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The coupling of spin waves in a finite Heisenberg system

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Abstract. The coupling of spin waves for a Heisenberg system with a finite size is studied. It is found that the final results are boundary condition dependent. For a periodic system, the finite-size effect weakens the coupling of spin waves and an additional term proportional to $-T^{7/2}/N$ appears with N, the system's size, in the expression for the number density of spin waves at temperature T. The free boundary greatly complicates the coupling of spin waves. There are three new kinds of scattering process: between spin waves parallel to the free surfaces of the system, between spin waves parallel to the surface and other waves, and between spin waves with the same components in the direction perpendicular to the free surface of the system. Unlike the others, the last effective interaction tends to depress the excitation of spin waves at low temperatures.

1. Introduction

Clusters made up of several hundred or thousand atoms show a different behaviour from that of an infinite system. The study of such clusters may give us an insight into how the properties of the system evolve from cluster to bulk on increasing the number of atoms in clusters, and the roles the finite size and surface play in the physical processes. Much effort, both theoretical [1–7] and experimental [8–13], has been devoted to the study of magnetic clusters. These studies have shown that the magnetization in a finite system is inhomogeneous. The magnetization of the surface layer of the cluster decreases more rapidly with increasing temperature T than does that of the inner layers [1–3, 7, 9, 10, 13]. An obvious deviation in the mean magnetization from the Bloch $T^{3/2}$ law was also found [7, 14].

In this paper, based upon the spin-wave theory, we shall study the influence of the size and surface of the system on the coupling of spin waves for two kinds of boundary condition: a periodic boundary condition and a free boundary condition. The coupling of spin waves has been a constant topic of research [15–18]. Dyson [15] proved that the coupling of spin waves promotes the excitation of spin waves and results in a term proportional to T^4 in the expression for the spontaneous magnetization. Wortis [17] discussed the possibility of the existence of bound states of two spin waves due to their coupling. In a finite system, the distribution of the spin deviation corresponding to a spin wave will be greatly different from that of an infinite system owing to the finite size and free surface. In this case, what will result from the coupling of spin waves is a question that needs be answering.

In this article, we report a numerical calculation which was performed for small clusters (composed of several hundred or fewer spins). For large but finite clusters, the asymptotic

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expression for the number density of spin waves was obtained. Combining both results, we try to give an idea about how the properties evolve from cluster to bulk.

The contents of this paper are arranged as following: in the next section, a general theory of the interaction of spin waves is presented on the basis of the perturbation theory. In section 3 and 4, the number densities of spin waves in clusters under the periodic boundary condition and the free boundary condition, respectively, are studied. In section 5, some conclusions are given.

2. The coupling of spin waves

The Hamiltonian of a Heisenberg system composed of $N \times N \times N$ spins is

$$\hat{H} = -2J \sum_{\langle i,j \rangle} \left(\frac{S_i^- S_j^+ + S_i^+ S_j^-}{2} + S_i^z S_j^z \right) + \gamma h \sum_i S_i^z \tag{1}$$

where S_i^+ (S_i^-) is the spin-raising (spin-lowering) operator, J is the ferromagnetic coupling parameter between spins, $\gamma = g\mu$ (with g the Landé g-factor, and μ the Bohr magneton) and h is the applied field. The sum $\sum_{\langle i,j \rangle}$ extends over all neighbouring pairs of spins. After the transformations

$$S_i^+ = \sqrt{2S - a_i^+ a_i} a_i$$

$$S_i^- = a_i^+ \sqrt{2S - a_i^+ a_i}$$

$$S_i^z = S - a_i^+ a_i$$

and

$$a_i^+ = \sum_q c_{iq}^* b_i^+$$

$$a_i = \sum_q c_{iq} b_i$$

equation (1) develops into the form

$$\hat{H} = \hat{H}_0 + \hat{H}_1 = \sum_q \bar{\varepsilon}_q b_q^+ b_q + \sum_{q_1 q_2 q_3 q_4} f(1234) b_{q_1}^+ b_{q_2}^+ b_{q_3} b_{q_4} + \mathrm{O}\left(\frac{1}{S}\right). \tag{2}$$

The first term corresponds to the free spin waves with

$$\bar{\varepsilon}_q = \varepsilon_q + \gamma h$$

and

$$f(1234) = \sum_{q_1 q_2 q_3 q_4} J \left(c_{iq_1}^* c_{iq_2}^* c_{iq_3} c_{jq_4} + c_{iq_1}^* c_{jq_2}^* c_{jq_3} c_{jq_4} - 2 c_{iq_1}^* c_{jq_2}^* c_{iq_3} c_{jq_4} \right). \tag{3}$$

Here, c_{iq} is given by the secular equations

$$\sum_q \left(\langle i | \hat{H}_0 | j \rangle - \delta_{i,j} \varepsilon_q \right) c_{iq} = 0 \tag{4}$$

for $i = 1, 2, \ldots, N$ with $|i\rangle = |0, 0, \ldots, n_i = 1, \ldots, 0\rangle$.

