PHYSICAL REVIEW B VOLUME 40, NUMBER 7 1 SEPTEMBER 1989

Absence of long-range order in three-dimensional Heisenberg models

E. Rastelli and A. Tassi
Dipartimento di Fisica dell'Università, 43100 Parma, Italy
(Received 16 May 1989)

The famous theorem of Mermin and Wagner excludes long-range order (LRO) in one- and two-dimensional Heisenberg models at any finite temperature if the exchange interaction is short ranged. Strong but nonrigorous indications exist about the absence of LRO even in three-dimensional Heisenberg models when suitable competing exchange interactions are present. We find, as a rigorous consequence of the Bogoliubov inequality, that this expectation is true. We find that for models where the exchange competition concerns at least two over three dimensions, a surface of the parameter space exists where LRO is absent. This surface meets at vanishing temperature the continuous phase-transition line which is the border line between the ferromagnetic and helical configuration.

Rigorous results are rare but welcome in statistical mechanics, as can be seen by the current theoretical effort[1] with particular emphasis on the Heisenberg model. In this context the theorem of Mermin and Wagner[2] is of primary importance because they were able to prove by means of the Bogoliubov inequality that long-range order (LRO) is absent at any finite temperature in isotropic one-dimensional (1D) and 2D Heisenberg models with finite-range exchange interaction.

The standard spin-wave analysis suggests that LRO can be destroyed even in 3D Heisenberg models in the presence of suitable competition of the exchange interaction. For instance, in some models on the boundary line between helical and ferromagnetic configurations the magnon dispersion curve shows a $k_\perp^4$ dependence, where $\mathbf{k}_\perp$ is the in-plane projection of the magnon wave vector. This obviously enters a catastrophic population number, which is a strong indication that no LRO is present. We notice that even if the magnon number is finite, LRO can be destroyed by different kinds of excitations, as is the case for the 1D Heisenberg model with uniaxial anisotropy. On the contrary, the divergence of the magnon number is usually sufficient to conclude that the system cannot have LRO.

In this Rapid Communication we give rigorous proof that in the presence of exchange competition, LRO is absent on a surface of the parameter space of 3D Heisenberg models in which the exchange competition extends over at least two independent directions. For instance, our statement applies when exchange competition up to third neighbors is present in *ab* planes of tetragonal or hexagonal lattices, independent of whether or not competition exists along the *c* axis. *A fortiori* the same conclusion is valid if the exchange competition extends over all the three main crystallographic directions. Moreover, we show that such a surface meets at vanishing temperature the continuous ferrohelical transition line.

The Hamiltonian of the model reads

$$H=-\sum_\alpha J_\alpha \sum_{i,\boldsymbol{\delta}_\alpha} \mathbf{S}_i\cdot\mathbf{S}_{i+\boldsymbol{\delta}_\alpha}, \tag{1}$$

where i labels the sites in which spins $\mathbf{S}_i$ are located, $\boldsymbol{\delta}_\alpha$ is a vector joining the site i with its neighbors of the αth shell, and J_α is half the exchange interaction between the spins in the sites i and $i+\boldsymbol{\delta}_\alpha$. Let us perform a local rotation of the reference axis given by

$$\begin{pmatrix} S_i^x \\ S_i^y \\ S_i^z \end{pmatrix} = \begin{pmatrix} 0 & -\sin\mathbf{Q}\cdot\mathbf{r}_i & \cos\mathbf{Q}\cdot\mathbf{r}_i \\ 0 & \cos\mathbf{Q}\cdot\mathbf{r}_i & \sin\mathbf{Q}\cdot\mathbf{r}_i \\ -1 & 0 & 0 \end{pmatrix} \begin{pmatrix} S_i^\xi \\ S_i^\eta \\ S_i^\zeta \end{pmatrix}, \tag{2}$$

where the x, y, and z axes are along the main crystallographic axes, while the ξ, η, and ζ axes are spiralling according to a helix characterized by the wave vector $\mathbf{Q}$. The Fourier transforms of the spin $\mathbf{S}_i$ and of the exchange interaction $J(\mathbf{r}_i)$ are defined in the usual way as

$$\mathbf{S}_i=\frac{1}{N}\sum_{\mathbf{k}}\mathbf{S}_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}_i}, \tag{3a}$$

$$J(\mathbf{r}_i)=\frac{1}{N}\sum_{\mathbf{k}}J(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{r}_i}. \tag{3b}$$

By application of Eqs. (2)–(3) to Eq. (1), one has

$$H=\frac{1}{N}\sum_{\mathbf{k}}\left[J(\mathbf{k})S_{\mathbf{k}}^\xi S_{-\mathbf{k}}^\xi+\tfrac{1}{2}[J(\mathbf{k}-\mathbf{Q})+J(\mathbf{k}+\mathbf{Q})](S_{\mathbf{k}}^\eta S_{-\mathbf{k}}^\eta+S_{\mathbf{k}}^\zeta S_{-\mathbf{k}}^\zeta)+\frac{1}{2i}[J(\mathbf{k}-\mathbf{Q})-J(\mathbf{k}+\mathbf{Q})](S_{\mathbf{k}}^\zeta S_{-\mathbf{k}}^\eta+S_{\mathbf{k}}^\eta S_{-\mathbf{k}}^\zeta)\right]. \tag{4}$$

