Longitudinal modes of spin fluctuations in iron-based superconductors

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Iron-based superconductors can exhibit different magnetic ground states and are in a critical magnetic region where frustrated magnetic interactions strongly compete with each other. Here we investigate the longitudinal modes of spin fluctuations in an unified effective magnetic model for iron-based superconductors. We focus on the collinear antiferromagnetic phase (CAF) and calculate the behavior of the longitudinal modes when different phase boundaries are approached. The results can help to determine the nature of the magnetic fluctuations in iron-based superconductors.

I. INTRODUCTION

Iron-based superconductors have very rich magnetic properties\textsuperscript{12}. They exhibit many intriguing magnetically ordered ground states, including stripe-like collinear antiferromagnetic (CAF) state\textsuperscript{6}, checkerboard-like antiferromagnetic (AFM) state\textsuperscript{5}, bi-collinear antiferromagnetic (BCAF) state\textsuperscript{5} and so on. The superconductivity appears to be linked to the magnetism, in particular, the CAF state\textsuperscript{6}. The origin of these magnetic states thus has been one of central focus in this field.

Theories and models based on different magnetic mechanisms including both itinerant magnetism and local spin moments have been deployed to explain magnetic properties. Although both theories are reasonably successful in explaining magnetic properties of some certain families of iron-based superconductors, it has been crystal clear that the magnetism is a hybrid with dual characters from both itinerant electrons and local spin moments. However, microscopically, the system can not be simply described by a Kondo lattice type of model because it is very difficult to separate itinerant electrons from localized ones.

A reasonable strategy is to seek an effective magnetic model. With the existence of local magnetic moments, we can still focus on the effective interactions between these local moments by integrating out of itinerant electrons to obtain a minimum magnetic effective model by keeping those Heisenberg-type leading interactions with the shortest distances. This approach has yielded a successful effective model, the $J_1 - J_2 - J_3 - K$ model\textsuperscript{2,14}, which can give a unified description of magnetic orders in all families of iron-based superconductors. In this model, the nearest neighbor (NN) magnetic interaction $J_1$ and the next NN one $J_2$ arise mainly through local magnetic direct exchange and magnetic superexchange mechanisms. The third NN interaction $J_3$ and the quartic interaction $K$ indicate the existence of the strong couplings to itinerant electrons. The phase diagram of the model has been studied extensively\textsuperscript{28}. It is found that the model can account for all magnetic phases and low energy magnetic excitations observed experimentally in iron-based superconductors. The magnetism in the effective model is extremely frustrated due to the strong competition among $J_1$, $J_2$ and $J_3$, which is also consistent with the fact that the long range magnetic order is absent in some iron-based superconductors.

In a model based on local magnetic moments, the spin waves are the low energy excitations in a given magnetically ordered state. The spin waves are the transverse modes, namely the magnetic fluctuations perpendicular to the direction of the ordered moment. The longitudinal modes, which are parallel to the ordered moments, are gapped out at low energy. However, if there are several competing magnetic states, the longitudinal modes can start to appear at low energy even at zero temperature. Therefore, the gaps of the longitudinal modes can provide us important information about the degree of magnetic frustration\textsuperscript{15,17}.

Recently, several polarized neutron scattering experiments have been carried out in the CAF state of iron-based superconductors\textsuperscript{16,19}. The gapped longitudinal modes have been observed. The observed gaps of these longitudinal modes are much lower than the band width of spin excitations. Thus, these measurements suggest the existence of strong magnetic frustration in the materials. The materials may be close to a quantum critical point or are located close to a spin liquid region\textsuperscript{20,21}.