According to perturbation theory, the grand partition function of the system can be written as

$$\Xi = \mathrm{Tr}\left[\exp(-\beta \hat{H}_0)\left(1 - \int_0^{\beta} \mathrm{d}s \exp(s\hat{H}_0)\hat{H}_1 \exp(-s\hat{H}_0)\right.\right.$$
$$\left.\left. + \int_0^{\beta} \mathrm{d}s \exp(s\hat{H}_0)\hat{H}_1 \exp(-s\hat{H}_0) \int_0^{s} \mathrm{d}t \exp(t\hat{H}_0)\hat{H}_1 \exp(-t\hat{H}_0) - \ldots\right)\right]$$

where $\beta = 1/k_{\mathrm{B}}T$ (k_{B} is the Boltzmann's constant and T the temperature). Further calculation gives the free energy of the system:

$$\Phi(N, \beta) = -\frac{1}{\beta}\ln \Xi = \frac{1}{\beta}\sum_{q\neq 0} \ln\left[1 - \exp(\beta\varepsilon_q)\right]$$
$$+ \sum_{q,q'} \frac{\left[F(q, q') + F(q', q)\right]/2}{\left[\exp(\beta\bar{\varepsilon}_q) - 1\right]\left[\exp(\beta\bar{\varepsilon}_{q'}) - 1\right]} + \mathrm{O}\left(\frac{1}{S}\right) \tag{5}$$

where $F(q, q') = f(q, q', q', q) + f(q, q', q, q')$. Following the familiar thermodynamic relation, the number density of spin waves is,

$$n(N, \beta) = -\frac{1}{N^3 S}\frac{\partial(\beta\Phi)}{\partial(\gamma h)}\bigg|_{h\to 0}$$
$$\simeq \frac{1}{N^3}\sum_{q\neq 0}\frac{1}{\exp(\beta\varepsilon_q) - 1}$$
$$+ \frac{\beta}{N^6}\sum_{q,q'}\frac{\exp(\beta\varepsilon_q)\left[F(q, q') + F(q', q)\right]}{\left[\exp(\beta\varepsilon_q) - 1\right]^2\left[\exp(\beta\bar{\varepsilon}_{q'}) - 1\right]} + \mathrm{O}\left(\frac{1}{S}\right). \tag{6}$$

Only a two-spin-wave process is explicitly considered in (6). A multiple-spin-wave process is relatively unimportant at low temperatures.

3. Clusters under the periodic boundary condition

Let us consider a system composed of $N \times N \times N$ spins that form a simple-cubic lattice. Under the periodic boundary condition, $c_{jq} = N^{-3/2}\exp(-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{j})$ where $\boldsymbol{j}$ denotes the coordinates of spin S_j and $\boldsymbol{q}$ the reciprocal vector. Substituting this expression for c_{jq} in (6), one obtains

$$n(N, \bar{\beta}) \simeq \frac{1}{N^3}\sum_{q\neq 0}\frac{1}{\exp(\beta\varepsilon_q) - 1} + \frac{\beta J}{N^6}\sum_{q\neq 0, q'\neq 0}\frac{\exp(\bar{\beta}q^2)\left(q_1^2 q_1'^2 + q_2^2 q_2'^2 + q_3^2 q_3'^2\right)}{\left[\exp(\bar{\beta}q^2) - 1\right]^2\left[\exp(\bar{\beta}q'^2) - 1\right]} \tag{7}$$

where $\bar{\beta} = 2SJ/k_{\mathrm{B}}T$.

The first sum in equation (7) corresponds to a free spin wave. If we denote this as $n_{\mathrm{f}}(N, \bar{\beta})$, for large N a direct calculation gives (see appendix B for details)

$$n_{\mathrm{f}}(N, \bar{\beta}) = \frac{1}{N^3}\sum_{q\neq 0}\frac{1}{\exp(\beta\varepsilon_q) - 1}$$
$$= -\frac{0.2268}{\bar{\beta}N} + \frac{\zeta\left(\frac{3}{2}\right)}{8\pi^{3/2}\bar{\beta}^{3/2}} + \frac{\zeta\left(\frac{5}{2}\right)}{128\pi^{3/2}\bar{\beta}^{5/2}}$$
$$+ \frac{33\zeta\left(\frac{7}{2}\right)}{4096\pi^{3/2}\bar{\beta}^{7/2}} + \mathrm{O}\left(\frac{1}{N^2}\right) + \mathrm{O}\left(\frac{1}{\bar{\beta}^{9/2}}\right). \tag{8}$$