The Bogoliubov inequality reads

$$\tfrac{1}{2}\langle\{A,A^\dagger\}\rangle\langle[[C,H],C^\dagger]\rangle\geq k_BT\,|\langle[A,C]\rangle|^2, \tag{5}$$

where $\langle \cdots \rangle$ means thermal average, and $\{\cdots\}$ and $[\cdots]$ are the anticommutator and commutator, respectively. If we choose $A=S^{\xi}_{-\mathbf{k}}$, $C=S^{\eta}_{\mathbf{k}}$, the inequality (5) becomes

$$\tfrac{1}{2}\langle S^{\xi}_{\mathbf{k}}S^{\xi}_{-\mathbf{k}}+S^{\xi}_{-\mathbf{k}}S^{\xi}_{\mathbf{k}}\rangle\left[\sum_{\alpha}2J_{\alpha}\sum_{\delta_\alpha}[(1-\cos\mathbf{k}\cdot\boldsymbol{\delta}_\alpha\cos\mathbf{Q}\cdot\boldsymbol{\delta}_\alpha)\langle S^{\xi}_{0}S^{\xi}_{\delta_\alpha}\rangle+(\cos\mathbf{Q}\cdot\boldsymbol{\delta}_\alpha-\cos\mathbf{k}\cdot\boldsymbol{\delta}_\alpha)\langle S^{\zeta}_{0}S^{\zeta}_{\delta_\alpha}\rangle]\right]\geq k_BTN\,|\langle S^{\zeta}_{0}\rangle|^2. \tag{6}$$

Summing both sides of (6) over $\mathbf{k}$, we may conclude that

$$S(S+1)\geq\frac{k_BT\,|\langle S^{\zeta}_{0}\rangle|^2}{N}\sum_{\mathbf{k}}\left[\sum_{\alpha}2J_{\alpha}\sum_{\delta_\alpha}[(1-\cos\mathbf{k}\cdot\boldsymbol{\delta}_\alpha\cos\mathbf{Q}\cdot\boldsymbol{\delta}_\alpha)\langle S^{\xi}_{0}S^{\xi}_{\delta_\alpha}\rangle+(\cos\mathbf{Q}\cdot\boldsymbol{\delta}_\alpha-\cos\mathbf{k}\cdot\boldsymbol{\delta}_\alpha)\langle S^{\zeta}_{0}S^{\zeta}_{\delta_\alpha}\rangle]\right]^{-1}. \tag{7}$$

If we choose $A=S^{\eta}_{-\mathbf{k}}$, $C=S^{\xi}_{\mathbf{k}}$, we obtain

$$S(S+1)\geq\frac{k_BT\,|\langle S^{\zeta}_{0}\rangle|^2}{N}\sum_{\mathbf{k}}\left[\sum_{\alpha}2J_{\alpha}\sum_{\delta_\alpha}\cos\mathbf{Q}\cdot\boldsymbol{\delta}_\alpha(1-\cos\mathbf{k}\cdot\boldsymbol{\delta}_\alpha)(\langle S^{\eta}_{0}S^{\eta}_{\delta_\alpha}\rangle+\langle S^{\zeta}_{0}S^{\zeta}_{\delta_\alpha}\rangle)\right]^{-1}. \tag{8}$$

When $\mathbf{Q}=0$, which corresponds to the ferromagnetic configuration, Eqs. (7) and (8) reduce to

$$S(S+1)\geq\frac{k_BT\,|\langle S^{\zeta}_{0}\rangle|^2}{N}\sum_{\mathbf{k}}\frac{1}{\sum_{\alpha}2\tilde{J}_{\alpha}(1-\cos\mathbf{k}\cdot\boldsymbol{\delta}_\alpha)}, \tag{9}$$

where

$$\tilde{J}_{\alpha}=J_{\alpha}(\langle S^{\xi}_{0}S^{\xi}_{\alpha}\rangle+\langle S^{\zeta}_{0}S^{\zeta}_{\alpha}\rangle), \tag{10}$$

because in this case $\langle S^{\xi}_{0}S^{\xi}_{\alpha}\rangle=\langle S^{\eta}_{0}S^{\eta}_{\alpha}\rangle$. The well-known Mermin and Wagner result [1] is obtained when we expand the denominator of Eq. (9) in the long-wavelength limit. In this limit it reduces to

$$Ak_{\perp}^2+Ck_z^2 \tag{11}$$

and no LRO is found in lesser than 3D because $C=0$ in 2D and $A=0$ in 1D.

