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► To cite this version:

C. Tsallis. Effects of external stresses on the metamagnetic transition of a highly anisotropic antiferromagnet. *Journal de Physique*, 1971, 32 (11-12), pp.903-911. 10.1051/jphys:019710032011-12090300 . jpa-00207190

HAL Id: jpa-00207190

<https://hal.science/jpa-00207190>

Submitted on 4 Feb 2008

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Classification
Physics Abstracts
17.60

EFFECTS OF EXTERNAL STRESSES ON THE METAMAGNETIC TRANSITION OF A HIGHLY ANISOTROPIC ANTIFERROMAGNET

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(Reçu le 21 janvier 1971, révisé le 15 mai 1971)

Résumé. — Dans le cadre d'une théorie de type Landau, on étudie les propriétés de la transition métamagnétique d'un cristal antiferromagnétique très anisotrope sous l'influence de contraintes extérieures. On donne l'expression de nombreuses grandeurs physiques intéressantes. Cette théorie est illustrée à l'aide d'un modèle d'Ising afin d'obtenir des résultats plus spécifiques. Certaines caractéristiques théoriques nouvelles sont obtenues et une comparaison avec la situation expérimentale est donnée.

Abstract. — Within the framework of a Landau-type theory we study the properties of the metamagnetic transition of a highly anisotropic antiferromagnet under external stresses. We give the expressions of a number of physical quantities of interest. This theory is illustrated with an Ising model in order to obtain more specific results. Some new theoretical features are obtained and a comparison with the experimental situation is given.

I. Introduction. — A number of materials exhibit first order magnetic transitions. Such a behavior is not expected to occur within the framework of the classical Weiss molecular field theory. However, Bean and Rodbell [1], assuming that the exchange energy is a function of interatomic spacing, were able to give a simple treatment of the first order ferromagnetic phase transition of MnAs. As a consequence of the magnetoelastic coupling the magnetic properties of MnAs are pressure dependent. In particular the order of the transition can be changed and in fact Bean and Rodbell predicted that the transition becomes of the second-order at high pressure.

The modification of the order of a transition under the influence of an external action is rather frequent. Metamagnetism is a well-known example. Field-induced transition from an antiferromagnetic state to a nearly saturated paramagnetic state are usually first order at very low temperature and second order at higher temperature. Here again magnetoelastic effects can be responsible for this behavior and it is of interest to study the properties of an antiferromagnet placed in a magnetic field and under external stresses. Various authors have investigated certain aspects of this problem ([2] to [17]).

In order to concentrate on the essential features of the problem and, at the same time, to simplify the calculations, we shall suppose the antiferromagnet to be highly anisotropic and consequently we shall not take into consideration a possible spin-flop phase.

We shall first give a Landau-type phenomenological theory [18] of this system and then illustrate more explicitly the various results on an Ising model with strain-dependent exchange integrals.

II. Thermodynamics. — **II.1 THERMODYNAMICAL THEORY.** — Let us consider a periodical tridimensional (*) array of spins $1/2$, and let us assume that their location enables us to distinguish two families, each one having the same number of spins. We shall also assume that each spin may be parallel or anti-parallel to a unique easy axis. If we want to study the behavior, as function of temperature T , of such a system under external stress t (the problem is easily generalized to more than one non vanishing stress-tensor component) and magnetic field H , we must consider the Gibbs thermodynamical potential (per site)

$$g = f(T, m_1, m_2, e) - H \frac{m_1 + m_2}{2} - te$$

where m_1, m_2 (reduced magnetic moment per site of each family respectively : $-1 \leq m_1, m_2 \leq +1$) and e (lattice-strain-tensor component associated to t) are the variational parameters. To precise the ideas let us have in mind a particular model (which will be treated later) corresponding to :

(*) Actually dimensionality does not appear explicitly in the model considered in this paper.

$$\begin{aligned}
 f = & -\frac{J_1(e)}{2} m_1 m_2 - \frac{J_2(e)}{4} (m_1^2 + m_2^2) + V(e) + \\
 & + \frac{T}{4} [(1 + m_1) \text{Log}(1 + m_1) \\
 & + (1 - m_1) \text{Log}(1 - m_1) \\
 & + (1 + m_2) \text{Log}(1 + m_2) \\
 & + (1 - m_2) \text{Log}(1 - m_2) - 4 \text{Log} 2] \quad (1)
 \end{aligned}$$

where the Boltzmann constant K_B has been chosen equal to unity, $J_2(e)$ and $J_1(e)$ are the exchange integrals between sites of the same and different family respectively, $V(e)$ is the crystalline potential energy supposed the same for all sites, and the entropy term is given by the Bragg-Williams approximation. Let us remark that such a model does not account for thermal expansion for $\partial^2 f / \partial e \partial T = 0$. We may define in general $m = \frac{1}{2}(m_1 + m_2)$ and $\eta = \frac{1}{2}(m_1 - m_2)$ where $2m$ is now the total magnetic moment per unit cell and η the antiferromagnetic order parameter. Hence we have :

$$g = f(T, \eta, m, e) - Hm - te,$$

where f is an even function of η and m . Minimizing g with respect to the variational parameters we obtain

$$\left. \frac{\partial f}{\partial \eta} \right|_{m,e} = 0 \quad \left. \frac{\partial f}{\partial m} \right|_{\eta,e} = H \quad \left. \frac{\partial f}{\partial e} \right|_{\eta,m} = t. \quad (2)$$

