

deg mole. This value is of the order of that observed by Ahlers and Orttung<sup>8</sup> for the entropy under the  $\lambda$  spike.

Temperatures<sup>5</sup> at which side peaks in the nuclear-magnetic-resonance lines of solid H<sub>2</sub> appear and disappear upon cooling and warming, respectively, are plotted also in Fig. 1. These temperatures appear to be those at which the H<sub>2</sub> crystal first starts to change from hexagonal to cubic on cooling or finally complete the transition from cubic to hexagonal on warming. When the points are shifted down in temperature by half the  $\Delta T$  over which the structure change takes place, then the agreement with  $T_{tr}$  and  $T_\lambda$  is quite satisfactory as shown by the arrows in Fig. 1. It is a reasonable conjecture that the nmr anomaly results because the cubic symmetry of the crystal favors a splitting of the energy levels.

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## ABSENCE OF FERROMAGNETISM OR ANTIFERROMAGNETISM IN ONE- OR TWO-DIMENSIONAL ISOTROPIC HEISENBERG MODELS\*

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It is rigorously proved that at any nonzero temperature, a one- or two-dimensional isotropic spin- $S$  Heisenberg model with finite-range exchange interaction can be neither ferromagnetic nor antiferromagnetic. The method of proof is capable of excluding a variety of types of ordering in one and two dimensions.

The number of exact results on the presence or absence of phase transitions in systems with realistically short-range interactions is small: Van Hove has proved that there are no phase transitions in a one-dimensional classical gas with hard-core and finite-range interactions, and Griffiths has proved that the Ising model is ferromagnetic in more than one dimension.<sup>1</sup> More recently Hohenberg<sup>2</sup> has shown that an inequality due to Bogoliubov<sup>3</sup> can be used to exclude conventional superfluid or superconducting ordering in one or two dimensions. We have found that a similar application of the Bogoliubov inequality leads to a surprisingly elementary but rigorous argument that the one-

and two-dimensional isotropic Heisenberg models with interactions of finite range can be neither ferromagnetic nor antiferromagnetic at nonzero temperature. These conclusions have long been suspected, being suggested by calculations of the elementary excitations from the ordered state,<sup>4</sup> as well as by considerations of the energetics of domain walls.<sup>5</sup> In view of the present degree of activity in critical-point studies, it nevertheless seems worth recording that these very plausible results can be proved rigorously.<sup>6</sup>

We prove that there can be no spontaneous magnetization or sublattice magnetization in an isotropic Heisenberg model with finite-range

interactions at temperature  $T$ , by showing that for sufficiently small fields  $h$ ,

$$|s_z| < \frac{\text{Const.}}{T^{1/2}} \frac{1}{|\ln|h||^{1/2}} \quad (2 \text{ dimensions}), \quad (1)$$

$$|s_z| |h| < \frac{\text{Const.}}{T^{2/3}} |h|^{1/3} \quad (1 \text{ dimension}), \quad (2)$$

where  $s_z$  can be taken as either the infinite-volume limit of the magnetization per particle in a uniform field  $h$ , or the infinite-volume limit of the difference of the two sublattice magnetizations per particle, in a field of magnitude  $h$  and opposite sign on the two sublattices.

The proof exploits Bogoliubov's inequality,<sup>3</sup>

$$\frac{1}{2} \langle [A, A^\dagger] \rangle \langle [[C, H], C^\dagger] \rangle \geq k_B T |\langle [C, A] \rangle|^2, \quad (3)$$

where  $H$  is the Hamiltonian and  $\langle X \rangle = \text{Tr} X e^{-\beta H} / \text{Tr} e^{-\beta H}$  with  $\beta = 1/k_B T$ . The following elementary proof of the inequality is designed to make it clear that (3) is valid provided only that the operators  $A$  and  $C$  are such that the ensemble averages on the left-hand side of (3) exist.

Define

$$(A, B) = \sum_{ij}' \langle i | A | j \rangle^* \langle i | B | j \rangle \frac{W_i - W_j}{E_j - E_i},$$

where the sum is over all pairs from a complete set of energy eigenstates, excluding pairs with the same energy;  $W_i = e^{-\beta E_i} / \text{Tr} e^{-\beta H}$ . Note first that

$$0 < (W_i - W_j) / (E_j - E_i) < \frac{1}{2} \beta (W_i + W_j),$$

and, therefore,

$$(A, A) \leq \frac{1}{2} \beta \langle [A, A^\dagger] \rangle. \quad (4)$$

It is also easily verified that  $(A, B)$  satisfies all the definitions of an inner product necessary to establish the Schwartz inequality

$$(A, A)(B, B) \geq |(A, B)|^2. \quad (5)$$

Finally, if one chooses  $B = [C^\dagger, H]$ , then

$$\begin{aligned} (A, B) &= \langle [C^\dagger, A^\dagger] \rangle, \\ (B, B) &= \langle [C^\dagger, [H, C]] \rangle. \end{aligned} \quad (6)$$

Equation (3) follows from (4)-(6).