The second sum in equation (7) represents the effect of the coupling of spin waves. A simple calculation shows that the change in the number density of spin waves due to the coupling is

$$n_c(N,\bar{\beta}) = \frac{\beta J}{N^6}\sum_{q\neq 0, q'\neq 0}\frac{\exp(\bar{\beta}q^2)\left(q_1^2 q'^2_1 + q_2^2 q'^2_2 + q_3^2 q'^2_3\right)}{\left[\exp(\bar{\beta}q^2)-1\right]^2\left[\exp(\bar{\beta}q'^2)-1\right]}$$

$$= -\frac{\beta J}{3}\frac{\partial}{\partial\bar{\beta}}\left[\frac{1}{N^3}\sum_{q\neq 0}\frac{1}{\exp(\bar{\beta}q^2)-1}\right]\left[\frac{1}{N^3}\sum_{q\neq 0}\frac{q'^2}{\exp(\bar{\beta}q'^2)-1}\right] \tag{9a}$$

$$= -\frac{0.0567}{8\pi^{3/2}NS\bar{\beta}^{7/2}} + \frac{3\zeta\left(\frac{3}{2}\right)\zeta\left(\frac{5}{2}\right)}{512\pi^3 S\bar{\beta}^4}. \tag{9b}$$

where (8) has been used. At a quick glance, one can see that the T^4 term is the same as obtained by Dyson except for a factor $0.3/S$, a result of the omission of non-diagonal scattering [15, 16]. Equation (9*b*) indicates that the finite size of the system results in a negative term proportional to $T^{7/2}$ and inversely proportional to N, the system's size.

Finally, for large N, one has

$$n\left(N,\bar{\beta}\right) \simeq -\frac{0.2268}{\bar{\beta}N} + \frac{\zeta\left(\frac{3}{2}\right)}{8\pi^{3/2}\bar{\beta}^{3/2}} + \frac{\zeta\left(\frac{5}{2}\right)}{128\pi^{3/2}\bar{\beta}^{5/2}} + \frac{1}{4096\pi^{3/2}\bar{\beta}^{7/2}}\left(33\zeta\left(\tfrac{7}{2}\right) - \frac{29.03}{NS}\right) + \frac{3\zeta\left(\frac{3}{2}\right)\zeta\left(\frac{5}{2}\right)}{512\pi^3\bar{\beta}^4}. \tag{10}$$

When N is small, no simple expression can be obtained. The interaction of spin waves in that case was studied numerically by directly calculating $n(N,\bar{\beta})$ from equation (6). Figure 1 gives the variation in $n_c(N,\bar{\beta})$ with N for different temperatures ($S = 1$). Data corresponding to large N are obtained from (9*b*). Two conclusions can be drawn from figure 1. Firstly the finite size weakens the effect of the coupling of spin waves. This is a result of the cut-off of the low lying energy modes of spin waves due to the finite size. Calculation also shows that the coupling effect is severely dependent on the system's size when N is small. It is expected that a finite-size effect is evident when N^3 is not more than several hundred. From (9*a*) we know that the coupling effect is proportional to the temperature gradient of $n_f(N,\bar{\beta})$. If we denote the energy gap between the ground state and the first excited state as ΔE, which has the form $\Delta E = 4JS[1-\cos(2\pi/N)]$, it is shown in figure 1 that ΔE drops rapidly with increasing N in the range $2^3 < N^3 < 10^3$. The decrease in ΔE makes it easy to excite a spin wave and then to increase the temperature gradient of $n_f(N,\bar{\beta})$. This explains the rapid change in the coupling effect for small N. Secondly $n_c(N,\bar{\beta}) > 0$, irrespective of the value of N, which means that the interaction between spin waves remains attractive if the periodic boundary condition is applied.

4. Clusters under the free boundary condition

Under the periodic boundary condition, the system is confined in a finite volume without a surface. Such a system is meaningful for theoretical study. However, for an isolated cluster, the free boundary condition is more natural. For simplicity, we shall consider first the case when the free boundary condition is imposed along only one direction (e.g., the $(1,0,0)$ direction) of a cubic Heisenberg system. It can be proved that in this case c_{jq} given

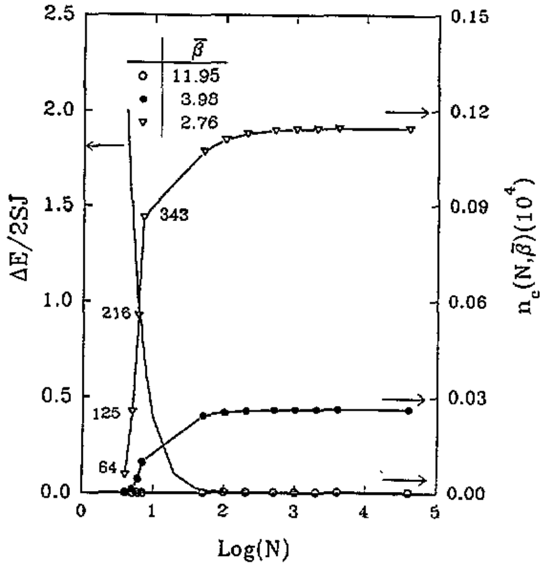

Figure 1. Contributions of the coupling of spin waves to its number density as functions of the system's size. The numbers in the figure are the sizes of the corresponding clusters. Accordingly, the change in ΔE is also given.

by equation (4) takes the form, [19]

$$C_{mq} = \left(\frac{2}{N^3(1+\delta_{q_1,0})}\right)^{1/2} \cos\left(\frac{\pi l_1}{N}\left(m_1 - \tfrac{1}{2}\right)\right) \exp\left(\mathrm{i}\frac{2\pi}{N}(l_2 m_2 + l_3 m_3)\right) \tag{11}$$

where $m = (m_1, m_2, m_3)$, $m_i = 1,\ 2,\ \ldots,\ N$, $l_i = 0, 1, \ldots,\ N-1$, $i = 1, 2, 3$, and $q = (q_1, q_2, q_3) = \pi(l_1/N_1,\ 2l_2/N_2,\ 2l_3/N_3)$. Spin waves in this case can also be identified by the reciprocal vector q in form.

