PHYSICAL REVIEW B VOLUME 24, NUMBER 9 1 NOVEMBER 1981

Spin-Peierls transition of the anisotropic XY model in a magnetic field: Thermodynamic properties

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(Received 26 May 1981)

The spin-Peierls phase transition for a quasi-one-dimensional XY chain is studied as a function of the degree of anisotropy of the exchange interaction (γ) and of the magnetic field (H). It is found that both anisotropy and magnetic field decrease the critical temperature. When γ and H progressively increase, the phase transition is first of second order, passes to first order, and finally the ordered phase completely disappears. The corresponding magnetization, magnetic susceptibility, and specific heat are given. In particular, a peak and a discontinuity in the magnetic susceptibility for finite magnetic field are found at the critical temperature, this discontinuity being maximum at the tricritical point separating the first- and second-order lines. For a second-order transition the magnetization behaves continuously through the critical point where a discontinuity is present only in the first derivative with respect to temperature. For the first-order case a discontinuity is present.

I. INTRODUCTION

An antiferromagnetic exchange interaction between the spins of singly occupied sites of a linear chain leads to a dimerization instability analog to that of the Peierls instability. When a large number of these chains form a crystal, a spin-Peierls phase transition, driven by the instability, can occur if the interchain magnetic coupling is weak enough[1,2] with respect to the intrachain coupling. This phase transition is predicted to happen as well for a XY or a Heisenberg interaction. However it is much easier to treat the XY case because the Hamiltonian can be exactly diagonalized in terms of quasifermion operators.[3] The Heisenberg interaction is much harder to treat because the quasifermion-quasifermion interaction terms must be treated approximately. Fortunately, the qualitative behavior predicted for the Heisenberg spin-Peierls transition is nearly identical to that of the XY case.[2]

The addition of an anisotropy in the XY or Heisenberg interaction is of interest because it adds a new degree of freedom to the parameters of the problem. This anisotropy, in real systems, can come either from the presence of a strong crystal field or from a large spin-orbit coupling. The anisotropic dimerized XY Hamiltonian with antiferromagnetic coupling was diagonalized by Dubois and Carton[4] without external fields and by Perk *et al.*[5] when a magnetic field is present. The behavior of the spin-Peierls transition in the presence of anisotropy only was investigated by Lépine and Caillé.[6] It was found that when the degree of anisotropy increases, the critical temperature decreases, that the initial second-order phase transition is transformed into a first-order one, and that finally for a large anisotropy the dimerized phase disappears for any temperature. This behavior as a function of the degree of anisotropy is very similar to that as a function of a magnetic field only as found by Tannous and Caillé.[7] Another interesting feature of the anisotropic Hamiltonian is that for the uniform phase a gap is present in the elementary magnetic excitations.[6] This gap decreases when the degree of dimerization increases and completely disappears when the degree of anisotropy is equal to the degree of dimerization. This gap subsequently increases for larger dimerization. The effect of this peculiar behavior on the thermodynamic properties of the system needs an investigation.

In this paper, I will concentrate on the thermodynamic properties of the anisotropic XY spin-Peierls phase transition, with a magnetic field applied. Within this model, two parameters can be varied to investigate the behavior of the phase transition. In the second section, I will present the Hamiltonian used, its eigenvalues, and discuss the energy levels found. In the third section, the behavior of the degree of dimerization as a function of temperature is derived. A phase diagram of critical temperature versus the degree of anisotropy is given for various magnetic fields. In the fourth section, the magnetization, the specific heat, and the magnetic susceptibility are given for different values of the parameters. Finally, a discussion of these results will be given that stresses the physical behavior of the system.

