^3/3)^2}{1 - \delta(T)^2} - 5, 
\]

where \( \delta(T) = \Delta(T) / \Delta(T) \). \( \Delta(T) = \lambda - 2/91 \cos(q/2) \) is the renormalized gap. Here we have assumed \( T \gg \Delta(T) \). Since \( B \) is positive for \( 0 < \delta(T) < 1 \), a sufficient condition for the collinear phase is \( A < 0 \) and a second order phase transition to the collinear state occurs at \( A = 0 \). Above \( T_{RVB} \), \( \delta(T) < 1 \) and hence the inequality \( A < 0 \) is not satisfied for small \( zJ_\perp \). This means \( T_N < T_{\text{BEC}} \). Between \( T_{RVB} \) and \( T_N \), there is an antiferromagnetic transition temperature. Further lowering the temperature with increasing \( z \), the gap collapses to result in BEC of SB. Therefore, we conclude \( T_{\text{BEC}} < T_N < T_{RVB} \). We have also checked the existence of the collinear phase for quasi-2D case by numerically solving the self-consistent equations without using the continuum model. In this way, the instability towards the collinear order is a robust feature of the strongly fluctuating quantum helimagnets, and is essentially different from that of classical system with an easy axis anistropy.

Neutron scattering spectra— Now we turn to the neutron scattering spectra in the helical phase. For simplicity, we shall focus on the quasi-1D case with the possible relevance to the recent experiment on LiCu_2O_2 [19]. The magnetic cross section for polarized neutron is given by the following correlation functions as \( \langle \sigma \rangle \propto \langle S^\alpha_s S^\alpha_t \rangle \), where the sign \((-\lambda)\) corresponds to the parallel (anti-parallel) neutron spin \( S^\alpha \) to the spin direction (see Fig. 1). To break the degeneracy of the helicity, we first set \( \langle b^{\alpha,\beta}_{i+1} \rangle \neq 0 \) and \( \langle b^{\alpha,\beta}_{i} \rangle = 0 \), and then the zero momentum spectra at zero energy are \( F_1(s) \) and \( F_2(s) \). Here we note that non-Bragg part is considered below, i.e., non-zero \( s \) component. For the low energy regime, using \( \alpha \) and \( \beta \), \( \langle S^\alpha_{i+1} S^\alpha_{i+2} \rangle \sim F_1(s) + F_2(s) = \frac{1}{N} \left( S + 1 \right) \frac{\Delta(T)^2}{16c^2} \).
where $q$ is assumed to be small. The difference $\langle S_{q+q}^x S_{-q}^y \rangle - \langle S_{q+q}^y S_{-q}^x \rangle$ is expressed by the vector spin chirality $(\langle S_{q+q}^x \times S_{-q}^y \rangle)/i$, and is directly related to the condensate fraction, i.e., the $F_1$ term. The crossover between the $F_1$ and $F_2$ terms occurs at $q_0 \sim \sqrt{2} \Delta(T)/\Lambda$. Another important correlation functions, $\langle S_q^x S_q^y \rangle$ ($\alpha = x, y$), can be observed by the $S_n^c$ setup. By a similar calculation, one can show that $\langle S_{Q+q}^x S_{-q}^y \rangle = \langle S_{Q+q}^y S_{-q}^x \rangle$ for the fluctuating part. In the experiment [19], $\langle S_{Q+q}^x S_{-q}^y \rangle$ suggests elliptic spiral while $\langle S_{Q+q}^y S_{-q}^x \rangle$ indicates circular one. This puzzling point would be resolved by our above analysis considering the quasi-elastic component [25].

Dielectric response—Finally, we examine the dynamical dielectric response both in the paramagnetic and helical phases of the quasi-1D model. Even in the paramagnetic and collinear phase, we assume that the fluctuating electric polarization is given by Eq. (1) [33]. We take the mean-field decoupling $S_i \times S_j = \langle \hat{b}_{i \mu} \eta_{\mu \nu} \hat{b}_{j \nu} \rangle - h.c.)/(4i)$ to the ferromagnetic bonds and $S_i \times S_j = \langle \eta_{\mu \nu} \hat{b}_{i \mu} \eta_{\nu \lambda} \hat{b}_{j \lambda} \rangle - h.c.)/(4i)$ to the antiferromagnetic bonds. We henceforth focus on the contribution from the antiferromagnetic ($J_2$) bonds since its fluctuation is stronger than that of the ferromagnetic one. From the geometry of the system (see Fig.1), the polarization along the $b$-axis $P^b$ is always zero. In the paramagnetic phase, $\text{Im} \varepsilon_{aa}(\omega) = \text{Im} \varepsilon_{cc}(\omega)$ due to the rotational symmetry in spin space. The expression for the polarization along the $a$-axis $P^a$ is given by

$$P^a \propto (\eta_2/J_2) \sum_k \cos(2k_x) (ib_{k_\sigma}b_{-k_\sigma} + h.c.).$$