From equation (3), one can directly write

$$\begin{aligned} F(q, q') = {} & \frac{J}{2N^3(1+\delta_{q_1,0})(1+\delta_{q'_1,0})}\{-12 + 6(\cos q_1 + \cos q_2 + \cos q_3) \\ & +2(\cos q'_1 + \cos q'_2 + \cos q'_3) - 4[\cos(q_1 - q'_1) + \cos(q_2 - q'_2) \\ & + \cos(q_3 - q'_3)] + 2[\cos(q_1 - q'_1) - \cos(q_1 + q'_1)] + \delta_{q_1,0}[-6 - 4\cos q'_1 \\ & +6(\cos q_2 + \cos q_3) + \cos(q_1 + q'_1) + \cos(q_1 - q'_1) - 4\cos(q_2 - q'_2) \\ & -4\cos(q_3 - q'_3) + 2(\cos q'_2 + \cos q'_3)] + \delta_{q'_1,0}[-10 - 4\cos q_1 \\ & +6(\cos q_2 + \cos q_3) + 3\cos(q_1 + q'_1) + 3\cos(q_1 - q'_1) \\ & -4\cos(q_2 - q'_2) - 4\cos(q_3 - q'_3) + 2(\cos q'_2 + \cos q'_3)] \\ & +(\delta_{q_1,q'_1} + \delta_{q_1,-q'_1} + \delta_{q_1,q'_1,2\pi})[-6 - 2\cos 2q'_1 + 3(\cos q_1 + \cos q_2 \\ & + \cos q_3) - 2\cos(q_2 - q'_2) - 2\cos(q_3 - q'_3) \\ & +(\cos q'_1 + \cos q'_2 + \cos q'_3)]\}. \end{aligned}$$

When $|q|$ is small, one has

$$\begin{aligned} & F(q', q) + F(q, 'q) \\ & \quad \simeq \frac{J}{N^3(1+\delta_{q_1,0})(1+\delta_{q'_1,0})}\Big[-4\left(q\cdot q' - q_1 q'_1\right) - \left(q_1^2 q'^2_1 + q_2^2 q'^2_2 + q_3^2 q'^2_3\right) \\ & \quad - \left(\delta_{q_1,0} + \delta_{q'_1,0}\right)\left(4q\cdot q' + q_2^2 q'^2_2 + q_3^2 q'^2_3\right) \end{aligned}$$

$$+(\delta_{q_1,-q_1'}+\delta_{q_1,q_1'}+\delta_{q_1+q_1',2\pi})\left(2q_1^2-\frac{q_2^2q_2'^2+q_3^2q_3'^2}{2}\right)\Bigg] \tag{12}$$

When $N \gg 1$, after tedious calculations one obtains (with formulae presented in appendix A)

$$n_{\mathrm{f}}(N,\bar{\beta}) \simeq -\frac{\ln[\bar{\beta}(\pi/N)^2]}{8\pi N\bar{\beta}} - \frac{0.1663}{N\bar{\beta}} + \frac{\zeta(\frac{3}{2})}{8\pi^{3/2}\bar{\beta}^{3/2}} - \frac{\zeta(2)}{512\pi N\bar{\beta}^2} + \frac{3\zeta(\frac{5}{2})}{128\pi^{3/2}\bar{\beta}^{5/2}}$$
$$-\frac{5\zeta(3)}{1536\pi N\bar{\beta}^3} + \frac{33\zeta(\frac{7}{2})}{4096\pi^{3/2}\bar{\beta}^{7/2}} \tag{13}$$

$$n_{\mathrm{c}}(N,\bar{\beta}) \simeq -\frac{0.13}{\pi^3 SN\bar{\beta}^{5/2}} + \frac{3\zeta(2)\ln N}{256\pi^2\bar{\beta}^3 SN^2} - \frac{0.0199\zeta(\frac{5}{2})}{128\pi^{5/2}SN\bar{\beta}^{7/2}}\ln\left(\frac{N^2}{\bar{\beta}}\right)$$
$$+\frac{1}{\pi^{5/2}SN\bar{\beta}^{7/2}}(0.00391+B) + \frac{3\zeta(\frac{3}{2})\zeta(\frac{5}{2})}{512\pi^3 S\bar{\beta}^4} \tag{14}$$

where

$$B = \frac{1}{256}\sum_{m_1=1}^{\infty}\sum_{m_2=1}^{\infty}\frac{1}{m_1 m_2^2\sqrt{m_1+m_2}} = 0.0126\ldots.$$