II. HAMILTONIAN AND ENERGY BANDS

The Hamiltonian used will be that of a linear alternating chain of $2N$ spins with an anisotropic antiferromagnetic XY interaction, as used by Dubois and Carton,[4] with a magnetic field added:

$$\hat{H} = J_1 \sum_{i=1}^{N} (1+\gamma) S_i^x T_i^x + (1-\gamma) S_i^y T_i^y + J_2 \sum_{i=1}^{N} (1+\gamma) T_i^x S_{i+1}^x + (1-\gamma) T_i^y S_{i+1}^y + \mu H \sum_{i=1}^{N} (S_i^z + T_i^z) + E_{\rm el} \ , \quad (1)$$

where $\vec{S}_j$ and $\vec{T}_j$ are the spin operators for the first and second electron of the jth link, respectively. J_1 and J_2 are the two values of the exchange parameters of a dimerized chain. A linear dependence of $J_{1,2}$ on δ, the degree of dimerization, will be used: $J_{1,2} = J(1 \pm \delta)$. γ is the degree of anisotropy for the exchange interaction: $\gamma = \pm 1$ is the Ising model and $\gamma = 0$ is the usual XY isotropic model. $E_{\rm el}$ is the elastic contribution of the lattice. In its simplest form it is quadratic: $E_{\rm el} = NC\delta^2$ because δ is directly proportional to ξ, the degree of distortion of the lattice. $\mu = 2\mu_B$ is twice the Bohr magneton, and H is the magnetic field.

The magnetic part of the Hamiltonian given in Eq. (1) can be diagonalized, following the procedure described by Perk *et al.*[5] (It is the same Hamiltonian, with some minor modifications.) The procedure consists in performing a Wigner-Jordan transformation to fermion operators and then to diagonalize the resulting Hamiltonian. The Hamiltonian obtained is

$$\hat{H} = \sum_{-\pi < ka < \pi} \epsilon_k' u_k^\dagger u_k + \epsilon_k'' v_k^\dagger v_k - \tfrac{1}{2}(\epsilon_k' + \epsilon_k'') \ , \quad (2)$$

where

$$\epsilon_k' = [c_2 + (c_2^2 - c_4)^{1/2}]^{1/2} \ ,$$

$$\epsilon_k'' = [c_2 - (c_2^2 - c_4)^{1/2}]^{1/2} \ ,$$

$$c_2 = (\mu H)^2 + J^2(1 + \delta^2\gamma^2)\cos^2 ka/2 + J^2(\delta^2 + \gamma^2)\sin^2 ka/2 \ ,$$

$$c_4 = [(\mu H)^2 - J^2(1 - \delta^2\gamma^2)\cos^2 ka/2 - J^2(\delta^2 - \gamma^2)\sin^2 ka/2]^2 + 4J^4(\gamma - \delta^2\gamma)^2 \sin^2 ka/2 \cos^2 ka/2,$$

a is the length of a dimer, and u_k and v_k are second quantization operators for two different quasifermion bands. The constant term corresponds to the energy of two filled quasifermion valence bands, that are mirror images of the conduction bands. If $H = 0$, Eq. (2) reduces to the expressions of the anisotropic XY Hamiltonian of Refs. 4 and 6. If $\gamma = 0$, Eq. (2) reduces to

$$\hat{H} = \sum_k (J_k + \mu H) u_k^\dagger u_k + (J_k - \mu H) v_k^\dagger v_k - \tfrac{1}{2} \sum_k (J_k + \mu H) + (J_k - \mu H) \ , \quad (3)$$

with

$$J_k = J[1 - (1 - \delta^2)\sin^2 ka/2]^{1/2} \ ,$$

a result equivalent to that found by Tannous and Caillé.[7] [Equation (3) is written in a particle-hole formalism.[5]]

The two bands are degenerate if $c_3 = (c_2^2 - c_4)^{1/2}$ is zero. This occurs in three cases: $H = 0$ and $\gamma = 0$, $H = 0$ and $\delta = 0$, or $\delta = 0$ and $\gamma = 0$. The energy gap determined from Eq. (2) is not always at the corner of the Brillouin zone. In the absence of magnetic field, it occurs between the ϵ'' bands and always at the zone corner: $k = \pi/a$. This gap is zero only when $\gamma = \delta$.[6] In the isotropic case ($\gamma = 0$), the gap is at the zone corner until $\delta J = \mu H$. For larger magnetic fields, the gap is no longer present and the minimum of the ϵ_k'' band occurs for $\mu H = J_k$, a value of k that can be far from the zone corner. If δ, γ, and H are different from zero, a mixture of these situations occurs and the gap can be anywhere in the Brillouin zone or can be zero.