In this paper, we calculate the longitudinal modes of the $J_1 - J_2 - J_3 - K$ model in the CAF state, in particular, in the parameter region near the phase boundary between the CAF and other magnetic ordered states. We find that the longitudinal modes become visible at low energy close to the phase boundary and they have different dispersion relations from the transverse spin wave modes. They disperse very rapidly along the antiferromagnetic direction and have very little dispersion along the FM direction in the CAF phase. This feature is absent in other magnetically ordered states so that it is unique for the CAF state. Therefore, our results suggest that the measurement of the longitudinal modes in a paramagnetic state that has a finite magnetic correlation length can be used to determine how the system is close to the CAF state.
II. THE J₁ - J₂ - J₃ - K MODEL

The J₁ - J₂ - J₃ - K model is described by

\[ H = \sum_{\langle i,j \rangle} J_1 \vec{S}_i \cdot \vec{S}_j + \sum_{\langle i,j \rangle} J_2 \vec{S}_i \cdot \vec{S}_j - \sum_{\langle i,j \rangle} J_3 \vec{S}_i \cdot \vec{S}_j + \sum_{\langle i,j \rangle} K(\vec{S}_i \cdot \vec{S}_j)^2, \] (1)

which includes the nearest (J₁), second (J₂) and third (J₃) nearest neighbor Heisenberg interactions and K(> 0) quartic term. To reduce the possible divergence during numerical calculations, we will also introduce anisotropy to gap the Goldstone mode by shifting the coupling to XXZ coupling \( \vec{S}_i \cdot \vec{S}_j = S_i^x S_j^y + S_i^y S_j^x + AS_i^z S_j^z \), with \( A = 1 + \delta_a \) the tiny anisotropy in spin z coupling. Various magnetic phases can be classified in the parameters space as seen in Fig.1. We will focus on the spin wave longitudinal modes in the CAF phase and the behavior of approaching the phase boundaries as illustrated by the arrows in Fig.1.

![Diagram](image_url)

**FIG. 1:** Various classical states (J₁ > 0, s = 1) from top left to bottom right: BCAF, In-Commensurate 3, staggered Dimmer(D1), In-Commensurate 1, AFM and CAF as classified in literature. The black-white dots represent the up-down spins in real space configuration for commensurate phases. The colored arrows indicate the path approaching the phases boundaries from CAF state. The left panel is for K/J₁ = 0.2 where incommensurate phases appear and the right is for K/J₁ = 0.5 where the commensurate phases expand and cover the whole parameter space.

This Hamiltonian can be solved within the standard spin wave (Holstein-Primakov) theory. Starting from the above depicted classical ground state, we rotate the down spins and then represent the spins \( \vec{S}_i \) with magnons \( a_i, a_i^\dagger \):

\[
S_i^z \rightarrow -S_i^z, \quad S_i^\pm \rightarrow -S_i^\mp, \\
S_i^x = s - a_i^\dagger, \quad S_i^y(S_i^z) = \chi a_i(a_i^\dagger \chi),
\] (2)

with \( \chi = \sqrt{2s - a_i a_i^\dagger} \). When the local environment for each spin is not identical, there are several spins in one magnetic cell and the site \( i = \{ e, a \} \) with \( c = 1, \cdots, N' \) counting for the cells and \( a = 1, \cdots, n \) for the magnon types. Up to two-magnon operators, the approximate Hamiltonian in momentum space is

\[
H = \sum_k \frac{1}{2} s \Psi_k^\dagger H_k \Psi_k - \sum_k \frac{1}{2} s \text{Tr}(H_k) + E_0,
\] (3)

with \( H_k \) the \( 2n \times 2n \) matrix, \( E_0 \) the classical ground state energy and \( \Psi_k = (a_k, a_k^\dagger) \), where \( a_k \) is a column collection of magnon annihilators and \( a_k^\dagger \) the matrix transpose. As derived in the appendix (A), this Hamiltonian can be diagonalized by Bogoliubov transformation and the spin wave spectra are the eigenvalues of \( \sigma_{zn} H_k \). Here the Hamiltonian and Pauli matrices are

\[
H_k = \begin{pmatrix} \omega_k & \gamma_k \\ \gamma_k^* & -\omega_{T-k} \end{pmatrix}, \quad \sigma_{zn} = \begin{pmatrix} I_n & 0 \\ 0 & -I_n \end{pmatrix}
\] (4)

with \( I_n \) the \( n \times n \) identical matrix and \( \omega_k = \omega_k^\dagger = \omega_{T-k}, \gamma_k = \gamma_{T-k} = \gamma_k^\dagger \).

