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MAGNETISM

Quantum Fluctuations in a Two-Dimensional Antiferromagnet with Four-Spin Interaction of Cubic Symmetry

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Abstract—The excitation spectrum of a non-Heisenberg 2D antiferromagnet with the four-spin interaction of cubic symmetry has been calculated in the first order of $1/2S$. It has been shown that, for weak anisotropy, the Néel state is destroyed by quantum fluctuations. The phase diagrams showing the stability regions of the Néel phase in the space of spin–anisotropy parameters have been plotted.

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1. INTRODUCTION

Discovery of high-T_c superconductors stimulated investigation of the properties of two-dimensional antiferromagnets (2D AFMs), since the magnetic properties of undoped high-T_c superconductors can be described by a spin Hamiltonian on a square lattice. One of the central questions of theory is associated with the effect of quantum fluctuations on the characteristics of 2D AFMs [1–12]. A special emphasis was put to studying the stability of the Néel phase. It turned out that the influence of quantum fluctuations in an isotropic 2D AFM with the interaction between the nearest neighbors is insignificant at $T = 0$ and expansion in powers of the parameter $1/2S$ is justified even for $S = 1/2$ [7].

For the Hamiltonian of cubic symmetry, the spin invariants that describe the pair interaction are reduced to the isotropic forms $I_{fm}(\mathbf{S}_f\mathbf{S}_m)$ and the Hamiltonian is invariant with respect to the transformations of group SU(2). The appearance of the Néel phase is accompanied by spontaneous symmetry breaking and, according to the Goldstone's theorem, the excitation spectrum is gapless ($\Delta \equiv \omega_{\mathbf{q}=0} = 0$). In this case, the question of the gap renormalization does not even arise.

The situation changes if the Hamiltonian includes the fourth-order invariants, whose symmetry is no higher than cubic. Spontaneous symmetry breaking upon the formation of the Néel phase concerns only the discrete group and the gap of the excitation spectrum is nonzero. Thus, quantum fluctuations lead to renormalization of the "bare" gap Δ_0 found in the harmonic approximation. In this case, the additional condition appears that results in a displacement of the stability boundary of the Néel phase. The situation when the Néel phase stable in the linear theory is destroyed by quantum fluctuations upon the inclusion of even a weak anisotropic interaction is of special interest. The present work is focused on studying this particular problem.

2. HAMILTONIAN OF THE NON-HEISENBERG ANTIFERROMAGNET

The effect of quantum fluctuations on the stability of the Néel phase of an anisotropic non-Heisenberg 2D AFM on a square lattice will be studied with the use of the Hamiltonian

$$H = -\frac{1}{2}\sum_{ff'} I_{ff'}(\mathbf{S}_f\mathbf{S}_{f'}) - \frac{1}{2}\sum_{gg'} I_{gg'}(\mathbf{S}_g\mathbf{S}_{g'}) + \sum_{fg} J_{fg}(\mathbf{S}_f\mathbf{S}_g) + H_{ms}, \quad (1)$$

$$\begin{aligned} H_{ms} = {} & \frac{K_1}{4S^2}\sum_{\langle fgf'g'\rangle}(S_f^xS_g^xS_{f'}^xS_{g'}^x + S_f^yS_g^yS_{f'}^yS_{g'}^y + S_f^zS_g^zS_{f'}^zS_{g'}^z) \\ & + \frac{K_2}{4S^2}\sum_{\langle fgf'g'\rangle}\{S_f^xS_{f'}^x(S_g^yS_{g'}^y + S_g^zS_{g'}^z) \\ & + S_f^yS_{f'}^y(S_g^xS_{g'}^x + S_g^zS_{g'}^z) + S_f^zS_{f'}^z(S_g^xS_{g'}^x + S_{g}^yS_{g'}^y)\} \\ & + \frac{K_3}{S^2}\sum_{\langle fgf'g'\rangle}(S_f^xS_{f'}^yS_g^xS_{g'}^y + S_f^yS_{f'}^zS_g^yS_{g'}^z + S_f^zS_{f'}^xS_g^zS_{g'}^x). \end{aligned} \quad (2)$$

