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Quantum fluctuations in antiferromagnets

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Abstract. Dynamic properties of antiferromagnets at zero temperature are explored. Going beyond RPA a nonlinear set of coupled equations for dynamic susceptibilities and correlation functions is derived. New results are presented for nondiagonal elements of the dynamic susceptibility tensor, longitudinal fluctuations and their influence on transverse fluctuations.

PACS. 75.10.Jm Quantized spin models – 75.30.Ds Spin waves

1 Introduction

This paper deals with a very old subject: fluctuations – or equivalently the excitation spectrum – of antiferromagnets. Standard spin-wave theory for antiferromagnets is textbook knowledge [1]. Dynamics of antiferromagnets at low temperature, in particular thermal fluctuations and their influence on spin wave damping and hydrodynamics, was studied in a classical paper by Harris, Kumar, Halperin and Hohenberg [2]. The advent of high T_c superconductivity triggered an extensive literature on quasi 2-dimensional antiferromagnets [2,3].

Nevertheless the effects of pure quantum fluctuations (at temperature $T = 0$), particularly important for small spin, on the excitation spectrum are not well studied, even in 3 dimensions. Very little is known, for example, about longitudinal quantum fluctuations. From general sum rules it follows, that the total weight of longitudinal quantum fluctuation is substantial [4], (for $S = \frac{1}{2}$ a sizable fraction of the transverse fluctuations), but their spectral distribution is largely unknown. Braune and Maleev [5] partly addressed this problem. A further subject – unexplored up to now – concerns the off-diagonal elements of the dynamic susceptibility tensor.

In the following we develop a selfconsistent scheme to describe longitudinal and transverse quantum fluctuations and their mutual influence, including all elements of the dynamic susceptibility tensor, diagonal and off-diagonal.

2 Preliminary remarks

The subject of discussion will be dynamical properties of antiferromagnets at zero temperature. To be specific we use the Heisenberg model

$$H = -\sum J_{AB}\mathbf{S}_A \cdot \mathbf{S}_B \tag{1}$$

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$\mathbf{S}_A$, $\mathbf{S}_B$ are spin operators, A and B lattice indices. The crystal lattice forms a simple "a–b structure", giving rise to a simple antiferromagnetic ground state, the modulation being described by a wavevector $\mathbf{Q}$. The ordered moment defines the z-direction

$$\langle \mathbf{S}_A \rangle = d\,\mathrm{e}^{\mathrm{i}\mathbf{Q}\cdot\mathbf{R}_A}\mathbf{e}_z \tag{2}$$

$\mathbf{e}_z$ is the unit vector into the z-direction; $\mathbf{R}_A$ is a lattice vector, $\exp\{\mathrm{i}\mathbf{Q}\mathbf{R}_A\} = \pm 1$; 2 $\mathbf{Q}$ equals a reciprocal lattice vector; the brackets $\langle\quad\rangle$ correspond to the (symmetry broken) ground state expectation value.

We will calculate the dynamic susceptibility tensor

$$\chi^{\nu\mu}_{AB}(t) \equiv -\mathrm{i}\Theta(t)\langle[S^\nu_A(t), S^\mu_B(0)]\rangle, \tag{3}$$

ν, μ are Cartesian indices (x, y, z).

Fourier transforms are defined as

$$\chi^{\nu\mu}_{\mathbf{k}\mathbf{k}'}(\omega) = \frac{1}{N}\sum_{A,B}\int \mathrm{d}t\mathrm{e}^{\mathrm{i}\omega t}\mathrm{e}^{-\mathrm{i}\mathbf{k}\mathbf{R}_A}\mathrm{e}^{\mathrm{i}\mathbf{k}'\mathbf{R}_B}\chi^{\nu\mu}_{AB}(t) \tag{4}$$

$$\chi^{\nu\mu}_{AB}(t) = \frac{1}{N}\sum_{\mathbf{k}\mathbf{k}'}\int \frac{\mathrm{d}\omega}{2\pi}\mathrm{e}^{-\mathrm{i}\omega t}\mathrm{e}^{+\mathrm{i}\mathbf{k}\mathbf{R}_A}\mathrm{e}^{-\mathrm{i}\mathbf{k}'\mathbf{R}_B}\chi^{\nu\mu}_{\mathbf{k},\mathbf{k}'}(\omega). \tag{5}$$

Due to the symmetry of the problem the components

$$\chi^{xx} = \chi^{yy} \equiv \chi^+, \tag{6}$$

$$\chi^{xy} = -\chi^{yx} \equiv \mathrm{i}\chi^-, \tag{7}$$

$$\chi^{zz} \equiv \chi^{(3)} \tag{8}$$

will be finite, while the other components of the susceptibility tensor vanish:

$$\chi^{xz} = \chi^{zx} = \chi^{yz} = \chi^{zy} = 0. \tag{9}$$

It is convenient to define 3 by 3 matrices,

$$P_1 = \frac{1}{2}\begin{pmatrix} 1 & \mathrm{i} & 0 \\ -\mathrm{i} & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{10}$$

$$P_2 = \frac{1}{2}\begin{pmatrix} 1 & -\mathrm{i} & 0 \\ \mathrm{i} & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{11}$$

$$P_3 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{12}$$

The P_i have projector properties:

$$P_i P_j = \delta_{ij} P_i, \tag{13}$$

$$\sum_{i=1}^{3} P_i = 1 \quad \text{(the } 3\times 3 \text{ unit matrix)}. \tag{14}$$

With their aid we decompose

$$\begin{aligned} \chi &= P_1\chi^{(1)} + P_2\chi^{(2)} + P_3\chi^{(3)} \\ &= (P_1+P_2)\chi^{+} + (P_1-P_2)\chi^{-} + P_3\chi^{(3)}; \end{aligned} \tag{15}$$

$$\chi^{\pm} = \frac{1}{2}(\chi^{(1)} \pm \chi^{(2)}). \tag{16}$$

For technical reasons we introduce the "vector" $\mathbf{L}$, whose components L_x, L_y, L_z are 3 by 3 matrices:

$$L_x = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & -1 & 0 \end{pmatrix}; L_y = \begin{pmatrix} 0 & 0 & -1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}; L_z = \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{17}$$

The commutation relations take the form:

$$[\mathbf{S}_A, \mathbf{S}_B] = \mathrm{i}\mathbf{S}_A \cdot \mathbf{L}\delta_{AB}. \tag{18}$$

We use units such that $\hbar = 1$.

