

CBPF-NF-068/83

ANISOTROPY, MAGNETIC FIELD AND STRESS IN-
FLUENCES ON THE PHASE TRANSITIONS OF SPIN-
FLOP-TYPE ANTIFERROMAGNETS

by

Sydney F. MACHADO *§

and

Constantino TSALLIS *

* Centro Brasileiro de Pesquisas Físicas/CNPq
Rua Dr. Xavier Sigaud, 150
22290 - Rio de Janeiro, RJ, BRAZIL

§ Departamento de Física e Química, Universidade Federal
do Espírito Santo
29000 - Vitória, ES, BRAZIL

ANISOTROPY, MAGNETIC FIELD AND STRESS INFLUENCES ON THE PHASE TRANSITIONS
OF SPIN-FLOP-TYPE ANTIFERROMAGNETS

SYDNEY F. MACHADO

Centro Brasileiro de Pesquisas Físicas - CBPF/CNPq
Rua Dr. Xavier, 150 - 22290 - Rio de Janeiro, RJ - BRAZIL and Departamento
de Física e Química, Universidade Federal do Espírito Santo - 29000 - Vitória,
ES - BRAZIL

CONSTANTINO TSALLIS

Centro Brasileiro de Pesquisas Físicas - CBPF/CNPq
Rua Dr. Xavier Sigaud, 150 - 22290 - Rio de Janeiro, RJ - BRAZIL

A B S T R A C T

We discuss, within a mean field approximation, the influences of anisotropy (in the spin space) and external uniaxial stress on the Heisenberg antiferromagnet in the presence of magnetic field. The phase diagram evolution (as function of anisotropy and stress) which is obtained, enables a satisfactory overall interpretation of recent experiments on $\text{Mn}(\text{Br}_{1-x}\text{Cl}_x)_2 \cdot 4\text{H}_2\text{O}$, $\text{K}_2[\text{FeCl}_5(\text{H}_2\text{O})]$, $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ and $(\text{C}_2\text{H}_5\text{NH}_3)_2\text{CuCl}_4$.

Key-words: Magnetic anisotropy; Stress influence; Spin-flop antiferromagnet.

I - INTRODUCTION

Since the pioneer work of Néel in 1932, a great amount of experimental and theoretical effort has been devoted to the understanding of antiferromagnetic systems. On experimental grounds we can mention, among many others, works on $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ ^[1], MnF_2 ^[2], $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ ^[3], $\text{MnBr}_2 \cdot 4\text{H}_2\text{O}$ ^[4] (see also references therein). More recently experiments have been performed analysing magnetoelastic effects on RbMnF_3 ^[5] and the pressure influence on $\text{K}_2[\text{FeCl}_5(\text{H}_2\text{O})]$ ^[6,7], $\text{Mn}(\text{Br}_{1-x}\text{Cl}_x)_2 \cdot 4\text{H}_2\text{O}$ ^[8], $(\text{C}_2\text{H}_5\text{NH}_3)_2\text{CuCl}_4$ ^[9] and $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ ^[10]; the effects of concentration have been systematically investigated on $\text{Mn}(\text{Br}_{1-x}\text{Cl}_x)_2 \cdot 4\text{H}_2\text{O}$ ^[11,12].

On theoretical grounds, most of the work has been done within mean field approximation (MFA) frameworks. This includes the study of several purely thermomagnetic aspects^[13-15], the influences of external stress^[16] and concentration of magnetic ions^[17,18] as well as effects due to the presence of an external magnetic field not necessarily parallel to the easy axis^[19]. Some work^[20] already exists which focuses the critical aspects of the phase transition, including the analysis of the bicritical point, where all three paramagnetic (P), antiferromagnetic (AF), and spin-flop (SF) phases join together.

By following along the lines of Ref.^[16] (which is herein recovered as the infinite anisotropy limit), we discuss, within a variational formulation of the MFA (Section II), the anisotropic Heisenberg antiferromagnet and exhibit the influences on the T (temperature) - H (magnetic field) phase diagram of anisotropy in the spin space (Section III) and of external uniaxial stress (Section IV). The comparison with experimental data is included in Sections III and IV, and reasonable agreement is verified.

II - MODEL AND FORMALISM

Let us consider a three-dimensional system of N spins $1/2$, half of them belonging to family A (oriented preferentially along the $+Z$ -axis), the other half belonging to family B (preferentially along $-Z$). The magnetic contribution to its associated Hamiltonian is given by

$$\mathcal{H}_m = \sum_{\langle a,b \rangle} J(e) (S_a^x S_b^x + S_a^y S_b^y + D S_a^z S_b^z) - H \left(\sum_{a=1}^{N/2} S_a^z + \sum_{b=1}^{N/2} S_b^z \right) \quad (1)$$

where the sum runs over first-neighbors (q will denote the coordination number), $J(e) > 0$, $D \geq 1$ (hence Z is the easy axis), $H \geq 0$ (convention), and e denotes the strain along the Z axis.

In order to treat the problem within the Variational Method of Statistical Mechanics, we adopt the following trial Hamiltonian:

$$\mathcal{H}_0 = - \sum_{a=1}^{N/2} (K_A^x S_a^x + K_A^y S_a^y + K_A^z S_a^z) - \sum_{b=1}^{N/2} (K_B^x S_b^x + K_B^y S_b^y + K_B^z S_b^z) \quad (2)$$

where the K 's are variational parameters to be determined by minimizing the variational free energy $\bar{F} \equiv F_0 + \langle \mathcal{H}_m - \mathcal{H}_0 \rangle_0$, where F_0 is the free energy associated with \mathcal{H}_0 , and $\langle \dots \rangle_0$ denotes the canonical thermal average performed with the states of \mathcal{H}_0 . It is well known that this procedure leads to the MFA equations. The replacement of the minimizing K 's into \bar{F} provides $\bar{F}(m_A^x, m_A^z, m_B^x, m_B^z; t, h)$ where

$$m_A^x \equiv 2 \langle S_A^x \rangle_0 \quad (3.a)$$

$$m_A^z \equiv 2 \langle S_A^z \rangle_0 \quad (3.b)$$

$$m_B^x \equiv 2 \langle S_B^x \rangle_0 \quad (3.c)$$

$$m_B^z \equiv 2 \langle S_B^z \rangle_0 \quad (3.d)$$

$$t \equiv 2k_B T / J(0) Dq \quad (4)$$

$$h \equiv H / J(0) Dq \quad (5)$$

and where we have taken advantage of the freedom to define the Y axis such that $m_A^y \equiv 2 \langle S_A^y \rangle_0 = 0$ and $m_B^y \equiv 2 \langle S_B^y \rangle_0 = 0$ (we recall that Hamiltonian (1)

is invariant through rotation in the XY plane). We obtain the following reduced magnetic free energy

$$f_m \equiv \frac{\bar{F}}{NJ(0)Dq} = \frac{j(e)}{4} \left(\frac{m_A^x m_B^x}{D} + m_A^z m_B^z \right) - \frac{h}{4} (m_A^z + m_B^z) + \frac{t}{8} \left[(1+m_A) \ln(1+m_A) \right. \\ \left. + (1-m_A) \ln(1-m_A) + (1+m_B) \ln(1+m_B) + (1-m_B) \ln(1-m_B) - 4 \ln 2 \right] \quad (6)$$

where

$$j(e) \equiv J(e)/J(0) \quad (7)$$

and

$$m_\alpha \equiv \left[(m_\alpha^x)^2 + (m_\alpha^z)^2 \right]^{1/2} \quad (\alpha=A,B) \quad (8)$$

