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We discuss, within a framework in which the structural degrees of freedom (assumed essentially three-dimensional) are adiabatically treated and the magnetic degrees of freedom are exactly treated, the stress-(magnetic)field-temperature phase diagram associated with the one-dimensional first-neighbour spin-$\frac{1}{2}$ magnetostrictive XY model. The important influence of the curvature of the coupling constant as a function of distance is exhibited and satisfactorily compared to experimental data.

The spin-Peierls phase transition is a magnetically-driven structural one which occurs in three-dimensional systems presenting quasi-one-dimensional magnetic interactions. On theoretical grounds the interactions that are commonly assumed are the Heisenberg and XY ones (Refs. [1-6]; for an excelent theoretical as well as experimental review see Ref. 7); also anisotropic interactions have been occasionally introduced[8]. Real substances presenting spin-Peierls instabilities include TTF-BDT, TTF-BDS, MEM $(TCNQ)_2$ and perhaps alkali-TCNQ. This type of systems typically present, in the H (external magnetic field along the Z axis)-T(temperature) space, a phase diagram which can be interpreted[9] as follows: a essentially high temperature region (where the magnetic chain is *uniform*; U phase) separated, through a second order phase transition, from the *ordered* region. The latter is further divided into two subregions, namely the low H one (corresponding to a *dimerized* (D) chain) and the high H one (corresponding to a complex *modulated* (M) chain), separated by a first-order critical line. All three U, D and M phases join at a structural Lifshitz point (for a typical theoretical phase diagram see the ($\nu=1$; $\sigma=0.2781$) curve of Fig.2).

The interesting influence (experimentally exhibited recently on TTF-Cu BDT[10]), on spin-Peierls systems, of pressure or of uniaxial external stress (along the Z axis) has already been predicted for the XY model[6] as well as for the Heisenberg one[11]. In both cases only the H=0 situation has been (preliminary) analyzed. In the present paper we discuss, with some detail and for all values of H, the influence of uniaxial stress τ (along the Z axis) on the T-H phase diagram associated with the one-dimensional first-neighbour spin-$\frac{1}{2}$ magnetostrictive XY model. The role played by the functional form of the coupling constant J with distance is analyzed as well by introducing a form which contains both linear and exponential dependences as particular cases; the drastic influence of J" is exhibited. Finally the variation, along the critical lines, of the crystalline parameter is presented as well.

We consider a cyclic chain of 2N spins; the magnetic contribution to its Hamiltonian is assumed to be

$$\mathcal{H}_m = -\frac{1}{2}\sum_{j=1}^{2N} J_j (S_j^x S_{j+1}^x + S_j^y S_{j+1}^y) - \mu H \sum_{j=1}^{2N} S_j^z \qquad (1)$$

where μ is the elementary magneton. By introducing, through the standard Jordan-Wigner transformation (see for example Ref. [9]), fermionic creation and annihilation operators we can rewrite this Hamiltonian as follows:

$$\mathcal{H}_m = -\frac{1}{2}\sum_{j=1}^{2N} J_j(a_j^+ a_{j+1} + a_{j+1}^+ a_j) - \mu H \sum_{j=1}^{2N} a_j^+ a_j \qquad (2)$$

where an additive constant has been ommited. Through the Fourier transformation characterized by

$$b_k = \frac{1}{\sqrt{2N}} \sum_{j=1}^{2N} e^{iajk}\, a_j \qquad (3)$$

and

$$J_q = \frac{1}{2N} \sum_{j=1}^{2N} e^{iajq}\, J_j \qquad (-\pi/a<k,q\leq\pi/a) \qquad (4)$$

($a \equiv 1+d$ being the reduced crystalline parameter and d a convenient variable) we can rewrite Hamiltonian (2) as follows:

$$\mathcal{H}_m = \mathcal{H}_o + V \qquad (5)$$

where

$$\mathcal{H}_o \equiv |J(0)| \sum_k \varepsilon_k\, b_k^+\, b_k \qquad (6)$$

and

$$V \equiv |J(0)| \sum_{\substack{k \\ q\neq 0}} \Lambda_{kq}\, b_k^+\, b_{k-q} \qquad (7)$$

with

$$\varepsilon_k \equiv h - \left|\frac{J(d)}{J(0)}\right| \cos ak \qquad (8)$$

$$h \equiv \mu H/|J(0)| \qquad (9)$$

$$\Lambda_{kq} \equiv \frac{1}{2}\, \frac{J_q}{J(0)}\, [e^{-ia(k-q)} + e^{iak}] \qquad (10)$$

$J(d)$ being the coupling constant between first-neighbouring spins (separated by a reduced distance $1+d$). Note that $\mathcal{H}_o$ is the

Hamiltonian of the U phase.