3 Derivation of the selfconsistency equations

In tensor form the equation of motion for the susceptibility

$$\chi_{AB}(t) = -\mathrm{i}\Theta(t)\langle[\mathbf{S}_A(t), \mathbf{S}_B(0)]\rangle \tag{19}$$

becomes

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}t}\chi_{AB}(t) &= -\mathrm{i}\delta(t)\langle[\mathbf{S}_A, \mathbf{S}_B]\rangle - \mathrm{i}\Theta(t)\langle\big[\mathrm{i}[H, \mathbf{S}_A(t)], \mathbf{S}_B\big]\rangle \\ &= -\mathrm{i}\delta(t)\langle[\mathbf{S}_A, \mathbf{S}_B]\rangle \\ &\quad - \mathrm{i}\Theta(t)\sum_C 2J_{CA}\langle[\mathbf{S}_C \cdot (\mathbf{S}_A\mathbf{L}), \mathbf{S}_B(-t)]\rangle. \end{aligned} \tag{20}$$

Operators $\mathbf{S}$ not containing an explicit time argument are to be taken at $t = 0$.

We define

$$K_{AB} \equiv \langle[\mathbf{S}_A, \mathbf{S}_B]\rangle = \mathrm{i}\langle\mathbf{S}_A\mathbf{L}\rangle\delta_{AB} \tag{21}$$

and

$$\widetilde{\chi}^{(3)}_{CAB}(t) \equiv -\mathrm{i}\Theta(t)\langle[\mathbf{S}_C \cdot (\mathbf{S}_A\mathbf{L}), \mathbf{S}_B(-t)]\rangle. \tag{22}$$

Equation (20) now takes the form:

$$\frac{\mathrm{d}}{\mathrm{d}t}\chi_{AB}(t) = -\mathrm{i}\delta(t)K_{AB} + 2\sum_C J_{CA}\widetilde{\chi}^{(3)}_{CAB}(t). \tag{23}$$

The equation of motion for $\widetilde{\chi}^{(3)}$ reads:

$$\frac{\mathrm{d}}{\mathrm{d}t}\widetilde{\chi}^{(3)}_{CAB}(t) = -\mathrm{i}\delta(t)R_{CAB} + 2\sum_D J_{DB}\chi^{(4)}_{CADB}(t), \tag{24}$$

where we introduced

$$R_{CAB} \equiv \langle[\mathbf{S}_C \cdot (\mathbf{S}_A\mathbf{L}), \mathbf{S}_B]\rangle, \tag{25}$$

and

$$\chi^{(4)}_{CADB}(t) \equiv \mathrm{i}\Theta(t)\langle[\mathbf{S}_C(t) \cdot (\mathbf{S}_A(t)\mathbf{L}), \mathbf{S}_D \cdot (\mathbf{S}_B\mathbf{L})]\rangle. \tag{26}$$

Fourier transformation with respect to time yields:

$$\omega\chi_{AB}(\omega) = K_{AB} + \mathrm{i}2\sum_C J_{CA}\widetilde{\chi}^{(3)}_{CAB}(\omega), \tag{27}$$

$$\omega\widetilde{\chi}^{(3)}_{CAB} = R_{CAB} + \mathrm{i}2\sum_D J_{DB}\chi^{(4)}_{CADB}(\omega). \tag{28}$$

Combining these two equations leads to

$$\begin{aligned} \omega^2\chi_{AB}(\omega) &= \omega K_{AB} + \mathrm{i}2\sum_C J_{CA}R_{CAB} \\ &\quad - 4\sum_{CD} J_{CA}J_{DB}\chi^{(4)}_{CADB}(\omega). \end{aligned} \tag{29}$$

The selfconsistency equations for χ are obtained by factorizing $\chi^{(4)}$ (Eq. 26) into products of spin-spin correlation functions and components of the susceptibility tensor itself.

Explicitly the (ν, μ) component of the susceptibility tensor $\chi^{(4)}_{CADB}$ is defined as

$$\chi^{(4)}_{\nu\mu} = \mathrm{i}\theta(t)\sum_{\substack{ii' \\ jj'}}\langle[S^i_C(t)S^j_A(t)L^j_{i\nu}, S^{i'}_D S^{j'}_B L^{j'}_{i'\mu}]\rangle. \tag{30}$$

If we factorize we obtain (we use $L^j_{i\nu} = -L^i_{j\nu}$, etc.):

$$\begin{aligned} \chi^{(4)}_{\nu\mu}(t) = -\sum_{jj'}\Big\{ &\langle(\mathbf{S}_C(t)\mathbf{L})_{j\nu}(\mathbf{S}_D\mathbf{L})_{j'\mu}\rangle\chi^{jj'}_{AB}(t) \\ &- \langle(\mathbf{S}_C(t)\mathbf{L})_{j\nu}(\mathbf{S}_B\mathbf{L})_{j'\mu}\rangle\chi^{jj'}_{AD}(t) \\ &- \langle(\mathbf{S}_D\mathbf{L})_{j'\mu}(\mathbf{S}_A(t)\mathbf{L})_{j\nu}\rangle\chi^{jj'}_{CB}(t) \\ &+ \langle(\mathbf{S}_B\mathbf{L})_{j'\mu}(\mathbf{S}_A(t)\mathbf{L})_{j\nu}\rangle\chi^{jj'}_{CD}(t)\Big\}. \end{aligned} \tag{31}$$

The correlation functions appearing as factors of the susceptibilities are connected to these *via* the fluctuation dissipation theorem. We define:

$$\Gamma_{AB}^{\nu\mu}(t) \equiv \langle S_A^{\nu}(t) S_B^{\mu} \rangle, \tag{32}$$

and its Fourier transform

$$\Gamma_{AB}^{\nu\mu}(\omega) = \int \mathrm{d}t \mathrm{e}^{\mathrm{i}\omega t} \Gamma_{AB}^{\nu\mu}(t). \tag{33}$$