For purely 1D case, $\text{Im} \varepsilon_{aa}(\omega) \propto (n(\omega/2) + 1/2) / (\sqrt{\omega^2 - 4\Delta^2(T)^2})$, where $n(\omega)$ is the Bose distribution function. Near the threshold of the absorption, the 1D van Hove singularity, $\text{Im} \varepsilon_{aa}(\omega) \propto 1/\sqrt{\omega - 2\Delta(T)}$, appears as schematically shown in Fig.4a. On the other hand, a drastic change of the absorption spectra occurs in the helical phase since the low-lying branch bosons become gapless (see Fig.4c). In this phase, the energy dispersions of the upper and lower branches are given by $\Omega_{\pm}(k_x) = \sqrt{c^2 + 2\Delta(0)^2}$ and $\Omega_{\pm}(k_x) = c/k_x$, respectively. We assume the BEC of SB by the weak interchain interaction. The schematic behavior at zero temperature is shown in Fig.4b. There are three contributions corresponding to the processes of two bosons i) in the upper branch, ii) in the gapless-lower branch and iii) in both the upper and lower branches, respectively. Finally, it should be noted that we cannot neglect the one-magnon contribution coming from the condensed portion in the helical phase. This contribution corresponds to that obtained in the previous analysis [34], but this is much smaller in the quantum limit.

The authors are grateful to S. Seki, Y. Yamasaki, N. Kida, S. Todo, and Y. Tokura for fruitful discussions. This work was supported in part by Grant-in-Aids (Grant No. 15104006, No. 16076205, and No. 17105002) and NAREGI Nanoscale Science Project from the Ministry of Education, Culture, Sports, Science, and Technology. H.K. and S.O. were supported by the Japan Society for the Promotion of Science.

![FIG. 4: Schematic plots of Im\varepsilon_{aa}(\omega) in a) the paramagnetic phase and b) the helical phase with T = 0 (\Delta = \sqrt{2}\Delta(0)). Behaviors near thresholds are indicated. Blue (solid and dotted) lines are the results for purely 1D model. Singularities are smeared out by the interchain interaction as shown by black lines. Insets are schematic boson dispersions in both the phases.](image)

---

[1] A. Yoshimori, J. Phys. Soc. Jpn 14, 807 (1959).
[2] V. Kalmeyer and R. B. Laughlin, Phys. Rev. Lett. 59, 2095 (1987); X.-G. Wen, F. Wilczek and A. Zee, Phys. Rev. B 39, 11413 (1989).
[3] P. Chandra, P. Coleman, and A.I. Larkin, J. Phys.: Condens. Matter 2, 7933 (1990).
[4] N. Read and S. Sachdev, Phys. Rev. Lett. 66, 1773 (1991).
[5] C. L. Henley, Phys. Rev. Lett. 62, 2056 (1989).
[6] M. Fiebig, J. Phys. D 38, R123 (2005).
[7] Y. Tokura, Science 312, 1481 (2006).
[8] H. Katsura, N. Nagaosa, and A.V. Balatsky, Phys. Rev. Lett. 95, 057205 (2005).
[9] I. A. Sergienko and E. Dagotto, Phys. Rev. B 73, 094434 (2006).
[10] C. Jia et al., Phys. Rev. B 76, 144424 (2007).
[11] M. Mostovoy, Phys. Rev. Lett. 96, 067601 (2006).
[12] A. B. Harris, Phys. Rev. B 76, 054447 (2007).
[13] T. Kimura et al., Nature 426, 55 (2003).
[14] T. Goto et al., Phys. Rev. Lett. 92, 257201 (2004).
[15] K. Noda et al., J. Appl. Phys. 97, 10C103 (2005).
[16] M. Kenzelmann et al., Phys. Rev. Lett. 95, 087206 (2005).
[17] G. Lawes et al., Phys. Rev. Lett. 95, 087205 (2005).
[18] S. Park et al., Phys. Rev. Lett. 98, 057601 (2007).
[19] S. Seki et al., arXiv:0801.2553 [cond-mat.str-el].
[20] Y. Naito et al., J. Phys. Soc. Jpn. 76, 023708 (2007).
[21] F. Cinti et al., Phys. Rev. Lett. 100, 057203 (2008).
[22] J. Villain, J. Phys. C 10, 4793 (1977).
[23] S. Miyashita and H. Shiba, J. Phys. Soc. Jpn. 53, 1145 (1984).
[24] S. Onoda and N. Nagaosa, Phys. Rev. Lett. 99, 027206 (2007).
[25] A.A. Nersesyan, A.O. Gogolin, and F.H.L. Essler, Phys. Rev. Lett. 81, 910 (1998).
[26] T. Hikihara et al., J. Phys. Soc. Jpn. 69, 259 (2000).
[27] Another approach to this problem can be found in S. Furukawa et al., arXiv:0802.3256 [cond-mat.str-el].
[28] A. Auerbach, Interacting Electrons and Quantum Magnetism, (Springer, New York, 1998).
[29] Although we can also apply this mean-field method to the 3D systems, the results are almost the same as those obtained by classical + spin wave theory.

[30] With moderate 3D couplings, our SB mean-field theory can give the helical magnetism even in quasi-1D half-integer spin systems.
[31] D. J. Scalapino, Y. Imry and P. Pincus, Phys. Rev. B 11, 2042 (1975).
[32] H. J. Schulz, Phys. Rev. Lett. 77, 2790 (1996).
[33] The magnetostriction (Jia et al. [10]) is another possible origin of the infrared absorption (S. Miyahara and N. Furukawa, private communication).
[34] H. Katsura, A. V. Balatsky, and N. Nagaosa, Phys. Rev. Lett. 98, 027203 (2007).