Three first terms of Hamiltonian (1) describe the isotropic Heisenberg exchange interaction within the F

and G sublattices and the exchange interaction between the spins of different sublattices. The subscripts f, f' and g, g' refer to the sites of the F and G sublattice, respectively. The Hamiltonian H_{ms} corresponds to the four-site spin–spin interaction of cubic symmetry, whereas the Heisenberg interaction is determined by three invariants with the constants K_1, K_2, and K_3. If $2K_3 = K_1 - K_2$, the four-spin Hamiltonian takes the isotropic form

$$H_{ms} = \frac{1}{4S^2}\sum_{\langle fgf'g'\rangle}\{(K_1 - K_2)(\mathbf{S}_f\mathbf{S}_g)(\mathbf{S}_{f'}\mathbf{S}_{g'}) + K_2(\mathbf{S}_f\mathbf{S}_{f'})(\mathbf{S}_g\mathbf{S}_{g'})\}. \quad (3)$$

The summation over the site indices in the non-Heisenberg part of H is performed in such a manner that four interacting spins situate at the sites forming the smallest square elementary placket. The said restriction on the indices f, g, f', g' in H_{ms} is formally denoted by the angular brackets. The factor $1/S^2$ in front of H_{ms} ensures that the dependence of the energy of the system on the magnitude of the spin is identical for Heisenberg and non-Heisenberg ($\sim S^2$) part in the $S \gg 1$ limit. The numerical coefficients in H_{ms} are chosen such that the energy per one placket for the allowed configuration be $\pm K_i S^2$ in the classical limit.

To solve the problem of the elementary excitation spectrum and stability of the Néel phase we use the Holstein–Primakoff transformation [13] taking into account the presence of F and G sublattices

$$\begin{aligned} S_f^+ &= (2S - a_f^+ a_f)^{\frac{1}{2}} a_f, \quad S_f^- = a_f^+(2S - a_f^+ a_f)^{\frac{1}{2}}, \\ S_f^z &= S - a_f^+ a_f, \\ S_g^+ &= b_g^+(2S - b_g^+ b_g)^{\frac{1}{2}}, \quad S_g^- = (2S - b_g^+ b_g)^{\frac{1}{2}} b_g, \\ S_g^z &= -S + b_g^+ b_g, \end{aligned} \quad (4)$$

where $a_f^+(a_f)$, $b_g^+(b_g)$ are the Bose quasiparticle creation (annihilation) operators in the sublattice F and G, respectively.

Using Eq. (4) and proceeding in a conventional manner we find that in the Hamiltonian of the system in the momentum representation takes the form

$$H = -NS^2(I_0 + J_0 - 2K_1) + H_{(2)} + H_{(4)} + H_{(6)} \ldots . \quad (5)$$

The part of the Hamiltonian that is quadratic in the operators is given by the expression

$$H_{(2)} = \sum_q \{\varepsilon_q(a_q^+ a_q + b_q^+ b_q) + \xi_q(a_q^+ b_{-q}^+ + b_q a_{-q})\}, \quad (6)$$

where

$$\begin{aligned} \varepsilon_q &= S(I_0 - I_q) + SJ_0 \\ &\quad - 4S\Big(K_1 - \frac{1}{2}K_2(\cos q_x + \cos q_y)\Big), \\ \xi_q &= SJ_q - 8SK_3\cos(q_x/2)\cos(q_y/2). \end{aligned} \quad (7)$$