Furthermore:

$$\chi_{AB}^{\prime\prime\nu\mu}(\omega) = \frac{1}{2\mathrm{i}} \big(\chi_{AB}^{\nu\mu}(\omega) - \chi_{BA}^{\mu\nu}(-\omega) \big). \tag{34}$$

Then the fluctuation dissipation theorem states:

$$\frac{2}{1 - \mathrm{e}^{-\beta\omega}} \chi_{AB}^{\prime\prime\nu\mu}(\omega) = -\Gamma_{AB}^{\nu\mu}(\omega). \tag{35}$$

This, together with equations (21, 25, 26, 29, 31) constitutes a set of selfconsistency equations for the dynamic susceptibility tensor and the corresponding spin-spin correlation functions. Equation (31) constitutes an approximation, its validity will be discussed later. A remark concerning previous derivations of selfconsistency equations for spin correlation functions: equation of motion techniques and decouplings of higher order functions to obtain a closed set of course exist in the literature [7]. Previous decoupling schemes, however, did not allow for a fully selfconsistent treatment of frequency dependencies including longitudinal fluctuations and off-diagonal elements of the susceptibility tensor.

4 RPA-approximation

Although the RPA is *not* adequate to describe quantum fluctuations, we nevertheless present it as a starting point. The purpose is twofold: we make contact to previous discussions (with partly well known results), and we demonstrate the inherent deficiencies of RPA. In the following Section 5 we will go beyond RPA to remedy these deficiencies.

The RPA is obtained from the selfconsistency equations by an additional drastic approximation: of the correlation functions appearing in equation (31) only the time independent part ($\omega = 0$) is retained, all time dependent contributions ($\omega \neq 0$) to the correlation functions are neglected. (A remark about the meaning of the word "static". Traditionally the "static susceptibility" means the $\omega = 0$ susceptibility, the "static correlation function" means the equal time ($t = 0$) function, *i.e.* the integral over all frequencies. Unfortunately this has become common use, we will try to avoid confusion by explicitly mentioning $t = 0$ or $\omega = 0$ whenever necessary.)

We have chosen the ordered moment to point in the z-direction (Eq. (2)), therefore the factorization of $\chi^{(4)}$ (Eq. (31)) will retain only the $\omega = 0$ part of the $S_z - S_z$ correlation function: RPA is obtained from equation (31) by substituting:

$$\langle S_A^j(t) S_B^{j'} \rangle \to \delta_{j,z}\delta_{j',z} d^2 \mathrm{e}^{\mathrm{i}\mathbf{Q}(\mathbf{R}_A - \mathbf{R}_B)}. \tag{36}$$

The spin-space matrix multiplications on the right hand side of equation (31) will then reduce to

$$\begin{aligned} \sum_{jj'} L_{j\nu}^z L_{j'\mu}^z \chi^{jj'} &= -(L^z \chi L^z)_{\nu\mu} \\ &= \Big((P_1 + P_2)\chi \Big)_{\nu\mu} \end{aligned} \tag{37}$$

(above we used $L^z = -\mathrm{i}(P_1 - P_2)$ and $\chi = P_1\chi^{(1)} + P_2\chi^{(2)} + P_3\chi^{(3)}$).

In RPA the quantity R defined in equation (25) takes the form (again only the $\omega = 0$ part of the $S_z - S_z$ correlation function is retained):

$$\begin{aligned} &2\mathrm{i} \sum_C J_{CA} R_{CAB} \to \\ &\quad - 2(P_1 + P_2) \sum_C (\delta_{CB} - \delta_{AB}) d^2 \mathrm{e}^{\mathrm{i}\mathbf{Q}(\mathbf{R}_C - \mathbf{R}_B)} J_{CA}. \end{aligned} \tag{38}$$

The quantity K defined in equation (21) does not require an approximation. According to equation (2) we have

$$K_{AB} = (P_1 - P_2)\delta_{AB} d \mathrm{e}^{\mathrm{i}\mathbf{Q}\mathbf{R}_A}. \tag{39}$$

The RPA-solution of equation (29) can now be obtained by Fourier transformation with respect to the lattice indices.

In general:

$$K_{\mathbf{k}\mathbf{k}'} = (P_1 - P_2) d \delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}. \tag{40}$$

We use the notation

$$(JR)_{AB} \equiv \sum_C J_{CA} R_{CAB}. \tag{41}$$

RPA according to equation (38) leads to

$$2\mathrm{i}(JR)_{\mathbf{k},\mathbf{k}'} = (P_1 + P_2) 2d^2 (J_{\mathbf{Q}} - J_{\mathbf{k}-\mathbf{Q}})\, \delta_{\mathbf{k},\mathbf{k}'}. \tag{42}$$

For the last term on the right hand side of equation (29) we use the notation

$$(JJ\chi^{(4)})_{AB} \equiv \sum_{CD} J_{CA} J_{DB} \chi_{CADB}^{(4)}. \tag{43}$$

Substituting the RPA approximation of equation (36) into the factorization equation (31) will enable us to carry out the Fourier transformation with respect to the lattice indices. The Fourier transform of χ itself will appear in the RPA form of $\chi^{(4)}$.

We define:

$$\chi_{\mathbf{k},\mathbf{k}'} \equiv \chi_{\mathbf{k}}^{(0)}\, \delta_{\mathbf{k},\mathbf{k}'} + \chi_{\mathbf{k}}^{(\mathbf{Q})} \delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}. \tag{44}$$

In RPA we obtain:

$$-4(JJ\chi^{(4)})_{\mathbf{k},\mathbf{k}'} = 4(P_1+P_2)d^2\Big\{\delta_{\mathbf{k}\mathbf{k}'}\chi^{(0)}_{\mathbf{k}+\mathbf{Q}}(J_{\mathbf{Q}}-J_{\mathbf{k}-\mathbf{Q}})^2 + \delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}\chi^{(\mathbf{Q})}_{\mathbf{k}+\mathbf{Q}}(J_{\mathbf{Q}}-J_{\mathbf{k}-\mathbf{Q}})(J_{\mathbf{Q}}-J_{\mathbf{k}})\Big\}. \quad (45)$$

