Coincidence inelastic neutron scattering for detection of two-spin magnetic correlations

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The inelastic neutron scattering (INS) is one powerful technique to study the low-energy single-spin dynamics of the magnetic materials. A variety of quantum magnets show novel magnetic correlations such as of quantum spin liquids. These novel magnetic correlations are beyond the direct detection of the INS. In this article we propose a coincidence technique, coincidence inelastic neutron scattering (cINS), which can detect the two-spin magnetic correlations of the magnetic materials. In the cINS there are two neutron sources and two neutron detectors with an additional coincidence detector. Two neutrons from the two neutron sources are incident on the target magnetic material and they are scattered by the electron spins of the magnetic material. The two scattered neutrons are detected by the two neutron detectors in coincidence with the coincidence probability described by a two-spin Bethe-Salpeter wave function. Since the two-spin Bethe-Salpeter wave function defines the momentum resolved dynamical wave function with two spins excited, the cINS can detect explicitly the two-spin magnetic correlations of the magnetic material. Thus it can be introduced to study the various spin valance bond states of the quantum magnets.

I. INTRODUCTION

The novel magnetic correlations in the various quantum magnets have attracted much attention in the condensed matter filed. The quantum spin liquids with strong frustration and quantum fluctuations are one special type of examples.1,2. One experimental technique in study of these novel magnetic correlations is the INS, which can provide the single-spin dynamical responses of the magnetic materials and thus can show the relevant physics of the single-spin excitations3,4,5,6. However, as most of the novel magnetic correlations in these quantum magnets are beyond that of the single-spin magnons, the spectrum of the INS can not provide the explicit information on these novel magnetic correlations. It is imperative to develop the experimental techniques which can detect explicitly these novel magnetic correlations.

Following a recent idea of the coincidence angle-resolved photoemission spectroscopy (cARPES) which is developed for detection of the two-particle correlations of the material electron5,11, in this article we will propose another coincidence technique, a cINS, which can detect explicitly the two-spin magnetic correlations of the magnetic materials. There are two neutron sources and two neutron detectors in the experimental instrument of the cINS with an additional coincidence detector. The two neutron sources emit two neutrons which are incident on the target magnetic material and are scattered by the material electron spins. These two scattered neutrons are then detected by the two neutron detectors in coincidence with the coincidence probability relevant to a two-spin Bethe-Salpeter wave function.

The two-spin Bethe-Salpeter wave function is defined as \( \phi_{\alpha\beta}^{(i)}(\mathbf{q}_1t_1, \mathbf{q}_2t_2) = \langle \Psi_\beta | T_i \hat{S}_t^{(i)}(\mathbf{q}_1t_1) \delta^{(i)}(\mathbf{q}_2t_2) | \Psi_\alpha \rangle \), where \( | \Psi_\alpha \rangle \) and \( | \Psi_\beta \rangle \) are the eigenstates of the electron spins of the target magnetic material, \( \hat{S}_t^{(i)}(\mathbf{q}, t) \) is the \( i \)-th component of the spin operator within a perpendicular plane normal to the momentum \( \mathbf{q} \), and \( T_i \) is a time-ordering operator. This Bethe-Salpeter wave function describes the time dynamical evolution of the magnetic material with two spins excited at times \( t_1 \) and \( t_2 \) in time ordering. The coincidence probability of the cINS can detect the Fourier transformation of the time dynamical Bethe-Salpeter wave function, with the center-of-mass frequency defined by the sum of the two transfer energies in the two-neutron scattering and the relative frequency defined by the difference of the two transfer energies. Therefore, the coincidence detection of the cINS can provide the momentum resolved dynamics of the two-spin magnetic correlations, with the physics both of the center-of-mass and of the relative degrees of freedom of two excited spins of the magnetic material. Thus it can be introduced to study the spin valence bond states of the quantum magnets.

Our article is arranged as follows. In Sec. II the theoretical formalism for the coincidence detection of the cINS will be provided. In Sec. III the coincidence probabilities of the cINS for a ferromagnet and an antiferromagnet with a long-range magnetic order will be presented. Discussion on the experimental detection of the cINS will be given in Sec. IV where a brief summary will also be provided.

II. THEORETICAL FORMALISM FOR cINS

In this section we will establish the theoretical formalism for the coincidence detection of the cINS. Firstly, we will review the principle of the single-spin INS in Sec. II A. We will then provide the theoretical formalism for the cINS in Sec. II B.

A. Review of INS

Suppose the incident neutrons have momentum \( \mathbf{q}_i \) and spin \( \beta_i \) with a spin distribution function \( P_i(\beta_i) \). The in-
incident neutrons interact with the electron spins of the target magnetic material via the electron-neutron magnetic interaction

$$\hat{V}_s = \sum_{q,\vec{q}} g(q) \hat{\sigma}_{q/\vec{q}} \cdot \mathbf{S}_\perp(q),$$  \hspace{1cm} (1)

where $g(q) \equiv gF_0(q)$ with $g$ an interaction constant and $F_0(q)$ a magnetic form factor, and $q = \vec{q} - \vec{q}$, with $\vec{q} = \frac{4}{\gamma}$. The operator $\hat{\sigma}_{q/\vec{q}}$ is defined for neutrons,

$$\hat{\sigma}_{q/\vec{q}} = \sum_{\beta \beta_f} d_{\beta_f}^\dagger \hat{\sigma}_{f\beta_f} d_{q/\beta},$$  \hspace{1cm} (2)

where $d_{q/\beta}$ and $d_{q/\beta}^\dagger$ are the respective neutron annihilation and creation operators, and $\hat{\sigma}$ is the Pauli spin operator. The electron spin operator $\mathbf{S}(q)$ is defined by

$$\mathbf{S}(q) = \sum_i S_i e^{-iqR_i},$$  \hspace{1cm} (3)

where $c_i$ and $c_i^\dagger$ are the annihilation and creation operators of the Wannier electrons at position $R_i$ respectively, and $\mathbf{S}$ is the spin angular momentum operator. Here we assume that the material electrons which have a dominant interaction with the incident neutrons are the local Wannier electrons. It is noted that $\mathbf{S}(\vec{q})$ is defined as

$$\mathbf{S}_\perp(\vec{q}) = \mathbf{S}(\vec{q}) - \hat{\mathbf{q}}(\mathbf{S}(\vec{q}) \cdot \hat{\mathbf{q}}),$$  \hspace{1cm} (4)

A simple review on the electron-neutron magnetic interaction $\hat{V}_s$ is given in Appendix A.