Solving this system we obtain

$$\eta = \eta(T, H, t), \quad m = m(T, H, t) \quad \text{and} \quad e = e(T, H, t).$$

We shall assume from now on that $m = 0$ for $H = 0$, this is to say no spontaneous ferromagnetic

order. In practice it is frequently impossible to solve exactly the system (2). That is why we shall limit ourselves to study the behavior of the system near the second order transition surface. In the disordered phase, system (2) reduces to

$$\left. \frac{\partial f}{\partial m} \right|_{\eta=0} = H \quad \text{and} \quad \left. \frac{\partial f}{\partial e} \right|_{m=0} = t;$$

for the first equation of (2) is identically satisfied. Let us then suppose a second order transition and expand the first equation of (2) near the transition surface (which is noted with the sub-index 0) :

$$\begin{aligned}
 \left. \frac{\partial^2 f}{\partial \eta^2} \right|_0 \eta + \left. \frac{\partial^3 f}{\partial \eta^2 \partial T} \right|_0 \eta(T - T_0) + \\
 + \left. \frac{\partial^3 f}{\partial \eta^2 \partial m} \right|_0 \eta(m - m_0) + \left. \frac{\partial^3 f}{\partial \eta^2 \partial e} \right|_0 \eta(e - e_0) + \\
 + \frac{1}{3!} \left. \frac{\partial^4 f}{\partial \eta^4} \right|_0 \eta^3 = 0.
 \end{aligned}$$

This equation has two solutions :

$$\begin{aligned}
 \eta = 0 & \quad \text{paramagnetic phase (P)} \\
 \eta \neq 0 & \quad \text{antiferromagnetic phase (AF)}.
 \end{aligned}$$

The transition surface is determined by

$$\left. \frac{\partial^2 f}{\partial \eta^2} \right|_0 = 0$$

in the case of a second order transition, and by $g(T, \eta, m, e) = g(T, 0, m, e)$ in the case of a first order transition.

If we expand also the second and third equations of (2) we obtain :

$$\begin{pmatrix} \frac{1}{3!} \left. \frac{\partial^4 f}{\partial \eta^4} \right|_0 & \left. \frac{\partial^3 f}{\partial \eta^2 \partial m} \right|_0 & \left. \frac{\partial^3 f}{\partial \eta^2 \partial e} \right|_0 \end{pmatrix} \begin{pmatrix} \eta^2 \\ m - m_0 \\ e - e_0 \end{pmatrix} = \begin{pmatrix} - \left. \frac{\partial^3 f}{\partial \eta^2 \partial T} \right|_0 (T - T_0) \\ - \left. \frac{\partial^2 f}{\partial m \partial T} \right|_0 (T - T_0) + (H - H_0) \\ - \left. \frac{\partial^2 f}{\partial e \partial T} \right|_0 (T - T_0) + (t - t_0) \end{pmatrix} \quad (3)$$

$$\begin{pmatrix} \frac{1}{2!} \left. \frac{\partial^3 f}{\partial \eta^2 \partial m} \right|_0 & \left. \frac{\partial^2 f}{\partial m^2} \right|_0 & \left. \frac{\partial^2 f}{\partial m \partial e} \right|_0 \\ \frac{1}{2!} \left. \frac{\partial^3 f}{\partial \eta^2 \partial e} \right|_0 & \left. \frac{\partial^2 f}{\partial m \partial e} \right|_0 & \left. \frac{\partial^2 f}{\partial e^2} \right|_0 \end{pmatrix} \begin{pmatrix} m - m_0 \\ e - e_0 \end{pmatrix} = \begin{pmatrix} - \left. \frac{\partial^2 f}{\partial m \partial T} \right|_0 (T - T_0) + (H - H_0) \\ - \left. \frac{\partial^2 f}{\partial e \partial T} \right|_0 (T - T_0) + (t - t_0) \end{pmatrix} \quad (3')$$

The system (3) gives the behavior in the (AF) phase ; and the system (3') gives the behavior in the (P) phase.

Solving for η^2 we obtain :

$$\eta^2 = \frac{N_T}{D} (T - T_0) + \frac{N_H}{D} (H - H_0) + \frac{N_t}{D} (t - t_0), \quad (4)$$

where D is the determinant of the left side matrix

of (3) and N_T, N_H and N_t are immediately obtained as functions of the derivatives of f . Obviously

$$N_T(T - T_0) + N_H(H - H_0) + N_t(t - t_0) = 0$$

determines the transition surface, whose slopes are given by :

$$\left. \frac{\partial H_0}{\partial T_0} \right|_{t_0} = - \frac{N_T}{N_H} \quad \text{and} \quad \left. \frac{\partial t_0}{\partial T_0} \right|_{H_0} = - \frac{N_T}{N_t}.$$

Therefore the zeros of N_T , N_H , N_t determine the singularities of the slopes. In particular N_H vanishes for $H_0 = 0$ and in general $(\partial H_0 / \partial T_0)|_{t_0}$ diverges.