Collecting the various contributions equation (29) becomes

$$\begin{aligned}\omega^2\chi_{\mathbf{k},\mathbf{k}'}(\omega) &= (P_1-P_2)d\omega\delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}} \\ &+ (P_1+P_2)2d^2(J_{\mathbf{Q}}-J_{\mathbf{k}-\mathbf{Q}})\delta_{\mathbf{k},\mathbf{k}'} \quad (46)\\ &+ 4(P_1+P_2)d^2\Big\{\chi^{(0)}_{\mathbf{k}+\mathbf{Q}}(J_{\mathbf{Q}}-J_{\mathbf{k}-\mathbf{Q}})^2\delta_{\mathbf{k},\mathbf{k}'} \\ &+ \chi^{(\mathbf{Q})}_{\mathbf{k}+\mathbf{Q}}(J_{\mathbf{Q}}-J_{\mathbf{k}-\mathbf{Q}})(J_{\mathbf{Q}}-J_{\mathbf{k}})\delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}\Big\}. \quad (47)\end{aligned}$$

From $\chi = P_1\chi^{(1)} + P_2\chi^{(2)} + P_3\chi^{(3)}$ we immediately obtain

$$\omega^2\chi^{(3)} = \omega^2\chi^{zz} = 0. \quad (48)$$

In RPA the longitudinal dynamic susceptibility vanishes identically! This actually is one of the essential deficiencies of RPA, we will have to correct this deficiency to obtain an internally consistent picture (this will be done in Sect. 5).

The RPA results for the susceptibilities in the plane perpendicular to the ordered moment are obtained from equation (29) by solving for $\chi^{\pm} = \frac{1}{2}(\chi^{(1)} \pm \chi^{(2)})$. We obtain two sets of 2 coupled equations by setting $\mathbf{k}' = \mathbf{k}$ and $\mathbf{k}' = \mathbf{k}+\mathbf{Q}$. The solutions are straightforward: the RPA result for the full susceptibility tensor is:

$$\begin{aligned}\chi_{\mathbf{k},\mathbf{k}'}(\omega) &= \delta_{\mathbf{k},\mathbf{k}'}(P_1+P_2)\frac{dE_{\mathbf{k}+\mathbf{Q}}}{\omega^2-\epsilon^2_{\mathbf{k}}} \\ &+ \delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}(P_1-P_2)\frac{\omega d}{\omega^2-\epsilon^2_k} \\ &= \delta_{\mathbf{k},\mathbf{k}'}\frac{dE_{\mathbf{k}+\mathbf{Q}}}{\omega^2-\epsilon^2_{\mathbf{k}}}\begin{pmatrix}1&0&0\\0&1&0\\0&0&0\end{pmatrix} \\ &+ \delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}\frac{d\omega}{\omega^2-\epsilon^2_k}\begin{pmatrix}0&\mathrm{i}&0\\-\mathrm{i}&0&0\\0&0&0\end{pmatrix}. \quad (49)\end{aligned}$$

Here the following definitions were used:

$$E_{\mathbf{k}} = 2d(J_{\mathbf{Q}}-J_{\mathbf{k}}); \quad (50)$$

$$\epsilon^2_k = E_{\mathbf{k}}\,E_{\mathbf{k}+\mathbf{Q}} = \epsilon^2_{\mathbf{k}+\mathbf{Q}}. \quad (51)$$

A few remarks: the diagonal elements of the tensor are standard and well-known; they are diagonal in wavevector; the response in the direction of the applied field (perpendicular to the ordered moment) is at the same wavevector as that of the applied field. The off-diagonal elements are not so well-known: *E.g.* an applied field of wave vector $\mathbf{k}$ in the x-direction also generates a response in the y-direction of wavevector $\mathbf{k}+\mathbf{Q}$. This describes a precession of the ordered moment around the applied field. To the authors knowledge these off-diagonal elements of the dynamic susceptibility had not been published before [7]. Real and imaginary parts of the dynamic susceptibility are obtained in the usual way by taking $\omega \to \omega + \mathrm{i}\eta$ (η being infinitesimally small and positive for real ω). The RPA results from equation (49) are:

$$\begin{aligned}\chi''^{xx}_{\mathbf{k},\mathbf{k}'} &= \chi''^{yy}_{\mathbf{k},\mathbf{k}'} \\ &= -\frac{\pi}{2}d\sqrt{\frac{E_{\mathbf{k}+\mathbf{Q}}}{E_{\mathbf{k}}}}(\delta(\omega-\epsilon_{\mathbf{k}})-\delta(\omega+\epsilon_{\mathbf{k}}))\,\delta_{\mathbf{k},\mathbf{k}'}; \quad (52)\\ \chi''^{xy}_{\mathbf{k},\mathbf{k}'} &= -\chi''^{yx}_{\mathbf{k},\mathbf{k}'} = -\mathrm{i}\frac{\pi}{2}d(\delta(\omega-\epsilon_{\mathbf{k}})+\delta(\omega+\epsilon_{\mathbf{k}}))\,\delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}; \quad (53)\end{aligned}$$

RPA gives χ'' as sharp δ-functions. For fixed $\mathbf{k}, \chi''$ is zero everywhere except for $\omega_k = \pm\epsilon_{\mathbf{k}}$, corresponding to an infinitely sharp spin wave excitation. The prefactors of the δ-functions (usually called "spin wave form factors") describe the coupling strength of the applied field to these excitations.

For the diagonal elements this strength vanishes as $|\mathbf{k}|$ for $|\mathbf{k}|$ going to zero and diverges as $\mathbf{k}$ approaches $\mathbf{Q}$ as $\frac{1}{|\kappa|}$, where $\kappa = \mathbf{k}-\mathbf{Q}$. For the offdiagonal elements RPA gives the coupling strength as $\mathbf{k}$-independent. For the latter $\mathrm{i}\chi''$ is real, the factor i signaling a phase shift by $\frac{\pi}{2}$.