By minimizing now f_m with respect to m_A^x , m_A^z , m_B^x and m_B^z we obtain the following equations of states:

$$m_A^i = - \frac{\delta_{xi} (m_B^x/D) + \delta_{zi} (m_B^z-h)}{\left[(m_B^x/D)^2 + (m_B^z-h)^2 \right]^{1/2}} \tanh \frac{\left[(m_B^x/D)^2 + (m_B^z-h)^2 \right]^{1/2}}{t} \\ \equiv k_i (m_B^x, m_B^z) \quad (i=x,z) \quad (9.a)$$

$$m_B^i = k_i (m_A^x, m_A^z) \quad (i=x,z) \quad (9.b)$$

where we have introduced Kronecker delta's for convenience. These equations of states provide three different phases, namely

$$P : m_A^x = m_B^x = 0 \quad \text{and} \quad m_A^z = m_B^z \geq 0 \quad (10.a)$$

$$AF : m_A^x = m_B^x = 0 \quad \text{and} \quad m_A^z \neq m_B^z \neq 0 \quad (10.b)$$

$$SF : m_A^x = -m_B^x > 0 \quad \text{and} \quad m_A^z = m_B^z > 0 \quad (10.c)$$

Before discussing the regions where one or the other phase is favored, let us introduce the elastic contribution. The total free energy F is given by

$$f \equiv \frac{F}{NJ(0)Dq} = f_m + u(e) - \theta e \quad (11)$$

where

$$\text{and} \quad u(e) \equiv U(e)/NJ(0)Dq \quad (12)$$

$$\theta \equiv \Theta/J(0)Dq \quad (13)$$

$U(e)$ being the total crystalline energy (herein treated within the standard adiabatic approximation, and assumed to satisfy $U''(e) > 0$ everywhere), and Θ being a uniaxial stress along the Z axis. The set of equations of thermodynamical states (Eqs.(9) and (10)) is completed by including that $(\partial f / \partial e)_{t,h,\theta} = 0$, i.e.

$$\left. \frac{\partial f_m}{\partial e} \right|_{t,h,\theta} + u'(e) = \theta \quad (14)$$

III - ANISOTROPY INFLUENCE

In this Section we discuss the influence of the anisotropy in spin space at constant axial strain (we shall assume $e = 0$ for convenience; we adopt $J_0 \equiv J(0)$).

III.1 - Theoretical results

By using Eqs.(9) and (10.b) in the limit $(m_A^Z - m_B^Z) \rightarrow 0$, we obtain the AF \leftrightarrow P critical line (which, within a straightforward analysis, turns out to be a second order one); it is given by

$$h^{\text{AF-P}} = \sqrt{1-t} + t \operatorname{arctanh} \sqrt{1-t} \quad (15)$$

hence

$$t^{\text{AF-P}}(h=0) = 1 \quad (15')$$

Note that this line, expressed in the t-h variables, is independent of D.

By using Eqs.(9) and (10.c) in the limit $m_A^X \rightarrow 0$ we obtain the SF \leftrightarrow P critical line (which also turns out to be a second order one); it is given by

$$t^{\text{SF-P}} = h/(D + 1) \operatorname{arctanh} \frac{Dh}{D + 1} \quad (16)$$

hence

$$h^{\text{SF-P}}(t=0) = \frac{D + 1}{D} \quad (16')$$

Note that also this line can be made independent of D , if we express it in the $(Dt) - Dh/(D + 1)$ variables.

By using Eqs.(9), (10.b) and (10.c) into Eq.(6), the free energies associated with both AF and SF phases can be numerically calculated: their equality determines the location of the phase boundary (which now turns out to be a first order one). In particular we obtain

$$h^{\text{AF-SF}}(t=0) = \sqrt{1 - \frac{1}{D^2}} \quad (17)$$

Our results are summarized in Figs. 1 and 2.

III.2 - Comparison with experimental results

Recently Westphal and Becerra (WB) [11] have measured the full phase diagram corresponding to the family $\text{Mn}(\text{Br}_{1-x}\text{Cl}_x)_2 \cdot 4\text{H}_2\text{O}$, which, for all values of x ($0 \leq x \leq 1$), presents a monoclinic crystallographic [11,21,22] and magnetic [21,23] unit cell containing 4 molecules (two pairs of spins $5/2$ per unit cell). These authors suggest that the relevant anisotropic mechanism would be of the type considered herein (see Hamiltonian (1)), i.e. anisotropic exchange (and not of the single-ion type). Although they speculate on the possibility of considering both intrasublattice and intersublattice exchanges, it seems worthy to try to interpret their results within the present theoretical framework. The effective values of J and D will naturally be x -dependent. From Eqs. (16') and (17) we obtain

$$\frac{h^{\text{SF-P}}(T=0, x)}{h^{\text{AF-SF}}(T=0, x)} = \sqrt{\frac{D + 1}{D - 1}} \quad (18)$$

where we have used definition (5). Through this equation and using the WB data, we obtain $D(x)$ (see Fig. 3)

From Eq. (16') and definition (5) we obtain

$$\frac{J_o(x)}{J_o(0)} = \frac{H^{\text{SF-P}}(T=0, x)}{H^{\text{SF-P}}(T=0, 0)} \frac{D(0) + 1}{D(x) + 1} \quad (19)$$

which, together with the above results for $D(x)$ and the WB [11] experimental data, determines $J_o(x)/J_o(0)$ (see Fig. 4).

Let us now test our theory: Eq. (15') together with definition (4) imply that

$$\frac{t_N(x)}{t_N(0)} = \frac{T_N(x)/J_o(x)D(x)}{T_N(0)/J_o(0)D(0)} = 1 \quad (20)$$

where T_N is the Néel temperature. The results are presented in Fig. 5.

Let us also test the location (t_b, h_b) of the bicritical point, whose D -dependence is obtained by simultaneously imposing Eqs. (15) and (16). The knowledge of $(t_b(D), h_b(D))$ together with definitions (4) and (5) and Eqs. (15') and (16') immediately provide

$$\frac{T_b(x)}{T_N(x)} = t_b(D) \quad (21)$$

and

$$\frac{H_b(x)}{H^{\text{SF-P}}(T=0, x)} = \frac{h_b(D) D}{D + 1} \quad (22)$$

The results are presented in Fig. 6. By using the WB experimental data [11], it would be possible in principle to calculate J_o and D ; however, this would be an unjustified overextension of the validity of the present theory which, besides the fact that it is a MFA one, has been devised for one pair of spins 1/2 per unit cell.

IV - STRESS INFLUENCE

IV.1 - Theoretical Results

We consider now the evolution of the system in the presence of a

(reduced) uniaxial stress θ . Eqs. (9), (10) and (14) univocally determine $(m_A^x, m_A^z, m_B^x, m_B^z, e)$ as functions of the external parameters (t, h, θ) for all three phases once D , $u(e)$ and $j(e)$ are known. In particular by considering the appropriate limits (specified in Section III.1) the AF-P and AF-SF critical surfaces can be determined (usually numerically); we respectively note them

$$\phi^{AF-P}(t, h, \theta) = 0 \quad \text{or} \quad \theta_o = \theta^{AF-P}(t_o, h_o) \quad (23.a)$$

$$\phi^{SF-P}(t, h, \theta) = 0 \quad \text{or} \quad \theta_o = \theta^{SF-P}(t_o, h_o) \quad (23.b)$$

we shall generically use (t_o, h_o, θ_o) to refer to points belonging to one or the other of these surfaces.