A. The Longitudinal Mode

Following Feynman’s approach to the helium superfluid, the longitudinal mode is approximated by applying the magnon density fluctuation on the spin wave (transverse) ground state:

\[
|L_q⟩ = X_q|0⟩ = \frac{1}{\sqrt{N}} \sum_i e^{i \vec{q} \cdot \vec{r}_i} S_i^z |0⟩, \quad q > 0.
\] (5)

With respect to the ground state, the longitudinal mode has the spectrum:

\[
E(q) = \frac{N(q)}{S(q)},
\] (6)

with \( N(q) \equiv \frac{1}{2} \langle 0| [X_{-q}, H, X_q] |0⟩ \) and \( S(q) \equiv ⟨ L_q | L_q ⟩ \) the structure factor. Separating the Hamiltonian by neighborhood \( \vec{m} \) coupling: \( H = \sum_m H_m + \sum_m H^K_m \), we have

\[
N(q) = \sum_m N_m(q) + \sum_m N^K_m(q)
\] (7)

with (dependent on whether the \( \vec{m} \) neighbor spins are anti-parallel or parallel (AP/P))

\[
N_m(q) = \begin{cases} \frac{1}{2n} \sum_a (\cos(qm) + 1) \Pi_{m+}, & AP \\ \frac{1}{2n} \sum_a (\cos(qm) - 1) \Pi_{m-}, & P \end{cases}
\] (8)

\[
N^K_m(q) = \begin{cases} \frac{K^2}{2n} \sum_a (\cos(qm) + 1) \Pi_{m+}, & AP \\ -\frac{K^2}{2n} \sum_a (\cos(qm) - 1) \Pi_{m-}, & P \end{cases}
\] (9)

where \( \sum_a \) sums over different magnon types and the correlation function \( \Pi \)'s are (see appendix A)

\[
\Pi_{m+} = 2s\Delta_{ab} - \Delta_{ab}(\rho_{aa} + \rho_{bb}) - \rho_{ab}(\Delta_{aa} + \Delta_{bb}),
\]

\[
\Pi_{m-} = \frac{s}{2}(\Delta_{ab} - \Delta_{bb} + \Delta_{aa}),
\]

\[
\Pi_{m+} = \frac{s}{2}(\Delta_{ab} - \Delta_{bb} + \Delta_{aa}),
\]
\[
\Pi_{m-} = 2s\rho_{ab} - \rho_{ab}(\rho_{aa} + \rho_{bb}) - \Delta_{ab}(\Delta_{aa} + \Delta_{bb}),
\]
\[
\Pi^K_{m+} = 8\Delta^2_{ab} + 4\Delta_{aa}\Delta_{bb} + A(2(s - 1)\rho_{ab} - 5\rho_{ab}(\rho_{aa} + \rho_{bb}) - 5/2\Delta_{ab}(\Delta_{aa} + \Delta_{bb})),\]
\[
\Pi^K_{m-} = 8\rho_{ab}^2 + 4\Delta_{aa}\Delta_{bb} + A(2(s - 1)\Delta_{ab} - 5\Delta_{ab}(\rho_{aa} + \rho_{bb}) - 5/2\rho_{ab}(\Delta_{aa} + \Delta_{bb})).
\]

Here \(\rho_{ab}(m) \equiv \langle a^i_i b_{i+m}\rangle\), \(\Delta_{ab}(m) \equiv \langle a_i b_{i+m}\rangle\) are the correlations of type \(a, b\) magnon at site \(i, i+m\). We have omitted the neighborhood \((m)\) to simplify the notation. The quartic \(K\) term modifies the exchange \(J\) by \(\pm 2AKs^2\) at the first order of large \(s\). The \(n\times n\) correlation matrices in momentum space \(\rho_k\), \(\Delta_k\) are the Fourier transformation of \(\rho(m)\), \(\Delta(m)\). The structure factor is
\[
S(q) = \frac{1}{n} Tr \left[ \rho(0) + \frac{1}{N^2} \sum_k (\rho_k\rho_{k+q} + \Delta_k\Delta_{k+q}) \right].
\]