D may vanish and in this case $D = 0$ determines the critical line which separates the transition surface in two or more regions : $D > 0$ (< 0) means second order (first order) transition. Clearly system (3) is not valid in the regions where $D < 0$. In the regions where $D > 0$ we obtain from (3), that

$$N_T(T - T_0) > 0, \quad N_H(H - H_0) > 0 \quad \text{and} \quad N_t(t - t_0) > 0$$

and hence the signs of N_T , N_H or N_t determine the side of the transition surface where the ordered phase appears. For example $N_T < 0$ (> 0) means that the ordered phase is on the low (high) temperature-side of the transition surface. Clearly the limiting situation $N_T = 0$, in general, means also that

$$\left. \frac{\partial H_0}{\partial T_0} \right|_{t_0} = \left. \frac{\partial t_0}{\partial T_0} \right|_{H_0} = 0.$$

If no magnetic field is applied and no magnetoelastic interaction is allowed then D reduces to :

$$\frac{1}{3!} \left. \frac{\partial^4 f}{\partial \eta^4} \right|_0 \left. \frac{\partial^2 f}{\partial m^2} \right|_0 \left. \frac{\partial^2 f}{\partial e^2} \right|_0,$$

and each factor being necessarily positive (because of the stability of the physical system) the transition is of the second order. If this is not the case, D will also include terms necessarily negative and first order transitions will then be possible.

II.2 THERMODYNAMICAL COEFFICIENTS. — Let us first introduce the entropy s per site of the system. Making use of $s = -\partial f / \partial T$ and expanding near the transition surface we obtain

$$-(s - s_0) = \left. \frac{\partial^2 f}{\partial T \partial m} \right|_0 (m - m_0) + \left. \frac{\partial^2 f}{\partial T \partial e} \right|_0 (e - e_0) + \frac{1}{2!} \left. \frac{\partial^3 f}{\partial T \partial \eta^2} \right|_0 \eta^2. \quad (5)$$

This expression is valid in both (P) and (AF) phases, where we must obviously use the corresponding

$(m - m_0)$, $(e - e_0)$ and η^2 . These three increments are related, in the case of an adiabatic process, by (5) where we impose $s - s_0 = 0$.

From system (3) all linear thermodynamical coefficients may be calculated, in both (P) and (AF) phases. The most usual are :

elastic coefficient	$\kappa = \frac{\partial e}{\partial t}$
magnetic susceptibility	$\chi = \frac{\partial m}{\partial H}$
heat capacity	$C = T \frac{\partial s}{\partial T}$
direct piezomagnetic coefficient	$d = \frac{\partial m}{\partial t}$
inverse piezomagnetic coefficient	$d^* = \frac{\partial e}{\partial H}$
expansion coefficient	$\alpha = \frac{\partial e}{\partial T}$
piezocaloric coefficient	$\alpha^* = \frac{\partial s}{\partial t}$
pyromagnetic coefficient	$q = \frac{\partial m}{\partial T}$
magnetocaloric coefficient	$q^* = \frac{\partial s}{\partial H}$

Simple thermodynamical considerations lead to $d = d^*$, $\alpha = \alpha^*$ and $q = q^*$.

Each coefficient of the set may be evaluated on any path in the (T, H, t) space. To define such a path two conditions are required, and usually two of the following six are adopted : isothermal ($T = T_0$), adiabatic ($s = s_0$), free crystal ($t = t_0$), clamped crystal ($e = e_0$), constant field ($H = H_0$), constant magnetization ($m = m_0$).

If we calculate, for example, the experimentally usual magnetic susceptibilities, in both (P) and (AF) phases, we obtain :

$$(\chi_{T,t}^P)^{-1} = \left\{ \frac{\partial^2 f}{\partial m^2} - \frac{\left(\frac{\partial^2 f}{\partial m \partial e} \right)^2}{\frac{\partial^2 f}{\partial e^2}} \right\}_0$$

$$(\chi_{T,t}^{AF})^{-1} = \left\{ \frac{\partial^2 f}{\partial m^2} - \frac{\left(\frac{\partial^2 f}{\partial m \partial e} \right)^2}{\frac{\partial^2 f}{\partial e^2}} - \frac{1 + \frac{\frac{1}{2!} \frac{\partial^2 f}{\partial e^2} \left(\frac{\partial^3 f}{\partial \eta^2 \partial m} \right)^2 - \frac{\partial^3 f}{\partial \eta^2 \partial m} \frac{\partial^3 f}{\partial \eta^2 \partial e} \frac{\partial^2 f}{\partial m \partial e}}{\frac{1}{3!} \frac{\partial^4 f}{\partial \eta^4} \left(\frac{\partial^2 f}{\partial m \partial e} \right)^2}} \right\}_0$$

For very weak fields, these reduce to

$$(\chi_{T,t}^P)^{-1} = (\chi_{T,t}^{AF})^{-1} = \frac{\partial^2 f}{\partial m^2} \Big|_0 > 0. \quad (6)$$

Quadratic thermodynamical coefficients may be calculated in a similar way. For example let us evaluate the magnetostrictive coefficient defined as

$$\lambda = \frac{1}{2} \frac{\partial^2 e}{\partial H^2}$$

for a vanishing magnetic field and at constant temperature and stress.