Quite tedious but straightforward expansions in the neighbourhood of the critical surfaces provide, for both AF-P and SF-P cases, the following thermodynamical stability conditions:

$$u_o'' > 0 \quad (24.a)$$

$$4u_o'' + j_o'' > \frac{(j_o')^2}{j_o} \geq 0 \quad (24.b)$$

The subscript (o) denotes the fact that these quantities are evaluated at $e_o \equiv e(t_o, h_o, \theta_o)$ (in order to make the connection with Section III, it is worthy to mention at this level that $e^{AF-P}(1, 0, 0) = 0$ by convention). We have also obtained

$$\left. \frac{\partial \theta^{AF-P}(t_o, h_o)}{\partial t_o} \right|_{h_o=0} = \frac{u_o''}{j_o'} \quad (25.a)$$

and

$$\left. \frac{\partial \theta^{AF-P}(t_o, h_o)}{\partial h_o} \right|_{t_o=0} = \frac{2(j_o'' + 4u_o'')}{j_o'} \quad (25.b)$$

We note that the conditions (24) imply that both limiting derivatives have the same sign as j_o' . Furthermore we have established that the AF-P transition will be a second order one if

$$1 + \frac{j_o''}{4u_o''} \left(1 - \frac{t_o}{j_o}\right) \geq \frac{(j_o')^2}{4u_o'' j_o} \left(4 - \frac{t_o}{j_o}\right) \quad (26)$$

and a first order one otherwise.

At $h_o = 0$ it is $t_o = j_o$ and therefore condition (26) implies

$$t_o \geq \frac{3}{4} \frac{(j_o')^2}{u_o''} \quad (27.a)$$

At the other end, if $t_o = 0$ it is $h_o = j_o$ and condition (26) implies

$$h_o \geq \frac{4(j_o')^2}{4u_o'' + j_o''} \quad (27.b)$$

In general Eqs. (27) do not provide a definite prescription. However in the most common cases, where $j(e)$ and $u(e)$ are respectively almost linear and almost quadratic in e (hence j_o' and u_o'' are almost constants and $j_o'' \approx 0$), Eqs.(27) imply $t_o > (\text{finite constant})$ and $h_o > (\text{finite constant})$. Consequently if a tricritical line exists on the AF-P critical surface, the second order region is located at the high temperatures and low fields side of it.

Let us now focus the SF-P critical surface. We obtain

$$\left. \frac{\partial \theta^{\text{SF-P}}(t_o, h_o)}{\partial t_o} \right|_{h_o=0} = D^2 \frac{u_o''}{j_o'} \quad (28.a)$$

and

$$\left. \frac{\partial \theta^{\text{SF-P}}(t_o, h_o)}{\partial h_o} \right|_{t_o=0} = \frac{D}{4(D+1)} \frac{4u_o'' + j_o''}{j_o'} \quad (28.b)$$

Once more both derivatives share the sign of j_o' . It can be shown that the phase transition will be a second order one if

$$\left\{ \left(1 + \frac{j_o'' m_o^2}{4u_o''}\right) \left[(1 - m_o^2) \operatorname{argtanh} m_o - m_o \right] - \frac{(j_o')^2}{2j_o u_o''} \left[(1 - m_o^2) \operatorname{argtanh} m_o - \frac{D+1}{D} m_o \right] \right\} /$$

$$\left\{ \left(1 + \frac{j_o'' m_o^2}{4u_o''}\right) \left[D(1 - m_o^2) \operatorname{argtanh} m_o + m_o \right] - \frac{D(j_o')^2}{2j_o u_o''} m_o^2 (1 - m_o^2) \operatorname{argtanh} m_o \right\} \geq 0 \quad (29)$$

and a first order one otherwise, where

$$m_o \equiv m(t_o, h_o, \theta_o) \equiv \frac{1}{2} (m_A^Z + m_B^Z)_o$$

At $h_o = 0$ it is $t_o = j_o/D$ and $m_o = 0$, and therefore condition (29) implies

$$t_o \geq \frac{(j_o')^2}{2u_o''} \quad (30.a)$$

At the other end, if $t_o = 0$ it is $h_o = j_o(D+1)/D$ and $m_o = 1$ and condition (29) implies

$$h_o \geq 2 \left(\frac{D+1}{D} \right)^2 \frac{(j_o')^2}{4u_o'' + j_o''} \quad (30.b)$$

As before, for linear $j(e)$ and quadratic $u(e)$, Eqs. (30) imply that, if a tricritical line exists on the SF-P critical surface, the second order region is located at the high temperatures and low fields side of it. This type of behaviour is similar to that observed previously^[14,16] in related systems.

The discussion of the evolution of the bicritical point as a function of θ is a quite complex one, and we shall restrict ourselves to the analysis within a rough approximation which consists in: (i) neglecting the differences in the strains associated with the AF-P and SF-P critical surfaces (i.e. we assume $e_o^{\text{AF-P}} \approx e_o^{\text{SF-P}}$); (ii) adopting linear θ -dependences for $t_o(\theta)$ at $h_o = 0$ and $h_o(\theta)$ at $t_o = 0$ for both AF-P and SF-P critical lines. In these circumstances it can be shown that, for increasing θ , the AF phase becomes predominant (in prejudice of the SF phase) if

$$\left. \frac{\partial^2 \text{SF-P}(t_o, h_o)}{\partial h_o} \right|_{t_o=0} < \frac{D^2(7D+8)}{8(D+1)^2} \left. \frac{\partial^2 \text{AF-P}(t_o, h_o)}{\partial t_o} \right|_{h_o=0} \quad (31)$$

hence

$$\frac{j_o''}{4u_o''} + 1 > \frac{D(7D+8)}{8(D+1)} \quad (32)$$

the opposite occurring in the opposite case. Both situations have been illustrated in Fig. 7 for the case $j_o' < 0$.

IV.2 - Comparison with experimental results

The present theory predicts almost linear dependences with stress for T_N and H^{SF-P} as long as $j'_O \neq 0$; the same most probably holds for H^{AF-SF} , although we have not checked it (numerically for instance). This is verified for $Mn(Br_{1-x}Cl_x)_2 \cdot 4H_2O$ ($x=0, 0.26$)^[8], $K_2[FeCl_5(H_2O)]$ ^[6] (orthorhombic crystallographic and cubic magnetic structure containing 4 molecules per unit cell; two pairs of spin 1/2 per unit cell^[24-27]), $CoCl_2 \cdot 6H_2O$ ^[10] (monoclinic crystallographic structure containing two molecules per unit cell; almost two-dimensional magnetic structure; one pair of spins per unit cell^[28]), and $(C_2H_5NH_3)_2CuCl_4$ ^[9] (face centered orthorhombic crystallographic structure containing 4 molecules per unit cell; almost two-dimensional magnetic structure; two pairs of spin 1/2 per unit cell^[9,29]). If we focus T_N and H^{SF-P} , all these substances correspond (at least for pressures above the atmospheric one) to $j'_O < 0$, i.e. a decrease of the coupling strength under expanding stress. But if we focus H^{AF-SF} then $K_2[FeCl_5(H_2O)]$ and $(C_2H_5NH_3)_2CuCl_4$ present one type of behaviour, whereas $CoCl_2 \cdot 6H_2O$ presents the opposite one. The present theory suggests that this is an effect of the anisotropy; however a more complex model (including possible strain-dependence of the anisotropy, i.e. $D.=D(e)$) would be necessary in order to attempt any detailed comparison; furthermore, the present theory strictly applies to uniaxial stress, whereas the experiments are usually performed under isotropic pressure.

