Infinitely Many Commensurate Phases in a Simple Ising Model

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On the basis of systematic low-temperature expansions "to all orders", it is shown that an infinite sequence of spatially modulated commensurate phases, with wave vectors $\pi j/(2j+1)a$ $(j=0,1,2,\ldots)$, occurs in simple, anisotropic Ising models with nn couplings J_0, $J_1>0$, in between spin-$\frac{1}{2}$ layers, and competing nnn interlayer couplings $J_2=-\kappa J_1$ along one axis. The free energies, interfacial tensions, and phase boundaries are found for low T in $d>2$ dimensions.

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The structure and behavior of spatially modulated phases of matter have recently attracted increasing theoretical attention.[1] The "sinusoidal" magnetic order observed in erbium and in other rare-earth elements provided an early challenge.[2] More recently, the complicated phase diagram of cerium antimonide (CeSb), which exhibits ordered magnetic layers with various different periods commensurate with the underlying lattice,[3] has stimulated theoretical attack.[4,5] Also striking is the phenomenon of "pure stage ordering", where, e.g., in graphite intercalation compounds, one observes[6] periodic structures of $j-1$ host layers followed by one intercalant layer, with j as high as 10; the corresponding phase diagrams have recently been studied theoretically by mean-field theory.[7]

The behavior observed, and that predicted by phenomenological theories, is often surprisingly complex (e.g., devil's staircases[4,5,8]), but there have been few definitive results for specific microscopic models that might serve as testing points for approximate but more general theories, or demonstrate unequivocally the range of phase behavior implicit in a given Hamiltonian—a matter also of intrinsic interest from the viewpoint of basic statistical mechanics. In this Letter we respond to this need by considering what are, perhaps, the simplest nontrivial models exhibiting periodically ordered phases, namely spin-$\frac{1}{2}$ Ising models with nearest-neighbor interactions augmented by competing next-nearest-neighbor couplings acting parallel to a single lattice axis.[2] We report calculations[9] which show that as a function of temperature these models can display an *infinite sequence of distinct spatially modulated phases*, each characterized by a commensurate wave vector

$$q_j=\pi j/(2j+1)a \quad (j=1,2,\ldots), \tag{1}$$

and having a definite periodic layered structure, denoted by $\langle 2^{j-1}3\rangle$, which means a sequence of $j-1$ pairs of lattice layers pointing (predominantly) two "up" and two "down", followed by three layers all pointing (predominantly) "up", or "down", to maintain an overall "antiphase" character, as illustrated for $\langle 2^2 3\rangle$ in Fig. 1 (a is the lattice spacing). At low temperatures where our analysis is valid,[9] it yields expressions for the free energies, and interfacial tensions, which demonstrate that no other structures occur[8,10] and that the transitions between adjacent phases are of first order although, as $j\to\infty$, the extent of each phase decreases and the wave vector ulti-

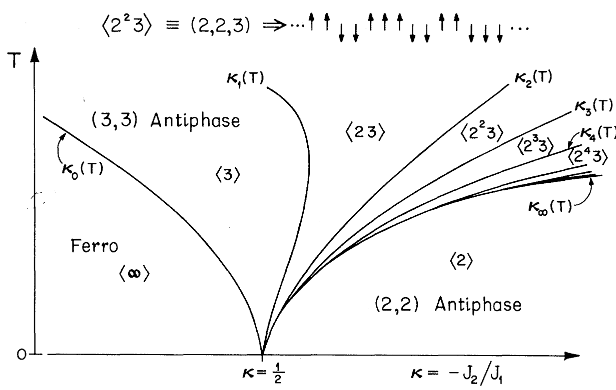

FIG. 1. Schematic phase diagram of the anisotropic next-nearest-neighbor Ising or ANNNI model in the plane of temperature T and parameter $\kappa=J_2/J_1$, exhibiting the infinite sequence of commensurate, layered antiphase states, $\langle 2^{j-1}3\rangle$, at low temperatures.

mately varies quasicontinuously according to

$$q_\infty - q(T,\kappa) \sim 1/\ln\{[\kappa_\infty(T)-\kappa]^{-1}\}, \qquad (2)$$

where $\kappa = -J_2/J_1$ measures the relative strength of the competing interaction constants, J_1 and J_2, between spins in nearest- and next-nearest layers, while $\kappa_\infty(T) \geqslant \kappa$ is a smooth function of T which delimits the $\langle 2^2\rangle \equiv (2,2)$ antiphase state (see Figs. 1 and 2).