One incident neutron with momentum $\vec{q}_i$ can be scattered by the material electrons into the state with momentum $\vec{q}_f$. The relevant scattering probability is defined by

$$\Gamma^{(1)}(\vec{q}_f, \vec{q}_i) = \frac{1}{Z} \sum_{\alpha \beta \beta_f} e^{-\beta E_\alpha} P_1(\beta_i) \times |\langle \Phi_\beta | \hat{S}^{(1)}(+\infty, -\infty) | \Phi_\alpha \rangle|^2,$$  \hspace{1cm} (5)

where the initial state $|\Phi_\alpha \rangle = |\Psi_\alpha; q_i, \beta_i \rangle$ and the final state $|\Phi_\beta \rangle = |\Psi_\beta; q_f, \beta_f \rangle$, and $|\Psi_\alpha \rangle$ and $|\Psi_\beta \rangle$ are the electron eigenstates whose eigenvalues are $E_\alpha$ and $E_\beta$, respectively. $\hat{S}^{(1)}(+\infty, -\infty)$ is the first-order expansion of the time-evolution $S$ matrix of the perturbation electron-neutron magnetic interaction $\hat{V}_s$ and is defined by

$$\hat{S}^{(1)}(+\infty, -\infty) = -i \frac{e^{iH_0 t/\hbar} \hat{V}_s e^{-iH_0 t/\hbar}}{\hbar},$$  \hspace{1cm} (6)

where $\hat{V}_s(t) = e^{iH_0 t/\hbar} \hat{V}_s e^{-iH_0 t/\hbar}$ with $H_0$ being the sum of the Hamiltonians of the material electrons and the neutrons. $F_0(t)$ defines the interaction perturbation time

$$F_0(t) = \theta(t + \Delta t/2) - \theta(t - \Delta t/2),$$  \hspace{1cm} (7)

where $\theta(t)$ is the step function.

It should be noted that in the above scattering probability, we have defined implicitly the initial and the final states by the density matrices as follows:

$$\hat{P}_I = \frac{1}{Z} \sum_{\alpha \beta_i} e^{-\beta E_\alpha} P_1(\beta_i) |\Psi_\alpha; q_i, \beta_i \rangle \langle \beta_i; \Psi_\alpha |,$$

$$\hat{P}_F = \sum_{\beta \beta_f} |\Psi_\beta; q_f, \beta_f \rangle \langle \beta_f; \Psi_\beta |.$$  \hspace{1cm} (8)

In this article we will focus on the cases where the incident neutrons are the thermal neutrons in the spin mixed state defined by

$$\sum_{\beta_i} P_1(\beta_i) |\beta_i \rangle \langle \beta_i | = \frac{1}{2} (| \uparrow \rangle \langle \uparrow | + | \downarrow \rangle \langle \downarrow |).$$  \hspace{1cm} (9)

Introduce an imaginary time Green’s function

$$G(q, \tau) = - \sum_{ij} (T \tau_{ij} S(q, \tau) \delta_{ij} - \hat{q} \hat{q} \delta_{ij} \langle \Phi_\beta | \hat{S}^{(1)}(\tau) | \Phi_\alpha \rangle),$$

its corresponding spectrum function $\chi(q, E)$ is defined as

$$\chi(q, E) = -2 \text{Im} G(q, i\hbar \omega - E + i0^+),$$

where $\omega$ is the time interval $\Delta t$ and $\text{Im} G(q, i\hbar \omega) \rightarrow \pi a \delta(x)$ when $a \rightarrow +\infty$.

Let us consider the scattering cross section. Define the incident neutron flux by $J_I = n_I v_I$, where the density $n_I = \frac{1}{V_I}$ ($V_I$ is the renormalization volume for one neutron) and the velocity $v_I = \frac{2k}{m}$. The scattering cross section per scatter $\sigma$ follows

$$J_I \sigma = \frac{1}{N_m \Delta t} \sum_{q_f, q_i} \Gamma^{(1)}(q_f, q_i),$$  \hspace{1cm} (10)

where $N_m$ is the number of the scatter electrons in the incident neutron beam. The double differential scattering cross section is shown to follow

$$\frac{d^2 \sigma}{d\Omega dE_f} = \frac{(\gamma R_e)^2 q_f}{2\pi N_m q_i} |F_0(q)|^2 \chi(q, E^{(1)} n_B(E^{(1)}),$$  \hspace{1cm} (11)

where $E_f$ is the energy of the scattered neutrons, $\gamma = 1.91$ is a constant for the neutron gyromagnetic ratio, and $R_e$ is the classical electron radius defined as

$$R_e = \frac{\mu_0 e^2}{4\pi m_e c^2} = \frac{e^2}{4\pi \varepsilon_0 m_e c^2}.$$  \hspace{1cm} (12)
with $\mu_0$ the free-space permeability and $\varepsilon_0$ the vacuum permittivity. This double differential cross section we have obtained is same to that from the Fermi’s Golden rule. Physically, the scattering probability and the scattering cross section of the INS come from the contribution of the first-order perturbation of the electron-neutron magnetic interaction.

**B. Theoretical formalism for cINS**

In this section we will present a coincidence technique, a coincidence inelastic neutron scattering which, we call “cINS”, can detect the two-spin magnetic correlations of the target magnetic material. The schematic diagram of the cINS is shown in Fig. 1. There are two neutron sources which emit two neutrons with momenta $q_{i1}$ and $q_{i2}$. These two neutrons are incident on the target magnetic material and interact with the electron spins. The two incident neutrons are then scattered outside of the material into the states with momenta $q_{f1}$ and $q_{f2}$. Two single-neutron detectors detect the two scattered neutrons, and a coincidence detector records the coincidence counting probability when each of the two single-neutron detectors detect one single neutron simultaneously.

The coincidence counting probability of the two scattered neutrons is described by

$$\Gamma(2)(q_{f1}, q_{f2}, q_{i1}, q_{i2}) = \frac{1}{Z} \sum_{\alpha\beta, \beta'} e^{-\beta E_0} P_2(\beta_{i1}, \beta_{i2})$$

$$\times |\langle \Phi_{\beta} | \hat{S}^{(2)}(+\infty, -\infty) | \Phi_{\alpha} \rangle|^2,$$

where the initial state $|\Phi_{\alpha}\rangle = |\Psi_{\beta}; q_{i1}, \beta_{i1}, q_{i2}; \beta_{i2}\rangle$ and the final state $|\Phi_{\beta}\rangle = |\Psi_{\beta}; q_{f1}, \beta_{f1}, q_{f2}; \beta_{f2}\rangle$. $P_2(\beta_{i1}, \beta_{i2})$ defines the spin distribution function of the incident thermal neutrons. In our following study, we will consider the cases with $P_2(\beta_{i1}, \beta_{i2}) = P_1(\beta_{i1})P_1(\beta_{i2})$. $\hat{S}^{(2)}(+\infty, -\infty)$ is the second-order expansion of the time-evolution $S$ matrix and is defined by

$$\hat{S}^{(2)}(+\infty, -\infty) = \frac{1}{2!} \left( -\frac{i}{\hbar} \right)^2 \int_{-\infty}^{+\infty} dt_1 dt_2 T_i [\hat{V}_1(t_1)\hat{V}_1(t_2)] F_\theta(t_1, t_2).$$

Here the time function $F_\theta(t_1, t_2)$ is defined as $F_\theta(t_1, t_2) = F_\theta(t_1) F_\theta(t_2)$. Physically, the coincidence probability of the cINS is determined by the second-order perturbation of the electron-neutron magnetic interaction.