V - CONCLUSION

We have discussed, in the mean field approximation language, the anisotropic Heisenberg antiferromagnet, where the anisotropy appears in the

coupling constant associated with the easy axis. The (temperature)-(magnetic field) phase diagram presents three phases, namely the paramagnetic (P), antiferromagnetic (AF) and spin-flop (SF) ones. The relative importance of the AF and SF phases is determined by the value of the anisotropy. Within this very simple picture, it has been possible to give a qualitatively (and to a certain extent quantitatively) satisfactory overall interpretation of recent experiments on $\text{Mn}(\text{Br}_{1-x}\text{Cl}_x)_2 \cdot 4\text{H}_2\text{O}$ ^[11]. This analysis provides, among others, concentration-dependences for both the coupling constant and the anisotropy, which could be checked with other experimental techniques. In particular, this would enlighten whether the strain-independence of the anisotropy D we have assumed is a reasonable first-order approximation. Finally, though well known, it is worthy to recall that the quantitative predictions of a mean field theory should not be overemphasized; furthermore the present calculation has been done assuming one pair of spins 1/2 per unit cell (which is not the case of $\text{Mn}(\text{Br}_{1-x}\text{Cl}_x)_2 \cdot 4\text{H}_2\text{O}$, for instance). Nevertheless the fact that we are dealing with three-dimensional systems, and the fact that all theory-experiment comparisons have been performed for conveniently reduced quantities (see Figs. 3-6) should satisfactorily minimize the errors, and extend the applicability of the theory. Obviously harder theories should be needed to extract reliable information on the absolute values of microscopic quantities such as J or D .

In addition to that we have presented a preliminary discussion of the influence of external uniaxial stress (along the easy axis), which can throw some light on the interpretation of recent pressure-dependent experiments on $\text{Mn}(\text{Br}_{1-x}\text{Cl}_x)_2 \cdot 4\text{H}_2\text{O}$ ^[8], $\text{K}_2[\text{FeCl}_5(\text{H}_2\text{O})]$ ^[6], $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ ^[10] and $(\text{C}_2\text{H}_5\text{NH}_3)_2\text{CuCl}_4$ ^[9].

On general grounds we can say that first order phase transitions (from the paramagnetic phase to the ordered ones) can appear at fixed stress. In particular, if a tricritical point exists, the common situation will be the exhibition of the second order phase transition at the high temperature and low magnetic field side of it. Furthermore, in the (temperature)-(magnetic field)-(stress) space, the bicritical line and the tricritical one (if it exists) may cross each other, leading to complex but interesting situations (see Fig. 7). Confirmation (or not) of this expectations within frameworks which are more reliable in what criticality is concerned, would be very wellcome.

We acknowledge C.C. Becerra, W.A. Ortiz and R.F.S. Andrade for communicating to us their results prior to publication as well as D.M.H da Silva, I.P. Fittilpaldi and E.M.F. Curado for useful discussions. One of us (SFM) acknowledges Fellowships from CAPES and CNPq (Brazilian agencies).

REFERENCES

- 1 - J. van den Handel, H.M. Gijssman and N.J. Poulis, Physica 18, 862 (1952)
- 2 - I.S. Jacobs and J.S. Kouvel, J.Appl. Phys 32, 615 (1961)
- 3 - I.E.Rives and S.N. Bhatia, Phys. Rev. B12, 1920 (1975)
- 4 - C.C. Becerra, PhD Thesis - University of S.Paulo, S.Paulo, Brazil (1974)
- 5 - Y. Shapira and N.F. Oliveira Jr, Phys. Rev. B18, 1425 (1978)
- 6 - W.A. Ortiz, A. Paduan-Filho and F. Missel, Phys. Let. A77, 183 (1980)
- 7 - W.A. Ortiz, A. Paduan-Filho and F. Missel, J. Mag. Mag. Mat. 24, 67 (1981)
- 8 - W.A. Ortiz and F. Missel, Phys Rev. B24, 915 (1981)
- 9 - M. Chikamatsu, H. Yamazaki, K. Yamagata and H. Abe, J. Mag. Mag. Mat 31-34, 1191 (1983)
- 10 - K. Takeda, M. Wada, A. Ohtani, A. Onodera and T. Haseda, J. Mag. Mag. Mat. 31-34, 1193 (1983)
- 11 - C.H. Westphal and C.C. Becerra, J. Phys C15, 6221 (1982)
- 12 - C.C. Becerra, Anais do VII Simpósio Brasileiro de Física Teórica - Rio de Janeiro, Brazil (1982); unpublished
- 13 - C.J. Gorter and T. van Peski-Timbergen, Physica 22, 273 (1956)
- 14 - R.B. Bidaux, P. Carrara et B. Vivet, J. Phys. Chem. Sol. 28, 2453 (1967)
- 15 - J.M. Kincaid and E.G.D. Cohen, Phys. Rep. 22, 57 (1975)
- 16 - C. Tsallis, J. Physique 32, 903 (1971)
- 17 - R.A. Tahir-Kheli, S.M. Rezende, F.G. Brady Moreira and I.P. Fittipaldi, J. Phys. C9, L403 (1976)
- 18 - F.G. Brady Moreira, I.P. Fittipaldi, S.M. Rezende, R.A. Tahir-Keli and B. Zēks, Phys. Stat. Sol. B80, 385 (1977)
- 19 - K.W. Blazey, H. Rohrer and R. Webster, Phys. Rev. B4, 2287 (1971); R.F.S. Andrade and S.R. Salinas, J. Phys. Chem. Solids 40, 687 (1979).
- 20 - A.D. Bruce and A. Aharony, Phys. Rev. B11, 478 (1975);
A. Aharony, Phys. Rev. B18, 3328 (1978), and references therein.

- 21 - R.F. Altman, S. Spooner, D.P. Landau and J.E. Rives, Phys. Rev. B11, 458 (1975)
- 22 - R.D. Spence and V. Nagarajan, Phys. Rev. 149, 191 (1966)
- 23 - Z.M. El Saffor and G.M. Brown, Acta Cryst. B27, 66 (1971)
- 24 - F. Palacio, A. Paduan-Filho and R. Carlin, Phys. Rev. B21, 296 (1979)
- 25 - C.J. O'Connor, B.S. Deaver Jr., and E. Sinn, J. Chem. Phys. 70, 5161 (1979)
- 26 - I. Lindqvist, Ark. Kemi Mineral. Geol. 24, 1 (1947)
- 27 - A. Bellanca, Period. Mineral. 17, 59 (1948)
- 28 - M. Date, J. Phys. Soc. Jap. 16, 1337 (1961)
- 29 - M. Chikamatsu, M. Tanaka and H. Yamazaki, J. Phys. Soc. Jap. 50, 2876 (1981)