By treating now V as a perturbation (up to second order) to $\mathcal{H}_o$ within the temperature-dependent Green function framework (see detailed procedure in Ref. [9]) we obtain the reduced free energy

$$f_m \equiv F_m/N|J(0)| = f_o + f_2 \qquad (11)$$

where

$$f_o \equiv -\frac{2ta}{\pi}\int_0^{\pi/a} dk\ \ln(2\cosh\frac{\varepsilon_k}{2t}) \qquad (12)$$

(U phase reduced free energy) and

$$f_2 \equiv -\sum_{q\neq 0}\left|\frac{J_q}{J(0)}\right|^2 \frac{a}{4}\int_0^{\pi/a} dk\ \cos^2 ak\ \frac{\tanh\frac{\varepsilon_{k+q/2}}{2t} - \tanh\frac{\varepsilon_{k-q/2}}{2t}}{\varepsilon_{k+q/2} - \varepsilon_{k-q/2}} \qquad (13)$$

with

$$t \equiv k_B T/|J(0)| \qquad (14)$$

and where the quasi-continuum limit has been used.

Let us next include, within both adiabatic and harmonic approximations, the elastic contribution F_e to the free energy of the system:

$$F_e = \frac{C}{2}\sum_{j=1}^{2N}(X_{j+1} - X_j - 1)^2 \qquad (15)$$

where C is the harmonic elastic constant and X_j is the reduced mean position of the j-th spin with respect to one of them chosen as origin. Through the relation

$$X_j = aj + u_j$$

we introduce the convenient variable u_j and its Fourier transformed variable u_q. Then it is straightforward to verify that expression (15) can be rewritten as follows:

$$F_e = NC\,d^2 + 2NC\sum_q (1-\cos aq)\,|u_q|^2 \qquad (16)$$

On the other hand the exchange integral $J_j = J(X_{j+1} - X_j - 1)$ can be linearly expanded as follows:

$$J_j = J(d) + J'(d)(u_{j+1} - u_j) \quad (17)$$

Replacing this expression into Eq.(4) and the result into Eq.(13) we finally obtain the total free energy $F = F_m + F_e$ of the system:

$$f \equiv \frac{F}{N|J(0)|} = f_o + \frac{1}{2} \sum_{q\neq 0} \omega_q^2 \eta_q^2 + KD^2 \quad (18)$$

where

$$\omega_q^2 \equiv (1-\cos q)(K-L_q) \quad (19)$$

$$K \equiv C|J(0)|/|J'(0)|^2 \quad (20)$$

$$D \equiv \frac{J'(0)}{|J(0)|} d \quad (21)$$

$$L_q \equiv \frac{1}{4\pi \sin(q/2)} \frac{[j'(D)]^2}{[j'(0)]^2 j(D)} \times$$

$$\int_0^\pi dk \frac{\cos^2 k}{\sin k} [\tanh \frac{h-j(D)\cos(k+q/2)}{2t} - \tanh \frac{h-j(D)\cos(k-q/2)}{2t}] \quad (22)$$

$$\eta_q \equiv 2|J'(0)u_q/J(0)| \quad (23)$$

$$j(D) \equiv J(|J(0)|D/J'(0))/|J(0)| \quad (24)$$

and where we have used the transformations $aq \to q$ and $ak \to k$.