These results are explained in more detail below, where we also present a brief sketch of the theoretical analysis,[9] which is based on a systematic low-temperature expansion "to all orders" in powers of $w = \exp(-2J_0/k_BT)$, where J_0 is the coupling strength of each spin to its $q_\perp$ nearest neighbors in its $(d-1)$-dimensional layer. Although the anisotropic next-nearest-neighbor Ising or ANNNI models are very simple, we believe many of their features will be reflected in real systems and more realistic models. Furthermore, the last decade in the experimental study of phase transitions has shown that many seemingly artificial microscopic models can be realized with surprising accuracy in appropriate physical systems.

The ANNNI model with *ferro*magnetic nn couplings ($J_0, J_1 > 0$) and competing *anti*ferromagnetic couplings ($\kappa > 0$) is of particular interest[4,5,10-12] and will be considered here. High-temperature expansions[11] for the sc lattice indicate a transition from paramagnetic to a sinusoidally magnetized phase for $\kappa > \kappa_L \simeq 0.27$. Monte Carlo studies[12] confirm the spatially modulated character of the ordered states for $\kappa > \kappa_L$ but show that the equilibrium wave vector varies strongly with both T and κ. The nature of this variation, however, cannot be established reliably by Monte Carlo work, particularly at lower temperatures. On the other hand, one may prove[12b] that for $\kappa < \frac{1}{2}$ the ground state is purely ferromagnetic, while for $\kappa > \frac{1}{2}$ the ground state is the fourfold-degenerate $(2,2) \equiv \langle 2\rangle$ "antiphase" state of wave vector $q = q_\infty \equiv 2\pi/4a$. However, the character of the low-temperature phases in the vicinity of the borderline $\kappa = \frac{1}{2}$ has been a matter of speculation.[5,10,12] In fact, as illustrated in Fig. 1, the point $(T=0, \kappa=\frac{1}{2})$ is a *multiphase point*[8] from which, when $d > 2$, spring the infinite number of distinct antiphase states $\langle 2^{j-1}3\rangle$, which interpolate discretely between the ferromagnetic and (2, 2) antiphase states. The jth phase is limited by first-order transition boundaries, $\kappa_{j-1}(T) < \kappa_j(T)$, where, with $\kappa = \frac{1}{2} + \delta$, $x = e^{-2K_1}$, $K_1 = J_1/k_BT$, the ferro-(3, 3) boundary is given by

$$\delta_0(T)K_1 = -\tfrac{1}{2}w^{q_\perp}(1-\tfrac{3}{2}x+\tfrac{1}{2}x^3) - \tfrac{1}{4}q_\perp w^{2q_\perp-2}(1-\tfrac{3}{2}x^2+\tfrac{1}{2}x^6) + \tfrac{1}{4}(q_\perp+6)w^{2q_\perp}[1+O(x)] + O(w^{3q_\perp-4}), \qquad (3)$$

while all subsequent boundaries may be described through the recursion relations

$$\kappa_{j+1} - \kappa_j \approx (\tfrac{1}{2}j + \tfrac{3}{4})[j+2-(j+3)vw^{q_\perp}]uv^j w^{(j+1)q_\perp}/K_1, \qquad (4)$$

in which $u(T) = (1-x)^2(1+\frac{1}{2}x)$, $v(T) \approx 1 - xe^{-4K_1\Delta_0}$, $K_1\Delta_0(T) = uw^{q_\perp}$.

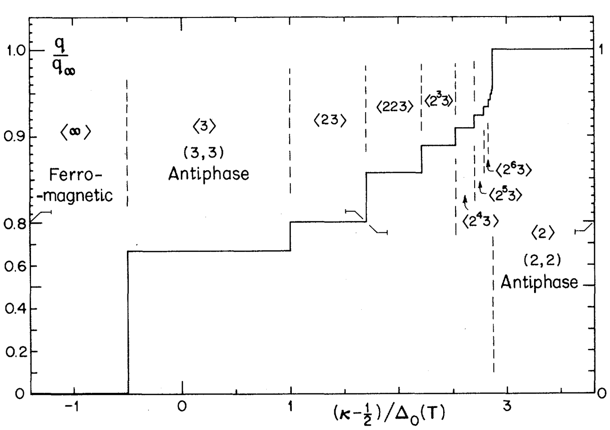

FIG. 2. Variation of the wave vector q for $\kappa \simeq \frac{1}{2}$. Note the break in the vertical scale at $q/q_\infty = 0.8$ ($q_\infty = \pi/2a$); the horizontal scale, set by $\Delta_0(T) \approx (k_BT/J_1)e^{-2q_\perp J_0/k_BT}$, is partially schematic [see (4) with $x \to 0$ and $2J_0q_\perp \approx k_BT$].