Following the theoretical treatment for the cARPES, we introduce a two-spin Bethe-Salpeter wave function,

$$\phi_{\alpha\beta}^{(i)}(q_{1}, q_{2}, t_2) = \langle \Psi_{\beta} | T_i S^{(i)}_{\perp}(q_{1}, t_{1}) S^{(j)}_{\perp}(q_{2}, t_{2}) | \Psi_{\alpha} \rangle.$$  

(18)

Obviously this two-spin Bethe-Salpeter wave function describes the time dynamical evolution of the magnetic material when two spins are excited in time ordering. Therefore, it describes the dynamical physics of the two-spin magnetic correlations of the target magnetic material.

With the two-spin Bethe-Salpeter wave function, we can show that the coincidence probability of the cINS can be expressed as

$$\Gamma(2) = \Gamma_1^{(2)} + \Gamma_2^{(2)},$$

(19)

where

$$\Gamma_1^{(2)} = \frac{1}{Z} \sum_{\alpha\beta, \beta'} e^{-\beta E_0} P_1(\beta_{i1})P_1(\beta_{i2})$$

$$\times \left| \int_{-\infty}^{+\infty} dt_1 dt_2 M_{\alpha, \beta, \beta', \beta} F_\theta(t_1, t_2) \right|^2,$$

$$\Gamma_2^{(2)} = \frac{1}{Z} \sum_{\alpha\beta, \beta'} e^{-\beta E_0} P_1(\beta_{i1})P_1(\beta_{i2})$$

$$\times \left| \int_{-\infty}^{+\infty} dt_1 dt_2 M_{\alpha, \beta, \beta, \beta, \beta} F_\theta(t_1, t_2) \right|^2.$$

Here the matrix elements $M_{\alpha, \beta, 1}$ and $M_{\alpha, \beta, 2}$ are defined as

$$M_{\alpha, \beta, 1} = \frac{g(q_{1})g(q_{2})}{\int_{-\infty}^{+\infty} dt_1 dt_2 M_{\alpha, \beta, \beta', \beta} F_\theta(t_1, t_2)}$$

$$\times \sigma_{\beta, \beta', \beta}^{(i)} \sigma_{\beta, \beta', \beta}^{(j)} \epsilon[(E_{i1}^{(2)} + E_{i2}^{(2)})/\hbar],$$

$$M_{\alpha, \beta, 2} = \frac{g(q_{1})g(q_{2})}{\int_{-\infty}^{+\infty} dt_1 dt_2 M_{\alpha, \beta, \beta, \beta, \beta} F_\theta(t_1, t_2)}$$

$$\times \sigma_{\beta, \beta', \beta}^{(i)} \sigma_{\beta, \beta', \beta}^{(j)} \epsilon[(E_{i1}^{(2)} + E_{i2}^{(2)})/\hbar],$$

where the transfer momenta are defined by

$$q_{1} = q_{f1} - q_{i1}, q_{2} = q_{f2} - q_{i2},$$

$$\mathbf{q}_{1} = q_{f1} - q_{i1}, \mathbf{q}_{2} = q_{f2} - q_{i1},$$

(20)

and the transfer energies are defined by

$$E_{i1}^{(2)} = E(q_{f1}) - E(q_{i1}), E_{i2}^{(2)} = E(q_{f2}) - E(q_{i2}),$$

$$\mathbf{E}_{1}^{(2)} = E(q_{f1}) - E(q_{i1}), \mathbf{E}_{2}^{(2)} = E(q_{f2}) - E(q_{i1}).$$

(21)
Physically, there are two different classes of the microscopic neutron scattering processes involved in the coincidence scattering. One is with the state change of the two neutrons as \( |\mathbf{q}_1, \beta_1\rangle \rightarrow |\mathbf{q}_1, \beta_1\rangle \rightarrow |\mathbf{q}_2, \beta_2\rangle \) and the other one is with \( |\mathbf{q}_1, \beta_1\rangle \rightarrow |\mathbf{q}_2, \beta_2\rangle \) and \( |\mathbf{q}_2, \beta_1\rangle \rightarrow |\mathbf{q}_1, \beta_1\rangle \). The matrix elements \( M_{\alpha \beta, 1} \) and \( M_{\alpha \beta, 2} \) and the corresponding coincidence probabilities \( \Gamma_1^{(2)} \) and \( \Gamma_2^{(2)} \) describe these two different classes of the microscopic neutron scattering processes, respectively. It should be noted that here we have ignored the quantum interference of these two different scattering contributions as they come from different scattering channels of energy transfer with the energy-conservation-like resonance features at different energies.

Define the center-of-mass time \( t_c = \frac{1}{2} (t_1 + t_2) \) and the relative time \( t_r = t_1 - t_2 \), and denote the two-spin Bethe-Salpeter wave function \( \phi_{\alpha \beta}^{(ij)} (q_1, q_2; t_c, t_r) = \phi_{\alpha \beta}^{(ij)} (q_{1t}, q_{2t}) \). We can introduce the Fourier transformations of \( \phi_{\alpha \beta}^{(ij)} (q_1, q_2; t_c, t_r) \) as below:

\[
\phi_{\alpha \beta}^{(ij)} (q_1, q_2; t_c, t_r) = \int \int d\Omega d\omega e^{i\Omega t_c - i\omega t_r},
\]

\[
\phi_{\alpha \beta}^{(ij)} (q_1, q_2; \Omega, \omega) = \int_0^{\infty} dt_c dt_r e^{i\Omega t_c + i\omega t_r}.
\]

For the incident thermal neutrons in the spin mixed state defined by Eq. 9, the coincidence probability is shown to follow

\[
\Gamma^{(2)} = 1 + \sum_{\alpha \beta ij} e^{-\beta E_\alpha} \times \left[ C_1 |\phi_{\alpha \beta, 1}^{(ij)} (q_1, q_2)|^2 + C_2 |\phi_{\alpha \beta, 2}^{(ij)} (\mathbf{q}_1, \mathbf{q}_2)|^2 \right],
\]

where the two factors are defined as

\[
C_1 = |g(q_1)g(q_2)|^2, C_2 = |g(\mathbf{q}_1)g(\mathbf{q}_2)|^2,
\]

and the two wave functions \( \phi_{\alpha \beta, 1}^{(ij)} (q_1, q_2) \) and \( \phi_{\alpha \beta, 2}^{(ij)} (\mathbf{q}_1, \mathbf{q}_2) \) are defined by

\[
\phi_{\alpha \beta, 1}^{(ij)} (q_1, q_2) = \int \int d\Omega d\omega e^{i\Omega t_c - i\omega t_r},
\]

\[
\phi_{\alpha \beta, 2}^{(ij)} (\mathbf{q}_1, \mathbf{q}_2) = \int \int d\Omega d\omega e^{i\Omega t_c - i\omega t_r}.
\]