The free boundaries complicate the coupling of spin waves greatly. Besides the effective interaction discussed by Dyson [15] and Oguchi, [16] there are three scattering processes: between spin waves whose wavevectors parallel to the free surfaces of the system (we call them two dimensional spin waves), described by terms associated with the symbol $\delta_{q_1,-q_1'}$ in (12); between waves parallel to the surfaces and other waves, corresponding to terms associated with the symbols $\delta_{q_1,0}$ and $\delta_{q_1',0}$; between waves with the same components in the direction perpendicular to the free surface of the system, represented by terms associated with the symbol $\delta_{q_1,q_1'}$.

The last effective interaction mentioned above gives a negative contribution to the number density of spin waves (the first term in (14)), which dominates the behaviour of the coupling at low temperatures and represents a repulsive interaction between special spin waves. This is different from either an infinite system or a finite system under the periodic boundary condition. Along the $(1,0,0)$ direction, a spin wave exists in the form of a standing wave. Spins with different positions in the $(1,0,0)$ direction find themselves special local environments, which influence the coupling of spin waves. A simple estimate shows that the total energy when two spin waves with wavevector $q=(q_1,\ 0,\ 0)$ are excited will be

$$E = 2\varepsilon_{q_1} + \frac{J}{N^3}(1-\cos q_1) \geqslant 2\varepsilon_{q_1}$$

This means that two such spin waves tend to repel each other.

The second term in (14) comes from the coupling of two dimensional spin waves. It decreases rapidly with increasing N. The third term is the result of enhancement of the number density of spin waves due to the free boundaries (see equation (13)). Part of term $1/\bar{\beta}^{7/2}$ comes from the coupling of two-dimensional and three-dimensional spin waves and part from the scattering corresponding to $(q_2^2q_2'^2+q_3^2q_3'^2)/2$ of (12) between spin waves with the same $(1,0,0)$ component. Corrections to the coupling effect mainly come from the first and second terms, especially the former. For small N, $n_{\mathrm{c}}(N,\bar{\beta})<0$ may appear.

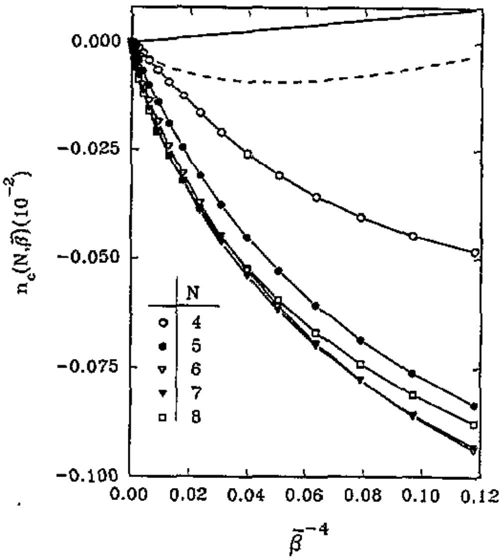

Figure 2. Negative contribution of the coupling of spin waves to its number density which results if the free boundary condition is applied: —, $N \to \infty$ with the second kind of boundary condition; - - -, $N^3 = 216$ with the second kind of boundary condition (see the text).

Combining (13) and (14), one has

$$n(N,\bar{\beta}) \approx -\frac{0.0398 \ln\left[\bar{\beta}(\pi/N)^2\right]}{N\bar{\beta}} - \frac{0.1663}{N\bar{\beta}} + \frac{\zeta\left(\frac{3}{2}\right)}{8\pi^{3/2}\bar{\beta}^{3/2}} - \frac{\zeta(2)}{512\pi N\bar{\beta}^2} + \frac{3\zeta\left(\frac{5}{2}\right) - 2.99/(SN)}{128\pi^{3/2}\bar{\beta}^{5/2}}$$
$$+ \frac{1}{256\pi N\bar{\beta}^3}\left(\frac{3\zeta(2)\ln(N)}{\pi NS} - \frac{5\zeta(3)}{6}\right) + \frac{\zeta\left(\frac{5}{2}\right)}{128\pi^{5/2}SN\bar{\beta}^{7/2}}\ln\left(\frac{N^2}{\bar{\beta}}\right)$$
$$+ \frac{1}{\pi^{3/2}\bar{\beta}^{7/2}}\left[\frac{33\zeta\left(\frac{7}{2}\right)}{4096} + \frac{0.00391 + B}{\pi SN}\right] + \frac{3\zeta\left(\frac{3}{2}\right)\zeta\left(\frac{5}{2}\right)}{512\pi^3 S\bar{\beta}^4} \tag{15}$$

When the free boundary condition is applied in three mutual orthogonal directions of the system, the calculation of $F(q, q')$ is direct but complex and provides no more information. We do not consider this case here.