Note that on heating the system from $T=0$ at fixed $\kappa \gtrsim \frac{1}{2}$, the (2, 2) antiphase state with $q = q_\infty$ remains locked in up to a finite temperature at which it "melts" abruptly, essentially by nucleating widely, but with equally spaced *bands* of three "up" and three "down" layers at increasing density. This corresponds to a solitonlike mechanism[1] for varying q: Thermodynamically, however, the melting once started proceeds through a series of discrete first-order transitions as illustrated in Fig. 2. The first-order character of the phase boundaries at low T can also be seen by calculating the interfacial tension between adjacent phases. For an interface parallel to the lattice layers on the boundary $\kappa_j(T)$, this is[9]

$$\Sigma_j(T) \approx \tfrac{1}{2}k_BT\, w^{(2j+1)q_\perp}x^{2-2\kappa_j}(1-x^{2\kappa_j})^{2(j+1)}, \qquad (5)$$

with corrections of relative order $w^{q_\perp-2}$. As T falls, $\Sigma_j(T)$ first rises, but then drops to zero in critical-point fashion when $T \to 0$. Since $(T=0,$

$\kappa=\frac{1}{2}$) is a multiphase point of infinite order this is not so surprising.

The main steps in establishing the above conclusions[9] are the following:

(i) Classification and systematic description of all possible ground states by the values of an infinite set of structural variables l_μ where μ labels finite sequences of "up" or "down" ferromagnetically ordered layers.

(ii) Expansion of the reduced free energy per spin, $f(\{l_\mu\}; T, \kappa) = -F_N/Nk_BT$, for *all* ground states in powers of the in-layer coupling parameter w for general x, by overturning spins in the ferromagnetically aligned layers: To ensure convergence of this layer-based expansion, $d>2$ is required.

(iii) Minimization of the free energy, $-f(\{l_\mu\})$, over the l_μ at low (but nonzero) T to determine the structures of the stable phases: Since the l_μ enter the expansion for $f(\{l_\mu\})$ *only linearly*, this leads to a linear-programming problem[13] in the space $\mathfrak{L}$ of the *standard structural variables* l_ν; the optimal vertices of the appropriate convex polytope, $\mathcal{P}$, in this space determine the discrete stable phases.[13]

(iv) Solution of the linear-programming problem in sequential stages, $n=1, 2, 3, \ldots$, corresponding to increasing powers of $w^{q_\perp}$ arising from overturning successively more spins; full calculations have been performed for 1, 2, and 3 overturned spins (involving analysis of 5, 19, and 96 distinct local configurations).[9] However, to elucidate the complete sequence of phases and demonstrate that the ith optimal vertex describes the periodic state $\langle 2^{j-1}3\rangle$, one must calculate the leading terms in the free-energy differences to orders $w^{(2j+1)q_\perp}$ for *all j*.

The interested reader may grasp the ideas via the initial details: Since $J_0>0$, all spins in a given layer are parallel in any ground state. A sequence of k adjacent layers oriented the same way but bordered by oppositely pointing layers defines a *k band:* For a system of length-L layers, let l_kL be the number of k bands; similarly, let $l_{kk'}L$ be the number of k bands which are followed consecutively by a k' band; likewise for $l_{kk'k''}$, etc. By their definition these variables satisfy linear *structural relations* such as

$$\textstyle\sum_k k l_k = 1, \quad \sum_{k'} l_{kk'} = l_k, \quad \text{etc.,} \tag{6}$$

which reduce the number of variables needed to describe a ground state to a standard set $\{l_\nu\}$. The inequalities $l_\mu \geqslant 0$ (all μ) then limit realizable ground states to a convex polytope, $\mathcal{P}$, in $\mathfrak{L}$.