III. PHASE DIAGRAM

As the number of quasifermions is not conserved (chemical potential equal to zero), the Gibbs free energy is obtained from

$$G = E_{\rm el} - k_B T \ln\left[\mathrm{tr}\exp\left[\frac{-\hat{H}}{k_B T}\right]\right] \ , \quad (4)$$

with the MH term included in $\hat{H}$. Substituting Eq. (2), we find for the free energy per dimer:

$$\frac{G}{NJ} = \frac{E_{\rm el}}{NJ} - \frac{k_B T}{NJ} \sum_k \ln\left(4\cosh\frac{\beta\epsilon_k'}{2}\cosh\frac{\beta\epsilon_k''}{2}\right) \quad (5)$$

with $\beta = 1/k_B T$. For the isotropic case ($\gamma = 0$), Eq. (5) can be written

$$\frac{G}{NJ} = \frac{E_{\rm el}}{NJ} - \frac{k_B T}{NJ} \sum_k \ln\left(4\cosh\frac{\beta(J_k + h)}{2}\cosh\frac{\beta(J_k - h)}{2}\right) \ , \quad (6a)$$

a result identical to that of Tannous and Caillé[7]:

$$\frac{G}{NJ}=\frac{E_{el}}{NJ}-\frac{k_BT}{NJ}\sum_k\{\ln[1+2\exp(-\beta\mu H)\cosh\beta J_k+\exp(-2\beta\mu H)]\}-h\ , \tag{6b}$$

where h is $2\mu_BH/J$.

When the magnetic field is absent, results of Ref. 6 are regained (anisotropic alternated XY model).

The phase diagram is obtained by a minimization of Eq. (5) with respect to δ for a given set of parameters $(C/J,\gamma,h)$. For a second-order phase transition, the critical temperature is obtained when δ_{min} passes from zero to a finite value. For a first-order phase transition, two minima are present in the neighborhood of the transition. The critical temperature is then obtained from a comparison of $G(0)$ with $G(\delta_{min})$. In Fig. 1 appears the behavior of δ_{min} (proportional to the degree of dimerization) as a function of temperature (kT/J) for $C/J=0.5$, $h=0.1$, and $\gamma=0$, 0.1, 0.125, 0.13, 0.135 and 0.139. It is seen that for small anisotropy ($\gamma<\gamma_t\approx0.125$), the transition is of second order, that for $\gamma_c>\gamma>\gamma_t$ it is of first order, and that for $\gamma>\gamma_c\approx0.1395$ the dimerized phase is no longer stable. For higher magnetic fields, the value of γ_t, the tricritical point, is smaller (γ_t becomes zero between $h=0.1$ and $h=0.2$). For smaller magnetic fields, γ_t is higher but the phase transition always becomes of first order before $\gamma=\gamma_c$ where the ordered phase disappears. The value of γ_c decreases when the magnetic field increases. The phase diagram, obtained from Fig. 1 and similar results for $h=0$ and 0.2, are shown in Fig. 2, where k_BT_c/J is plotted as function of γ for $C/J=0.5$ and $h=0$, 0.1, 0.2. The full drawn lines are second-order lines and the dotted ones are first-order lines. From this figure, we observe the following: T_c is maximum for the isotropic chain without magnetic field; T_c decreases when the magnetic field (h) increases; and T_c decreases when the degree of anisotropy (γ) increases. For large values of h or γ, the transition passes from second to first order. For still larger values of h or γ, the phase transition disappears: the dimerized phase no longer exists. When $\gamma\neq0$ and $h\neq0$ both at the same time, the rate of decreasing of T_c is larger and the phase transition becomes of first order and disappears sooner than with $\gamma\neq0$ or $h\neq0$ alone. The effect of anisotropy or magnetic field is thus to reduce the critical temperature.