System (3) is no more sufficient because all odd derivatives of f with respect to m vanish, so we must expand (2) to the second order in m . We obtain

$$\begin{aligned} \frac{1}{3!} \frac{\partial^4 f}{\partial \eta^4} \Big|_0 \eta^2 + \frac{1}{2!} \frac{\partial^4 f}{\partial \eta^2 \partial m^2} \Big|_0 m^2 + \frac{\partial^3 f}{\partial \eta^2 \partial e} \Big|_0 (e - e_0) + \frac{\partial^3 f}{\partial \eta^2 \partial T} \Big|_0 (T - T_0) &= 0 \\ \frac{1}{2!} \frac{\partial^4 f}{\partial \eta^2 \partial m^2} \Big|_0 \eta^2 + \frac{\partial^2 f}{\partial m^2} \Big|_0 m + \frac{\partial^3 f}{\partial m^2 \partial e} \Big|_0 m(e - e_0) + \frac{\partial^3 f}{\partial m^2 \partial T} \Big|_0 m(T - T_0) &= H \\ \frac{1}{2!} \frac{\partial^3 f}{\partial e \partial \eta^2} \Big|_0 \eta^2 + \frac{1}{2!} \frac{\partial^3 f}{\partial e \partial m^2} \Big|_0 m^2 + \frac{\partial^2 f}{\partial e^2} \Big|_0 (e - e_0) + \frac{\partial^2 f}{\partial e \partial T} \Big|_0 (T - T_0) &= t - t_0. \end{aligned}$$

Solving this system we obtain (for $t = t_0$)

$$\lambda_{T,t_0}^\varphi = \lambda_{T_0,t_0}^\varphi \left\{ 1 - 2 \frac{\frac{\partial}{\partial T} (\chi_{T,t_0}^\varphi)^{-1} \Big|_{T=T_0}}{(\chi_{T_0,t_0}^\varphi)^{-1}} (T - T_0) \right\} \quad (\varphi = P, AF)$$

where $(\chi_{T_0,t_0}^\varphi)^{-1}$ is given by (6) and

$$\begin{aligned} \frac{\partial}{\partial T} (\chi_{T,t_0}^P)^{-1} \Big|_{T=T_0} &= \left(\frac{\partial^3 f}{\partial m^2 \partial T} - \frac{\frac{\partial^2 f}{\partial e \partial T} \frac{\partial^3 f}{\partial m^2 \partial e}}{\frac{\partial^2 f}{\partial e^2}} \right) \Big|_{T=T_0} \\ \frac{\partial}{\partial T} (\chi_{T,t_0}^{AF})^{-1} \Big|_{T=T_0} &= \left(\frac{\partial^3 f}{\partial m^2 \partial T} - \frac{\frac{\partial^2 f}{\partial e \partial T} \frac{\partial^3 f}{\partial m^2 \partial e}}{\frac{\partial^2 f}{\partial e^2}} - \frac{1 - \frac{\frac{1}{2!} \frac{\partial^4 f}{\partial \eta^2 \partial m^2} \frac{\partial^3 f}{\partial \eta^2 \partial e}}{\frac{1}{3!} \frac{\partial^4 f}{\partial \eta^4} \frac{\partial^3 f}{\partial m^2 \partial e}}}{\frac{\partial^3 f}{\partial m^2 \partial T} - \frac{\frac{\partial^2 f}{\partial e \partial T} \frac{\partial^3 f}{\partial m^2 \partial e}}{\frac{\partial^2 f}{\partial e^2}}} + \frac{1}{2!} \frac{\partial^3 f}{\partial \eta^2 \partial T} \frac{\frac{\partial^3 f}{\partial e \partial \eta^2} \frac{\partial^3 f}{\partial e \partial m^2} - \frac{\partial^4 f}{\partial \eta^2 \partial m^2} \frac{\partial^2 f}{\partial e^2}}{\frac{\partial^3 f}{\partial m^2 \partial T} - \frac{\frac{\partial^2 f}{\partial e \partial T} \frac{\partial^3 f}{\partial m^2 \partial e}}{\frac{\partial^2 f}{\partial e^2}}} \right) \Big|_{T=T_0} \\ \lambda_{T_0,t_0}^P &= - \frac{\frac{1}{2!} \frac{\partial^3 f}{\partial e \partial m^2} \Big|_0}{\left(\frac{\partial^2 f}{\partial m^2} \right)_0 \frac{\partial^2 f}{\partial e^2} \Big|_0} \\ \lambda_{T_0,t_0}^{AF} &= \lambda_{T_0,t_0}^P \frac{1 - \frac{\frac{1}{2!} \frac{\partial^4 f}{\partial \eta^2 \partial m^2} \Big|_0 \frac{\partial^3 f}{\partial e \partial \eta^2} \Big|_0}{\frac{1}{3!} \frac{\partial^4 f}{\partial \eta^4} \Big|_0 \frac{\partial^3 f}{\partial e \partial m^2} \Big|_0}}{1 - \frac{\frac{1}{2!} \left(\frac{\partial^3 f}{\partial e \partial \eta^2} \right)_0}{\frac{1}{3!} \frac{\partial^4 f}{\partial \eta^4} \Big|_0 \frac{\partial^2 f}{\partial e^2} \Big|_0}}. \end{aligned}$$