Now the energy of a spin in any configuration can be determined, given the orientations of its nearest and next-nearest axial neighbors. The ground-state energy can hence be written

$$E_0 = -\tfrac{1}{2}q_\perp J_0 - \tfrac{1}{2}J_1 - J_1\delta[2l_2 + l_3 - \sum_{k\geq 4}^{\infty}(k-4)l_k], \tag{7}$$

with $l_1 \equiv 0$. For $\delta>0$ this is minimal for $l_2=\frac{1}{2}$ and $l_k=0$ ($k\neq 2$) which uniquely describes the state $\langle 2\rangle$; conversely, for $\delta<0$ the minimum specifies the ferromagnetic state. The excited states corresponding to single overturned spins likewise yield the first-order free-energy contribution

$$\Delta f^{(1)} = w^{q_\perp}\{2x^{1+2\delta}l_2 + (2+x^{3+2\delta})l_3 + \sum_{k\geq 4}^{\infty}[2+(k-4)x^{1-2\delta}+2x^2]l_k\}, \tag{8}$$

which is again linear in the l_μ. Now combine with (7), eliminate l_3 by (6), and examine $f(\{l_\mu\})$ for small δ: One sees that any state with $l_4, l_5, \ldots >0$ cannot have minimal free energy. Thus near $\kappa=\frac{1}{2}$ the *allowed phases* arise from ground states containing *only* 2 bands or 3 bands. Furthermore, *to this order* and for a range of $\delta=O(w^{q_\perp})$ a new phase with $l_2=0$ and, hence, $l_3=\frac{1}{3}$ becomes stable: This is just the state $\langle 3\rangle$.

When two spins are overturned the structural varaibles $l_{23}=l_{32}$, $l_{22}=l_2-l_{23}$ and $l_{33}=\frac{1}{3}-\frac{2}{3}l_2-l_{23}$ are needed. With l_2 and l_{23} as standard variables,[9] the free-energy expansion reads

$$f(\{l_\mu\}; T, \kappa) = a_0(w, x, \delta) + (\tfrac{4}{3}K_1\delta + w^{q_\perp}b_2)l_2 + w^{2q_\perp}b_{23}l_{23} + \sum_{n=3}^{\infty} w^{nq_\perp}\sum_{\nu(n)} b_\nu(x,\delta)l_\nu, \tag{9}$$

where the $b_\nu \approx -\frac{4}{3}$ and $b_{23}\approx 3$ as $w, x\to 0$. The allowed region of the (l_2, l_{23}) plane is the triangle $\mathcal{P}_2 \equiv (\langle 2\rangle, \langle 3\rangle, \langle 23\rangle)$, whose third vertex at $l_2=l_{23}=\frac{1}{5}$ specifies the (2, 3) state which then appears as a new stable phase between phases $\langle 3\rangle$ and $\langle 2\rangle$ but with $\delta_2-\delta_1=O(w^{2q_\perp})$. Likewise one discovers that *no new phases* appear between the ferromagnetic and $\langle 3\rangle$ phases: One may then calculate the leading term in the interfacial tension, $\Sigma_0(T)$ [see (5)].

At the third stage, two new standard variables, l_{223} and l_{233}, are required; the corresponding polytope, $\mathcal{P}_3$, has new vertices describing states

$\langle 233\rangle$, $\langle 2233\rangle$, and $\langle 223\rangle$ but, because[9] $b_\nu \approx (j+2) \times (1-x^2)(1-x^{1+2\delta})^{j+1}$ for $\nu = 2^j 3$ and $b_\nu \approx -x^{1-2\delta} \times (1-x^{1+2\delta})^{2(j+1)}$ for $\nu = 2^j 3 2^{j-i} 3$, as $w^{q_\perp} \to 0$, only $\langle 223\rangle$ appears as a stable phase, between $\langle 23\rangle$ and $\langle 2\rangle$ with $\delta_3 - \delta_2 = O(w^{3q_\perp})$. More generally, by using these results in (9) at stages $n = j+1$ and $n = 2j+1$, respectively, an inductive argument can be constructed which yields the remaining conclusions.

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Raman Scattering from Nonequilibrium LO Phonons with Picosecond Resolution

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A novel technique is described for time-resolved Raman scattering for studying the dynamics of nonequilibrium excitations on a picosecond time scale. The generation and the decay of nonthermal LO phonons in GaAs is measured, and $\tau' = 7 \pm 1$ ps is obtained for the relaxation time of the phonon population at 77 K.

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This paper describes a novel technique for time-resolved spontaneous Raman scattering. As a first application we investigate the generation and relaxation of nonequilibrium optical phonons in a solid. The relaxation of optical phonons occurs on a picosecond time scale and is still inadequately understood. The information on the phonon relaxation processes obtained from the analysis of the broadening of Raman and infrared spectra is rather indirect and incomplete. Direct measurements in the time domain, on the other hand, have provided a detailed picture of the dephasing of molecular vibrations[1] and of the decay of *coherent* optical phonons.[2-5] Here we present, for the first time, a measurement of the time revolution of nonequilibrium *incoherent* optical phonons. We observe the generation of optical phonons during the interaction of photoexcit-

