Phonon-induced phase transition in a classical Heisenberg chain*

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The linear chain of classical Heisenberg spins coupled to phonons is studied. The coupling arises from the phonon-modulation of the nearest-neighbor exchange integral and the cases of both direct exchange and superexchange are treated. The phonons lead to an effective biquadratic interaction between spins, with important physical consequences. Thermodynamic functions at finite temperatures and zero field are evaluated exactly and discussed. It is shown that antiferromagnetically coupled spins in an external field at $T=0°K$ can undergo a first-order phase transition; as the field crosses a critical value, the magnetization and lattice spacing change discontinuously.

I. INTRODUCTION

Although models exhibiting spin-phonon interactions have been studied for several years, exact solutions so far have been confined to Ising systems.[1-5] The present paper obtains the thermodynamic properties and correlation functions of a linear chain of classical Heisenberg spins whose coupling is modulated by phonons. Bolton and Lee[6] have noted that with classical Heisenberg spins and a spin-phonon interaction linear in the phonon operators, the Hamiltonian may be transformed into parts which separately depend on spin and lattice degrees of freedom. However, the transformed spin Hamiltonian is not usually exactly solvable. Part of the interest of the present model stems from its being an exception in this respect.

The method of solution employs a unitary transformation which is a direct generalization of that used by Mattis and Shultz[1] for the corresponding spin-$\frac{1}{2}$ Ising problem. However, unlike the transformed Hamiltonian obtained by Mattis and Schultz,[1] that obtained here exhibits biquadratic interactions—and these have important physical consequences. Besides affecting the thermodynamic properties and correlation functions (which can be calculated exactly at all temperatures in zero magnetic field[7,8]), the interactions can lead to a first-order phase transition at zero temperature; both the magnetization and the lattice spacing change discontinuously as the field crosses a critical value.

It is shown that these results hold even when the magnetic coupling arises through superexchange. A rough estimate of the phonon-induced interaction shows that it may well be large enough for the phase transition to be observable.

II. HAMILTONIAN AND UNITARY TRANSFORMATION

The Hamiltonian under study here is

$$\mathcal{H}=J\sum_i \hat{S}_i\cdot\hat{S}_{i+1}-H\sum_i S_i^z -\gamma\sum_i(x_{i+1}-x_i)\hat{S}_i\cdot\hat{S}_{i+1}+\frac{1}{2m}\sum_i p_i^2+\frac{k}{2}\sum_i(x_{i+1}-x_i)^2. \tag{1}$$

$\hat{S}_i$ is a classical unit vector, S_i^z is its z component, and H is the external field applied along the z axis. x_i is the displacement of site i from its mean position with $\gamma=0$ and p_i is its momentum. x_i and p_i are quantum-mechanical operators obeying the commutation rule $[x_j,p_l]=i\delta_{jl}$. k (>0) is the spring constant between adjacent sites and m is the mass of each site.

The interactions in Eq. (1) are envisaged to arise from direct exchange. The spin-phonon coupling arises as the strength of the exchange coupling $\tilde{J}(r)$ between two sites is a function of their separation r. Assuming that expanding $\tilde{J}(r)$ as a function of r and then retaining terms linear in site displacements is justified, the Hamiltonian in Eq. (1) results, with the identification $J=\tilde{J}(a)$ and $\gamma=-[\partial\tilde{J}(r)/\partial r]_{r=a}$, where a is the equilibrium separation of the sites in the absence of spin-phonon coupling. The form of the spin-phonon interaction, when $\tilde{J}(r)$ arises via superexchange, is discussed in Sec. V.

Salinas[4] has pointed out that the Ising chain interacting with phonons exhibits different properties under different constraints. Here free boundary conditions are assumed, corresponding to fixing the applied pressure at zero and allowing the length of the chain to vary.

Defining

$$U=\prod_i U_i, \tag{2}$$

with

$$U_i=\exp\left(i\frac{\gamma}{k}p_i\sum_{j<i}\hat{S}_j\cdot\hat{S}_{j+1}\right),$$

one finds

$$Ux_iU^{-1}=x_i+\frac{\gamma}{k}\sum_{j<i}\hat{S}_j\cdot\hat{S}_{j+1}, \tag{3}$$

$$U\mathcal{H}U^{-1}=\mathcal{H}_S+\mathcal{H}_{\text{ph}}, \tag{4}$$

where

$$\mathcal{H}_S=J\sum_i\hat{S}_i\cdot\hat{S}_{i+1}-H\sum_i S_i^z - A\sum_i(\hat{S}_i\cdot\hat{S}_{i+1})^2, \tag{5}$$