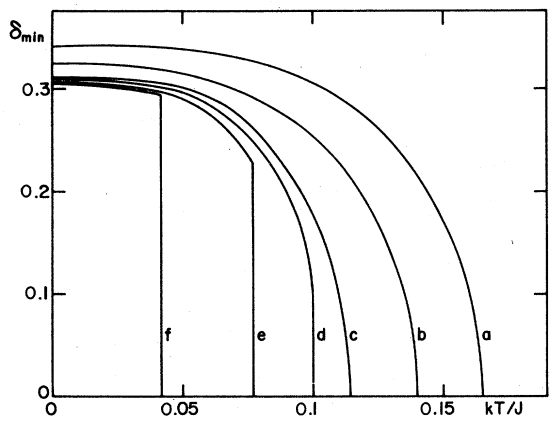

FIG. 1. Degree of dimerization (δ_{min}) vs kT/J for $C/J=0.5$ and $h=0.1$. a: $\gamma=0$, b: $\gamma=0.1$, c: $\gamma=0.125$, d: $\gamma=0.13$, e: $\gamma=0.135$, and f: $\gamma=0.139$.

IV. MAGNETIC SUSCEPTIBILITY, MAGNETIZATION, AND SPECIFIC HEAT

From the phase diagram obtained in Sec. III, physical quantities that can be measured are obtained, apart from the distortion of the lattice. Of these, I will consider the magnetization, the magnetic susceptibility, and the specific heat.

These thermodynamic quantities are obtained directly from their definition as applied to Eq. (5)[8]

$$M=-\left(\frac{\partial G}{\partial H}\right)_T\ , \tag{7}$$

$$\chi=-\left(\frac{\partial M}{\partial H}\right)_T=-\left(\frac{\partial^2G}{\partial H^2}\right)_T\ , \tag{8}$$

$$C_H=-T\left(\frac{\partial^2G}{\partial T^2}\right)_H=\left(\frac{\partial H'}{\partial T}\right)_H\ . \tag{9}$$

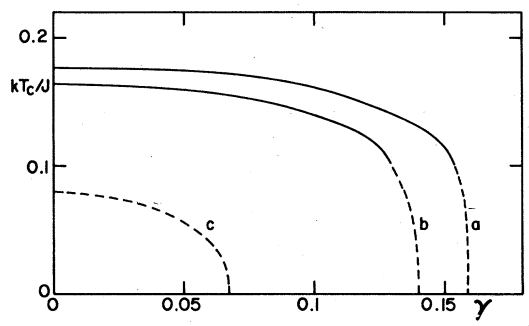

FIG. 2. Phase diagram of kT_c/J vs γ for $C/J=0.5$. The full drawn lines are second-order transition lines and the dotted ones are first order. a: $h=0$, b: $h=0.1$, and c: $h=0.2$.

H' is the enthalpy of the system, obtained from Eq. (2) by replacing the expectation value of the number operators by the Fermi-Dirac distribution:

$$H' = -\frac{1}{2}\sum_k \epsilon_k' \tanh\left(\frac{\beta\epsilon_k'}{2}\right) + \epsilon_k'' \tanh\left(\frac{\beta\epsilon_k''}{2}\right) . \quad (10)$$

The actual numerical calculations work as follows: First we obtain a curve of the degree of dimerization (δ) as a function of temperature by a minimization of Eq. (5). Then M, χ, and C_H are obtained directly by numerical differentiation, either from Eq. (10) or of Eq. (5). The relation $\chi = M/H$ cannot be used here because the dependence of M on H is not linear except in the limit when H goes to zero. In the derivatives, care is taken of the variation of δ with T or H and the term in $C\delta^2$ is always present, describing the change in energy of the lattice. This variation of δ with H makes the difference with the results of Perk *et al.*[5] for the magnetic susceptibility. (Note that the variation of δ with T or H is negligible when we compute the magnetization because the value of δ is that of a minimum of G.)