In the large but finite \( \Delta t \), we can make an approximation that \( \Delta t / \Delta t \approx \Delta t / \Delta t \), and \( \Delta t / \Delta t \approx \Delta t / \Delta t \). In this case the functions \( Y_1(\Omega, \omega) \) and \( Y_2(\Omega, \omega) \) can be approximated as

\[
Y_1(\Omega, \omega) = \frac{\sin((\bar{E}_1^2 - \bar{E}_2^2) / h + \bar{E}_1^2 / h - \bar{E}_2^2 / h) \Delta t / 2}{(\bar{E}_1^2 - \bar{E}_2^2) / h - \bar{E}_1^2 / h - \bar{E}_2^2 / h) / 2}
\]

where the transfer frequencies are defined by

\[
\Omega_1 = \frac{1}{h} (E_1^2 + E_2^2), \omega_1 = \frac{1}{2h} (E_1^2 - E_2^2); \Omega_2 = \frac{1}{h} (\bar{E}_1^2 + \bar{E}_2^2), \omega_2 = \frac{1}{2h} (\bar{E}_1^2 - \bar{E}_2^2).
\]
function:

\[ \phi^{(ij)}_{\alpha\beta}(\mathbf{q}_1, \mathbf{q}_2; \Omega, \omega) = 2\pi \delta(\Omega + (E_\beta - E_\alpha)/\hbar) \phi^{(ij)}_{\alpha\beta}(\mathbf{q}_1, \mathbf{q}_2; \omega), \]

where \( \phi^{(ij)}_{\alpha\beta}(\mathbf{q}_1, \mathbf{q}_2; \omega) \) follows

\[ \phi^{(ij)}_{\alpha\beta}(\mathbf{q}_1, \mathbf{q}_2; \omega) = \sum_\gamma \left[ \frac{i\langle \Psi_\gamma | S^{(i)}(\mathbf{q}_1) | \Psi_\gamma \rangle \langle \Psi_\gamma | S^{(j)}(\mathbf{q}_2) | \Psi_\gamma \rangle}{\omega + i\delta^+ + (E_\alpha + E_\beta - 2E_\gamma)/2\hbar} - \frac{i\langle \Psi_\gamma | S^{(i)}(\mathbf{q}_2) | \Psi_\gamma \rangle \langle \Psi_\gamma | S^{(j)}(\mathbf{q}_1) | \Psi_\gamma \rangle}{\omega - i\delta^+ + (E_\alpha + E_\beta - 2E_\gamma)/2\hbar} \right]. \]

Obviously the frequency Bethe-Salpeter wave function involves the dynamical magnetic physics of two spins of the target magnetic material: (1) The center-of-mass dynamics of the two spins described by \( \delta(\Omega + (E_\beta - E_\alpha)/\hbar) \), which shows the energy transfer conservation with the center-of-mass degrees of freedom involved; (2) The two-spin relative dynamics defined by \( \phi^{(ij)}_{\alpha\beta}(\mathbf{q}_1, \mathbf{q}_2; \omega) \), which has the resonance structures peaked at \( \pm(E_\alpha + E_\beta - 2E_\gamma)/2\hbar \) with the weights \( \langle \Psi_\gamma | S^{(i)}(\mathbf{q}_1) | \Psi_\gamma \rangle \langle \Psi_\gamma | S^{(j)}(\mathbf{q}_2) | \Psi_\gamma \rangle \) and \( \langle \Psi_\gamma | S^{(j)}(\mathbf{q}_2) | \Psi_\gamma \rangle \langle \Psi_\gamma | S^{(i)}(\mathbf{q}_1) | \Psi_\gamma \rangle \), respectively. Therefore, the cINS can provide the momentum resolved dynamical two-spin magnetic correlations of the target magnetic material.

III. COINCIDENCE PROBABILITIES OF FERROMAGNET AND ANTIFERROMAGNET

In this section we will study the coincidence probabilities of the cINS for a ferromagnet and an antiferromagnet which have a long-range magnetic order with well-defined magnon excitations.

Provided that (1) the two incident neutrons are independent with the spin distribution function following \( P_2(\beta_1, \beta_2) = P_1(\beta_1)P_1(\beta_2) \), and (2) the single-spin magnetic excitations of the target material have well-defined momenta and are decoupled from each other. Under these conditions, the coincidence probability of the cINS has a simple product behavior, which can be expressed mathematically as

\[ \Gamma^{(2)} = \Gamma^{(1)}(\mathbf{q}_f, \mathbf{q}_i) \cdot \Gamma^{(1)}(\mathbf{q}_f, \mathbf{q}_i) + \Gamma^{(1)}(\mathbf{q}_f, \mathbf{q}_i) \cdot \Gamma^{(1)}(\mathbf{q}_f, \mathbf{q}_i). \]

This is a general result which can be exactly proven from the definition of the scattering probability of the INS Eq. [1] and the definition of the coincidence probability of the cINS Eq. [10].

We will consider the local spin magnetic systems with a cubic crystal lattice, the Hamiltonians of which are defined by

\[ H = \frac{J}{2} \sum_{i\delta} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta}, \]

where \( \delta = \pm \mathbf{e}_x, \pm \mathbf{e}_y, \pm \mathbf{e}_z \). The local spins are in a low-temperature ordered state with the magnetic moments ordered along \( \mathbf{e}_z \) axis.

A. Ferromagnet

Let us consider a ferromagnet with \( J > 0 \). Introduce the Holstein-Primakoff transformations, \( S^+_l = \sqrt{2S-a^+_l a_l}, S^-_l = a^+_l \sqrt{2S-a^+_l a_l}, S^z_l = S - a^+_l a_l \), where \( a^+_l \) and \( a_l \) are the bosonic magnon operators. In a linear spin-wave theory, the spin Hamiltonian can be approximated as

\[ H_{FM} = \sum_k \varepsilon_k a_k^\dagger a_k, \]

where \( \varepsilon_k = |J| z S (1 - \gamma_k) \) with \( \gamma_k = \frac{1}{2} \sum_l e^{ik \cdot R_l} \) and the coordination number \( z = 6 \). Here \( a_k^\dagger = \frac{1}{\sqrt{N}} \sum_l a^+_l e^{-ik \cdot R_l} \).