CAPTION FOR FIGURES

- FIG. 1 - Phase diagram corresponding to a fixed value of D (P, AF, SF respectively denote the paramagnetic, antiferromagnetic and spin-flop phase). The AF-P and SF-P critical lines are second order ones; the AF-SF is a first order one. The dashed lines (unphysical) indicate the analytic extensions of Eqs. (15) and (16). (t_b, h_b) denotes the location of the bicritical point.
- FIG. 2 - Evolution of the t - h phase diagram as a function of anisotropy. The dot-dashed line is the bicritical one; the dashed line is the AF-SF critical one.
- FIG. 3 - Concentration-dependence of D obtained through the present Eq. (18) and WB ^[11] data on $\text{Mn}(\text{Br}_{1-x}\text{Cl}_x)_2 \cdot 4\text{H}_2\text{O}$ (the dashed line is a guide to the eye).
- FIG. 4 - Concentration-dependence of $J_o(x)/J_o(0)$ obtained through the present Eq. (19) and WB ^[11] experimental data on $\text{Mn}(\text{Br}_{1-x}\text{Cl}_x)_2 \cdot 4\text{H}_2\text{O}$ (the dashed line is a guide to the eye).
- FIG. 5 - Concentration-dependence of $t_N(x)/t_N(0) = T_N/J_o D$ from experimental data ^[11] (the present theory predicts a constant).
- FIG. 6 - Location of the bicritical point (t_b, h_b) as a function of D (full line: present theory; dots: from experimental data ^[11]).
(a) $T_b(x)/T_N(x)$; (b) $H_b(x)/H^{\text{SF-P}}(T=0, x)$
- FIG. 7 - Indicative possible θ (stress)-evolutions of the t (temperature)- h (magnetic field) phase diagram: (a) the AF phase becomes predominant; (b) the SF phase becomes predominant. The dashed lines is the AF-SF critical ones, the dot-dashed line is the bicritical one, and the dotted line is a possible tricritical one. Both (a) and (b) correspond to $j_o' < 0$.