$$\mathcal{H}_{\text{ph}}=\sum_i\frac{p_i^2}{2m}+\frac{k}{2}\sum_i(x_{i+1}-x_i)^2, \tag{6}$$

and

$$A=\gamma^2/2k. \tag{7}$$

The effect of U is to uncouple the Hamiltonian into parts which depend separately on the spin and lattice degrees of freedom. The transformation exploits the classical (i.e., commuting) nature of the spins; although a similar transformation may be used for a pair of quantum-mechanical Heisenberg spins,[9] the generalization to a chain is not possible.

In contrast to the Ising system studied by Mattis and Schultz,[1] here there is a phonon-induced biquadratic coupling between the spins. With $H=0$, the linear chain of classical spins with isotropic bilinear and biquadratic coupling has been studied previously[4,5] and the partition function and correlation functions have been found. The thermodynamic properties are briefly discussed in Sec. III. As A is always positive, the possibility of competition[7] between the bilinear (BL) and biquadratic (BQ) terms does not arise. The situation is quite different with nonzero H, and in Sec. IV it is shown that a first-order phase transition can occur at $T=0$ °K as H is changed, as a result of different types of ordering favored by the BL and BQ terms.

The partition function Z factors:

$$Z\equiv\text{Tr}e^{-\beta\mathcal{H}}=Z_SZ_{\text{ph}}, \tag{8a}$$

where Tr denotes integration over the spin variables together with a trace over phonon states and

$$Z_S=\int\prod_{i=1}^N\frac{d\hat{S}_i}{4\pi}e^{-\beta\mathcal{H}_S}, \tag{8b}$$

$$Z_{\text{ph}}=\text{tr}\,e^{-\beta\mathcal{H}_{\text{ph}}}. \tag{8c}$$

Here $\beta=T^{-1}$ (T is the temperature and Boltzmann's constant has been set equal to unity). The integrals in Eq. (8b) are over all orientations of the vectors $\hat{S}_i$, and the trace in Eq. (8c) is over phonon states. Let $\langle\cdots\rangle$ denote $(1/Z)\text{Tr}e^{-\beta\mathcal{H}}\ldots$, and let $\langle\cdots\rangle_0$ denote $(1/Z)\text{Tr}e^{-\beta(\mathcal{H}_S+\mathcal{H}_{\text{ph}})}\ldots$.

Using Eq. (3) and the invariance of the trace under unitary transformations, one then has

$$\zeta\equiv\langle x_{i+1}-x_i\rangle=(\gamma/k)\langle\hat{S}_i\cdot\hat{S}_{i+1}\rangle_0. \tag{9}$$

The linear chain is thus seen to exhibit magnetostriction, and for small fields H the distance between nearest neighbors is less (greater) than the value with $\gamma=0$, provided that γ has a sign which is the same as (opposite to) J. As the temperature rises, $\langle\hat{S}_i\cdot\hat{S}_{i+1}\rangle_0$ monotonically decreases in absolute magnitude and the chain correspondingly expands (contracts) to its length with $\gamma=0$.

III. FINITE-TEMPERATURE PROPERTIES IN ZERO FIELD

H is set equal to zero throughout this section. With this constraint, Z_S has been evaluated previously[7,8] and straightforward differentiation yields the corresponding spin specific heat per site,

$$C_S=\beta^2[A^2(\tilde{I}_4-\tilde{I}_2^2)-2JA(\tilde{I}_3-\tilde{I}_2\tilde{I}_1)+J^2(\tilde{I}_2-\tilde{I}_1^2)], \tag{10}$$

where

$$\tilde{I}_n=I_n/I_0 \tag{11}$$

and

$$I_n=\int_{-1}^1 x^ne^{\beta(Ax^2-Jx)}\,dx. \tag{12}$$

The specific heat of the system is the sum of C_S and the phononic contribution.

Using the expression for the correlation function $\langle\hat{S}_i\cdot\hat{S}_j\rangle_0$ obtained in Refs. 7 and 8 in Eq. (9), one finds the change in the intersite spacing is

$$\zeta=(\gamma/k)\tilde{I}_1 \tag{13}$$

and the temperature derivative of ζ is

$$\alpha\equiv\frac{d\zeta}{dT}=\frac{\gamma\beta^2}{k}[J(\tilde{I}_2-\tilde{I}_1^2)-A(\tilde{I}_3-\tilde{I}_1\tilde{I}_2)]. \tag{14}$$

For the Ising model considered by Mattis and Schultz,[1] α is proportional to C_s. From Eqs. (10) and (13) it is seen that such a relation holds for the Heisenberg chain only in the limit of weak spin-phonon coupling ($\gamma, A\to 0$).