To apply the inequality to the Heisenberg model, let

$$\begin{aligned} H &= - \sum_{\vec{R}\vec{R}'} J(\vec{R}-\vec{R}') \vec{S}(\vec{R}) \cdot \vec{S}(\vec{R}') \\ &\quad - h \sum_{\vec{R}} s_z(\vec{R}) e^{-i\vec{K} \cdot \vec{R}}. \end{aligned} \quad (7)$$

To rule out ferromagnetism we will take  $\vec{K}$  to be 0, and to exclude antiferromagnetism, we chose it such that  $e^{i\vec{K} \cdot \vec{R}} = 1$  when  $\vec{R}$  connects sites in the same sublattice, and  $-1$  when it connects sites in different sublattices;  $\vec{R}$  and  $\vec{R}'$  run over the sites of a Bravais lattice with the usual periodic boundary condition to insure the presence of just  $N$  spins;  $J(0) = 0$ ,  $J(-\vec{R}) = J(\vec{R})$ , and  $J$  is of finite range.<sup>7</sup> Define

$$\vec{S}(\vec{k}) = \sum_{\vec{R}} e^{-i\vec{k} \cdot \vec{R}} \vec{S}(\vec{R}), \quad \vec{S}(\vec{R}) = (1/N) \sum_{\vec{k}} e^{i\vec{k} \cdot \vec{R}} \vec{S}(\vec{k}),$$

$$J(\vec{k}) = \sum_{\vec{R}} e^{-i\vec{k} \cdot \vec{R}} J(\vec{R}), \quad J(\vec{R}) = (1/N) \sum_{\vec{k}} e^{i\vec{k} \cdot \vec{R}} J(\vec{k})$$

(where sums over  $\vec{k}$  are always restricted to the first Brillouin zone).

If we take  $C = S_+(\vec{k})$ ,  $A = S_-(-\vec{k}-\vec{K})$ , then (3) gives

$$\begin{aligned} \frac{1}{2} \langle \{ S_+(\vec{k}+\vec{K}), S_-(-\vec{k}-\vec{K}) \} \rangle &\geq N k_B T s_z \left\{ (1/N) \sum_{\vec{k}} [J(\vec{k}) - J(\vec{k}-\vec{K})] \right. \\ &\quad \left. \times \langle S_z(-\vec{k}') S_z(\vec{k}') + \frac{1}{4} \{ S_+(\vec{k}'), S_-(-\vec{k}') \} \rangle + (N/2) h s_z \right\}^{-1}, \end{aligned} \quad (8)$$

where  $s_z = (1/N) \sum_{\vec{R}} \langle S_z(\vec{R}) e^{i\vec{K} \cdot \vec{R}} \rangle$ . The denominator on the right-hand side of (8) is positive, being

of the form  $(B, B)$ , and is therefore less than

$$\begin{aligned} (1/N) |\sum_{\vec{R}} J(\vec{R}) (1 - e^{i\vec{k} \cdot \vec{R}}) \sum_{\vec{k}'} e^{-i\vec{k}' \cdot \vec{R}} \langle S_z(-\vec{k}') S_z(\vec{k}) + \frac{1}{4} \{S_+(\vec{k}'), S_-(-\vec{k}')\} \rangle| \\ + \frac{1}{2} N h s_z | \langle N \sum_{\vec{R}} J(\vec{R}) (1 - \cos \vec{k} \cdot \vec{R}) S(S+1) + \frac{1}{2} N h s_z | \\ \langle \frac{1}{2} N [\sum_{\vec{R}} R^2 |J(\vec{R})| S(S+1) k^2 + |h s_z|] \rangle. \end{aligned} \quad (9)$$

We have used the fact that

$$\sum_{\vec{k}'} \langle S_i(\vec{k}') S_i(-\vec{k}') \rangle = N \sum_{\vec{R}} \langle S_i(\vec{R}) S_i(\vec{R}) \rangle = N^2 \langle S_i(\vec{R}_0) S_i(\vec{R}_0) \rangle \quad (10)$$

independent of  $\vec{R}_0$ . Replacing the denominator by this upper bound and summing both sides of (8) over  $\vec{k}$ , we may conclude that<sup>8</sup>

$$S(S+1) > 2k_B T s_z^2 (1/N) \sum_{\vec{k}} [S(S+1) \sum_{\vec{R}} R^2 |J(\vec{R})| k^2 + |h s_z|]^{-1} \quad (11)$$

In the infinite-volume limit (11) becomes

$$S(S+1) > \frac{2k_B T s_z^2}{\rho} \int_{\text{first zone}} \frac{d\vec{k}}{(2\pi)^d} [S(S+1) \sum_{\vec{R}} R^2 |J(\vec{R})| k^2 + |h s_z|]^{-1} \quad (12)$$

where  $1/\rho$  is the volume per spin and  $d$  is the number of dimensions. This inequality is strengthened if we integrate only over a sphere contained in the first Brillouin zone, so if  $k_0$  is the distance of the nearest Bragg plane from the origin in  $\vec{k}$  space, then

$$s_z^2 < \frac{2\pi\rho S(S+1)}{k_0^2} \frac{\omega}{kT \ln(1 + \omega/h s_z)} \quad (13)$$

(2 dimensions),

$$|s_z|^3 < h |\omega| \left( \frac{S(S+1)}{2kT \tan^{-1}[\omega/h s_z]} \right)^2 \quad (14)$$

(1 dimension),

$$\omega = \sum_{\vec{k}} S(S+1) k_0^2 R^2 |J(\vec{R})|.$$

In the limit of small  $h$  these reduce to (1) and (2).