III. Model. — **III.1 GENERAL CONSIDERATIONS.** — Let us study the characteristics of the transition surface corresponding to the free energy (1), which may be re-written as follows

$$f = -\frac{\tau_F(e)}{2} m^2 - \frac{\tau_{AF}(e)}{2} \eta^2 + V(e) + \frac{T}{4} [(1+m+\eta) \text{Log}(1+m+\eta) + (1+m-\eta) \text{Log}(1+m-\eta) + (1-m+\eta) \text{Log}(1-m+\eta) + (1-m-\eta) \text{Log}(1-m-\eta) - 4 \text{Log} 2],$$

where $\tau_F = J_2 + J_1$ and $\tau_{AF} = J_2 - J_1$ are the ferromagnetic and antiferromagnetic (respectively) second order transition temperatures at vanishing external field, as may be seen in the following equations :

$$\begin{aligned} \frac{2\eta}{1-m^2+\eta^2} &= \text{th} \frac{2\tau_{AF}\eta}{T} \\ \frac{2m}{1+m^2-\eta^2} &= \text{th} \frac{2(\tau_F m + H)}{T} \\ 2V' &= \tau'_F m^2 + \tau'_{AF} \eta^2 + 2t. \end{aligned} \quad (7)$$

The antiferromagnetic second order transition temperature T_0 is readily obtained from the first equation by considering the limit $\eta \rightarrow 0$:

$$T_0 = \tau_{AF0}(1 - m_0^2).$$

Using this and the second equation we obtain that at 0 °K the second order transition field (considering the positive-field portion of the transition surface) is $H_0 = -\tau_{F0}$. As we are interested in a (P) $\leftrightarrow$ (AF) transition we must suppose $\tau_{AF} > \tau_F$, hence $J_1 < 0$. We shall treat, for simplicity, the case $J_2 \equiv 0$. If all the partial derivatives of f required to use (4) are evaluated we obtain

$$\eta^2 = 3 \left[\tau_{AF0} - \frac{\tau_{AF0}^2 (4\tau_{AF0} - T_0)}{\tau_{AF0}'' m_0^2 + 2V_0''} \right]^{-1} \cdot \left[\left(H_0 m_0 - \tau_{AF0} + \frac{H_0 m_0 T_0 \tau_{AF0}^2}{\tau_{AF0}^2 (\tau_{AF0}'' m_0^2 + 2V_0'')} \right) (T - T_0) - m_0 T_0 \left(1 + \frac{T_0 \tau_{AF0}^2}{\tau_{AF0}^2 (\tau_{AF0}'' m_0^2 + 2V_0'')} \right) (H - H_0) + \frac{2 T_0 \tau_{AF0}'}{\tau_{AF0}'' m_0^2 + 2V_0''} (t - t_0) \right]. \quad (8)$$

The critical line on the second order transition surface is determined by making the first and the second expressions between keys equal to zero. Let us study the consequences of system (8).

III.2 VANISHING MAGNETOSTRICTION. — We shall study the plane $t = t_0$ for which τ'_{AF0} vanishes (assuming it does for some value of e) ; hence (8) becomes :

$$\eta^2 = \frac{3}{\tau_{AF0}^2} \times [(\tau_{AF0} - H_0 m_0)(T_0 - T) + m_0 T_0(H_0 - H)].$$

We see that considering the point where $\tau'_{AF0} = 0$ is equivalent to assume that τ_{AF0} is a constant, that the transition is of the second order, and that

$$\left. \frac{\partial H_0}{\partial T_0} \right|_{t=t_0} = \frac{H_0 m_0 - \tau_{AF0}}{m_0 T_0}.$$

Using (7) it is readily obtained that

$$\left. \frac{\partial H_0}{\partial T_0} \right|_{t=t_0} = \begin{cases} -\infty & \text{for } m_0 = 0 \\ & (\text{hence } H_0 = 0 \text{ and } T_0 = \tau_{AF0}) \\ +\infty & \text{for } m_0 = 1 \\ & (\text{hence } H_0 = \tau_{AF0} \text{ and } T_0 = 0). \end{cases}$$

The transition line is given on figure 1.

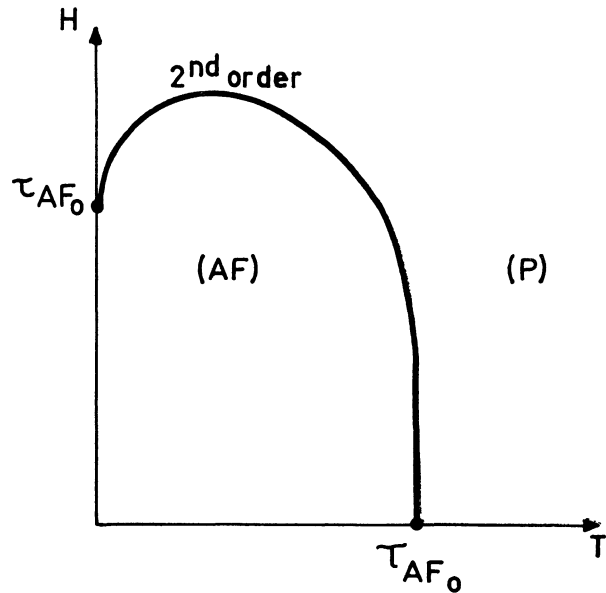


FIG. 1. — Transition line for vanishing magnetostriction.