Numerical calculations for clusters of $N \times N \times N$ with $N = 4, 5, 6, 7, 8$ were also carried out by directly diagonalizing equation (4). In our calculation, two cases with the free boundary condition along three mutual orthogonal directions $(1,0,0)$, $(0,1,0)$ and $(0,0,1)$ and with the free boundary condition along $(1,0,0)$ and the periodic boundary condition along $(0,1,0)$ and $(0,0,1)$ (we shall call them the first kind of boundary condition and second kind of boundary condition, respectively) were considered. Typical results for the former are shown in figure 2. As a comparison, two curves corresponding to $N \to \infty$ (solid curve) and $N^3 = 216$ (broken curve) with the second kind of boundary condition are also presented. As expected, the negative effect of the coupling is greatly enhanced for small N, which causes an obvious modification to the number density of spin waves (figure 3) and cannot be neglected in a accurate analysis. It is shown by figure 3 that the ratio $|n_c(N,\bar{\beta})/n_f(N,\bar{\beta})|$ at $\bar{\beta} = 2$ is 2.1% for a cluster with $N^3 = 216$, while for an infinite system it is about 0.1%. Calculation also shows that $n_c(N,\bar{\beta}) < 0$ even for the second kind of boundary condition, although $|n_c(N,\bar{\beta})|$ is much smaller than that for the first kind of boundary condition. Combining the numerical results and equation (14), we can investigate

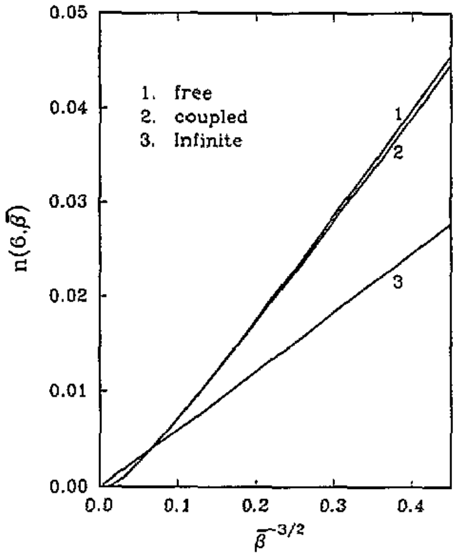

Figure 3. The number density of spin waves as a function of temperature for a cluster with size $N^3 = 216$.

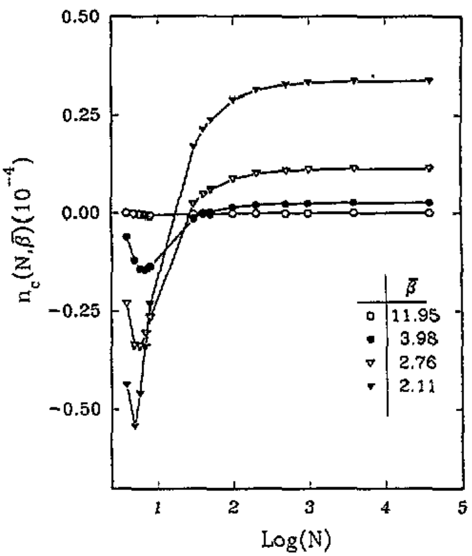

Figure 4. Contributions of the coupling of spin waves to its number density as a functions of the system's size. Data for small N are numerically calculated with the free boundary condition along $(1, 0, 0)$ and the periodic boundary condition along $(0, 1, 0)$ and $(0, 0, 1)$. Data for large N are from equation (14).

the change in $n_c(N, \bar\beta)$ against N (figure 4). Initially $n_c(N, \bar\beta)$ decreases smoothly with increasing N and has a minimum at N = 5–6 (temperature dependent). Subsequently an increase in N leads to a rapid increase in $n_c(N, \bar\beta)$ up to $N \simeq 30$. At this point, the behaviour of the cluster is very near to that of an infinite system. Different from the case of the periodic boundary condition, the most obvious finite-size effect for the coupling of spin waves takes place when N takes certain values. As mentioned above, there is a repulsive interaction between certain spin waves, which negatively contributes to $n_c(N, \bar\beta)$. At the same time, the free boundary condition makes it easy to excite a spin waves (compare curves 2 and 3 in figure 3), which in turn enhances the positive contribution from the coupling of spin waves. The negative and positive contributions relate to the system's size differently. This may be the reason for the emergence of the minimum in figure 4 when N is varied.

5. Conclusions

(1) For a system under the periodic boundary condition, the coupling of spin waves contributes a term proportional to $-T^{7/2}/N$ to its number density when N is large. The coupling effect of spin waves is obviously weakened only in a system whose size N^3 is not more than a few hundred. No new scattering processes are introduced in this case.