The processes involving four magnons are represented by the operator

$$\begin{aligned} H_{(4)} = \frac{1}{N}\sum_{1234}\Big\{ &\Gamma^{(0)}_{abba}(12;34)a_1^+ b_2^+ b_3 a_4 \\ &+ \Gamma^{(0)}_{aaaa}(12;34)[a_1^+ a_2^+ a_3 a_4 + b_1^+ b_2^+ b_3 b_4] \\ &+ \frac{1}{2}\Gamma^{(0)}_{aabb}(123;4)[a_1^+ b_2^+ b_3^+ b_4 + b_1^+ a_2^+ a_3^+ a_4 + \text{H.c.}] \\ &+ \frac{1}{4}\Gamma^{(0)}_{aabb}(12;34)[a_1^+ a_2^+ b_3 b_4 + \text{H.c.}] \\ &+ \frac{1}{4}\Gamma^{(0)}_{aabb}(1234)[a_1 a_2 b_3 b_4 + \text{H.c.}]\Big\}. \end{aligned} \quad (8)$$

For the convenience of the notation, the wave vectors, over which the summation is performed, are denoted by the digits 1, 2, 3, 4. The x and y components of these vectors are denoted respectively as 1_x, 1_y, etc. The bare amplitudes $\Gamma^{(0)}$ entering $H_{(4)}$ are given in the Appendix.

In the approximation considered in this work, only the terms up to the fourth order are significant and we therefore omit the expression for $H_{(6)}$.

3. THE LINEAR THEORY OF THE EXCITATION SPECTRUM OF THE NON-HEISENBERG 2D ANTIFERROMAGNET

In the harmonic approximation, scattering of quasiparticles is ignored and the spin-wave spectrum is determined solely by $H_{(2)}$. Making use of the Bogoliubov transformation $a_q = u_q\alpha_q + v_q\beta_{-q}^+$, $b_q = u_q\beta_q + v_q\alpha_{-q}^+$, we find the expression for the energy of spin excitations $\omega_q = \sqrt{\varepsilon_q^2 - \xi_q^2}$. In this case, the energy gap has the form

$$\begin{aligned} \Delta &\equiv \omega(\mathbf{q} \equiv 0) \\ &= 2S\sqrt{2(2K_3 + K_2 - K_1)(J_0 - 4K_3 - 2K_1 + 2K_2)}. \end{aligned} \quad (9)$$

The condition of the positive definiteness of the spectrum at $\mathbf{q} = 0$ imposes the restrictions (the necessary conditions of the stability of the structure under consideration) on the relations between the parameters

$$K_1 - K_2 \geq \begin{cases} 2K_3, & \text{if} \quad K_3 \leq J_0/8 \\ J_0/2 - 2K_3, & \text{if} \quad K_3 \geq J_0/8. \end{cases} \tag{10}$$

At $2K_3 = K_1 - K_2$, the excitation spectrum becomes gapless, which is associated with a higher symmetry of the Hamiltonian.

Another stability condition follows from the analysis of the excitation energy at the point $\mathbf{q} = (\pi, \pi)$. Thus, assuming that $I_q = 2I(\cos q_x + \cos q_y)$, $J_q = 4J\cos\frac{q_x}{2}\cos\frac{q_y}{2}$, we find

$$K_2 \leq -K_1 + 2I + J. \tag{11}$$

The energy due to zero-point quantum oscillations per site is given by the expression $\Delta\varepsilon = \frac{1}{N}\sum_q(\omega_q - \varepsilon_q)$.

If $\Delta \neq 0$, the Néel phase remains stable in a certain finite temperature range. In this case, we consider a decrease in the sublattice magnetization with an increase in temperature. In the first order in $1/2S$, the magnetization is

$$\sigma = S - \frac{1}{N}\sum_q \left(\frac{\varepsilon_q - \omega_q}{2\omega_q}\right) - \frac{1}{N}\sum_q \left(\frac{\varepsilon_q}{\omega_q}\right) n_q, \tag{12}$$

where $n_q = \left(\exp\left(\frac{\omega_q}{T}\right) - 1\right)^{-1}$ is the Bose–Einstein distribution function. The temperature dependence is given by the last term, which at $T \ll T_N$ can be expressed as

$$\begin{aligned} \sigma_1(T) &= \frac{1}{N}\sum_q \left(\frac{\varepsilon_q}{\omega_q}\right) n_q \\ &= -\frac{Q}{8\pi S}\left(\frac{T}{J}\right) \ln\left(1 - \exp\left(-\frac{\Delta}{T}\right)\right), \end{aligned} \tag{13}$$

where

$$Q = \frac{J(J - K_1 + K_2)}{(J - K_1 + K_2)(I - K_2) + (J - 2K_3)^2}. \tag{14}$$