At this stage we can finally assume that the system is in the presence of an external stress τ; therefore the Gibbs energy G associated with the U phase is given by

$$g \equiv \frac{G}{N|J(0)|} = f_o + KD^2 - 2\sigma D \quad (25)$$

where $\sigma \equiv \tau / J'(0)$. At thermodynamical equilibrium, $\left.\frac{\partial g}{\partial D}\right|_{t,h,\sigma} = 0$; consequently the use of Eqs. (12) and (25) leads to the equation of states

$$\sigma = \frac{1}{2\pi}\frac{dj}{dD}\int_0^{\pi/2} dk\left[\tanh\frac{h-j(D)\cos k}{2t} - \tanh\frac{h+j(D)\cos k}{2t}\right] + KD \tag{26}$$

This equation implicitely determines $D(t,h,\sigma)$. The critical surface separating, in the (t,h,σ) space the U phase from the polymerized ones (D and M phases) is determined (see Ref.[9]) by the soft mode condition $\omega_{q_c}(t,h,D(t,h,\sigma))=0$ hence (by using Eq.(19)) $L_{q_c}(t,h,D(t,h,\sigma)) = K$ where q_c is the wave vector of the structural mode which freezes at the phase transition (ω_q is minimal at $q=q_c$). Let us stress that the results we are looking for (critical surface and $D(t,h,\sigma)$ in the U phase) are, except for the adiabatic assumption, *exact* in spite of the perturbative procedure we have followed (see Eqs.(5) and (18)); this procedure is in fact nothing but an operational conveniency within a quite complex set of equations.

In order to exhibit concrete results we have assumed the following family of functional forms:

$$j(D) = -[(1-\nu)(1-D) + \nu e^{-D}] \quad (0 \le \nu \le 1) \tag{27}$$

which in the limits $\nu = 0$ and $\nu = 1$ reproduces respectively the standard linear and exponential antiferromagnetic proposals (notice also that, in the limit $D \to 0$, $j \sim -1+D-\frac{\nu}{2}D^2$, hence $\nu = -j''(0)$).

Typical results for $H = 0$ are presented in Figs. 1 and 2. The confluence points of Figs. 1(a) and 1(b) correspond to D=0 which is the case (fixed strain instead of the present cases which are fixed stress ones) we have discussed in Ref.[9]; we observe now the drastic effect of ν. To be more precise let us consider the typical situation $J(0) < 0$ and $J'(0) > 0$: if $\nu = 1$ (exponential law) the critical temperature *decreases* with stress (or equivalently *increases* under pressure-like action), whereas if $\nu=0$ (linear law) the critical temperature paradoxally *increases* with stress (or equivalently *decreases* under pressure-like action). *This paradoxal behaviour is precisely the one observed experimentally*[10] and had been previously predicted[6]. It can be un derstood if one takes into account the fact that the critical temperature is determined by Eq.(22) where we remark the presence of the factor $[j'(D)]^2/j(D)$ whose slope (as a function of D) is very sensitive to ν.

In Fig.2 we exhibit typical T-H phase diagrams; they are in fact quite similar in shape to those obtained[9] for D=0, presenting in particular Lifshitz points (full dots; *inflexion points*) as well as "starting points" (open dots). The $t \to 0$ asymptotic behaviour of the critical lines is given by

$$h \sim |j(D)| - \frac{t}{2} \ln t \qquad (28)$$

where

$$|j(D)| = (1-\nu)(1+\frac{\sigma}{K}) + \nu e^{\sigma/K} \qquad (29)$$

For $\nu=1$ (exponential law) the critical lines associated with different values of σ *do not cross each other*; *this is not true* for $\nu=0$ (linear law) as a consequence of the fact that *increasing* σ leads, for let us say $J(0) < 0$ and $J'(0) > 0$, to an *increasing* H=0 critical temperature but to a *decreasing* T=0 critical field. Also the "re-entrances" of the critical lines are much more pronounced for $\nu=1$ than for $\nu=0$.

It should be very interesting to experimentally exhibit all these effects. In particular TTF - Cu BDT presents[10] a 1.1K *decrease* in the H=0 critical temperature if a 6Kbar pressure is applied; this fact characterizes a nearly linear J(d) law ($\nu \cong 0$) and therefore critical T-H lines associated with different values of pressure-like action are in principle expected to cross each other.

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CAPTION FOR FIGURES

FIG. 1 - Results for vanishing magnetic field, a typical value for the reduced elastic constant $K = 0.4$, and several values of ν ($\nu = 0$ and $\nu = 1$ respectively correspond to linear and exponential dependences of the coupling constant with distance): (a) uniform-(non uniform) critical lines in the reduced uniaxial stress (σ)-temperature (t) space ($-\sigma$ is a compressing-like stress for the standard antiferromagnetic case $J(0) < 0$ and $J'(0) > 0$) and (b) the corresponding reduced uniaxial strains D.