(2) For a system with free boundaries, because of the reflection of the boundary, spin deviations exist in the form of a standing wave in the direction perpendicular to the surface and are position dependent. This introduces new scattering mechanisms into the interaction of spin waves. There are four kinds of scattering process between spin waves in a small Heisenberg system. The first is the conventional scattering discussed by Dyson [15]; it

gives a contribution to the number density of spin waves which is proportional to T^4. The second is the scattering between the spin waves whose wavevectors are parallel to the free surface of the system. The third is the scattering between the spin waves parallel to the surface and others. The last is the scattering between spin waves whose wavevectors have the same component in the direction perpendicular to the free surface of the system.

(3) Different from the other cases, some spin waves with the same components in directions perpendicular to the free surfaces of the system show a tendency to repel each other. As a result, the contributions from the coupling of spin waves to its number density is negative at low temperatures.

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Appendix A

In the following, we give the calculation of a sum with the form

$$G(N,\alpha) = \frac{1}{N}\sum_{l=0}^{N-1} f\left(\frac{\alpha\pi l}{N}\right). \tag{A1}$$

Doubling the size of the system, one has

$$G(2N,\alpha) = \frac{1}{2N}\sum_{l'=0}^{2N-1} f\left(\frac{\alpha\pi l'}{2N}\right). \tag{A2}$$

$\pi l'/(2N)$ can be alternatively expressed as

$$\frac{\pi l'}{2N} = \frac{\pi l}{N} + \frac{\pi k}{2N}$$

where $k = 0, 1$. Expanding $f(\pi l'/(2N))$ at $\pi l/N$ gives rise to

$$f\left(\frac{\alpha\pi l'}{2N}\right) = f\left(\frac{\alpha\pi l}{N}\right) + \alpha f'\left(\frac{\alpha\pi l}{N}\right)\frac{\pi k}{2N} + \frac{\alpha^2}{2!}f''\left(\frac{\alpha\pi l}{N}\right)\left(\frac{\pi k}{2N}\right)^2$$
$$+\frac{\alpha^3}{3!}f'''\left(\frac{\alpha\pi l}{N}\right)\left(\frac{\pi k}{2N}\right)^3 + \dots \tag{A3}$$

where $f^{(m)}(x)$ represents the mth derivative of $f(x)$ with respect to x. From equations (A1)–(A3) one obtains

$$G(N,\alpha) - G(2N,\alpha)$$
$$= -\frac{1}{2N}\sum_{l=0}^{N-1}\sum_{k=0}^{1}\left[\alpha f'\left(\frac{\alpha\pi l}{N}\right)\frac{\pi k}{2N} + \frac{\alpha^2}{2!}f''\left(\frac{\alpha\pi l}{N}\right)\left(\frac{\pi k}{2N}\right)^2 + \dots\right]. \tag{A4}$$

The first term of equation (A4) gives a correction proportional to $1/N$, others correspond to higher order corrections. Substituting the first sum by an integral, one has

$$G(N,\alpha) - G(2N,\alpha) = -\frac{1}{4N}\left[f(\alpha\pi) - f(0)\right] + O\left(\frac{1}{N^2}\right)$$

or

$$
\begin{aligned}
G(N,\alpha) - G(\infty,\alpha) \\
&= [G(N,\alpha) - G(2N,\alpha)] + \left[G(2N,\alpha) - G(2^2N,\alpha)\right] + \ldots \\
&\simeq -\frac{1}{2N}\left[f(\alpha\pi) - f(0)\right] \qquad \text{(A5)}
\end{aligned}
$$

where

$$(\infty,\alpha) = \frac{1}{\pi\alpha}\int_0^{\alpha\pi} f(x)\,\mathrm{d}x.$$

Similarly, the sum for multiple variables can be obtained. Firstly,

$$
\begin{aligned}
G\,(N_1, N_2, N_3, \alpha) \\
&= \frac{1}{N_1N_2N_3}\sum_{l_1=0}^{N_1-1}\sum_{l_2=0}^{N_2-1}\sum_{l_3=0}^{N_3-1} f\left(\frac{\alpha\pi l_1}{N_1}, \frac{\alpha\pi l_2}{N_2}, \frac{\alpha\pi l_3}{N_3}\right) \\
&= \frac{1}{\pi^3}\int_\Omega f(\alpha\boldsymbol{x})\,\mathrm{d}\boldsymbol{x} - \frac{1}{4\pi^3}\int_\Omega (\boldsymbol{\Delta}\cdot\nabla) f(\alpha\boldsymbol{x})\,\mathrm{d}\boldsymbol{x} + \mathrm{O}\left(\frac{1}{N_1^2}, \frac{1}{N_2^2}, \frac{1}{N_3^2}\right) \qquad \text{(A6)}
\end{aligned}
$$

where

$$\Omega = \{(x_1, x_2, x_3)\,,\ x_1 \in [0,\pi]\,,\ x_2 \in [0,\pi]\,,\ x_3 \in [0,\pi]\,\}$$