III.3 VANISHING APPLIED MAGNETIC FIELD. — In the plane $H = 0$ we have $T_0 = \tau_{AF0}$ and (8) becomes :

$$\eta^2 = \frac{(T_0 - T) + \frac{\tau'_{AF0}}{V_0''} (t - t_0)}{\frac{\tau_{AF0}}{3} - \frac{\tau_{AF0}^2}{2V_0''}}.$$

We see that it may exist a critical point determined by

$$\frac{\tau_{AFc}}{3} - \frac{\tau_{AFc}^{'2}}{2 V_c''} = 0,$$

and in that case first order transitions will appear on the low temperature side of the critical point. The reason for this is that V_0'' (which is positive because of stability) and τ_{AF0}' are, in all usual physical situations, only slightly dependent on strain e . This point is however discussed in some more detail in section III.4. We see also that $(\partial t_0 / \partial T_0) |_{H=0} = V_0'' / \tau_{AF0}'$, hence the sign of the slope is determined by the sign of τ_{AF0}' . Hence, if the transition temperature behaves as indicated on figure 2, the transition line (with possible critical points) will be as indicated on figure 3.

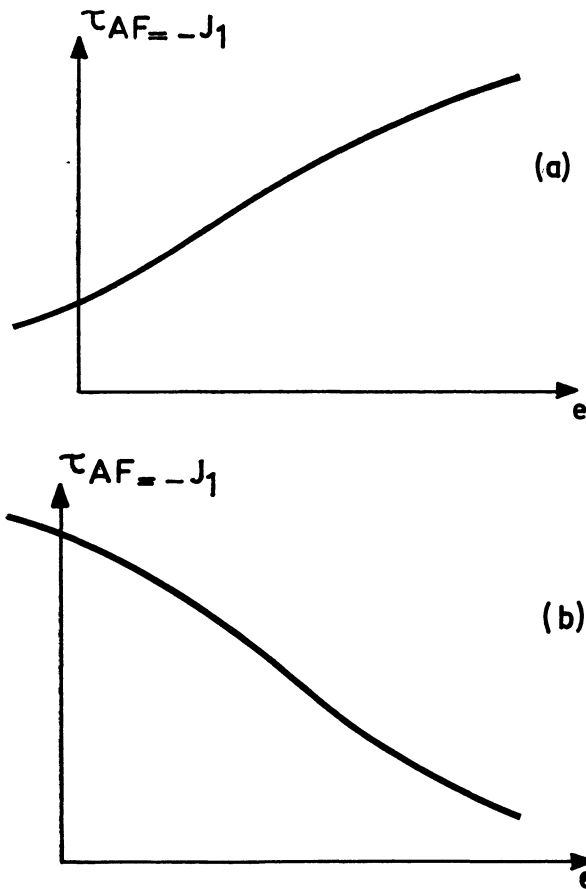


FIG. 2. — Two possible strain-dependences of the transition temperature. a) $\tau_{AF}' > 0$ [2] and b) $\tau_{AF}' < 0$ [4], [11] (*).

III.4 STABILITY, CRITICAL LINE AND ORDER OF THE TRANSITION. — Let us study in this section some characteristics of the transition surface ($\eta = 0$). First of all, the stability of the system requires the eigenvalues of the matrix on the left hand side of (3') to be positive. Hence the trace and determinant of

(*) In a similar way, both signs for τ_F' exist in actual ferromagnetic substances : $\tau_F' > 0$ [3], [15] and $\tau_F' < 0$ [10], [15].

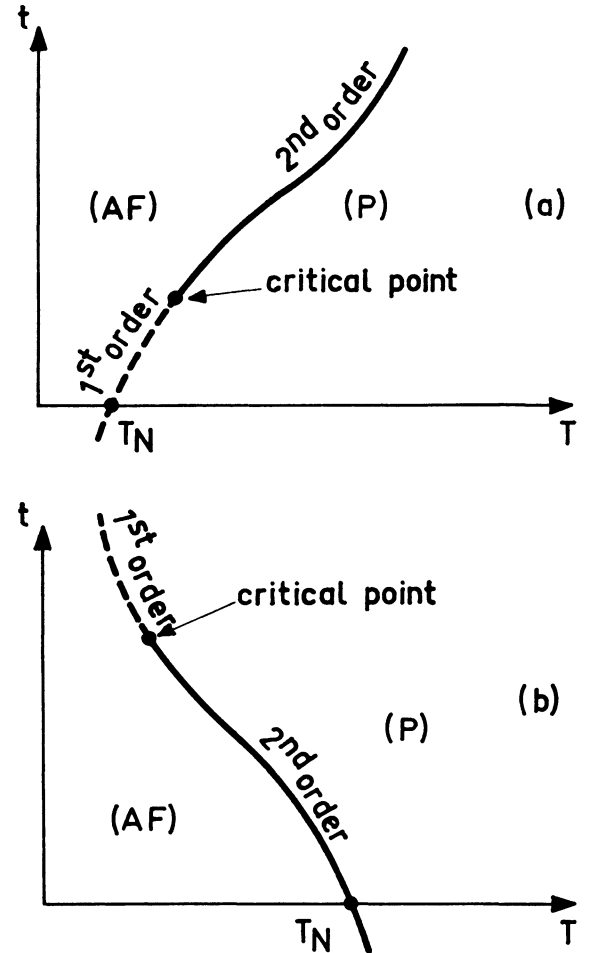


FIG. 3. — Two possible transition lines (corresponding respectively to the two cases in figure 2, for vanishing H , where the existence of critical points has been assumed).

this matrix must be positive. This leads to the following requirements (besides τ_{AF0} and V_0'' positive) :

$$-1 < B_0 \equiv \frac{\tau_{AF0}^{'2}}{2 V_0''}$$

$$0 \leq A_0 \equiv \frac{\tau_{AF0}^{'2}}{2 \tau_{AF0} V_0''} < 1 + B_0,$$

where A_0 and B_0 are (through τ_{AF} and V) functions of e .