5 Beyond RPA

RPA as derived in the previous chapter is internally inconsistent. The sum rule

$$\sum_\nu \int \frac{\mathrm{d}\omega}{2\pi}\Gamma^{\nu\nu}_{AA}(\omega) = \sum_\nu \langle S^\nu_A S^\nu_A\rangle = S(S+1), \quad (54)$$

and the fluctuation dissipation theorem (Eq. (35)) cannot be reconciled with $\chi^{zz} = 0$ for finite ω (Eq. (48)). The ordered moment d for the antiferromagnetic ground state is necessarily smaller than S and this unambiguously requires a finite longitudinal susceptibility [4]. This is most easily seen for $S = \frac{1}{2}$, where 3 independent sum rules exist for the 3 components due to $\langle S^\nu_A S^\nu_A\rangle = \frac{1}{4}$.

The contribution of $\Gamma^{zz}(\omega)$ to the sum rule coming from $\omega = 0$ (describing the ordered moment) is d^2, necessarily smaller than $(\frac{1}{2})^2$, therefore there must be a finite ω contribution to satisfy the sum rule.

The necessary existence of longitudinal quantum fluctuations will also have a strong influence on the spectral functions for the transverse susceptibilities: the single δ-functions obtained in equations (52, 53) are an artefact of RPA.

To correct these deficiencies, we go back to the set of selfconsistency equations derived in Section 2. In a first step we do not attempt full selfconsistency, but we use a type of perturbation theory beyond RPA: for the time dependent correlation functions in equation (31) and the equal time correlation functions in equation (25) we use RPA results. With their aid we then obtain the dynamic susceptibilities from equation (29).

The quantity R (Eq. (25)) contains equal time correlation functions of the type

$$\langle S_A^\nu S_B^\mu \rangle \equiv \gamma_{AB}^{\nu\mu} = \Gamma_{AB}^{\nu\mu}(t=0). \tag{55}$$

To *obtain* RPA results the approximation equation (42) only retained the $S^z - S^z$ correlation function (furthermore its $\omega = 0$ part only). However, the RPA results generate finite $\gamma^{xx} = \gamma^{yy}$ as well as $\gamma^{xy} = -\gamma^{yx}$ through application of the fluctuation dissipation theorem. These contributions originate from finite frequencies.

General symmetry requires:

$$\gamma^{xx} = \gamma^{yy}; \gamma^{xy} = -\gamma^{yx}; \gamma^{xz} = \gamma^{yz} = \gamma^{zx} = \gamma^{zy} = 0. \tag{56}$$

This leads to

$$(JR)_{AB}^{xx} = -\mathrm{i} \sum_C J_{CA}(\delta_{AB} - \delta_{CB})(\gamma_{CA}^{xx} + \gamma_{CA}^{zz}) \equiv (JR)_{AB}^{+}; \tag{57}$$

$$(JR)_{AB}^{xy} = \mathrm{i} \sum_C J_{CA}(\delta_{AB} + \delta_{CB})\gamma_{CA}^{xy} \equiv \mathrm{i}(JR)_{AB}^{-}; \tag{58}$$

$$(JR)_{AB}^{zz} = -\mathrm{i} \sum_C J_{CA}(\delta_{AB} - \delta_{CB})\, 2\gamma_{CA}^{xx} \equiv (JR)_{AB}^{(3)}. \tag{59}$$

In compact form:

$$(JR) = (P_1 + P_2)(JR)^+ + (P_1 - P_2)(JR)^- + P_3(JR)^{(3)}. \tag{60}$$

Concerning the decomposition of $\chi^{(4)}$ (Eq. (31)) we use the symmetry conditions:

$$\Gamma_{AB}^{xx}(t) = \langle S_A^x(t) S_B^x \rangle = \Gamma_{AB}^{yy}(t) \equiv \Gamma_{AB}^{+}(t); \tag{61}$$

$$\Gamma_{AB}^{xy}(t) = -\Gamma_{AB}^{yx}(t) \equiv \mathrm{i}\Gamma_{AB}^{-}(t). \tag{62}$$

It follows:

$$\begin{aligned}\sum_{jj'} \langle (\mathbf{S}_C(t)\mathbf{L})_{j\nu} (\mathbf{S}_D\mathbf{L})_{j'\mu} \rangle \chi_{AB}^{jj'}(t) = \\ \Gamma_{CD}^{+}(t) \Big\{ 2\chi_{AB}^{+}(t)(P_3)_{\nu\mu} + \chi_{AB}^{(3)}(t)(P_1 + P_2)_{\nu\mu} \Big\} \\ - \Gamma_{CD}^{-}(t) \Big\{ 2\chi_{AB}^{-}(t)(P_3)_{\nu\mu} - \chi_{AB}^{(3)}(t)(P_1 - P_2)_{\nu\mu} \Big\} \\ + \Gamma_{CD}^{(3)}(t) \Big\{ \chi_{AB}^{+}(t)(P_1 + P_2)_{\nu\mu} + \chi_{AB}^{-}(t)(P_1 - P_2)_{\nu\mu} \Big\};\end{aligned} \tag{63}$$

$$\begin{aligned}\sum_{jj'} \langle (\mathbf{S}_B\mathbf{L})_{j'\mu} (\mathbf{S}_A(t)\mathbf{L})_{j\nu} \rangle \chi_{CD}^{jj'}(t) = \\ \Gamma_{BA}^{+}(-t) \Big\{ 2\chi_{CD}^{+}(t)(P_3)_{\nu\mu} + \chi_{CD}^{(3)}(t)(P_1 + P_2)_{\nu\mu} \Big\} \\ + \Gamma_{BA}^{-}(-t) \Big\{ 2\chi_{CD}^{-}(t)(P_3)_{\nu\mu} - \chi_{CD}^{(3)}(t)(P_1 - P_2)_{\nu\mu} \Big\} \\ + \Gamma_{AB}^{(3)}(-t) \Big\{ \chi_{CD}^{+}(t)(P_1 + P_2)_{\nu\mu} + \chi_{AB}^{-}(t)(P_1 - P_2)_{\nu\mu} \Big\}.\end{aligned} \tag{64}$$

Several general conclusions are already possible concerning the general structure of the Fourier-transformed χ

$$\chi_{\mathbf{k},\mathbf{k}'} = (P_1 + P_2)\chi_{\mathbf{k},\mathbf{k}'}^{+} + (P_1 - P_2)\chi_{\mathbf{k},\mathbf{k}'}^{-} + P_3\chi_{\mathbf{k},\mathbf{k}'}^{(3)}. \tag{65}$$