If J, $|I| \gg K_1$, K_2, K_3, then $Q = J/(J + I)$. At the ultralow temperature $T \ll \Delta$,

$$\sigma_1(T) = \frac{Q}{8\pi S}\left(\frac{T}{J}\right)\exp\left(-\frac{\Delta}{T}\right). \tag{15}$$

In the intermediate temperature range $\Delta \ll T \ll T_N$

$$\sigma_1(T) = \frac{Q}{8\pi S}\left(\frac{T}{J}\right)\ln\left(\frac{T}{\Delta}\right). \tag{16}$$

4. INCLUSION OF QUANTUM FLUCTUATIONS IN THE FIRST ORDER IN $1/2S$

To study the influence of quantum fluctuations on the stability of the antiferromagnetic phase we include in the Hamiltonian the terms that describe the interaction between magnons. After the Bogoliubov transformation, these terms expressed via the new operators acquire the structure, in which some operator expressions do not have a form of the normal product of creation and annihilation operators (in the normal form, all annihilation operators appear to the right of creation ones). The use of the standard commutation relations reduces these expressions to the normal form. However, there appear the contributions of the zero and first order in the second-quantization operators. The zero-order terms contribute to the ground-state energy, whereas the quadratic terms determine renormalization of the energy expressions of the Hamiltonian $H_{(2)}$. As a result of such a procedure, we find that excitation spectrum at zero temperature is given by the expression

$$\Omega_q^2 = A_q^2 - B_q^2, \tag{17}$$

where $A_q = \varepsilon_q + \delta\varepsilon_q$, $B_q = \xi_q + \delta\xi_q$. The quantities ε_q and ξ_q are specified by Eq. (7) and $\delta\varepsilon_q$ and $\delta\xi_q$, according to the above-described algorithm, are expressed in terms of bare amplitudes and the transformation parameters

$$\begin{aligned} \delta\varepsilon_q &= \frac{1}{N}\sum_p \{2\Gamma^{(0)}_{abbb}(p, q, -p; q)u_p v_p \\ &+ [\Gamma^{(0)}_{aaaa}(p, q; p, q) + \Gamma^{(0)}_{abba}(q, p; p, q)]v_p^2\}, \\ \delta\xi_q &= \frac{1}{N}\sum_p \{2\Gamma^{(0)}_{abbb}(q, p, -q; p)v_p^2 \\ &+ [\Gamma^{(0)}_{abba}(p, -p; q, -q) + \Gamma^{(0)}_{aabb}(p, q; -p, -q)]u_p v_p\}. \end{aligned} \tag{18}$$

In this approach, finding the parameters of the Bogoliubov transformation taking into account the contributions quantum fluctuations is reduced to solving the integral equation for the function Φ_p, which specifies u_p and v_p,

$$\begin{aligned} u_p &= \cosh\Phi_p, \quad v_p = \sinh\Phi_p, \\ \cosh 2\Phi_p &= A_p/\Omega_p, \quad \sinh 2\Phi_p = -B_p/\Omega_p. \end{aligned} \tag{19}$$

The use of the specific quasi-momentum dependence of the bare scattering amplitudes allows us to reduce the solution of the integral equation to the system of three transcendental equations with respect to L, M and R, in terms of which the quasi-momentum depen-