Let us firstly study the scattering probability in the single-spin INS. Suppose the incident thermal neutrons are in the spin mixed state defined by Eq. [9]. It can be shown from Eq. [11] that the scattering probability \( \Gamma^{(1)} \) follows

\[ \Gamma^{(1)}_{FM}(\mathbf{q}_f, \mathbf{q}_i) = \frac{|g(\mathbf{q})|^2 \Delta \lambda}{\hbar} n_B(E^{(1)}) \left[ \chi_{xx}(\mathbf{q}, E^{(1)}) (1 - q_x^2) + \chi_{yy}(\mathbf{q}, E^{(1)}) (1 - q_y^2) + \chi_{zz}(\mathbf{q}, E^{(1)}) (1 - q_z^2) \right], \]

where the spin spectrum functions \( \chi_{ii}(\mathbf{q}, E) \) are given by

\[ \chi_{xx}(\mathbf{q}, E) = \chi_{yy}(\mathbf{q}, E) = \pi N S [\delta(E - \varepsilon_\mathbf{q}) - \delta(E + \varepsilon_\mathbf{-q})], \]

\[ \chi_{zz}(\mathbf{q}, E) = 2\pi \sum_k [n_B(\varepsilon_k) - n_B(\varepsilon_{k+\mathbf{q}})] \delta(E + \varepsilon_k - \varepsilon_{k+\mathbf{q}}). \]

Here the transfer momentum \( \mathbf{q} \) and the transfer energy \( E^{(1)} \) are defined in Eq. [12]. While the transverse spin flips lead to the single-magnon peak structures in the scattering probability, the longitudinal spin fluctuations contribute the magnon density fluctuations. Besides these inelastic scattering contributions, there is one additional elastic scattering contribution from the magnon
condensation, which gives
\[ \Gamma_{FM,c}^{(1)} = \frac{2\pi |g(q)|^2 \Delta t}{\hbar} (Nm_{FM})^2 \delta(E^{(1)}) \delta_{q,0}(1 - \frac{q^2}{4}) , \]
where \( m_{FM} = \frac{1}{N} \sum_i \langle S_i^z \rangle \) is the ordered spin magnetic moment per site. It is noted that in experiment, \( N \) is the number of the local Wannier electron spins in the incident neutron beam. When only consider the single-magnon contributions without that of the magnon density fluctuations, the inelastic scattering probability of the INS for the ordered ferromagnet follows
\[ \Gamma_{FM}^{(1)}(q_f, q_i) = \frac{\pi NS|g(q)|^2 \Delta t}{\hbar} n_B(E^{(1)})(1 + \frac{q^2}{4}) \times [\delta(E^{(1)} - \epsilon_q) - \delta(E^{(1)} + \epsilon_{-q})] . \]

Now let us study the coincidence probability of the cINS for the ordered ferromagnet. Suppose the two incident thermal neutrons with momenta \( q_1 \) and \( q_2 \) are scattered into the states with momenta \( q_{f1} \) and \( q_{f2} \). Consider the incident neutrons are in the spin states with \( P_2(\beta_1, \beta_2) = P_1(\beta_1)P_1(\beta_2) \) and \( P_1(\beta_1) \) defined by Eq. (33). Since the magnons are well-defined single-spin excitations with the momentum as a good quantum number, the coincidence probability of the cINS for the ordered ferromagnet has a general behavior described by Eq. (34), i.e.,
\[ \Gamma_{FM}^{(2)}(q_f, q_i) = \Gamma_{FM}^{(1)}(q_{f1}, q_i) \cdot \Gamma_{FM}^{(1)}(q_{f2}, q_i) + \Gamma_{FM}^{(1)}(q_{f1}, q_i) \cdot \Gamma_{FM}^{(1)}(q_{f2}, q_i) \]
where the four \( \Gamma_{FM}^{(1)}(q_f, q_i) \)'s are the scattering probabilities of the single-spin INS defined in Eq. (33), with the relevant transfer momenta and energies defined in Eq. (30) and (31).

### B. Antiferromagnet

Now let us consider an antiferromagnet in a cubic crystal lattice with a long-range magnetic order. It has a spin lattice Hamiltonian defined by Eq. (35) with \( J > 0 \). Introduce the spin rotation transformations as \( \tilde{S}_i^z = e^{iQ \cdot R_i} S_i^z \), \( \tilde{S}_i^y = S_i^y \), \( \tilde{S}_i^x = e^{iQ \cdot R_i} S_i^x \), where \( Q = (\pi/a, \pi/a, \pi/a) \) is the characteristic antiferromagnetic momentum. Introduce the Holstein-Primakoff transformations for the new spin operators, the spin Hamiltonian can be approximated in a linear spin-wave theory as
\[ H_{AF} = \sum_k \psi_k^\dagger \begin{pmatrix} A & B_k \\ B_k & A \end{pmatrix} \psi_k = \begin{pmatrix} c_k^\dagger \\ c_{-k} \end{pmatrix} \begin{pmatrix} \beta_k \\ \beta_{-k}^{\dagger} \end{pmatrix} , \]
where \( A = J_z S \) and \( B_k = -J_z S \gamma_k \), and \( \psi_k \) is a bosonic Nambu spinor operator. Here the sum over \( k \) involves each pair \((k, -k)\) once. With the canonical transformations
\[ \begin{pmatrix} c_k \\ c_{-k}^\dagger \end{pmatrix} = \begin{pmatrix} u_k & v_k \\ v_k & u_k \end{pmatrix} \begin{pmatrix} \beta_k \\ \beta_{-k}^{\dagger} \end{pmatrix} , \]
the Hamiltonian can be diagonalized into the form:
\[ H_{AF} = \sum_k E_k (\beta_k^\dagger \beta_k + \beta_{-k} \beta_{-k}^{\dagger}) , \]
where \( E_k = \sqrt{A^2 - B_k^2} \). Here \( u_k^2 = \frac{A + E_k}{2E_k} \), \( v_k^2 = \frac{A - E_k}{2E_k} \), and \( u_k v_k = -\frac{B_k}{2E_k} \).

It can be easily shown that the neutron scattering probability of the INS for the ordered antiferromagnet, \( \Gamma_{AF}^{(1)}(q_f, q_i) \), follows a similar expression to Eq. (37) of \( \Gamma_{FM}^{(1)}(q_f, q_i) \), with the corresponding spin spectrum functions \( \chi_{ii}(q, E) \) given by
\[ \chi_{zz}(q_f, E) = \chi_{yy}(q_f, E) = \pi NS \frac{A + Bq}{E_q} \frac{\delta(E - E_q) - \delta(E + E_q)}{1} , \]
and
\[ \chi_{zz}(q, E) = 2\pi \sum_k \left\{ \left[ C_{kq}^{(1)} \delta(E - \epsilon_k - \epsilon_{k+q} + Q) - C_{kq}^{(4)} \delta(E - \epsilon_k - \epsilon_{k+q} + Q) \right] n_B(\epsilon_k) - n_B(\epsilon_{k+q} + Q) \right\} + \left[ C_{kq}^{(2)} \delta(E - \epsilon_k - \epsilon_{k+q} + Q) - C_{kq}^{(3)} \delta(E + \epsilon_k + \epsilon_{k+q} + Q) \right] [n_B(\epsilon_k) + n_B(\epsilon_{k+q} + Q)] \}
\]