In RPA $\chi^{(3)}$ vanished and for $\chi^{\pm}$ we obtained

$$\chi_{\mathbf{k},\mathbf{k}'}^{+} \sim \delta_{\mathbf{k},\mathbf{k}'}; \chi_{\mathbf{k},\mathbf{k}'}^{-} \sim \delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}. \tag{66}$$

Beyond RPA the full selfconsistency equations will maintain the property of equation (66) and furthermore $\chi^{(3)}$, now finite, will be

$$\chi_{\mathbf{k},\mathbf{k}'}^{(3)} \sim \delta_{\mathbf{k},\mathbf{k}'}. \tag{67}$$

This follows from equations (40, 57–59, 63, 64) from which the general structure of $K_{\mathbf{k},\mathbf{k}'}, (JR)_{\mathbf{k},\mathbf{k}'}$ and the factorized form of $\chi_{\mathbf{k},\mathbf{k}'}^{(4)}$ can be extracted.

From equation (29) we can project out the factors of $(P_1 + P_2)$ and P_3: we either have contributions containing the diagonal elements of correlation functions ($\Gamma^{\nu\nu}$) and susceptibilities ($\chi^{\nu\nu}$), which are $\sim \delta_{\mathbf{k},\mathbf{k}'}$, or even powers of correlation functions (Γ^{xy}) and susceptibilities (χ^{xy}). Γ^{xy} and χ^{xy} being proportional to $\delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}$, the even powers will give a translationally invariant quantity again.

Projecting out the factors of $(P_1 - P_2)$ we obtain a contribution from $K_{\mathbf{k},\mathbf{k}'} \sim \delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}$; the other contributions contain odd powers of the correlation function Γ^{xy} or the offdiagonal part of the susceptibility χ^{xy}, which are proportional to $\delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}}$.

Using this general property equations (66, 67) we define:

$$\begin{aligned}\chi_{\mathbf{k},\mathbf{k}'} = (P_1 + P_2)\chi_{\mathbf{k}}^{+}\, \delta_{\mathbf{k},\mathbf{k}'} + (P_1 - P_2)\chi_{\mathbf{k}}^{-}\delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}} \\ + P_3\chi_{\mathbf{k}}^{(3)}\delta_{\mathbf{k},\mathbf{k}'};\end{aligned} \tag{68}$$

and similarly:

$$\begin{aligned}\Gamma_{\mathbf{k},\mathbf{k}'} = (P_1 + P_2)\Gamma_{\mathbf{k}}^{+}\, \delta_{\mathbf{k},\mathbf{k}'} + (P_1 - P_2)\Gamma_{\mathbf{k}}^{-}\delta_{\mathbf{k},\mathbf{k}'-\mathbf{Q}} \\ + P_3\Gamma_{\mathbf{k}}^{(3)}\delta_{\mathbf{k},\mathbf{k}'}.\end{aligned} \tag{69}$$

The equal time correlation functions $\gamma^+, \gamma^-, \gamma^{(3)}$ are defined equivalently ($\gamma = \Gamma(t=0)$).

The different components of the susceptibility tensor are obtained from equation (29) with the aid of equations (31, 59, 64) for the factorized form of $\chi^{(4)}$: we obtain 3 coupled equations by projecting out the factors of $(P_1 + P_2), (P_1 - P_2)$, and P_3: Fourier transformation yields:

$$\begin{aligned}
\omega^2\chi_k^+ &= 2\sum_{\mathbf{q}}(\gamma_{\mathbf{q}}^+ + \gamma_{\mathbf{q}}^{(3)})(J_{\mathbf{q}} - J_{\mathbf{k-q}}) \\
&+ 4\sum_{q}\Big\{(\Gamma_q^+\chi_{k-q}^{(3)} + \Gamma_q^{(3)}\chi_{k-q}^+)J_{\mathbf{q}}(J_{\mathbf{q}} - J_{\mathbf{k-q}}) \\
&- (\widehat{\Gamma}_q^+\chi_{k-q}^{(3)} + \widehat{\Gamma}_q^{(3)}\chi_{k-q}^+)J_{\mathbf{k-q}}(J_{\mathbf{q}} - J_{\mathbf{k-q}})\Big\}; \\
\omega^2\chi_k^- &= \omega d + 2\mathrm{i}\sum_{\mathbf{q}}\gamma_{\mathbf{q}}^-(J_{\mathbf{q}} + J_{\mathbf{k-q-Q}}) \qquad (70)\\
&+ 4\sum_{q}\Big\{\Gamma_q^{(3)}\chi_{k-q}^- J_{\mathbf{q}}(J_{\mathbf{q}} - J_{\mathbf{k-q-Q}}) \\
&- \widehat{\Gamma}_q^{(3)}\chi_{k-q}^- J_{\mathbf{k-q}}(J_{\mathbf{q}} - J_{\mathbf{k-q-Q}}) \\
&+ \Gamma_q^-\chi_{k-q}^{(3)}J_{\mathbf{q}}(J_{\mathbf{q+Q}} - J_{\mathbf{k-q}}) \\
&- \widehat{\Gamma}_q^-\chi_{k-q}^{(3)}J_{\mathbf{k-q}}(J_{\mathbf{q+Q}} - J_{\mathbf{k-q}})\Big\}; \qquad (71)\\
\omega^2\chi_k^{(3)} &= 4\sum_{\mathbf{q}}\gamma_{\mathbf{q}}^+(J_{\mathbf{q}} - J_{\mathbf{k-q}}) \\
&+ 8\sum_{q}\Big\{\Gamma_q^+\chi_{k-q}^+ J_{\mathbf{q}}(J_{\mathbf{q}} - J_{\mathbf{k-q}}) \\
&- \widehat{\Gamma}_q^+\chi_{k-q}^+ J_{\mathbf{k-q}}(J_{\mathbf{q}} - J_{\mathbf{k-q}}) \\
&- \Gamma_q^-\chi_{k-q}^- J_{\mathbf{q}}(J_{\mathbf{q-Q}} - J_{\mathbf{k-q-Q}}) \\
&+ \widehat{\Gamma}_q^-\chi_{k-q}^- J_{\mathbf{k-q}}(J_{\mathbf{q-Q}} - J_{\mathbf{k-q-Q}})\Big\}. \qquad (72)
\end{aligned}$$