The zero-field magnetic susceptibility may be evaluated, expressing it in terms of spin correlation functions and noting that

$$\langle\hat{S}_i\cdot\hat{S}_j\rangle=\langle\hat{S}_i\cdot\hat{S}_j\rangle_0.$$

The result is

$$\chi^{zz} = \frac{\beta\mu^2}{3} \frac{1-\tilde{I}_1}{1+\tilde{I}_1}, \qquad (15)$$

where μ is the magnetic moment of the electron.

To analyze the low-temperature behavior of various thermodynamic quantities, one may use the readily proved recursion relation

$$2\beta A I_{n+1} = e^{\beta A}[e^{-\beta J} + (-1)^{n+1} e^{\beta J}] + \beta J I_n - n I_{n-1} \qquad (16)$$

together with the asymptotic form of I_0. Writing $\tilde{I}_0$ in terms of an error function of complex argument,[7] and using the asymptotic form of the latter[10] one finds

$$I_0 \xrightarrow[\beta\to\infty]{} \frac{e^{\beta(A+|J|)}}{2\beta(A+\frac{1}{2}|J|)} \left[1 + \frac{1}{2\beta A(1+|J|/2A)^2} + \frac{3}{4\beta^2 A^2 (1+|J|/2A)^4} + \cdots\right]. \qquad (17)$$

In the absence of spin-phonon coupling, it is known[11] that $C_S \to 1$ as $T \to 0\,°\text{K}$—a reflection of the classical nature of the spins. With $A \neq 0$, C_S approaches the same value, as can be shown from Eqs. (10), (16), and (17). α approaches the value, $(\text{sgn}\,J)\gamma/[k(|J|+2A)]$, as $T \to 0\,°\text{K}$.

With ferromagnetic coupling between spins ($J<0$), Eqs. (15)–(17) yield

$$\chi^{zz} \xrightarrow[T\to 0\,°\text{K}]{} \tfrac{2}{3}\beta^2\mu^2(|J|+2A). \qquad (18)$$

The amplitude of the divergence is enhanced over its value in the absence of coupling to phonons.

With antiferromagnetic coupling ($J>0$),

$$\chi^{zz} \xrightarrow[T\to 0\,°\text{K}]{} \frac{\mu^2}{6(J+2A)}. \qquad (19)$$

The coupling to phonons in this case acts to diminish the susceptibility.

In either case (i.e., J positive or negative) the effect of the phonon-induced BQ interactions as $T \to 0\,°\text{K}$ is to effectively increase the value of $|J|$. This seems intuitively plausible (if $H=0$) as the BQ terms in $\mathcal{H}_S$ do not distinguish between configurations in which adjacent spins are parallel or antiparallel; in either case they tend to reinforce the effect of the BL interactions.

This is no longer necessarily true in the presence of an applied magnetic field; the types of ordering favored by the BQ and BL terms may not be identical and, as shown in Sec. IV, a first-order phase transition can result.

IV. PHASE TRANSITION WITH CHANGING H AT T=0°K

In this section, the ground state of the spin system described by the Hamiltonian $\mathcal{H}_S$ [Eq. (5)] is studied. The results are valid for arbitrary lattice dimensionality for the analogous Hamiltonian with nearest-neighbor BL and BQ interactions, although for dimensions larger than one, such a Hamiltonian does not result from spin-phonon interactions.

In order to understand why the phase transition occurs, consider the case of antiferromagnetic J with an applied magnetic field. (J will be restricted to be positive throughout this section.) For small fields, the ground state of the system is as illustrated in Fig. 1(a). The larger the value of J, the smaller will be the bending of the spins, and the smaller the susceptibility. In this regime it is clear that the BQ terms assist the BL terms in opposing the applied field.

However, when θ is close to 90°, the BQ terms side with the magnetic field in tending to align the spins parallel. It is this ambivalence in the ordering tendency of the BQ terms which leads to a first-order phase transition. The latter occurs when the state with $\theta = 90°$ [Fig. 1(b)] becomes energetically favorable over one with a small value of θ [Fig. 1(a)].