The following additional points are of some interest:

(1) If the coupling is anisotropic the argument is inconclusive, but if  $J_y = J_z \neq J_x$ , then the same conclusions are reached for aligning fields in the  $z$  direction.

(2) Our inequality rules out only spontaneous magnetization or sublattice magnetization. It does not exclude the possibility of other kinds

of phase transitions. For example, a state with  $s_z = 0$  but  $(\partial s_z / \partial h)_T = \infty$  as  $h \rightarrow 0$  is not inconsistent with (1) or (2).

(3) A very similar argument<sup>9</sup> rules out the existence of long-range crystalline ordering in one or two dimensions, without making the harmonic approximation.

(4) Since our conclusions hold whatever the magnitude of  $S$ , one would expect them to apply to classical spin systems. We can, in fact, prove them directly by purely classical arguments in such cases.

This study was stimulated by our hearing of P. C. Hohenberg's argument in the superfluid case. We have also had very useful conversations with G. V. Chester, M. E. Fisher, and J. Langer.

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<sup>8</sup>We have not hesitated to sacrifice better bounds for simpler expressions [e.g.,  $\langle S_x^2(\vec{R}_0) + S_y^2(\vec{R}_0) \rangle < S(S+1)$ ], but such crudities affect only the constants in (1) and (2) and not the dependence on  $\hbar$  and  $T$ .

<sup>9</sup>N. D. Mermin and H. Wagner, to be published.

## EVIDENCE FOR RESONANT-MODE SIDEBANDS IN ALKALI HALIDES\*

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Recently sidebands of a local-mode absorption peak were observed in alkali halides doped by a very light negative impurity.<sup>1,2</sup> They were recognized to be produced by the combination of the local mode itself with an odd mode of the vibrational quasicontinuum,<sup>2,3</sup> through the anharmonic interaction. Hereafter, we call odd the modes whose displacements are antisymmetrical with respect to the defect site, i.e.,  $\vec{u}(-l) = -\vec{u}(l)$ . The experimental observation of the sidebands represents a direct method to detect the spectral density of the impurity-induced odd modes. Nevertheless, this method has been applied only in the case of the *U* center, the only case as yet known in which the defect is able to produce a local mode.

Our purpose here is to give theoretical support to the existence of sidebands for low-frequency resonant modes also, and to suggest how to detect them by stress experiments. We will show, indeed, that the application of an external stress makes one able to divide out the harmonic lattice absorption in the total absorption spectrum. One should then be able to extend to a good number of impurity-host lattice systems the experimental study on the odd-mode spectral densities. Since the coupling coefficients responsible for the sideband absorption are simply related to the cubic anharmonic coefficients in the defect-host-lattice interaction, stress experiments on the resonant as well as local-mode absorption peak are also suggested in order to deduce such coupling coefficients.

Consider an infrared (IR)-active resonant mode ( $\Gamma_{15}$  symmetry) which produces a narrow

absorption peak in the low acoustic-frequency region (for example,<sup>4</sup>  $^6\text{Li}^+$  in KBr). The two-phonon absorption processes which can occur below the reststrahl region are schematically shown in Table I.  $\Omega$  denotes the frequency of the absorbed light;  $\omega_e$  and  $\omega_o$ , the frequency of the even ( $\Gamma_{15}$ ) and odd phonons, respectively. The statistical weight for the process is reported in the last column, where  $n(\omega) = [\exp(\hbar\omega/kT) - 1]^{-1}$  denotes the phonon occupation number at absolute temperature  $T$ . Since we are concerned with low-temperature absorption spectra, we consider the contribution coming from the first process (Stokes sideband) and the small contribution coming from the second process (photon-resonant phonon annihilation with odd-symmetry phonon production), while we neglect the third process (anti-Stokes reststrahl sideband). Furthermore, we assume that the resonant mode does not extend appreciably beyond the first neighbors of the impurity, and we retain only terms of the type  $\Phi^{(3)}(0, 0, l) : \vec{u}(0)\vec{u}(0)\vec{u}(l)$  in the local anharmonic interaction [ $0$  denotes the impurity site;  $l$ , the nearest neighbors (nn)].

By using the method given by Nguyen,<sup>5</sup> the sideband contribution to the IR absorption co-

Table I. Two-phonon absorption processes generating sidebands below the reststrahl region.

Process	Weight
$\Omega = \omega_o + \omega_e$ (resonance)	$1 + n(\omega_e) + n(\omega_o)$
$\Omega = \omega_o - \omega_e$ (resonance)	$n(\omega_e) - n(\omega_o)$
$\Omega = \omega_e$ (reststrahl) $-\omega_o$	$n(\omega_o) - n(\omega_e)$