In the case of one type magnon, the spin wave spectrum and the correlators can be solved analytically:
\[
\epsilon_k = s\sqrt{\omega_k^2 - \gamma_k^2},
\]
\[
\rho_k = (\sqrt{\omega_k^2/(\omega_k^2 - \gamma_k^2)} - 1)/2,
\]
\[
\Delta_k = -\gamma_k/(2\sqrt{\omega_k^2 - \gamma_k^2}).
\]

The CAF phase with ordered momentum \(Q = (\pi, 0)\) is an example of the exactly solvable case of Eq. (12) with the Hamiltonian matrix elements
\[
\omega_k = 2(J_1 - 2AK^2)\cos k_y + 4AJ_2 + 8A^2Ks^2
+ 2J_3(\cos 2k_x + 2\cos k_y - 2A),
\]
\[
\gamma_k = -2(J_1 + 2AK^2 - 2J_2\cos k_y)\cos k_x.
\]

The spin wave spectra are depicted in the upper panels (a) of Fig. 2, in the unit of \(J_1\) without further specification. The Goldstone modes appear at \((0, 0)\) and \((\pi, \pi)\) dependent on \(K\) and the coupling anisotropy \(A\). Zooming into \(Q\), the spin wave has oval-shape equal energy line as seen from Fig. 2(c) which has been measured experimentally.

The longitudinal mode spectra are depicted in the upper panels (b) of Fig. 2. It has a deep valley structure along the \(k_x = \pi\) line. The dispersion is flat in \((\pi, \pm\delta_y)\) direction, but it is steep in \((\pi, \pm\delta_y, 0)\) as shown in Fig. 2(b,d). The valley structure from \(q_y\) and \(q_x\) directions are manifested in Fig. 2(d). It is interesting to point out that at \(Q = (0, \pi)\), the structure factor \(S(Q) = 0\). Thus, at this point, the energy of the longitudinal mode is undefined in Eq. (12). However, except this special point, Eq. (12) is a smooth function.

The dispersion of the spin wave transverse and longitudinal modes along some high symmetry directions are shown in Fig. 2(e). As expected, the transverse mode converges to almost zero and its linear dispersion shows slight anisotropy. The longitudinal mode at this point is gapped and strongly anisotropic. The dispersion is flat in \(q_y (X\text{M})\) direction and sharp in \(q_x (X\Gamma)\) direction.

As approaching the phases boundaries, the frustration arises due to the competition of different magnetic orders. The longitudinal gap would decrease to appear in low energy excitation. We will use the energy limit approached from the flat \(k_y\) direction as the longitudinal gap \(\Delta\). The decrease of the longitudinal gap is depicted in Fig. 3. The approaching routes are indicated by the color arrows in Fig. 3. The gap \(\Delta\) drops to zero near the phases boundaries, appearing as low energy excitation in the neutron scattering experiment. We also noticed that the gaps of the three empty-dots marked routes approaching BCAF, DI-horizontally and DI-vertically respectively drop faster to zero in front of the boundaries, as a possible sign of stronger frustration.

No big difference is found for the paths approaching the DI phases horizontally and vertically.
FIG. 3: The gap $\Delta$ changes as approaching the phases boundaries marked by color arrows paths in Fig. 1. The filled and empty dots mark $K/J_1 = 0.2$ and $K/J_1 = 0.5$ respectively. $J_c$ represents the parameter on the phase boundary and $\delta J_c$ measures the distance from the phase boundary.