Writing $\mathcal{H}_S = \sum_i h_{i,i+1}$, it follows that

$$E_S \geq \sum_i (E_{i,i+1}), \qquad (20)$$

where E_S and $E_{i,i+1}$ are the ground-state energies of $\mathcal{H}_S$ and $h_{i,i+1}$, respectively. The equality holds in (20) provided the ground states of $h_{i,i+1}$ for different i are compatible. This proves to be the case[12] if

$$h_{i,i+1} = J\hat{S}_i\cdot\hat{S}_{i+1} - A(\hat{S}_i\cdot\hat{S}_{i+1})^2 - \tfrac{1}{2}h(S_i^z + S_{i+1}^z). \qquad (21)$$

Thus it is only necessary to find the configuration which minimizes $h_{1,2}$. It is easily seen that in the

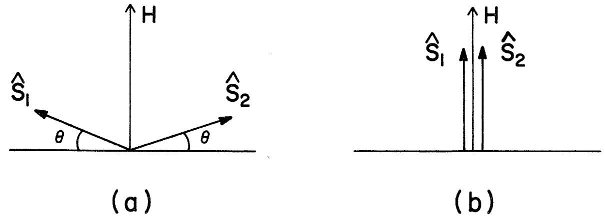

FIG. 1. (a) Ground-state configuration with antiferromagnetic coupling is illustrated for the case $H < H_c$. (b) When the field increases and crosses the critical value H_c, the spins suddenly line up along the field.

lowest-energy configuration, the magnetic field bisects the angle between $\hat{S}_1$ and $\hat{S}_2$. Defining θ to be the angle made by each of the spins with the axis normal to the magnetic field [Fig. 1(a)] the problem reduces to finding the value of θ which maximizes

$$f(\theta) = J\cos 2\theta + A\cos^2 2\theta + H\sin\theta \tag{22}$$

in the domain $0 \leq \theta \leq 90°$.

Setting the first derivative equal to zero, one obtains

$$\cos\theta = 0 \tag{23a}$$

or

$$\sin\theta\cos 2\theta = H/8A - (J/2A)\sin\theta. \tag{23b}$$

Equation (23b) may have zero, one, or two roots in the domain of interest depending on the relative magnitudes of J, A, and H. The two-root case is of interest here. Figure 2 shows a plot of the left- and right-hand sides of Eq. (23b) (the solid and dashed curves, respectively), with $J/A = 4$ and $H/A = 10.52$. The intersections of the curves determine the locations of the local maximum M_1 (say at θ_0) and the local minimum M_2 of $f(\theta)$. The function $f(\theta)$ is plotted in Fig. 3 for the same values of J/A and H/A. It is seen that $f(\theta_0)$ equals $f(90°)$ for these values of the parameters. Were H slightly smaller, $f(\theta_0) > f(90°)$ would hold; if slightly larger, $f(90°) > f(\theta_0)$ would hold. Thus, holding J fixed at $4A$, as H crosses the critical value $H_c = 10.52A$, there is a first-order phase transition as the spins move from the spin-flopped phase [Fig. 1(a)] to one in which they are aligned along the field [Fig. 1(b)].

The staggered magnetization M_s normal to the field is a measure of the antiferromagnetic order in the system. As H increases and crosses H_c, M_s jumps from $\cos\theta_0$ to zero. There is a corresponding discontinuous change in the inter-site spacing, as the value of the spin correlation $\langle \hat{S}_1 \cdot \hat{S}_{i+1} \rangle_0$ changes abruptly. Using Eq. (9), one has

$$\zeta_{H \to H_c(+)} - \zeta_{H \to H_c(-)} = (2\gamma/k)\cos^2\theta_0. \tag{24}$$

A necessary condition for the transition to occur is that the solid and dashed curves (see Fig. 2) intersect at two points. As H is changed, the dashed curve is displaced vertically. Increasing J/A corresponds to making the dashed curve steeper. It is clear that when J/A is sufficiently large, there can be only one point of intersection. Consequently, $f(\theta)$ has only one maximum point in $0 \leq \theta \leq 90°$ and no discontinuous phase transition is possible if A is too small.[18]

The first-order transition discussed here is quite different from that discussed by Bean and Rodbell,[13] with increasing magnetic field. The transition considered by them occurs in a ferromagnet above the Curie temperature; as the field increases, the ferromagnetic-order parameter increases discontinuously. In contrast, in the phase transition in the antiferromagnet discussed here, the order parameter M_s decreases discontinuously to zero as the field increases.