However, it may happen that for a particular choice of the competing effective couplings $\tilde{J}_\alpha$, A or C, or both are zero. Indeed if we assume that there is a competition along the c axis we may have $C=0$. This occurs in the so-called axial next-nearest-neighbor Heisenberg model,[3] the quantum version of the well-known axial next-nearest-neighbor Ising model of Fisher and Selke.[4] In this case, however, the sum in Eq. (9) does not diverge in 3D and LRO should survive. On the contrary, if we consider models where the competition is in the *ab* plane, we may have $A=0$ and in this case LRO is destroyed in 3D at any finite temperature. Notice that real compounds exist, as the helimagnets $NiBr_2$,[5] NiI_2,[6] CoI_2,[6] well described by the Heisenberg Hamiltonian with competing exchange interactions in the *ab* plane of a hexagonal lattice for which Eq. (11) becomes

$$3(\tilde{J}_1+3\tilde{J}_2+4\tilde{J}_3)(ak_{\perp})^2+2\tilde{J}'(ck_z)^2, \tag{12}$$

where a and c are the in-plane and out-of-plane lattice constants. When

$$\tilde{J}_1+3\tilde{J}_2+4\tilde{J}_3=0, \tag{13}$$

the denominator of Eq. (9) has the form

$$-\tfrac{3}{16}(\tilde{J}_1+9\tilde{J}_2+16\tilde{J}_3)(ak_{\perp})^4+2\tilde{J}'(ck_z)^2, \tag{14}$$

which makes the sum in Eq. (9) logarithmically divergent and leads to the conclusion that LRO is destroyed on the surface (13). At low temperature, where

$$\langle S^{\xi}_{0}S^{\xi}_{\alpha}\rangle\simeq 0,\quad \langle S^{\zeta}_{0}S^{\zeta}_{\alpha}\rangle\simeq S^2, \tag{15}$$

Eq. (13) reduces to the well-known ferro-helix transition line.[7]

In a tetragonal lattice we find that the surface where LRO is destroyed is given by

$$\tilde{J}_1+2\tilde{J}_2+4\tilde{J}_3=0. \tag{16}$$

Even stronger divergence of the sum in Eq. (9) is found when competition is extended to all three dimensions, so that we may have a surface where both A and C vanish.

So we conclude that for any Heisenberg model where the exchange competitions affect at least two independent directions, LRO is lost in 3D on a surface whose equation is given by the condition that the coefficient of $k_{\perp}^2$ in Eq. (11) vanish. At low temperature this surface reduces to the line of second-order ferro-helix phase transition. This boundary line has been obtained in classical approximation,[7] but we have shown that its location is unchanged for a substantial part when quantum fluctuations are taken into account.[8]

An interesting result of the simple spin-wave theory is the possible absence of LRO even on the *infinite degeneration line* corresponding to the boundary line between two different helical configurations, where in classical approximation infinite inequivalent helices whose wave vectors span all the in-plane directions, minimize the energy of the model.[9] Unfortunately, the argument based on the Bogoliubov inequality that we have developed here is inconclusive concerning the infinite degeneration line, as one can see by examining Eq. (7). Indeed, the existence of quantum fluctuations prevents the sum over $\mathbf{k}$ on the right-hand side of Eq. (7) from diverging so that one cannot conclude that the order parameter vanishes. On the contrary, a divergence could occur in the classical limit because at vanishing temperature $\langle S^{\xi}_{0}S^{\xi}_{\delta_\alpha}\rangle$ vanishes as well as the prefactor of $\langle S^{\zeta}_{0}S^{\zeta}_{\delta_\alpha}\rangle\simeq S^2$ which vanishes for all $\mathbf{k}$ belonging to the locus $\mathcal{L}_{\mathbf{Q}}$ of the infinite degeneration line.[9]

In summary, we have rigorously proved that no LRO exists in 3D on a surface of the parameter space where the ferromagnetic configuration continuously meets the helical configuration in agreement with what one should expect from the simple spin-wave theory. We notice that this absence of LRO on the second-order ferro-helix transition surface could be tested by the absence of the elastic neutron scattering.

[1]K. Kubo and T. Kishi, Phys. Rev. Lett. **61**, 2585 (1988), and references therein.
[2]N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966).
[3]E. Rastelli and A. Tassi, J. Phys. C **19**, 1993 (1986).
[4]See, for instance, W. Selke, Phys. Rep. **170**, 213 (1988).
[5]P. Day, M. W. Moore, T. E. Wood, D. McK. Paul, K. R. A. Ziebeck, L. P. Regnault, and J. Rossat-Mignod, Solid State Commun. **51**, 627 (1984).
[6]S. R. Kuindersma, J. P. Sanchez, and C. Haas, Physica B **111**, 231 (1981).
[7]See, for instance, E. Rastelli, A. Tassi, and L. Reatto, Physica B **97**, 1 (1979).
[8]E. Rastelli and A. B. Harris, Phys. Rev. B (to be published).
[9]E. Rastelli, L. Reatto, and A. Tassi, J. Phys. C **16**, L331 (1983).