In the preceding equations we use:

$$q \equiv (\mathbf{q}, q_0); k \equiv (\mathbf{k}, \omega); \sum_q \equiv \sum_{\mathbf{q}}\int\frac{\mathrm{d}q_0}{2\pi}\cdot \qquad (73)$$

For example: χ_{k-q} is an abbreviation for $\chi_{\mathbf{k-q}}(\omega - q_0)$. Furthermore we introduced:

$$\widehat{\Gamma}_{\mathbf{q}}^+(q_0) \equiv \Gamma_{-\mathbf{q}}^+(-q_0); \qquad (74)$$

$$\widehat{\Gamma}_{\mathbf{q}}^{(3)}(q_0) \equiv \Gamma_{-\mathbf{q}}^{(3)}(-q_0); \qquad (75)$$

$$\widehat{\Gamma}_{\mathbf{q}}^-(q_0) \equiv -\Gamma_{-\mathbf{q}-\mathbf{Q}}^-(-q_0). \qquad (76)$$

6 Longitudinal fluctuations

Equations (70–72) constitute a set of nonlinear selfconsistency equations for the susceptibilities

$$\begin{aligned}\chi^+ &= \chi^{xx} = \chi^{yy}\\ \chi^- &= -\mathrm{i}\chi^{xy} = \mathrm{i}\chi^{yx};\\ \chi^{(3)} &= \chi^{zz},\end{aligned}$$

and the correlation functions

$$\begin{aligned}\Gamma^+ &= \Gamma^{xx} = \Gamma^{yy};\\ \Gamma^- &= -\mathrm{i}\Gamma^{xy} = \mathrm{i}\Gamma^{yx},\\ \Gamma^{(3)} &= \Gamma^{zz}.\end{aligned}$$

Recall that correlation functions and susceptibilities are connected *via* the fluctuation-dissipation theorem (Eq. (35)).

In this paper we are not attempting to reach full selfconsistency. Instead on the right hand side of equations (70–72) we are going to use results obtained in perturbation theory. This may be viewed as the beginning of an iteration procedure.

We start with equation (72), which gives the longitudinal dynamic susceptibility in terms of $\Gamma^\pm$ and $\chi^\pm$. In a first step we use results for $\Gamma^\pm$ and $\chi^\pm$ obtained in RPA approximation.

For $T = 0$ and $\omega > 0$ RPA according to equations (52, 53) yields:

$$\Gamma_{\mathbf{k}}^+(\omega) = \pi d\sqrt{\frac{E_{\mathbf{k+Q}}}{E_{\mathbf{k}}}}\delta(\omega - \epsilon_k); \qquad (77)$$

$$\Gamma_{\mathbf{k}}^-(\omega) = \pi d\delta(\omega - \epsilon_k). \qquad (78)$$

Inserting the RPA results into the right hand side of equation (72) we obtain the spectral function $\chi^{(3)\prime\prime}$:

$$\begin{aligned}
&\omega^2\chi_{\mathbf{k}}^{(3)\prime\prime}(\omega) = \\
&\quad -\pi 2d^2\sum_{\mathbf{q}}\{(\delta(\omega - \epsilon_{\mathbf{q}} - \epsilon_{\mathbf{k-q}}) - \delta(\omega + \epsilon_{\mathbf{q}} + \epsilon_{\mathbf{k-q}}))\} \\
&\Big\{J_{\mathbf{q}}(J_{\mathbf{q}} - J_{\mathbf{k-q}})\sqrt{\frac{E_{\mathbf{q+Q}}\,E_{\mathbf{k-q+Q}}}{E_{\mathbf{q}}\,E_{\mathbf{k-q}}}} + J_{\mathbf{q}}(J_{\mathbf{q+Q}} - J_{\mathbf{k-q+Q}})\Big\}.
\end{aligned} \qquad (79)$$

The result for $\chi^{(3)\prime\prime}$ constitutes a continuum, which appears in addition to the sharp δ-functions obtained in RPA for $\chi^{(+)\prime\prime}$. The sharp δ-functions of $\chi^{(+)\prime\prime}$ are the usual single spin wave excitations. Perturbation theory, which yielded equation (79), in low order then gives a two spin wave continuum. Its general properties are:

i) the continuum starts at the single spin wave spectrum, extending to higher energies;
ii) the width in energy depends on wavevector k; the maximum width is reached for $\mathbf{k}$ tending towards zero and towards $\mathbf{Q}$ (the magnetic Bragg vector); the minimum in width is reached for $\mathbf{k}$ tending towards $\mathbf{Q}/2$;
iii) the strength of the continuum for a given $\mathbf{k}$, *i.e.* the integral over ω,

$$\gamma_{\mathbf{k}}^{(3)} \sim \int_0^\infty \frac{\mathrm{d}\omega}{2\pi}\chi_{\mathbf{k}}^{(3)\prime\prime}(\omega)$$

is maximum for $\mathbf{k}$ towards $\mathbf{Q}/2$, where the width is smallest, leading to reasonably sharp excitation in that part of $(\mathbf{k}, \omega)$ space. The strength $\gamma_{\mathbf{k}}^{(3)}$ vanishes for $\mathbf{k}$ tending towards zero and $\mathbf{Q}$, where the width in ω is maximum, both effects combine to give negligible contributions for $\mathbf{k} \approx (0, \mathbf{Q})$. For $\mathbf{k} = 0$ this is due to the conservation law for the total spin, for $\mathbf{k} = \mathbf{Q}$ longitudinal fluctuations vanish due to the existence of the (macroscopic) order parameter (the staggered magnetization);
iv) concerning the frequency dependence of the continuum: close to the spinwave energy ϵ_k the continuum starts from zero, increasing towards higher frequencies as $(\omega - \epsilon_k)^2$. This is due to phase-space factors

(we discuss 3 dimensions) in the integral over $\mathbf{q}$ in equation (79), where an overall downward curvature in the dispersion relation $\epsilon_{\mathbf{k}}$ is assumed.