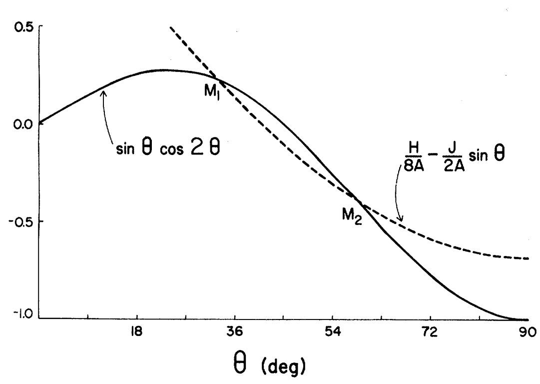

FIG. 2. Left- and right-hand sides of Eq. (23b) are plotted with $J = 4A$ and $H = H_c = 10.52A$. The points of intersections of the two curves determine the positions of two of the extrema of $f(\theta)$. With sufficiently large J/A, there is at most one point of intersection and no phase transition is possible.

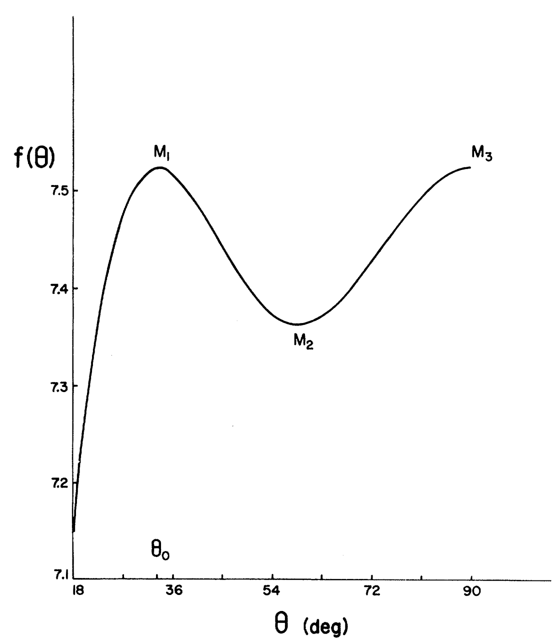

FIG. 3. $f(\theta)$ is plotted with $J = 4A$ and $H = H_c = 10.52A$. The maximum of $f(\theta)$ determines the ground-state configuration. As H increases and crosses H_c, the location of the maximum jumps from θ_0 to 90°.

V. PHONON MODULATION OF SUPEREXCHANGE COUPLING

It is interesting that the generalization of the results of Sec. II–IV to the case of *super*exchange coupling presents no additional difficulty. Such a generalization is useful if one wants to estimate the magnitude of the phonon-induced BQ coupling.

Suppose that there is a nonmagnetic (Y) site between every pair of magnetic (X) sites. Further suppose that the only nonzero overlap is between wave functions on adjacent X and Y sites and that this overlap occurs in the tail regions of exponentially decaying wave functions. Let σ and τ be the inverse decay lengths of the exponentials corresponding to X and Y sites, respectively.

Let x_i, x_{i+1} be the instantaneous longitudinal displacements of succesive X sites and let y_i be the displacement of the Y site between them. Then the hopping integral $\tilde{b}$ between the two magnetic (X) sites is proportional to

$$\int dr\, e^{-\sigma(r-x_i)+\tau(r-y_i)} \int dr'\, e^{-\tau(r'-y_i)+\sigma(r'-x_{i+1})},$$

and consequently

$$\tilde{b} = b e^{-\sigma(x_{i+1}-x_i)},$$

where b is the hopping integral with the magnetic sites at their equilibrium positions. Assuming that the instantaneous value of the superexchange coupling is proportional to $\tilde{b}^2$ and expanding to first order in the x_i, one obtains the total Hamiltonian

$$\mathcal{H}' = J\sum_i \hat{S}_i \cdot \hat{S}_{i+1} - H\sum_i S_i^z - \gamma\sum_i (x_{i+1}-x_i)\hat{S}_i \cdot \hat{S}_{i+1} + \mathcal{H}'_{\rm ph}, \tag{25}$$

where

$$\gamma = 2\sigma J \tag{26}$$

and

$$\mathcal{H}'_{\rm ph} = \frac{k}{2}\sum_i [(y_i - x_i)^2 + (x_{i+1}-y_i)^2] + \sum_i \left(\frac{(p_i^x)^2}{2m} + \frac{(p_i^y)^2}{2M}\right). \tag{27}$$

p_i^x and p_i^y are the momenta of the X and Y sites, and m and M are their respective masses.