In Fig. 3, I plot the magnetic susceptibility $\chi J/N\mu^2$ versus the temperature parameter kT/J for $\gamma = 0$, 0.125, 0.13, 0.135, and 0.139 and for $H = 0.1$. In each case, a discontinuity is observed in χ at T_c, due to the abrupt change of slope of the dependence of δ on magnetic field (δ passes from zero to finite values at T_c). This behavior is different from that obtained in the isotropic case by Tannous and Caillé,[7] who obtained χ from the magnetization M divided by H. Such a result is only valid for small magnetic fields or for a linear dependence of χ on H. Their results did not exhibit the discontinuity because M involves only the first derivative of G with respect to h and

$$M = \frac{\partial G}{\partial H} = \frac{\partial G}{\partial \delta}\frac{\partial \delta}{\partial H} + \left(\frac{\partial G}{\partial H}\right)_\delta . \quad (11)$$

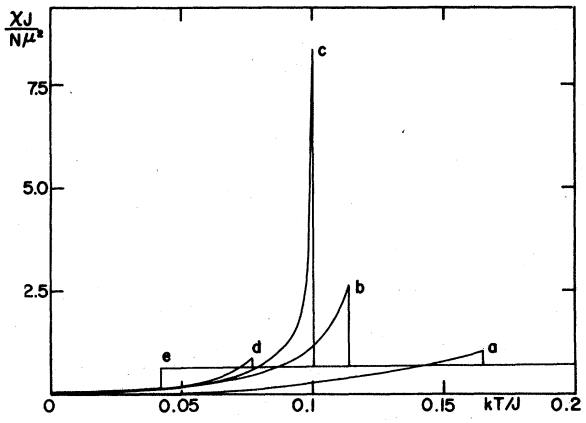

FIG. 3. Magnetic susceptibility vs kT/J for $C/J = 0.5$ and $h = 0.1$. a: $\gamma = 0$, b: $\gamma = 0.125$, c: $\gamma = 0.13$, d: $\gamma = 0.135$, and e: $\gamma = 0.139$.

The first term, which involves the first derivative of δ with respect to h, is zero because G is at a minimum with respect to δ. The discontinuity is thus absent. Also it can be observed that the discontinuity is at a maximum at the tricritical point ($\gamma = 0.13$ for $h = 0.1$). It is in this case that the change of slope of δ vs T at T_c is the highest (one passes from a zero slope to an infinite one). It can also be observed from Fig. 3 that $\chi(T=0) = 0$ if $\gamma = 0$ but is finite for finite values of γ.

Simple analytical expressions can be obtained for limiting cases. For $H = 0$, $\chi = M/H$ and the dependence of δ on H can be neglected because $\partial G/\partial\delta = 0$. The magnetic susceptibility is then

$$\frac{\chi J(H=0)}{N\mu^2} = \lim_{H\to 0}\frac{M}{H} = \frac{1}{\pi}\int_0^{\pi/2} dk \left[\frac{\tanh\beta\epsilon_k'/2}{\epsilon_k'}\left(1 + \frac{\cos^2 k + \delta^2\sin^2 k}{\gamma\delta}\right) + \frac{\tanh\beta\epsilon_k''/2}{\epsilon_k''}\left(1 - \frac{\cos^2 k + \delta^2\sin^2 k}{\gamma\delta}\right)\right] , \quad (12)$$

where ϵ_k' and ϵ_k'' are evaluated at $H = 0$. For $\delta = 0$ and $H = 0$, the limit of Eq. (12) gives

$$\frac{\chi J}{N\mu^2} = \frac{2}{\pi}\int_0^{\pi/2} dk \frac{\tanh\beta(\cos^2 k + \gamma^2\sin^2 k)^{1/2}/2}{(\cos^2 k + \gamma^2\sin^2 k)^{1/2}}\left(1 - \frac{\cos^2 k}{\cos^2 k + \gamma^2\sin^2 k}\right)$$

$$+ \frac{\beta}{\pi}\int_0^{\pi/2} dk \left[\frac{\mathrm{sech}^2\beta(\cos^2 k + \gamma^2\sin^2 k)^{1/2}/2}{\cos^2 k + \gamma^2\sin^2 k}\right]\cos^2 k\, dk \quad (13)$$

and for $H=0$ and $\gamma=0$, we obtain

$$\frac{\chi J}{N\mu^2}=\frac{\beta}{\pi}\int_0^{\pi/2} dk\ \mathrm{sech}^2\beta(\cos^2 k+\delta^2\sin^2 k)^{1/2}/2\ . \tag{14}$$