These general conclusions become rather obvious when we adopt the simplest model for the exchange constants J_{AB}, retaining one nearest neighbor interaction J only. For this simple case the expression in the second curly brackets of equation (79) reduces to

$$J_{\mathbf{q}}(J_{\mathbf{q}} - J_{\mathbf{k-q}}) \left\{ \sqrt{\frac{(J_{\mathbf{Q}} + J_{\mathbf{q}})((J_{\mathbf{Q}} + J_{\mathbf{k-q}})}{(J_{\mathbf{Q}} - J_{\mathbf{q}})(J_{\mathbf{Q}} - J_{\mathbf{k-q}}))}} - 1 \right\}. \quad (80)$$

The prefactor of the curly bracket above vanishes for $\mathbf{k} \to 0$, the curly bracket itself vanishes for $\mathbf{k} \to \mathbf{Q}$.

Away from these special values the curly bracket becomes singular as $|\mathbf{q}|^{-1}$ or $|\mathbf{k} - \mathbf{q}|^{-1}$ towards $\mathbf{Q}$. In three dimensions these singularities do not lead to any problems in the integration over $\mathbf{q}$, due to small phase space. The marginal dimension is two: at $T = 0$ (here we are discussing quantum fluctuations only) the integral over $\mathbf{q}$ still remains finite. The divergence of the integral in one dimension signals the usual instability of long range order.

The longitudinal fluctuations will in turn change the spectral function of transverse fluctuations. Perturbatively we may insert the result obtained in (79) into equations (70, 71): this will yield a continuum (in addition to the sharp δ-functions of Eq. (51)):

$$\begin{aligned} \delta\chi_{\mathbf{k}}^{(+)\prime\prime}(\omega) = -4\pi d^3 \sum_{\mathbf{q},\mathbf{q}'} \sqrt{\frac{E_{\mathbf{q+Q}}}{E_{\mathbf{q}}}} G_{\mathbf{q}',\mathbf{k-q}}(J_{\mathbf{q}} - J_{\mathbf{k-q}})^2 \\ \times \frac{1}{\omega^2 - \epsilon_k^2} \{\delta(\omega - \epsilon_{\mathbf{q}} - \epsilon_{\mathbf{q}'} - \epsilon_{\mathbf{k-q-q}'}) \\ -\delta(\omega + \epsilon_{\mathbf{q}} + \epsilon_{\mathbf{q}'} + \epsilon_{\mathbf{k-q-q}'})\}, \end{aligned} \quad (81)$$

where we have used

$$\begin{aligned} G_{\mathbf{q},\mathbf{k}} = J_{\mathbf{q}}(J_{\mathbf{q}} - J_{\mathbf{k-q}}) \sqrt{\frac{E_{\mathbf{q+Q}} E_{\mathbf{k-q+Q}}}{E_{\mathbf{q}} E_{\mathbf{k-q}}}} \\ + J_{\mathbf{q}}(J_{\mathbf{q+Q}} - J_{\mathbf{k-q+Q}}). \end{aligned} \quad (82)$$

Similarly we obtain:

$$\begin{aligned} \delta\chi_{\mathbf{k}}^{(-)\prime\prime}(\omega) = -4\pi d^3 \sum_{\mathbf{q},\mathbf{q}'} (J_{\mathbf{q}} - J_{\mathbf{k-q}})(J_{\mathbf{q}} - J_{\mathbf{k-q-Q}}) G_{\mathbf{q}',\mathbf{q}} \\ \times \frac{1}{\omega^2 - \epsilon_k^2} \{\delta(\omega - \epsilon_{\mathbf{q}'} - \epsilon_{\mathbf{q-q}'} - \epsilon_{\mathbf{k-q}}) \\ +\delta(\omega + \epsilon_{\mathbf{q}'} + \epsilon_{\mathbf{q-q}'} + \epsilon_{\mathbf{k-q}})\}. \end{aligned} \quad (83)$$

7 Discussion

In the preceding chapters selfconsistency equations were derived and analyzed to describe the full dynamic susceptibility tensor of quantum antiferromagnets. As a starting point "complete" RPA was introduced, the addition "complete" referring to the off-diagonal elements of the susceptibility tensor, which had not been included in previous publications. The diagonal elements (of standard and well-known form) in RPA contain obvious deficiencies: longitudinal quantum fluctuations are absent, and transverse fluctuations are described by sharp δ-functions in frequency ω only.

To remedy these deficiencies a nonlinear set of selfconsistency equations for longitudinal and transverse quantum fluctuations was obtained in Section 5. In this paper full selfconsistency was not attempted, instead a perturbation expansion beyond RPA was used. The main results are as follows.

Longitudinal fluctuations consist of a continuum in energy starting at the single spin wave energy, extending to higher energies. The transverse fluctuations, too, acquire an additional continuum, above the single spin wave energies, the latter still being described by sharp δ-functions. The question remains open, whether the sharp δ-functions only result from finite order perturbation expansion beyond RPA (– where phase space arguments lead to the conclusion that the continuum starts with zero weight at the sharp spin wave energy –), whereas a full selfconsistent solution might lead to peaks in frequency of finite width. General symmetry considerations require that for wavevector $\mathbf{k}$ in the vicinity of $\mathbf{Q}$ (the magnetic Bragg vector) and zero well defined collective excitations exist: around $\mathbf{Q}$ this will be the Goldstone mode, whose existence is required due to the broken continuous symmetry. For the wavevector tending towards zero the vanishing width is due to conservation of the total magnetization in the Heisenberg model used. For general wavevector and small spin (where $1/s$ expansion is questionable) however, it cannot be excluded that a fully selfconsistent treatment leads to peaks of finite width instead of the sharp δ-functions.

A final remark concerning the range of applicability of the approximation used to obtain the selfconsistency equations in Section 5: the decoupling procedure should be applicable in the vicinity of $\mathbf{Q}$ and zero: as pointed out above in that region of $\mathbf{k}$ space the collective excitations will dominate. For small spin, however, there is no valid expansion parameter applicable to all of $(\mathbf{k}, \omega)$ space.

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