Here \( C_{kq}^{(1)} = u_k^2 + Q u_k^2 + u_{k+q}^2 + Q u_{k+q}^2 u_k^2 + u_{k+q}^2 + Q u_{k+q}^2 u_k^2 \), \( C_{kq}^{(2)} = u_k^2 + Q u_k^2 + u_{k+q}^2 + Q u_{k+q}^2 u_k^2 \), \( C_{kq}^{(3)} = u_k^2 + Q u_k^2 + u_{k+q}^2 + Q u_{k+q}^2 u_k^2 \), \( C_{kq}^{(4)} = u_k^2 + Q u_k^2 + u_{k+q}^2 + Q u_{k+q}^2 u_k^2 \). Similar to the ordered ferromagnet, there is also one additional elastic scattering contribution due to the magnon condensation,
\[ \Gamma_{AF,c}^{(1)} = \frac{2\pi |g(q)|^2 \Delta t}{\hbar} (Nm_{AF})^2 \delta(E^{(1)}) \delta_{q,0}(1 - \frac{q^2}{4}) , \]
where $m_{AF} = \sum_i e^{\mathbf{Q} \cdot \mathbf{R}_i} \langle S_i^z \rangle$ is the ordered antiferromagnetic moment per site. Here the transfer momentum $\mathbf{q}$ and the transfer energy $E^{(1)}$ are also defined in Eq. (12). In the approximation with only the single-magnon contributions, the inelastic scattering probability of the INS for the ordered antiferromagnet follows

$$
\Gamma_{AF}^{(1)}(\mathbf{q}_1, \mathbf{q}_2) = \frac{\pi N S |g(\mathbf{q})|^2 \Delta t A + B_{\mathbf{q}}}{E_{\mathbf{q}}} n_{B}(E^{(1)}(1 + \mathbf{q}^2)[\delta(E^{(1)} - E_{\mathbf{q}}) - \delta(E^{(1)} + E_{\mathbf{q}})]).
$$

Now let us consider the cINS with the thermal neutrons which have initial incident momenta $\mathbf{q}_1$ and $\mathbf{q}_2$ and final scattered momenta $\mathbf{q}_1'$ and $\mathbf{q}_2'$. The incident neutrons are independent with the spin state defined by Eq. (4). In the linear spin-wave theory defined by the approximate Hamiltonian Eq. (22), the Nambu spinor operators with different momenta are decoupled. This manifests that the single-spin magnon excitations in the ordered antiferromagnet are decoupled. Therefore the conditions for the product behavior of the coincidence probability in Eq. (34) are also satisfied in the ordered antiferromagnet. The coincidence probability of the cINS for the ordered antiferromagnet follows a similar product behavior defined by

$$
\Gamma_{AF}^{(2)} = \Gamma_{AF}^{(1)}(\mathbf{q}_1, \mathbf{q}_1') \cdot \Gamma_{AF}^{(1)}(\mathbf{q}_2, \mathbf{q}_2')
$$

where the four $\Gamma_{AF}^{(1)}(\mathbf{q}_i, \mathbf{q}_i')$’s are the scattering probabilities of the single-spin INS, which are similarly defined to $\Gamma_{FM}^{(1)}(\mathbf{q}_i, \mathbf{q}_i)$ in Eq. (37) with the relevant spin spectrum functions defined by Eq. (45) and (46) and the transfer momenta and energies defined in Eq. (20) and (21).

IV. DISCUSSION AND SUMMARY

In this article we have proposed a coincidence technique, the cINS, which has two neutron sources and two neutron detectors with an additional coincidence detector. The two neutron sources emit two neutrons which are scattered by the electron spins of the magnetic material and are then detected by the two neutron detectors. The coincidence detector records the coincidence probability of the two scattered neutrons, which shows the information on a two-spin Bethe-Salpeter wave function. This two-spin Bethe-Salpeter wave function defines the momentum resolved dynamical wave function of the magnetic material with two spins excited. Thus the cINS can detect explicitly the two-spin magnetic correlations of the magnetic material. The coincidence probabilities of the cINS for a ferromagnet and an antiferromagnet with a long-range magnetic order are calculated, which show a general product behavior contributed from the single-spin INS’s.

On the experimental instrument of the cINS, we remark that the two incident neutrons can come from one neutron source. In this case the initial momenta of the two incident neutrons follow $\mathbf{q}_1 = \mathbf{q}_1 + \delta_q$ with $\delta_q \to 0$. The theoretical formalism for the cINS with one neutron source can be similarly established following the one we have established in Sec. (113) for the cINS with two neutron sources.

The cINS we have proposed is one potential technique to study the novel magnetic correlations which are far beyond the physics of the single-spin magnons. For example, the long-sought quantum spin liquids from strong frustration and quantum fluctuations show novel physics, such as the various spin valance bond states and the novel quantum criticality. Experimental study on the spin valance bond states by the cINS would provide new insights on the quantum spin liquids. The various quantum magnetic materials with spin dimers, such as TiCuCl$_2$, SrCu$_2$(BO$_3$)$_2$ and BaCuSi$_2$O$_6$, can be the first focus in the cINS experiment. The quantum spin liquid materials in triangular, kagome, and hyperkagome lattices (e.g., the materials reviewed in Ref. [4]) are also the interesting target materials for the cINS experiment.

As a summary, we have proposed a coincidence technique, the cINS, which can detect explicitly the two-spin magnetic correlations of the magnetic materials. It can be introduced to study the dynamical physics of the spin valance bond states of the quantum magnets.

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Appendix A: Electron-neutron magnetic interaction

Let us review the electron-neutron magnetic interaction. Define the neutron spin magnetic moment as $\mu_n = -\gamma \mu_N \sigma$, where $\gamma = 1.91$ is a constant for the neutron gyromagnetic ratio, $\mu_N = \frac{e \gamma}{2m_p}$ is the nuclear magneton with $m_p$ the proton mass, and $\sigma$ is the Pauli spin operator. Define the electron spin magnetic moment as $\mu_e = -g_e B \mathbf{S}$ and the electron orbital magnetic moment as $g_i = -g_i B \mathbf{L}$, where the $g$-factors are set as $g_e = 2$ and $g_i = 1$, and $\mu_B = \frac{e\hbar}{2m_e}$ is the Bohr magneton. The spin angular momentum operator $\mathbf{S}$ has eigenvalues $\pm \frac{1}{2}$ and the orbital angular momentum
operator is defined as \( \mathbf{L} = \frac{1}{2} \mathbf{r}_e \times \mathbf{p}_e \). Suppose there is an electron at position \( \mathbf{r}_e \), which can produce a magnetic field at position \( \mathbf{r}_n \) as

\[
\mathbf{B} = \frac{\mu_0}{4\pi} \nabla \times \left[ (\mu_n + \mu_l) \times \frac{\mathbf{R}}{R^3} \right],
\]

where \( \mu_0 \) is the free-space permeability and \( \mathbf{R} = \mathbf{r}_n - \mathbf{r}_e \).

The electron-neutron magnetic interaction can be defined by \( V = -\mathbf{\mu}_n \cdot \mathbf{B} \), which follows

\[
V = \frac{\mu_0}{4\pi} \gamma \mu_N \mu_B \sigma \cdot \nabla \times \left[ (q_e \mathbf{S} + gi \mathbf{L}) \times \frac{\mathbf{R}}{R^3} \right].
\]

(A2)

Here we have introduced the orbital angular momentum \( \mathbf{L} \) to describe the orbital motions of the electrons. It is more convenient in study of the orbital motions of the electrons in the compounds with transition metal and/or rare earth ions.