In all cases, the magnetic susceptibility decays exponentially when the temperature decreases to zero (the only exception is when $\delta=\gamma=0$, the uniform case, for which the susceptibility slowly saturates to a finite value). The limiting value is 0 for the isotropic case ($\gamma=0$), but is finite as soon as a small anisotropy is present. From the numerical calculations we can extend this result to nonzero magnetic fields and conclude that as soon as an anisotropy is present ($\gamma\neq 0$), $\chi(0)$ is finite and if $\gamma=0$, $\chi(0)$ is always zero. This can be understood as follows. When $\gamma=0$ and H increases, the single band of dimerized quasifermions is split into two bands, one going upward and the other downward, but each band moves by the same amount of energy. In this case, the magnetic susceptibility is zero because the rate of change of the total energy of the system is zero. In the case when $\gamma\neq 0$, the movement of the bands is not the same for all k's. This produces a rate of change of G which is not zero.

In Fig. 4 appears the graph of the magnetization ($MJ/N\mu^2H$) of the system divided by H as a function of temperature for $h=0.1$, $C/J=0.5$, and $\gamma=0$, 0.1, 0.125, 0.130, 0.135, and 0.139. Those results are essentially the same as those of Tannous and Caillé[7] for the magnetic susceptibility in the isotropic case ($\gamma=0$) (they used the relation $\chi=M/H$ which is valid for $H\to 0$). A discontinuity in the derivative of M is present at T_c for the second-order-transition case. A discontinuity in M is present for the first-order case. The value of M at 0 K is finite for $\gamma\neq 0$ but is zero for the isotropic case, the explanation being the same as that of the preceding paragraph concerning the magnetic susceptibility. At high temperature, the behavior of M is that of a uniform chain. At very low temperature, when the dimerization parameter, δ, is at its saturation value, the behavior is that of a dimerized chain. The decreasing rate of M is then exponential [$M\propto\exp(-D/T)$, D being a constant]. When $H=0$, $M=0$: No magnetic moment is present because of the antiferromagnetic exchange interaction.

In Fig. 5, the constant-magnetic-field specific heat (C_H/Nk_B) is plotted as a function of T for $h=0.1$, $C/J=0.5$, and $\gamma=0$, 0.125, 0.13, and 0.139. These curves have been obtained from Eqs. (9) and (10) but a verification has been made with the definition of C_H as the second derivative of Gibbs free energy with respect to temperature. For each curve, a discontinuity is observed at T_c, the discontinuity having different values or sign, depending on γ. For a second-order phase transition, this discontinuity is predicted to occur in mean-field theory.[8] It is predicted to be infinite at the tricritical point. For a first-order transition, the curves of Fig. 5 have, in addition to what is drawn, a delta-function-type singularity at T_c. This singularity gives rise to a nonzero latent heat. However, it is well known that the critical fluctuations are particularly important for the calculation of the specific heat. The analytical behavior near T_c must not be taken too seriously. It is expected that fluctuations will smooth out the second-order jump and that a peak can appear in C_H near T_c to account for the latent heat in the first-order case.

V. CONCLUSIONS

The spin-Peierls phase transition of an anisotropic XY chain in a magnetic field has been investigated. Concerning the transition itself, it has been found that when the degree of anisotropy (γ) or the magnetic field (H) increases, the phase transition passes

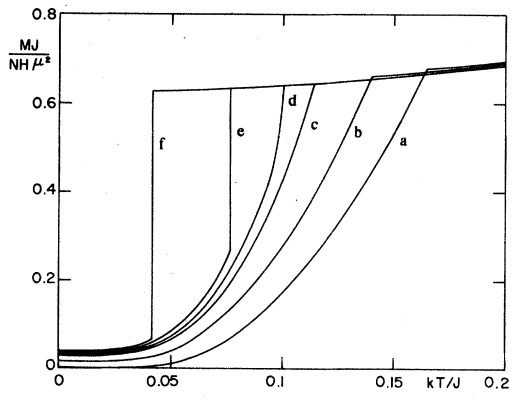

FIG. 4. Magnetization divided by H vs kT/J for $C/J=0.5$ and $h=0.1$. a: $\gamma=0$, b: $\gamma=0.1$, c: $\gamma=0.125$, d: $\gamma=0.13$, e: $\gamma=0.135$, and f: $\gamma=0.139$.