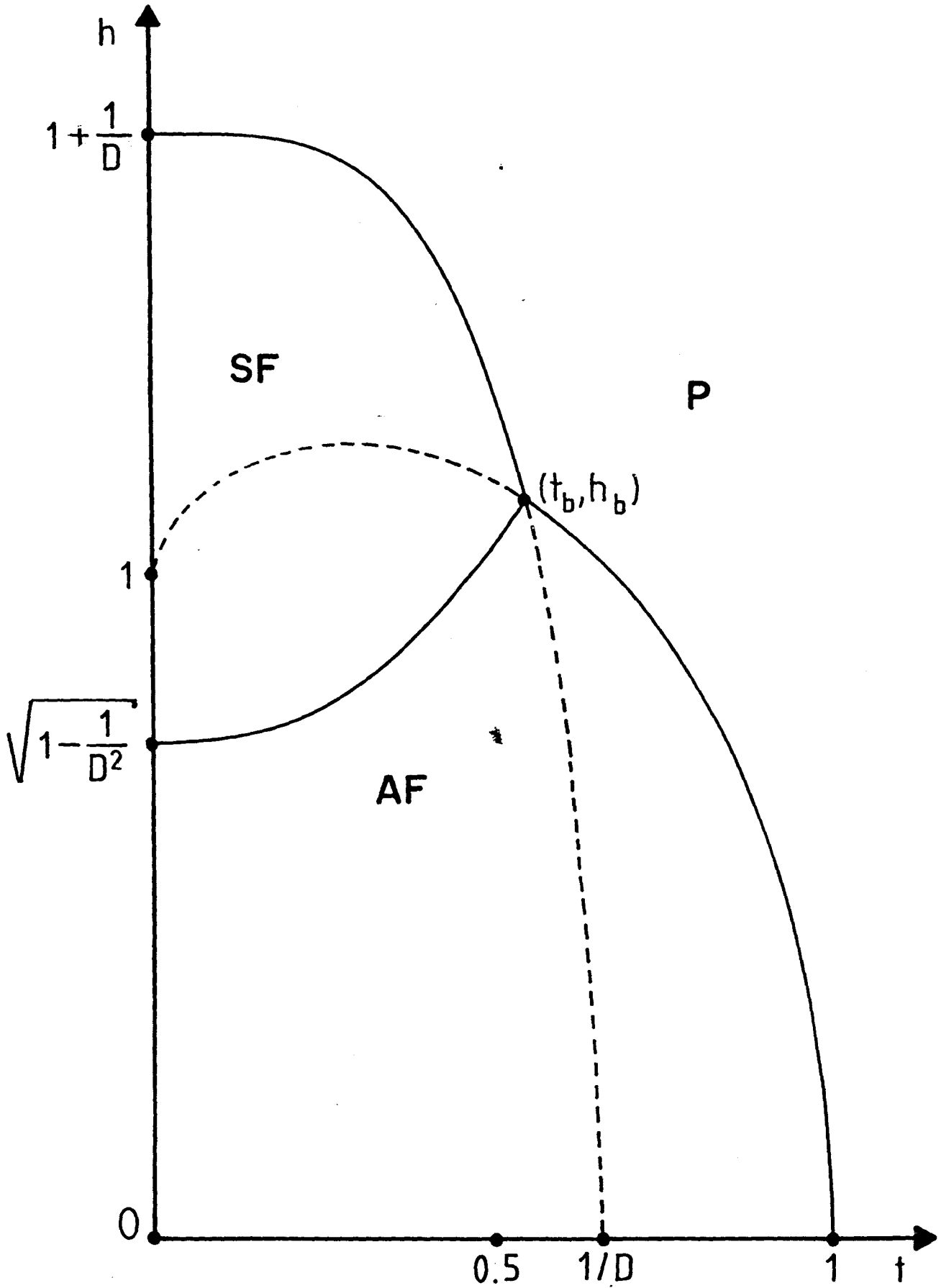


FIG. 1

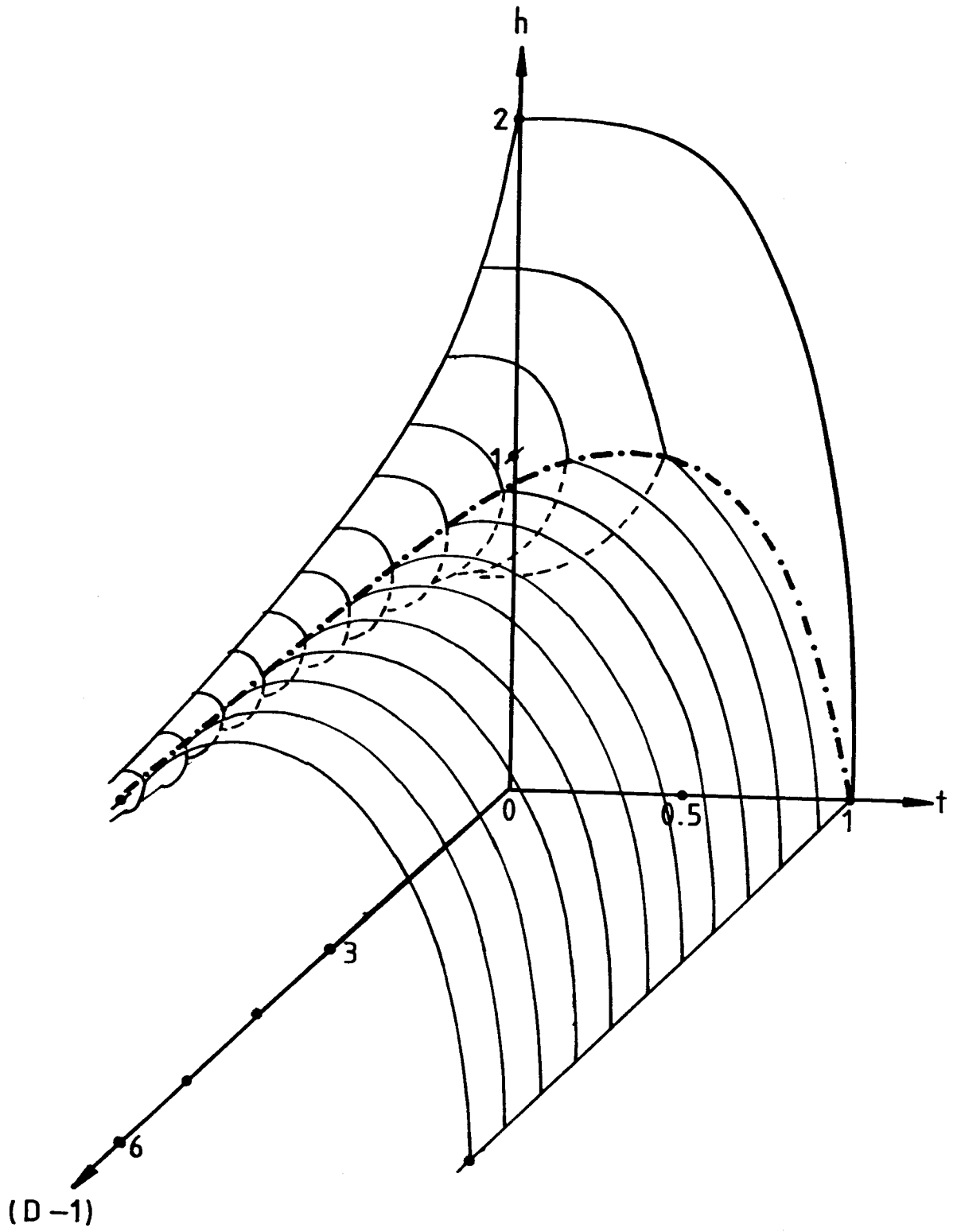


FIG.2

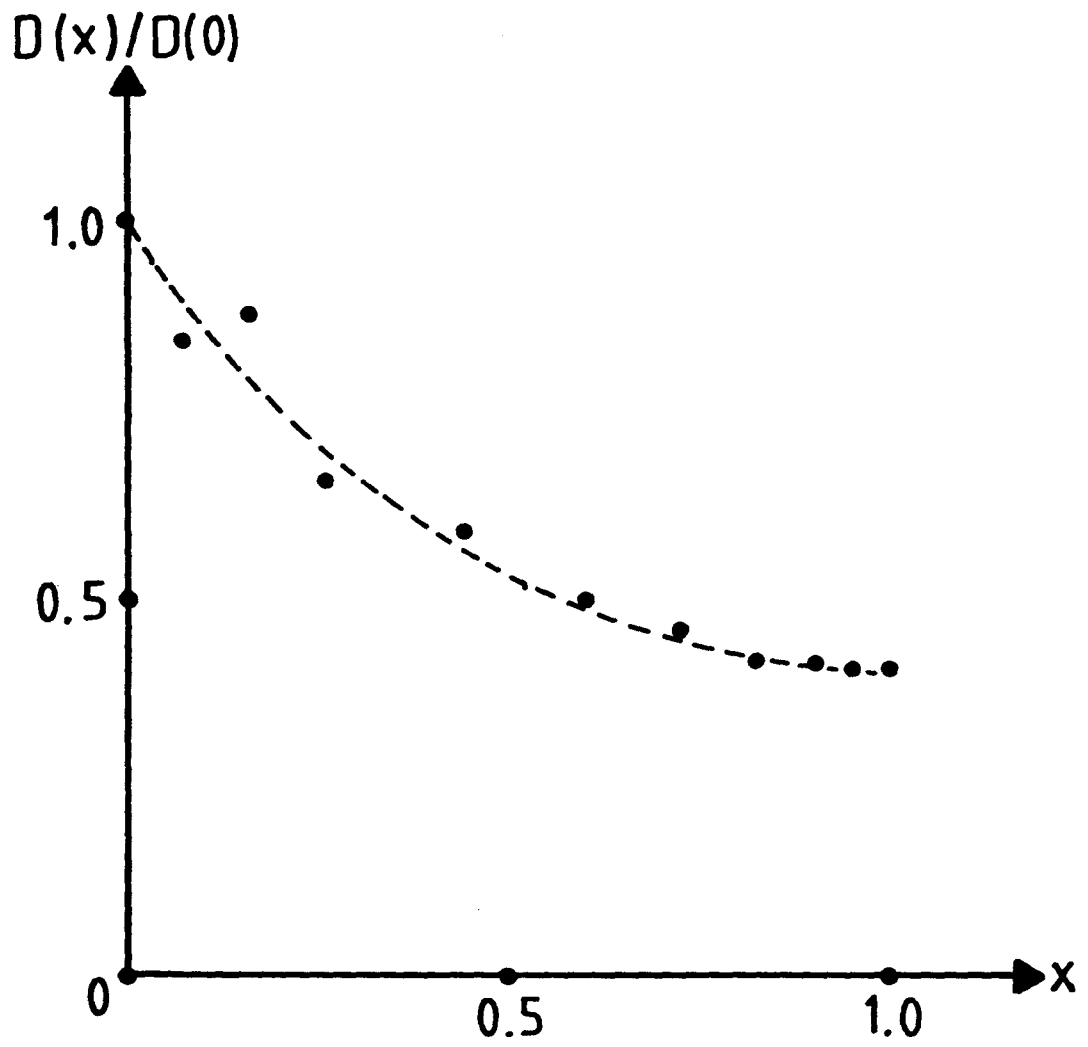