Let us present the second quantization of the electron-neutron magnetic interaction. Introduce the single-neutron states \( \{q_{\beta}\} \) where \( q \) is the neutron momentum and \( \beta \) defines the neutron spin, and the single-electron states \( \{|\lambda\} \) where \( \lambda \) involves the momentum, orbital, and spin degrees of freedom, etc. Let us introduce the following identities:

\[
1 = \frac{1}{V_1} \int d\mathbf{r}_e |\mathbf{r}_e\rangle \langle \mathbf{r}_e|
\]

for the electrons, and

\[
1 = \frac{1}{V_2} \int d\mathbf{r}_n |\mathbf{r}_n\rangle \langle \mathbf{r}_n|
\]

for the neutrons. Here \( V_1 \) and \( V_2 \) are the renormalization volumes for the single-electron and single-neutron states, respectively. The electron-neutron magnetic interaction in second quantization can be expressed as

\[
\hat{V} = \hat{V}_s + \hat{V}_l,
\]

(A3)

where

\[
\hat{V}_s = \frac{4\pi A_s}{V_2} \sum_{\mathbf{q}, \mathbf{q}'} \hat{\sigma}_{\mathbf{q}, \mathbf{q}'} \cdot [\hat{\mathbf{q}} \times (\mathbf{D}^s(\mathbf{q}) \times \hat{\mathbf{q}})],
\]

(A4)

\[
\hat{V}_l = \frac{4\pi A_l}{V_2} \sum_{\mathbf{q}, \mathbf{q}'} \hat{\sigma}_{\mathbf{q}, \mathbf{q}'} \cdot [\hat{\mathbf{q}} \times (\mathbf{D}^l(\mathbf{q}) \times \hat{\mathbf{q}})].
\]

(A5)

Here the momentum \( \mathbf{q} = \mathbf{q}_l - \mathbf{q}_e \) and \( \hat{\mathbf{q}} = \frac{\mathbf{q}}{q} \). It is noted that \( \hat{\mathbf{q}} \times (\mathbf{D}(\mathbf{q}) \times \hat{\mathbf{q}}) \) can be reexpressed as \( \mathbf{D}_\perp(\mathbf{q}) \):

\[
\mathbf{D}_\perp(\mathbf{q}) = \mathbf{D}(\mathbf{q}) - \hat{\mathbf{q}}(\mathbf{D}(\mathbf{q}) \cdot \hat{\mathbf{q}}).
\]

(A6)

In the electron-neutron magnetic interaction \( \hat{V} \), the constants \( A_s \) and \( A_l \) are defined as

\[
A_s = -\frac{\mu_0}{4\pi} \gamma g_s \mu_N \mu_B, \quad A_l = -\frac{\mu_0}{4\pi} \gamma g_l \mu_N \mu_B,
\]

(A7)

and the operator \( \hat{\sigma}_{\mathbf{q}, \mathbf{q}'} \) is defined by

\[
\hat{\sigma}_{\mathbf{q}, \mathbf{q}'} = \sum_{\beta, \beta'} d_{\beta \beta'}^\dagger \mathbf{\sigma}_{\beta \beta', \beta' \beta} d_{\beta' \beta},
\]

(A8)

where \( d_{\beta \beta'} \) and \( d_{\beta' \beta}^\dagger \) are the annihilation and creation operators for the neutrons. The operators \( \mathbf{D}_s \) and \( \mathbf{D}_l \) in \( \hat{V} \) are defined by

\[
\mathbf{D}^s(\mathbf{q}) = \sum_{\lambda_1 \lambda_2} c_{\lambda_2 \lambda_1}^s \mathbf{M}^{(s)}(\lambda_2 \lambda_1)(\mathbf{q}) c_{\lambda_1 \lambda_2}, \quad (A9)
\]

\[
\mathbf{D}^l(\mathbf{q}) = \sum_{\lambda_1 \lambda_2} c_{\lambda_2 \lambda_1}^l \mathbf{M}^{(l)}(\lambda_2 \lambda_1)(\mathbf{q}) c_{\lambda_1 \lambda_2}, \quad (A10)
\]

where \( c_{\lambda} \) and \( c_{\lambda}^\dagger \) are the annihilation and creation operators for the electrons, and

\[
\mathbf{M}^{(s)}(\lambda_2 \lambda_1)(\mathbf{q}) = \frac{1}{V_1} \int d\mathbf{r}_e [\psi^{*}_{\lambda_2}(\mathbf{r}_e) \mathbf{S} \psi_{\lambda_1}(\mathbf{r}_e)] e^{-i\mathbf{q} \cdot \mathbf{r}_e},
\]

\[
\mathbf{M}^{(l)}(\lambda_2 \lambda_1)(\mathbf{q}) = \frac{1}{V_1} \int d\mathbf{r}_e [\psi^{*}_{\lambda_2}(\mathbf{r}_e) \mathbf{L} \psi_{\lambda_1}(\mathbf{r}_e)] e^{-i\mathbf{q} \cdot \mathbf{r}_e}.
\]

Here \( \psi_{\lambda}(\mathbf{r}_e) \) is the single-electron wave function.

Let us focus on the spin degrees of freedom of the electrons and ignore the orbital ones. Consider the electrons are in the local Wannier states \( \{|\alpha\} \) with position \( \mathbf{R}_l \) and spin \( \alpha \). \( \mathbf{D}^s(\mathbf{q}) \) can be approximately defined as

\[
\mathbf{D}^s(\mathbf{q}) = F_0(\mathbf{q}) \mathbf{S}(\mathbf{q}),
\]

(A11)

where the spin operator \( \mathbf{S}(\mathbf{q}) \) is defined by

\[
\mathbf{S}(\mathbf{q}) = \sum_{\alpha} \mathbf{S}_\alpha e^{-i\mathbf{q} \cdot \mathbf{R}_l}, \quad \mathbf{S}_\alpha = \sum_{\alpha\beta} c_{\alpha \beta}^\dagger \mathbf{S}_{\alpha \beta} c_{\beta \alpha}, \quad (A12)
\]

and the magnetic form factor \( F_0(\mathbf{q}) \) is given by

\[
F_0(\mathbf{q}) = \frac{1}{V_1} \int d\mathbf{a} \psi^{*}_\alpha(\mathbf{a}) \psi_\alpha(\mathbf{a}) e^{-i\mathbf{q} \cdot \mathbf{a}}, \quad \mathbf{a} = \mathbf{r}_e - \mathbf{R}_l.
\]

(A13)