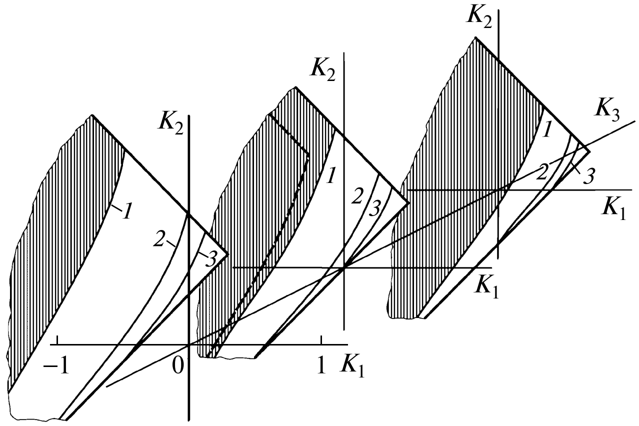

Fig. 1. Stability regions of the Néel phase of a 2D non-Heisenberg antiferromagnet for a constant parameter I.

dence of the renormalization corrections $\delta\varepsilon_q$ and $\delta\xi_q$ are expressed

$$\frac{1}{4}\delta\varepsilon_q = (I-3K_2)R-(J+I-3K_1)M-(J-6K_3)L$$
$$-\frac{1}{2}[(I-2K_1-K_2)R-(I-3K_2)M-4K_3L]$$
$$\times(\cos q_x+\cos q_y), \quad (20)$$
$$\frac{1}{4}\delta\xi_q = [(4K_1-4K_2+K_3-J)L$$
$$-(J-6K_3)M+4K_3R]\cos\frac{q_x}{2}\cos\frac{q_y}{2}.$$

In this case, the equations for L, M, and R follow from the expressions

$$L = \frac{1}{N}\sum_p \cos\frac{p_x}{2}\cos\frac{p_y}{2}u_p v_p, \quad M = \frac{1}{N}\sum_p v_p^2,$$
$$R = \frac{1}{N}\sum_p \cos p_x v_p^2. \quad (21)$$

Using the derived relations, let us analyze the renormalized excitation spectrum and find the displacement of the stability boundaries of the Néel phase due to the quantum fluctuations.

The numerical results are shown in Figs. 1 and 2. The Cartesian axes in Fig. 1 are the four-site exchange interaction parameters K_1, K_2 and K_3 in the units of J. The modification of the stability region of the Néel phase is shown for three values of K_3: $K_3 = -0.2J$, 0, and $0.2J$. For each value, there is a region limited by two straight lines

$$K_3 \geq \frac{1}{2}(K_1-K_2), \quad K_2 \leq J-K_1. \quad (22)$$

These lines result from the condition of the positive definiteness of the spectrum at $q = 0$ and $q = (\pi, \pi)$, respectively. In Fig. 1, these correspond to two perpendicular lines. Thus, the stability region of the Néel phase does not change with the spin S in the absence of quantum fluctuations.