FIG. 3

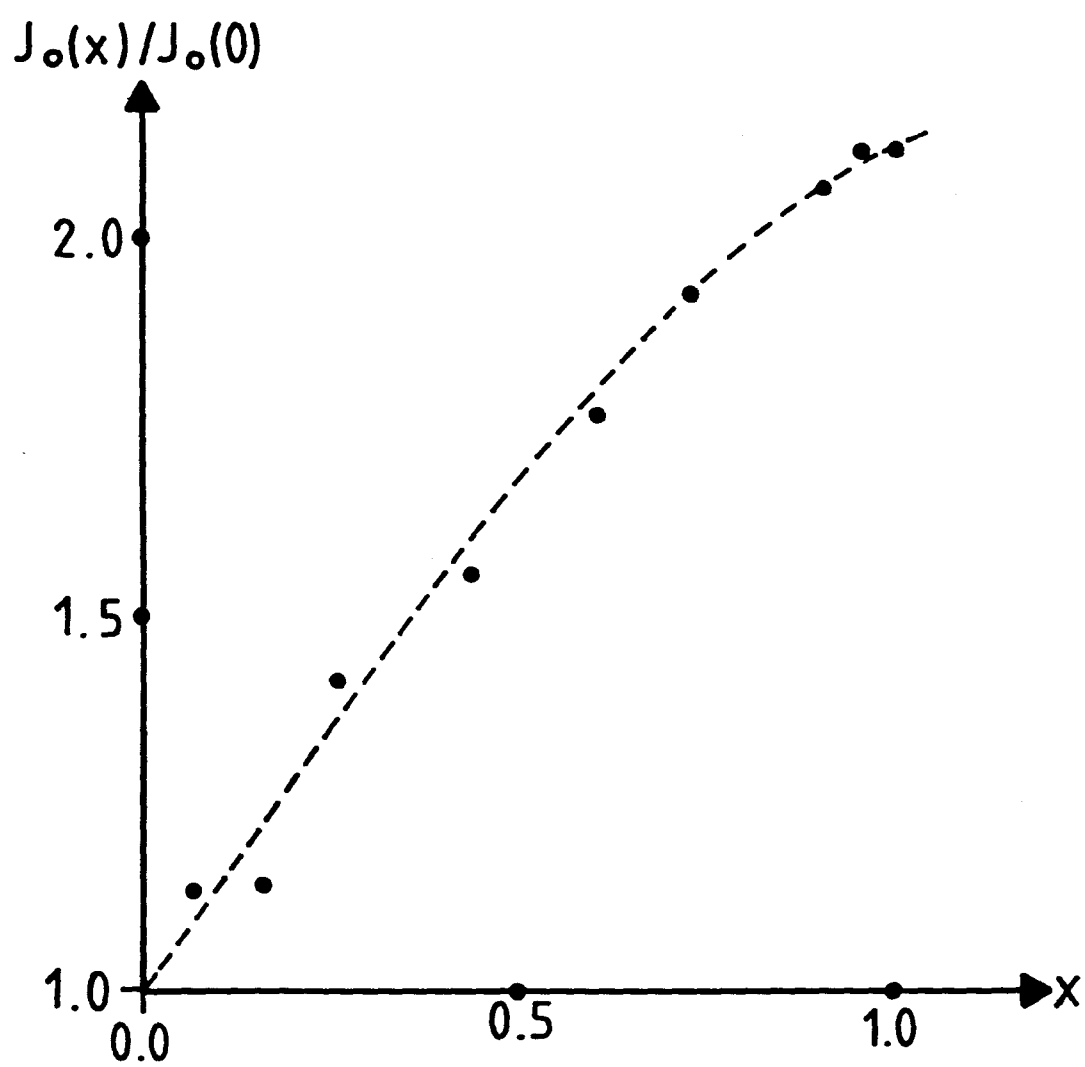


FIG. 4

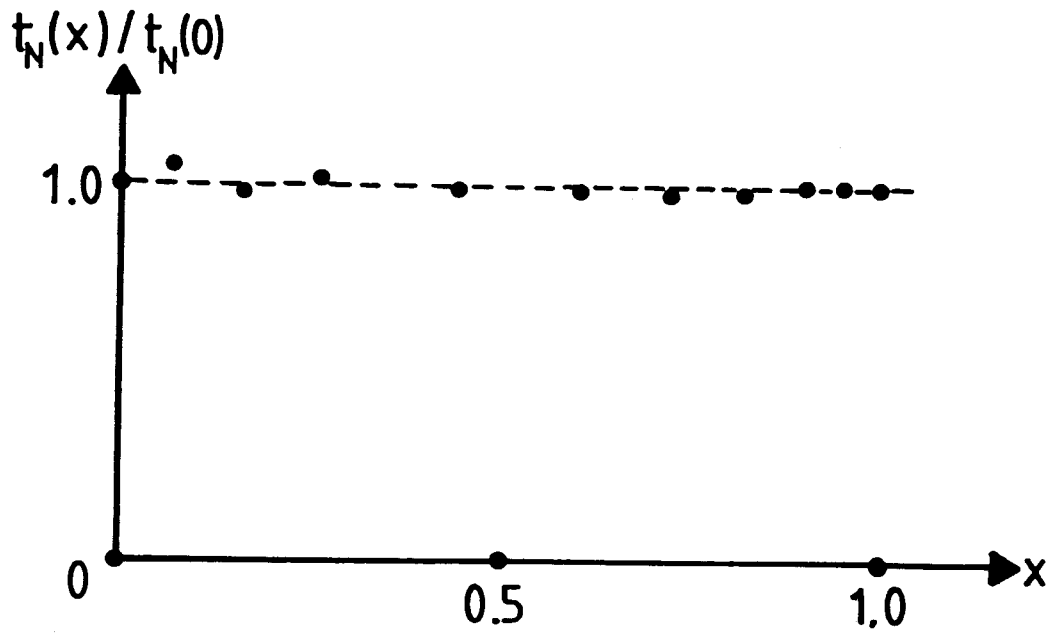
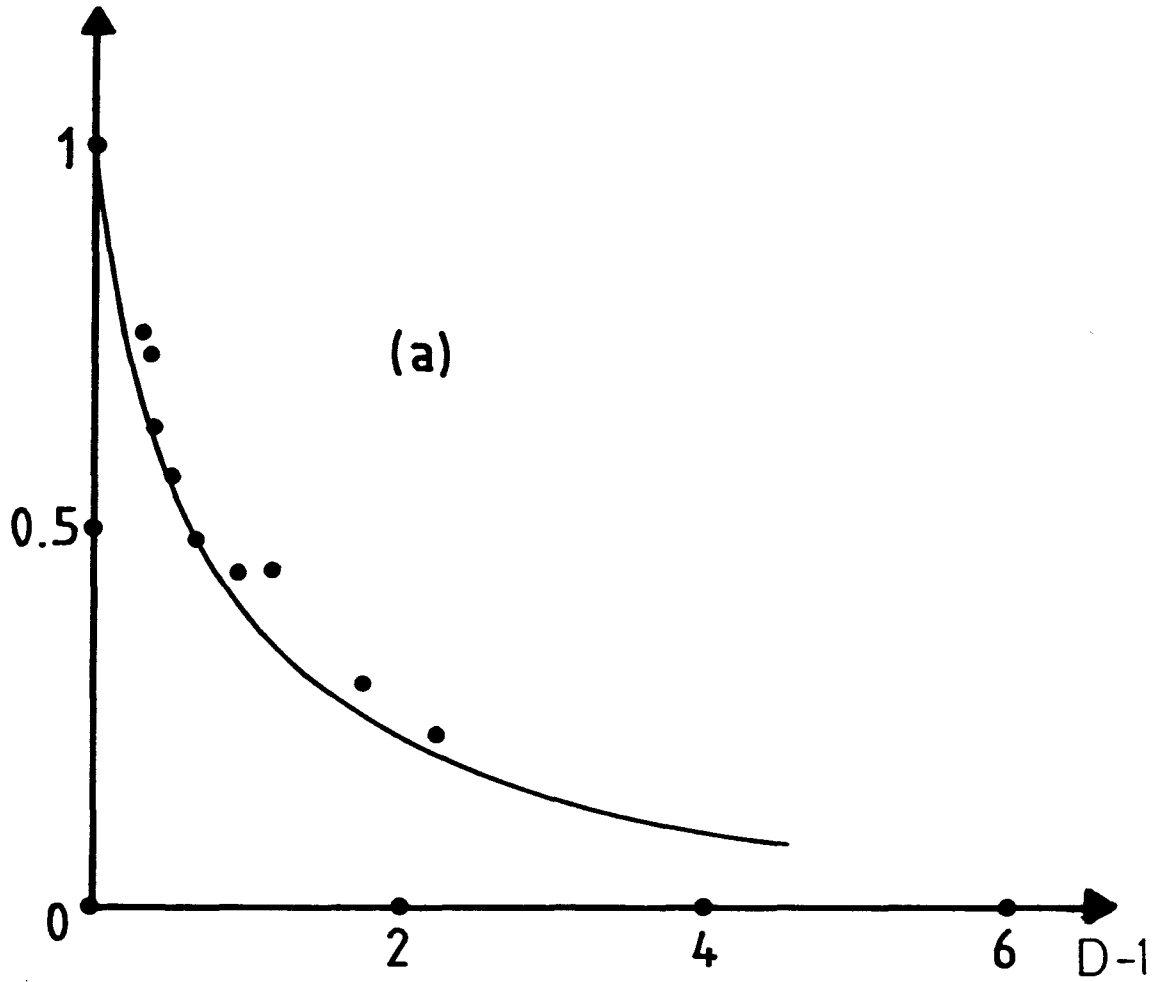