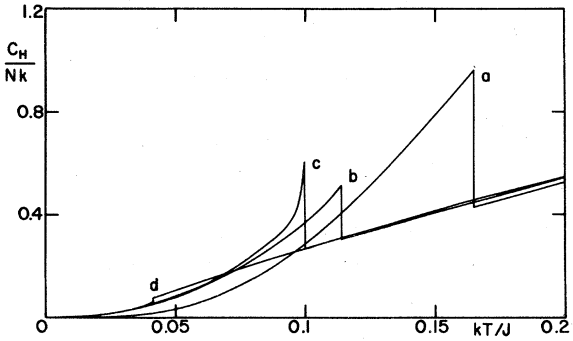

FIG. 5. Specific heat vs kT/J for $C/J=0.5$ and $h=0.1$. a: $\gamma=0$, b: $\gamma=0.125$, c: $\gamma=0.13$, and d: $\gamma=0.139$.

from a second-order one to the first-order one [at a tricritical point $\gamma_t(H,C)$] and finally that the ordered phase disappears, even at $T=0$ K. During this process, the critical temperature (T_c) decreases continuously. This effect has been found to be enhanced when both γ and H are different from zero. I have also calculated the magnetization, the magnetic susceptibility, and the specific heat for the transition. For the magnetization, I have obtained results similar to those of Tannous and Caillé[7]: a quasilinear magnetization at high temperature and an exponential decay to zero if $\gamma=0$ or to a constant for $\gamma\neq 0$ at low temperature. The curve is continuous for a second-order transition and a jump exists at T_c for the first-order one. The behavior of the magnetic susceptibility at finite field is quite different because of the variation of δ with H. At low and high temperature, the behavior is very similar to that of the magnetization. Near T_c, however, a jump is always present, even for second-order transitions. The size of the jump is largest at the tricritical point where it becomes infinite. It has also been found that the specific heat at constant magnetic field falls exponentially to zero at low temperature. It has a linear behavior in the high-temperature phase. The behavior near T_c is governed mostly by fluctuations and the mean-field results are not reliable in this neighborhood, particularly for the first-order transition where a peak should be seen near T_c. The physical meaning of this lowering of T_c with γ or H has been described previously.[6,7] It can be summarized as follows: Finite values of γ or H limit the freedom of the electronic spin in the xy plane. The spins lose their ability to orient themselves and the energy lowering due to dimerization decreases when γ or H increases. Finally, for large γ or H, not enough energy for dimerization can be gained from the spins and the ordered phase disappears. The joint effect of γ and H is additive.

From a microscopic point of view, the elementary excitations of the magnetic system, named quasifermions, are responsible for this Peierls distortion. In the absence of magnetic fields, a gap is present in these excitations, always at the Brillouin zone (this gap disappears only at $\gamma=\delta$). In the absence of anisotropy, this gap, if present, is at the Brillouin-zone boundary. If $\mu H > J_k$ at the zone boundary, the gap is absent and the bands touch somewhere inside the Brillouin zone. If both γ and H are finite, the gap, if present, can be anywhere inside the Brillouin zone. However, this vanishing of the energy gap does not seem to have a significant effect on the thermodynamic response functions. In actual cases, it occurs at temperatures just below T_c where δ varies very rapidly, with the temperature. The variation of the thermodynamic functions is then very pronounced and highly dependent on δ. The effect of the disappearance of the gap is hidden in this variation. This should be more easily seen at very low temperature when δ has saturated. Unfortunately, at low temperature the value of δ is much higher than the value for the disappearance of the gap. If one then tries to increase γ, the ordered phase will disappear before the gap vanishes. It seems that at low temperature, the vanishing of the gap is incompatible with the stability of the ordered phase. Perhaps an external stress, allowing the disappearance of the gap at low temperature, can make this effect visible in the susceptibility or specific heat.

ACKNOWLEDGMENTS

This research was partially supported by the Natural Sciences and Engineering Research Council of Canada and le Ministère de l'Education du Québec. I would also like to thank D. Forgues for help in the numerical calculations.

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