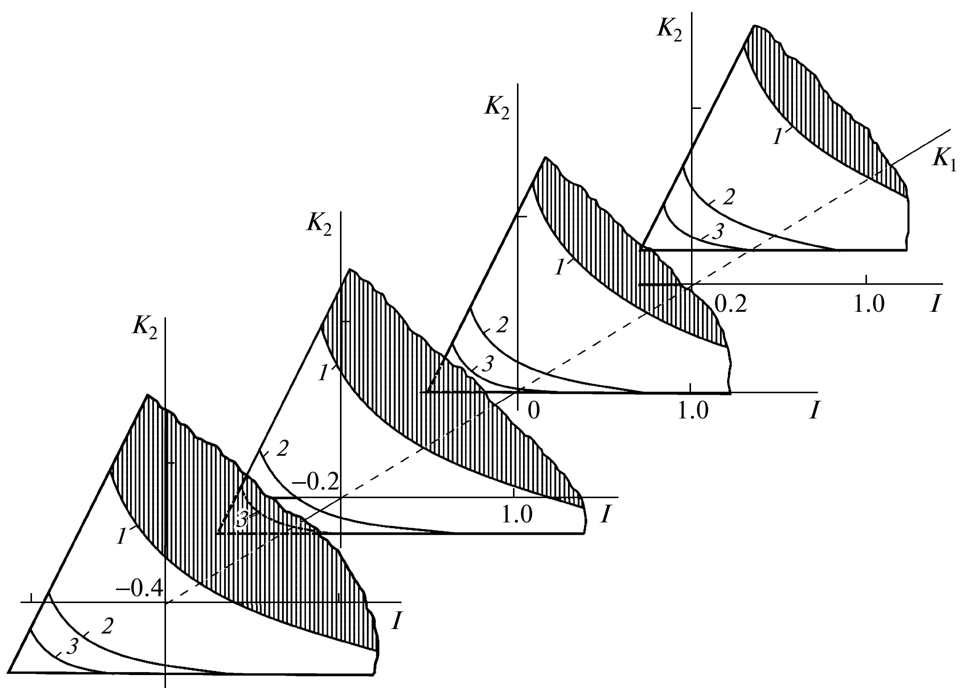

Fig. 2. Stability regions of the Néel phase of a 2D non-Heisenberg antiferromagnet in the space of the parameters K_1, K_2, and I.

When the quantum fluctuations are taken into account, one of the stability boundaries is given by a more complicated nonlinear relation of the parameters K_1, K_2, and K_3 rather than the equation $2K_3 - K_1 + K_2 = 0$. As a result, the stability boundary of the Néel phase is substantially modified in the case of weak anisotropy (Figs. 1 and 2) and becomes dependent on the magnitude of spin S. This modification (change in shape and displacement) of the boundary is most pronounced at small spin. The stability boundary calculated for $S = 1/2$, 1 and 3/2 is shown in Figs. 1 and 2 by curves *1*, *2*, and *3*, respectively. As is seen, the quantum renormalization is quite significant at $S = 1/2$ and 1. The stability region of the Néel phase at $S = 1/2$ is marked by hatching. Remarkably, the quantum fluctuations destroy the Néel phase at infinitesimal anisotropy of the four-spin interaction. The dashed line in the central section in Fig. 1 shows the stability region of the Néel phase at $I/J = -0.2$ and $S = 1/2$. As one would expect, frustration leads to the reduction of the stability region.

Figure 2 shows the cross sections of the stability region of the Néel phase in the space of the parameters K_1, K_2, and I for four values of $K_1 = -0.4J$, $-0.2J$, 0, and $0.2J$. The hatched regions of the cross sections in Fig. 2 correspond to the stability regions for $S = 1/2$. In the cases of $S = 1$ and 3/2, the hatching (like in Fig. 1) should continue to the lines *2* and *3*, respectively. Figure 2 also demonstrates instability of the Néel phase of weakly anisotropic 2D AFM with four-spin interaction with respect to quantum fluctuations. The Néel phase is not destroyed by quantum fluctuations only at a relatively large anisotropy of the multi-spin exchange interaction.

5. CONCLUSIONS

In this work, we have developed the quantum theory of a non-Heisenberg 2D AFM on a square lattice to the first order in the parameter $1/2S$ inclusive. The quasi-momentum dependence of the bare vertices taking into account four-spin interactions has been calculated in the general form (at three cubic invariants). The use of these dependences allowed us to switch from the integral equation or the renormalized characteristics of the spectrum to the system of three transcendental equations. As a result, the closed expressions for the spectrum with the contributions of quantum fluctuations have been obtained in the compact form.