FIG. 5



$$H_b(x)/H^{SF-P}(T=0, x)$$

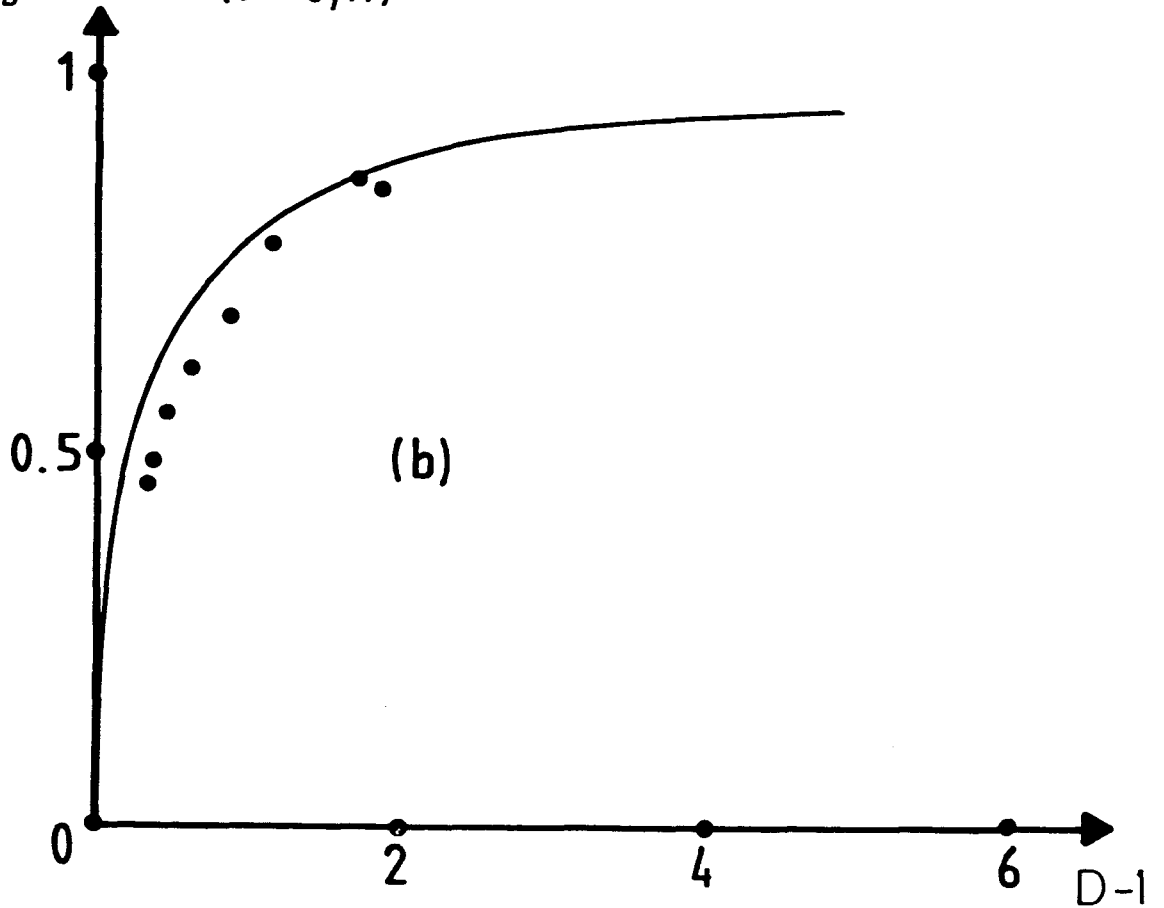


FIG. 6

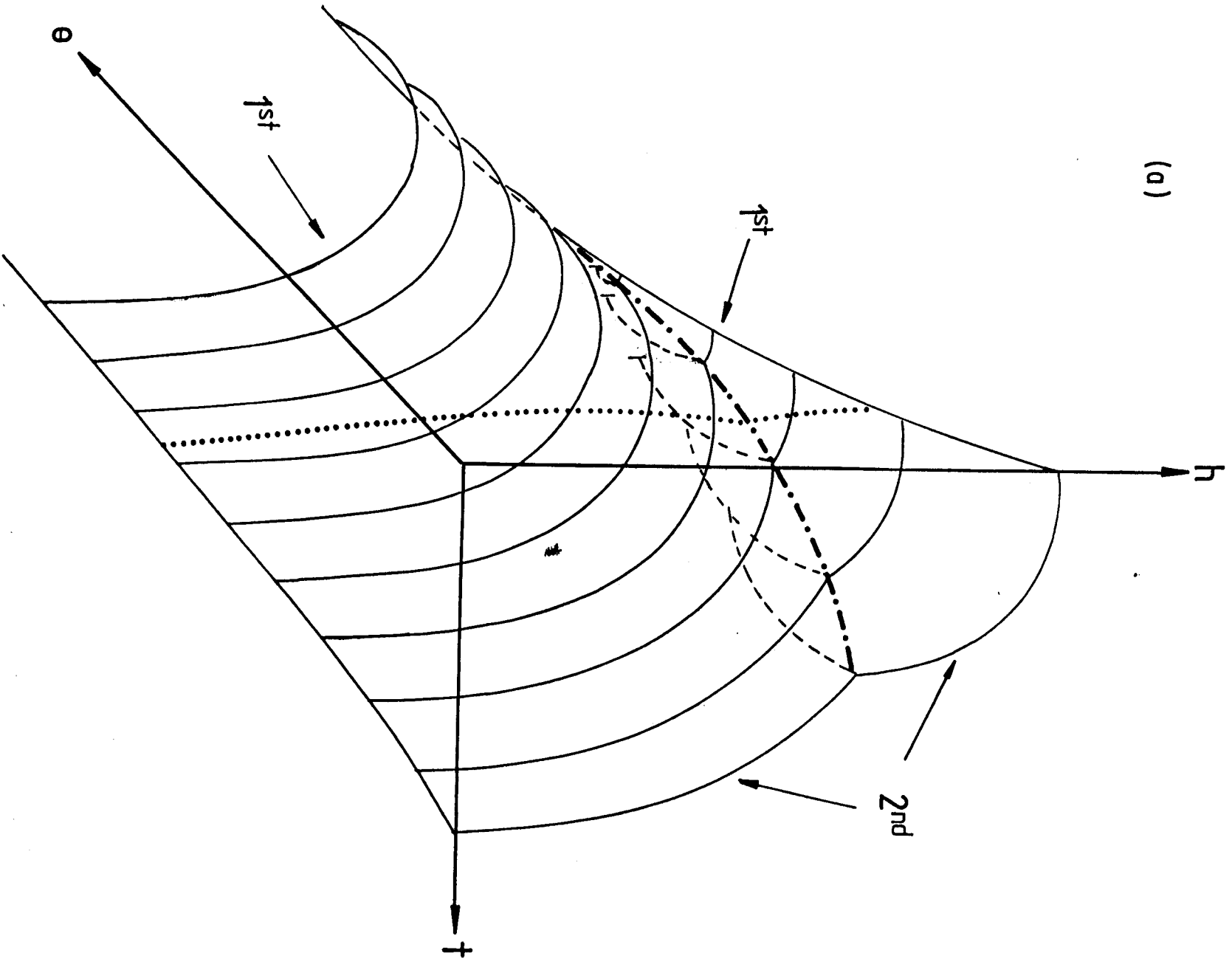


FIG. 7