FIG. 2 - Typical phase diagrams in the reduced magnetic field (h) - temperature (t) space for a reduced elastic constant $K = 0.4$ and both linear ($\nu = 0$) and exponential ($\nu = 1$) dependences of the coupling constant with distance. All uniform (U) - (non-uniform) critical lines are second order ones; within the non uniform region, the critical lines separating the dimerized (D) and modulated (M) phases are first order ones (herein indicated only for the case ($\nu = 1$, $\sigma = 0.2781$); dotted line). $\sigma = 0.2781$ corresponds to the confluence point of Fig. 1. The full (open) circles correspond to the Lifshitz points ("starting points"); see details in Ref. [9].

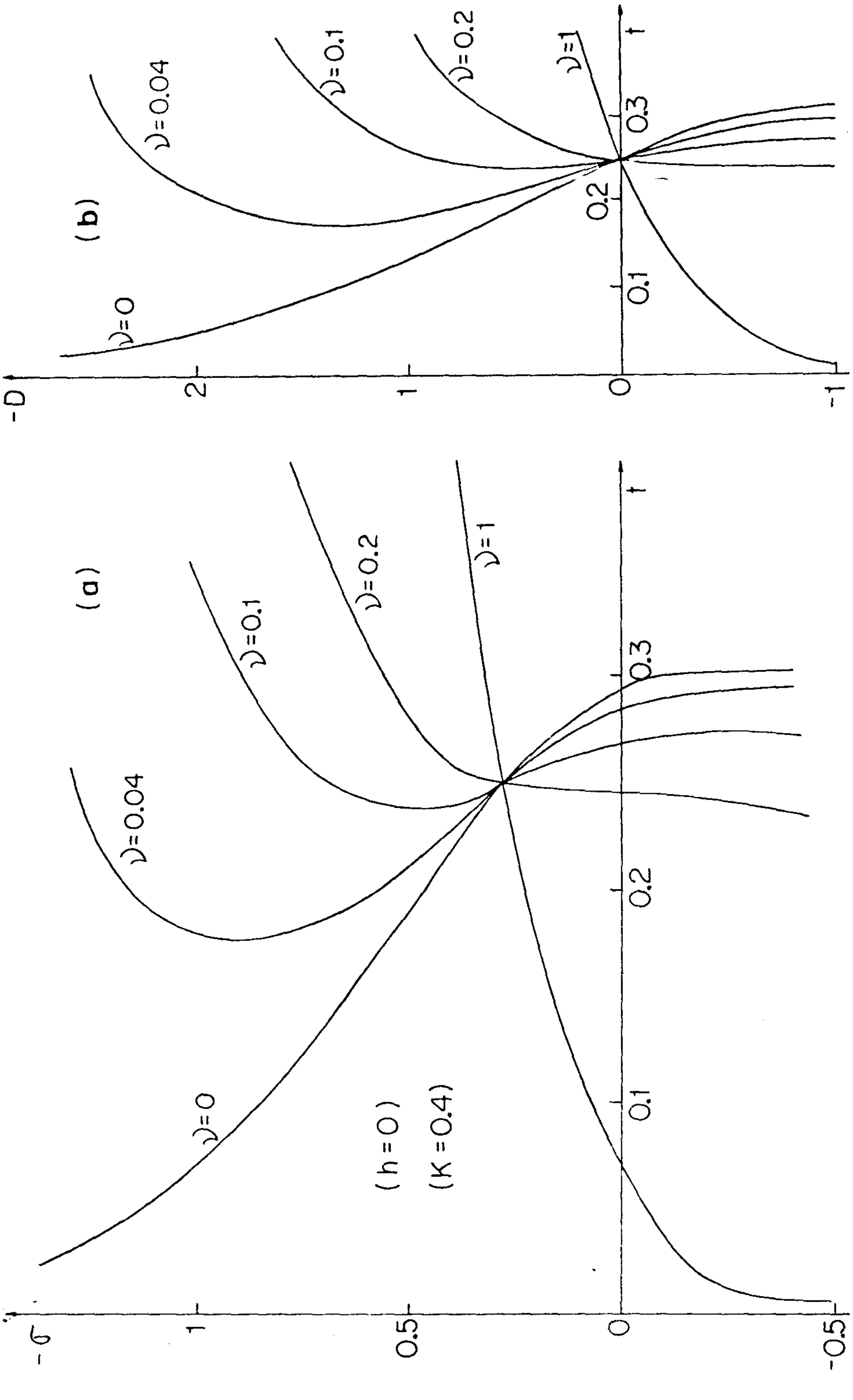
(a)
-σ
1
0.5
0
-0.5
0.1
0.2
0.3
t
ν=0
ν=0.04
ν=0.1
ν=0.2
ν=1
(h=0)
(K=0.4)
(b)
-D
2
1
0
-1
0.1
0.2
0.3
t
ν=0
ν=0.04
ν=0.1
ν=0.2
ν=1