C. Other Magnetic Phase $(\pi, \pi)$

Similar work can be done for other magnetic phases represented in Fig. 1, following the method generally described in Appendix. For instance, the $J_1$ dominating AF state with $Q = (\pi, \pi)$, the spin wave the longitudinal mode along the high symmetry points are in Fig. 1. Now $M$ is the ordered momentum, where the Goldstone modes appears. Similar to the CAF phase, the longitudinal mode is gapped at $M$. The magnetic environment is $C_4$ symmetric and both the spin wave and longitudinal dispersion are isotropic. Approaching to the phases boundaries, the longitudinal gaps also decreases to appear as low excitations due to phases competition. The longitudinal modes are always more sensitive to the quantum frustration. Near the quantum critical point, the interaction among magnons in longitudinal mode breaks the ground state.

FIG. 4: The longitudinal spectrum of the AF state along the high symmetry points with parameters $K = 0.2$, $J_2 = 0.2$, $J_3 = 0.05$ and $A = 1.0001$.

III. DISCUSSION AND SUMMARY

Recently, the inelastic neutron scattering experiments have already measured the longitudinal modes in iron-based superconducting materials. These results are typically considered as evidence to support pure itinerant magnetism in these materials. However, as we have mentioned in the introduction, a pure itinerant magnetism can not explain observed magnetic properties in iron-based superconductors.

Combining with the experimental results, our results strongly support that iron-based superconductors are strongly frustrated magnetic systems in a vicinity to many different magnetic phases. In particular, the CAF order is very close to quantum critical transitions to other magnetic orders. The nature of the frustration stems from the competitions between short range local magnetic exchange couplings and other effective magnetic interactions through couplings with itinerant electrons.

In summary, we derived the longitudinal excitation for the general $J_1 - J_2 - J_3 - K$ magnetic model. Specifically, the analytic solution in the CAF state is given. The dispersion of the longitudinal modes in the CAF state near the quantum critical point is very different from the transverse spin wave modes.

Acknowledgement: the work is supported by the Ministry of Science and Technology of China 973 program (Grant No. 2015CB921300), National Science Foundation of China (Grant No. NSFC-11334012, No. NSFC-11534014), and the Strategic Priority Research Program of CAS (Grant No. XDB07000000).

Appendix A: Derivation

1. Spin Wave spectrum

The magnon satisfies the bosonic commute relation:

$$\Psi_k \Psi_q^\dagger - ((\Psi_q^\dagger)^T \Psi_k^T)^T = \delta_{kq}\sigma_{zn}. \quad (A1)$$

With the help of Bogoliubov transformation $\Psi_k = T_k \Phi_k$, where $\Phi_k = (d_k^\dagger, d_k^T)$, the Hamiltonian can be diagonalized as $T_k^\dagger H_k T_k = \sigma_{zn} \Lambda_k$, when the transformation matrix satisfy

$$\sigma_{zn} = T_k \sigma_{zn} T_k^\dagger,$$

$$\Lambda_k = T_k^\dagger (\sigma_{zn} H_k) T_k, \quad (A2)$$

where $\Lambda_k$ is the diagonalized matrix of $\sigma_{zn} H_k$ by similarity transformation and $T_k$ is the Bogoliubov matrix:

$$T_k = \begin{pmatrix} \mu_k & \nu_k \\ \mu^*_k & \nu^*_k \end{pmatrix}, \quad (A3)$$

where $\mu_k, \nu_k$ are the $n \times n$ coherent phase matrices. One can collect the eigenvectors of matrix $\sigma_{zn} H_k$ and then normalize them by equation. (A2). It is proven that for
the eigenvalue $\epsilon_k$ with eigenvector $\mathbf{v}_k$, there exists a dual eigenvalue $-\epsilon_k^*$ with eigenvector $\sigma_{2n}\mathbf{v}_k^*$. Thus by properly order the eigenvalues and adjust the relative phases, this $T_k$ is the Bogoliubov matrix to diagonalize the Hamiltonian via congruence transformation. In algorithm, there could be phase freedom for those eigenvectors, making the matrix non-Bogoliubov, i.e. $T \rightarrow T e^{i\Theta}$ with $\Theta = \text{diag}\{b_1, \cdots, b_{2n}\}$ the phases matrix. It’s easy to remove these phases and, moreover, this phase freedom does not affect the spin wave spectra and the longitudinal mode.