Let us now define, for a given plane $t = t_0$, the function

$$\theta_0 \equiv 1 - \frac{\tau_{AF0}^{'2} (4 \tau_{AF0} - T_0)}{\tau_{AF0}^2 (\tau_{AF0}^{'2} m_0^2 + 2 V_0'')} = 1 - \frac{A_0 \left(4 - \frac{T_0}{\tau_{AF0}}\right)}{1 + B_0 \left(1 - \frac{T_0}{\tau_{AF0}}\right)}, \quad (9)$$

where we have used $T_0 = \tau_{AF0}(1 - m_0^2)$. Clearly $\theta_0 = 0$ determines the critical point (if such a point

does exist at $t = t_0$) and $\theta_0 > 0$ (< 0) means second order (first order) transition. It follows that a critical point exists if and only if the values of A_0 and B_0 determine, on a (A_0, B_0) plane, a point laying between the straight lines $A_0 = \frac{1}{3}$ (which corresponds to a critical temperature $T_c = \tau_{AFc}$) and $A_0 = \frac{1}{4}(1 + B_0)$ (which corresponds to $T_c = 0$). Clearly the stability requirements must be simultaneously satisfied.

Let us now make a first order development of θ_0 in the vicinity of a critical point. Assuming

$$V(e_0) \simeq V(e_c) + V'(e_c)(e_0 - e_c) + \frac{V''(e_c)}{2}(e_0 - e_c)^2$$

$$\tau_{AF}(e_0) \simeq \tau_{AF}(e_c) + \tau'_{AF}(e_c)(e_0 - e_c) + \frac{\tau''_{AF}(e_c)}{2}(e_0 - e_c)^2$$

and using the third equation of (7) (which gives us $d\theta_0/dT_0|_{T_c}$) we obtain for (9)

$$\begin{aligned} \theta_0 &\simeq \left(\frac{\partial \theta_0}{\partial e_0} \Big|_{e_c} \frac{de_0}{dT_0} \Big|_{T_c} + \frac{\partial \theta_0}{\partial T_0} \Big|_{T_c} \right) (T_0 - T_c) \\ &= \frac{d\theta_0}{dT_0} \Big|_{T_c} (T_0 - T_c), \end{aligned}$$

where the coefficient

$$\frac{d\theta_0}{dT_0} \Big|_{T_c}$$

is a known function which we may discuss. It follows that

$$-1 < B_c \equiv \frac{\tau''_{AFc}}{2V''_c} < \frac{1}{3} \left(> \frac{1}{3} \right)$$

implies that

$$\frac{d\theta_0}{dT_0} \Big|_{T_c} > 0 \text{ } (< 0),$$

which means that the transition on the high temperature side of the critical point is a second order (first order) one (as a matter of fact the limiting value is not exactly $1/3$ but depends slightly on the actual critical temperature T_c). This example shows that both situations are thermodynamically possible. However it is clear that, in all usual physical cases, B_c is expected to be almost vanishing, which enables us to understand (at least for this particular model) why the experimental evidence seems to be that, if a critical point exists, the second order transition is always on the high temperature side.

In section III.2, we saw an example of lowering of the symmetry with increasing temperature through a second order transition. Let us study in a more complete way this problem. For this we define the function

$$\psi \equiv H_0 m_0 - \tau_{AF_0} + \frac{H_0 m_0 T_0 \tau_{AF_0}^2}{\tau_{AF_0}^2 (\tau_{AF_0}^2 m_0^2 + 2V_0'')};$$

$\psi = 0$ determines the maxima line (noted M-line) which separates the transition surface in two regions corresponding to $\psi > 0$ (ordered phase at the high temperature side of the second order transition temperature) and $\psi < 0$ (ordered phase at the low temperature side) respectively. We shall note T_M the temperature values of the M-line. On the critical line we have :

$$\psi_c = H_c m_c - \tau_{AFc} + \frac{H_c m_c T_c}{4\tau_{AFc} - T_c}.$$

$\psi_c > 0$ (< 0) means that $T_c < T_M$ ($> T_M$).

If we use equations (7) we may re-write ψ_c :

$$\begin{aligned} \psi_c &= \tau_{AFc} \left\{ \left[m_c^2 + \frac{1 - m_c^2}{2} m_c \arg \operatorname{th} \frac{2m_c}{1 + m_c^2} \right] \times \right. \\ &\quad \left. \times \frac{4}{3 + m_c^2} - 1 \right\}. \end{aligned}$$

Hence for $m_c = 0$ (which means $H_c = 0$ and $T_c = \tau_{AFc|H=0}$) we obtain $\psi_c = -\tau_{AFc|H=0}$ and $d\psi_c/dm_c = 0$, and for $m_c = 1$ (which means $T_c = 0$ and $H_c = \tau_{AFc|T=0}$) we obtain $\psi_c = 0$ and $d\psi_c/dm_c = -\infty$. It follows that, if a critical line exists, there is necessarily a domain of stresses where $T_c > T_M$ and another where $T_c < T_M$. As a consequence of continuity of the transition surface and its slopes (see for example [19]), this second situation means that a lowering of the symmetry, with increasing temperature is possible also through a first order transition.