Here we have made an approximation to consider only the on-site intra-orbital integrals and ignore all other contributions. For the itinerant electrons in the Bloch states \( \{|\mathbf{k}\alpha\} \), the operator \( \mathbf{D}^s(\mathbf{q}) \) can be given by

\[
\mathbf{D}^s(\mathbf{q}) = \sum_{\mathbf{k}, \mathbf{k}'} F_{\mathbf{k}, \mathbf{k}'}(\mathbf{q}) \mathbf{S}_{\mathbf{k}, \mathbf{k}'}(\mathbf{q}),
\]

(A14)

where the spin operator is defined by

\[
\mathbf{S}_{\mathbf{k}, \mathbf{k}'} = \sum_{\alpha\beta} c_{\alpha \beta}^\dagger \mathbf{S}_{\alpha \beta} c_{\beta \alpha},
\]

(A15)

and the form factor \( F_{\mathbf{k}, \mathbf{k}'}(\mathbf{q}) \) is given by

\[
F_{\mathbf{k}, \mathbf{k}'}(\mathbf{q}) = \frac{1}{V_1} \int d\mathbf{r}_e \psi^{*}_{\mathbf{k}'}(\mathbf{r}_e) \psi_{\mathbf{k}}(\mathbf{r}_e) e^{-i\mathbf{q} \cdot \mathbf{r}_e}.
\]

(A16)
Here $\psi_k(r_e)$ is the Bloch-state wave function. In the approximation with $\psi_k(r_e) = e^{ik\cdot r_e}$, $D^s(q)$ can be simplified as

$$D^s(q) = \sum_k S_{k,k+q}. \quad (A17)$$

As a summary, the electron-neutron magnetic interaction with only the spin degrees of freedom of the electrons can be given as follows. For the local Wannier electrons,

$$\hat{V}_s = \sum_{q_i,q_j} g(q) \hat{\sigma}_{q_i,q_j} \cdot S(q). \quad (A18)$$

where $g(q) \equiv gF_0(q)$ with $g = \frac{4\pi A}{\nu^2}$. $\hat{\sigma}_{q_i,q_j}$ is defined in Eq. (A8) and $S(q)$ is defined in Eq. (A12). For the itinerant Bloch electrons,

$$\hat{V}_s = \sum_{q_i,q_j,k_{1},k_{2}} g_{k_{1}k_{2}}(q) \hat{\sigma}_{q_i,q_j} \cdot S_{k_{1}k_{2},\perp}. \quad (A19)$$

where $g_{k_{1}k_{2}}(q) \equiv gF_{k_{1}k_{2}}(q)$ and $S_{k_{1}k_{2}}$ is defined in Eq. (A15). Here $S_{\perp} = S - \bar{q}(S \cdot \bar{q})$ with $q = q_f - q_i$ and $\bar{q} = \frac{q_f + q_i}{2}$. It should be noted that the form factors $F_F(q)$ and $F_{k_{1}k_{2}}(q)$ defined in Eq. (A13) and (A16), have strong $q$ dependence.

One remark is that in the above electron-neutron magnetic interaction, the contributions from the spin and orbital magnetic moments are independently derived. This is for the case where the spin-orbit coupling is weak such as for the electrons of the transition metal ions. In the case with a strong spin-orbit coupling such as for the electrons of the rare earth ions, it is the total angular momentum $\mathbf{J}$ that is conserved. In this case we can introduce the total magnetic moment $\mu_{J} = -g(JLS)\mu_B\mathbf{J}$ with the Landé $g$-factor $g(JLS)$ defined following $g_L = g_s = g(JLS)\mathbf{J}$. A similar derivation can give us an electron-neutron magnetic interaction in this case. Another remark is that the Debye-Waller factor from the crystal lattice effects is ignored here.

Appendix B: Calculations for scattering probability of INS

Let us introduce the imaginary time Green’s functions $G_{ij}(q,\tau) = -(T\tau S_i(q,\tau)S_j^\dagger(q,0))$ with $i,j = x, y, z$. The corresponding spectrum functions are defined as $\chi_{ij}(q,E) = -2\text{Im}G_{ij}(q,i\nu_n \rightarrow E + i\delta^+)$). Then we have

$$G(q,\tau) = -\sum_{ij} G_{ij}(q,\tau)(\delta_{ij} - \hat{q}_i\hat{q}_j). \quad (B1)$$

and

$$\chi(q,E) = -\sum_{ij} \chi_{ij}(q,E)(\delta_{ij} - \hat{q}_i\hat{q}_j). \quad (B2)$$

Firstly let us consider the ferromagnet in a cubic crystal lattice with a long-range magnetic order. Introduce the imaginary time Green’s function for the ferromagnetic magnons, $G_a(q,\tau) = -(T\tau \psi_k(\tau)\psi_k^\dagger(0))$. Its frequency Fourier transformation is given by

$$G_a(k,i\nu_n) = \frac{1}{i\nu_n - \varepsilon_k}. \quad (B3)$$

where the magnon energy dispersion $\varepsilon_k$ is defined in Eq. (36). It can be shown that in the linear spin-wave approximation,

$$G_{xx}(q,i\nu_n) = G_{yy}(q,i\nu_n) = \frac{NS}{2}[G_a(q,i\nu_n) + G_a(-q,-i\nu_n)], \quad (B4)$$

and

$$G_{zz}(q,i\nu_n) = -\frac{1}{\beta} \sum_{k,i\nu_{1}} G_a(k+q,i\nu_1 + i\nu_n)G_a(k,i\nu_1). \quad (B5)$$

The other Green’s functions follow

$$G_{ij}(q,i\nu_n) = 0, \text{ for } i \neq j. \quad (B6)$$

From these results, we can obtain the spectrum functions $\chi_{ij}(q,E)$ in Eq. (38) for the ordered ferromagnet.

Now let us consider the antiferromagnet in a cubic crystal lattice with a long-range magnetic order. Introduce the imaginary time Green’s function of a Nambu spinor operator,

$$G_{\psi}(k,\tau) = -(T\tau \psi_k(\tau)\psi_k^\dagger(0)), \quad (B7)$$

where $\psi_k$ is defined in Eq. (42). It can be shown that the frequency Green’s function follows

$$G_{\psi}(k,i\nu_n) = \frac{i\nu_n \tau_3 + A - B_k \tau_1}{(i\nu_n)^2 - E_k}, \quad (B8)$$

where $A$ and $B_k$ are defined in Eq. (42) and the magnon energy $E_k$ is given in Eq. (44). Here $\tau_i$ ($i = 1, 2, 3$) are the Pauli matrices.

It can be shown that

$$G_{xx}(q,i\nu_n) = \frac{NS}{2}\text{Tr}[G_{\psi}(q + Q,i\nu_n) + G_{\psi}(q + Q,i\nu_n)]$$

$$G_{yy}(q,i\nu_n) = \frac{NS}{2}\text{Tr}[G_{\psi}(q,i\nu_n) - G_{\psi}(q,i\nu_n)]$$

and
The other Green’s functions \( G_{ij}(q, \nu_n) = 0 \) for the cases with \( i \neq j \). With these results, we can obtain the spectrum functions \( \chi_{ij}(q, E) \) in Eq. (45) and (46) for the ordered antiferromagnet.