Define $U' = \prod_i U_i'$,

$$U_i' = \exp\left(i(p_i^x + p_i^y)\frac{2\gamma}{k}\sum_{j<i} \hat{S}_j \cdot \hat{S}_{j+1} + i p_i^Y \frac{\gamma}{k}\hat{S}_i \cdot \hat{S}_{i+1}\right).$$

Then

$$U'\mathcal{H}'U'^{-1} = \mathcal{H}'_S + \mathcal{H}'_{\rm ph},$$

where $\mathcal{H}'_{\rm ph}$ has been defined in Eq. (27) and

$$\mathcal{H}'_S = J\sum_i \hat{S}_i \cdot \hat{S}_{i+1} - H\sum_i S_i^z - A'\sum_i (\hat{S}_i \cdot \hat{S}_{i+1})^2. \tag{28}$$

Here

$$A' = \gamma^2/k \tag{29}$$

and equals twice A, the value of the biquadratic interaction resulting from the phonon modulation of direct exchange.

The value of A' is now estimated, using parameters relevant to manganous oxide (MnO).

The dispersion of the system described by $\mathcal{H}'_{\rm ph}$ is known.[14] In the long-wavelength limit,

$$k = [(m+M)/2\pi^2]R^2, \tag{30}$$

where R is the $q \to 0$ slope of the acoustic branch in the plot of frequency versus the wave vector q in units of π/a. Neutron-scattering experiments[15] yield the value $R = 1.4 \times 10^{13}$ Hz, which implies $k = 1100$ erg cm^{-2}.

The radial Hartree-Fock wave function[16] for the manganese ion is the sum of differently decaying functions. The two decay rates of importance at distances of interest (i.e., 1–2 Å) are $\sigma_1 = 1.46$ (a.u.)$^{-1}$ and $\sigma_2 = 2.41$ (a.u.)$^{-1}$.

The next-nearest-neighbor exchange in MnO (which is the result of 180° superexchange) has been estimated[17] to be 3.5 °K. Since this is the interaction between two $S = \frac{5}{2}$ spins, the coupling J between unit vectors in the Hamiltonian is ≈ 30 °K.

Equations (26) and (29) imply

$$A'/J = 4\sigma^2 J/k.$$

With the values of J and k given above and $\sigma = \sigma_1$, it is found that $A'/J \approx 1$. This value of A'/J is large enough[18] for the phase transition described in Sec. IV to occur with a critical field of several hundred kilogauss. Using σ_2 instead of σ_1, one would estimate a larger value of A'/J and a smaller H_c.

The aim has been to get some idea of the sizes of the parameters involved, rather than to predict the occurrence of the transition described in Sec. IV in a specific substance. In MnO, nearest-neighbor direct-exchange interactions compete with the next-nearest-neighbor superexchange interactions which were used as a basis for the estimates. A more realistic model than that considered here would also include the effects of anisotropic terms in the spin Hamiltonian and account for the nonharmonicity of the lattice to begin with.

One may speculate on the persistence of the phase transition as the temperature is raised. One expects that the one-dimensional system at $T \neq 0$ °K will not exhibit the discontinuity while a three-dimensional system probably will.

VI. SUMMARY

The effects of the coupling of a linear chain of Heisenberg spins to phonons have been studied. The direct-exchange case was treated first, and a displaced-oscillator transformation was used to decouple the Hamiltonian into parts which separately depended on the spin and lattice variables. The lattice underwent magnetostriction, and the transformed spin Hamiltonian contained biquadratic terms. Thermodynamic functions were evaluated exactly and studied in zero external field.

Next the effect of an external magnetic field was investigated at $T=0\,^{\circ}$K. It was found that provided the induced biquadratic coupling was large enough, antiferromagnetically coupled spins underwent a first-order phase transition as the field crossed a critical value, with the magnetization changing discontinuously. The phase transition was a consequence of the form of the resultant spin Hamiltonian $\mathcal{H}_s$ and occurred in all dimensions in systems described by a similar Hamiltonian, whether or not the Hamiltonian resulted from phonon effects. The length of the compressible linear chain was seen to change abruptly at the phase transition.

Finally, it was shown that it was straightforward to extend the results to a simplified model of superexchange. A rough estimate of the phonon-induced biquadratic coupling was made, and it was found to be large enough in order for the phase transition in an applied field to occur.

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[18]The critical value of A is $A_c=J/10$. The phase diagram and a discussion of the region $A\approx A_c$ will be published elsewhere.