In linear spin wave theory, the Hamiltonian matrix elements are the Fourier transformation of the neighborhood interaction in real space, so $\omega_k, \gamma_k, \epsilon_k, \mu_k, \nu_k$ all satisfy $f^*(k) = f(-k)$ and the diagonalized Hamiltonian $\sigma_{2n}\Lambda_k$ is real and doubly degenerate.

2. Longitudinal Spectrum

The essence of calculating the ground state expectation of the commutators $N_m(q)$ and $N^K_m(q)$ in equation is to calculate the correlation of spins, up to second (SS) and forth order (SSSS) system. Define the magnon correlators first:

$$\langle \Psi_k | \Psi_q \rangle = \left( \frac{\mu_k \mu_k^T}{\nu_k \nu_k^T} \right) \delta_{kq} = \left( \frac{\rho_k^T + 1}{\rho_k} \right) \delta_{kq}.$$  

It is easy to see $\rho_k^+ = \rho_k$ and $\Lambda_k = \Delta^T_k$. In linear spin theory, $f(-k) = f^*(k)$ also works for $\rho_k$ and $\Lambda_k$. The real space correlations

$$\rho(m) \equiv a_{l+m}^T a_{l+m} = \frac{1}{N} \sum_k e^{-ikm} \rho_k,$$

$$\Delta(m) \equiv \langle a_l a_{l+m}^T \rangle = \frac{1}{N} \sum_k e^{-ikm} \Lambda_k$$

satisfy $\rho^*(m) = \rho^*(m) = \rho(m)$ and $\Delta^*(m) = \Delta^*(m) = \Delta(m)$. Thus the correlation among different magnons is the elements of the correlation matrices $\rho(m)$ and $\Delta(m)$:

$$\langle a_{l+m}^T a_l \rangle = \rho_{ab}(m) = \langle a_{l+m}^T a_l \rangle - \delta_{m0},$$

$$\langle a_{l+m}^T a_l \rangle = \Delta_{ab}(m) = \langle a_{l+m}^T a_l \rangle,$$

For higher (even) operators correlation, thereafter, the contraction rules can be concluded $\langle c_1 c_2 \cdots c_{2n-1} c_{2n} \rangle (c_i = a_i, a_i^\dagger)$:

1. Put the $c$’s operators in pairs, all possible combinations;

2. Refer to the matrix element of $\rho(m)$ and $\Delta(m)$, write all the operator pairs as two-operator correlation functions in real space. Referring this contraction rule, the following four operators (and their conjugate) correlation is needed:

$$\langle S_{i}^+ S_{j}^- \rangle = 2s \rho_{ab} - \rho_{ab} (\rho_{aa} + \rho_{bb}) - \Delta_{ab} (\Delta_{aa} + \Delta_{bb})/2,$$

$$\langle S_{i}^- S_{j}^+ \rangle = 2s \Delta_{ab} - \rho_{ab} (\rho_{aa} + \rho_{bb}) - \rho_{ab} (\Delta_{aa} + \Delta_{bb})/2,$$

and in the quartic term $N^K_m(q)$

$$\langle S_{i}^+ S_{j}^+ S_{k}^- S_{l}^- \rangle \approx (2s)^2 (2\Delta_{ab}^2 + \Delta_{aa} \Delta_{bb}),$$

$$\langle S_{i}^- S_{j}^- S_{k}^+ S_{l}^+ \rangle \approx (2s)^2 (2\Delta_{ab}^2 + \Delta_{aa} \Delta_{bb}),$$

We omit the variables $(j-i)$ within $\rho_{ab}, \Delta_{ab}$ for convenience. Relevant to the longitudinal mode up to the four operators correlation, for $N_m(q)$, we need to estimate:

$\langle S_{i}^+ S_{j}^- S_{k}^- S_{l}^+ \rangle \approx \Delta_{ab} (\Delta_{aa} + \Delta_{bb})/2,$

With the above correlators derived, the correlation function II’s in $N(q)$ can be obtained.

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