The results of this and preceeding sections enable us to present on figure 5 a typical transition surface which corresponds to a behavior of τ_{AF} as indicated on figure 4 (this is to say in the neighborhood of a

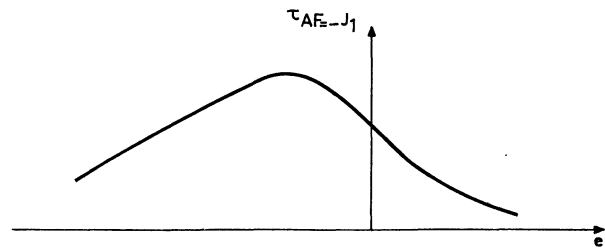


FIG. 4. — Possible strain dependence of the transition temperature.

maximum of τ_{AF} , whose experimental observation should allow useful comparison with theory). We assume in figure 5 the existence of a critical line and that $(\tau_{AF_0}^2/2V_0'') < 1/3$ for all stresses t corresponding to this critical line.

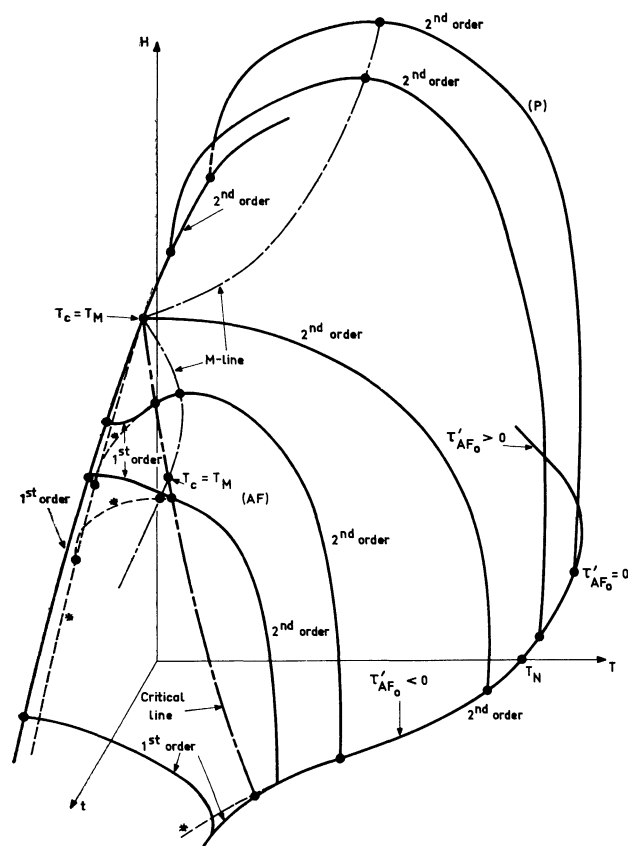


FIG. 5. — Example of transition surface (The *-lines indicate the location of the 2nd order transition, if no 1st order transition had been « preferred » by the system).

IV. Conclusion. — Let us make some comments on phase transition lines in a (H, T) plane, that is to say at constant stress t :

a) Experimental results indicate that, if a critical point exists, the transitions are of the second order on the high temperature side of the line. We have shown in Section III, that the opposite situation is not theoretically forbidden, however it seems very unlikely to be observed experimentally.

b) Usually the ordered phase is stable at the low

temperature side of the transition point. This is not always true : our model allows for the opposite case and this may occur not only for a second order transition (see for example [20]) but also for a first order one. It should be interesting to obtain experimental confirmation of these possibilities.

c) Rather general thermodynamic considerations (see Section II.1) indicate that, for a second order phase transition, the threshold magnetic field H_0 increases very sharply immediately below the Néel temperature T_N . This fact is experimentally confirmed in the case of FeCl_2 (second order transition) [21] and MnBr_2 (almost second order transition) [17].

When, for a fixed magnetic field, there is a critical point on the transition line in a (t, T) plane, the phase transitions are of the second order on the high temperature side of the line if the transition temperature τ_{AF} as a function of strain has no minimum. The NiAs — type NiS [2] seems to be an experimental confirmation of this result. It is finally worth while to remark that the part of figure 5 corresponding to $\tau'_{AF} < 0$ gives a good qualitative representation of the actual transition surfaces of FeCl_2 and FeBr_2 , considering that they do have a negative τ'_{AF} [11] and that the second order transitions occur, for constant stress, at the high temperature side of their critical points ([21], [22], [23] for FeCl_2 ; [21], [24] for FeBr_2).

In conclusion, a great variety of physical situations can be met with a simple model involving a small number of parameters. In particular we have pointed out that the assumption of a non vanishing J_2 is not an essential one : however it modifies the very low temperature behavior of second order transitions [20] and it allows, at non vanishing temperatures, for first order transitions from the antiferromagnetic to other magnetic (for example ferromagnetic) phases.

The author wishes to express his sincere gratitude to Dr. N. Boccara who suggested him to study this problem and gave him constant and valuable advices. He would like also to thank Dr. R. Bidaux and Dr. J. Hamman for useful discussions.

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