FIG.1

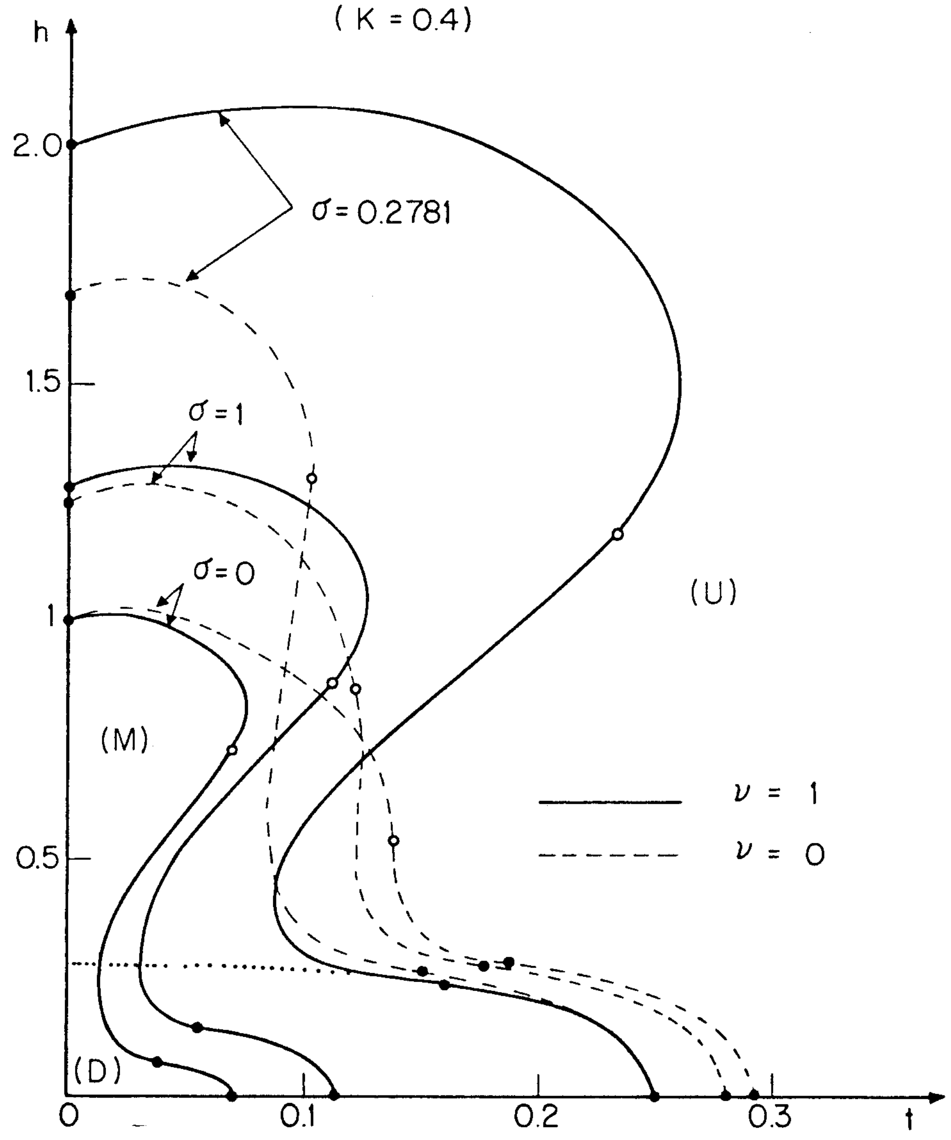

FIG.2