Based on the analysis of the elementary excitation spectrum, the stability conditions of the Néel phase with respect to quantum fluctuations have been found. It turn out that, under the inclusion of all fluctuations, the stability regions of the Néel phase strongly depend on the magnitude of spin at small S. This qualitatively new effect is due to the fact that quantum fluctuations destroy the Néel phase as soon as the four spin interaction becomes weakly anisotropic. This effect is the most pronounced at small S. This can be qualitatively interpreted as follows. The anisotropy of cubic-symmetry leads to the appearance of a gap in the elementary excitation spectrum. From this point of view, the anisotropy stabilizes the Néel phase. However, the appearance of anisotropic invariants simultaneously results in the terms, which induce quantum fluctuations. These terms form the tendency of breaking the antiferromagnetic long-range order. Apparently, the latter processes prevail at small anisotropy and the magnetic order disappears. The effective field of anisotropy increases with anisotropy, the quantum fluctuations are suppressed and the long-range magnetic order becomes stable at $T = 0$.

APPENDIX

The bare amplitudes $\Gamma^{(0)}$ entering $H_{(4)}$ are

$$\begin{aligned}\Gamma^{(0)}_{abba}(12;34) = \Big\{&(8K_1 - 4J)\cos\frac{1_x - 4_x}{2}\cos\frac{1_y - 4_y}{2} \\ &+ 2(K_1 + K_2)\Big[\cos\frac{2_x + 3_x}{2}\cos\frac{1_y + 4_y}{2} + \cos\frac{2_y + 3_y}{2}\cos\frac{1_x + 4_x}{2}\Big] \\ &- 4K_2\Big[\cos\frac{2_x - 3_x}{2}\Big(\cos\frac{1_y + 4_y}{2} + \cos\frac{2_y + 3_y}{2}\Big) \\ &+ \cos\frac{2_y - 3_y}{2}\Big(\cos\frac{1_x + 4_x}{2} + \cos\frac{2_x + 3_x}{2}\Big)\Big]\Big\} \\ &\times \Delta(1 + 2 - 3 - 4),\end{aligned} \tag{23}$$

$$\begin{aligned}\Gamma^{(0)}_{aaaa}(12;34) = \Big\{&(I - K_2)\Big(1 + \frac{1}{8S}\Big)(\cos 1_x + \cos 2_x \\ &+ \cos 3_x + \cos 4_x + \cos 1_y + \cos 2_y + \cos 3_y + \cos 4_y) \\ &+ (K_1 - I)[\cos(1_x - 3_x) + \cos(1_x - 4_x) \\ &+ \cos(2_x - 3_x) + \cos(2_x - 4_x) + \cos(1_y - 3_y) \\ &+ \cos(1_y - 4_y) + \cos(2_y - 3_y) + \cos(2_y - 4_y)]\Big\} \\ &\times \Delta(1 + 2 - 3 - 4),\end{aligned} \tag{24}$$

$$\Gamma^{(0)}_{abbb}(123;4) = \left\{8K_3\left[\cos\frac{1_x}{2}\cos\frac{4_y}{2}\cos\frac{2_y-3_y}{2}\right.\right.$$

$$\left.+\cos\frac{1_y}{2}\cos\frac{4_x}{2}\cos\frac{2_x-3_x}{2}\right]-2\left(1+\frac{1}{8S}\right) \quad (25)$$

$$\left.\times(J-2K_3)\cos\frac{1_x}{2}\cos\frac{1_y}{2}\right\}\Delta(1+2+3-4),$$

$$\Gamma^{(0)}_{aabb}(12;34) = 2(K_1-K_2-2K_3)$$

$$\times\left[\cos\frac{1_x-2_x}{2}\cos\frac{3_y-4_y}{2}+\cos\frac{1_y-2_y}{2}\cos\frac{3_x-4_x}{2}\right] \quad (26)$$

$$\times\Delta(1+2-3-4),$$

$$\Gamma^{(0)}_{aabb}(1234) = 2(K_1-K_2+2K_3)$$

$$\times\left[\cos\frac{1_x-2_x}{2}\cos\frac{3_y-4_y}{2}+\cos\frac{1_y-2_y}{2}\cos\frac{3_x-4_x}{2}\right] \quad (27)$$

$$\times\Delta(1+2+3+4).$$

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