$$\boldsymbol{\Delta} = (\pi/2)(x_1/N_1, x_2/N_2, x_3/N_3).$$

Secondly,

$$
\begin{aligned}
G\,(N_1, N_2, N_3, \alpha) \\
&= \frac{1}{N_1N_2N_3}\sum_{l_1=0}^{N_1-1}\sum_{l_2=-[N_2/2]}^{[N_2/2]-1}\sum_{l_3=-[N_3/2]}^{[N_3/2]-1} f\left(\frac{\alpha\pi l_1}{N_1}, \frac{2\alpha\pi l_2}{N_2}, \frac{2\alpha\pi l_3}{N_3}\right) \\
&= \frac{1}{4\pi^3}\int_\Omega f(\alpha\boldsymbol{x})\,\mathrm{d}\boldsymbol{x} - \frac{1}{8\pi^2N_1}\int_{\Omega_1}\mathrm{d}x_2\,\mathrm{d}x_3\left[f(\alpha\pi, \alpha x_2, \alpha x_3)\right. \\
&\quad \left.- f(0, \alpha x_2, \alpha x_3)\right] + \mathrm{O}\left(\frac{1}{N_1^2}, \frac{1}{N_2^2}, \frac{1}{N_3^2}\right) \qquad \text{(A7)}
\end{aligned}
$$

where $[x]$ means an integer equal to or smaller than x and

$$\Omega = \{(x_1, x_2, x_3)\,,\ x_1 \in [0,\pi]\,,\ x_2 \in [-\pi,\pi]\,,\ x_3 \in [-\pi,\pi]\}$$

$$\Omega_1 = \{(x_2, x_3)\,,\ x_2 \in [-\pi,\pi]\,,\ x_3 \in [-\pi,\pi]\}\,.$$

Thirdly,

$$
\begin{aligned}
G\,(N_1, N_2, N_3, \alpha) \\
&= \frac{1}{N_1N_2N_3}\sum_{l_1=-[N_1/2]}^{[N_1/2]-1}\sum_{l_2=-[N_2/2]}^{[N_2/2]-1}\sum_{l_3=-[N_3/2]}^{[N_3/2]-1} f\left(\frac{2\alpha\pi l_1}{N_1}, \frac{2\alpha\pi l_2}{N_2}, \frac{2\alpha\pi l_3}{N_3}\right) \\
&= \frac{1}{8\pi^3}\int_\Omega f(\alpha\boldsymbol{x})\,\mathrm{d}\boldsymbol{x} + \mathrm{O}\left(\frac{1}{N_1^2}, \frac{1}{N_2^2}, \frac{1}{N_3^2}\right) \qquad \text{(A8)}
\end{aligned}
$$

where

$$\Omega = \{(x_1, x_2, x_3)\,,\ x_1 \in [-\pi,\pi]\,,\ x_2 \in [-\pi,\pi]\,,\ x_3 \in [-\pi,\pi]\}\,.$$

Appendix B

It is easy to see that

$$n_{\mathrm{f}}(N,T) = \frac{1}{N^3}\sum_{q\neq 0}\frac{1}{\exp(\beta\varepsilon_q)-1}$$
$$= \frac{1}{N^3}\sum_{q\neq 0}\frac{1}{\exp(\bar{\beta}q^2)-1} - \frac{\beta}{N^3}\sum_{q\neq 0}\frac{\exp(\bar{\beta}q^2)\,\Delta\varepsilon_q}{\left[\exp(\bar{\beta}q^2)-1\right]^2} + \ldots \tag{B1}$$

where

$$\Delta\varepsilon_q = -4JS\left((q_1^4+q_2^4+q_3^4)/4! - (q_1^6+q_2^6+q_3^6)/6! + \ldots\right).$$

In the following, we give the details of the calculation of the first term of (B1). Let

$$G(N,T) = \frac{1}{N^3}\sum_{q\neq 0}\frac{1}{\exp(\beta q^2)-1}$$
$$= \frac{1}{8\pi^3}\int\frac{\mathrm{d}q}{\exp(\bar{\beta}q^2)-1} + \frac{1}{\bar{\beta}}\left(\frac{1}{N^3}\sum_{q\neq 0}\frac{1}{q^2} - \frac{1}{8\pi^3}\int\frac{\mathrm{d}q}{q^2}\right)$$
$$-\left(\frac{1}{N^3}\sum_{q\neq 0}\frac{1/2+\bar{\beta}q^2/3!+\ldots}{1+\bar{\beta}q^2/2+\bar{\beta}^2q^4/3!+\ldots}\right.$$
$$\left.-\frac{1}{8\pi^3}\int\frac{1/2+\bar{\beta}q^2/3!+\ldots}{1+\bar{\beta}q^2/2+\bar{\beta}^2q^4/3!+\ldots}\,\mathrm{d}q\right)$$
$$= -\frac{0.2268}{\bar{\beta}N} + \frac{\zeta\left(\frac{3}{2}\right)}{8\pi^{3/2}\bar{\beta}^{3/2}} + \mathrm{O}\left(\frac{1}{N^2}\right). \tag{B2}$$

The term in the first pair of parenthesis is numerically calculated. According to equation (A8), contribution from the second pair of parenthesis is of the order of $1/N^2$. Other terms in (B1) can be calculated directly from (A8).

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