

CBPF-NF-005/81
DIMERIZATION IN THE MAGNETOSTRICTIVE
ONE-DIMENSIONAL XY MODEL

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ABSTRACT

We discuss the one-dimensional first-neighbour $\frac{1}{2}$ -spin magnetostrictive XY model (where the crystalline degrees of freedom are assumed to be three-dimensional), and exhibit that, for all temperatures below T_c , no other contributions to the structural order appear than the pure dimerization one. The influences of temperature and elastic constant on the order parameter and sound velocity are analyzed as well.

In the last decade increasing effort has been dedicated to the study of the so called spin-Peierls instability (SPI) which induces structural phase transitions in systems which are magnetically quasi-one-dimensional although three-dimensional in what concerns crystalline interactions. The system typically presents an uniform (or disordered) phase (equidistant atoms along the chain) at high temperatures and a more complex (or ordered) phase (a structurally dimerized or polymerized chain) at low temperatures. These facts have been tested^[1-7] on several substances like TTF-BDT and alkali-TCNQ salts. The relevant models that have been used are the magnetostrictive XY^[8-13] and Heisenberg^[14-19] ones. The XY model has the advantage of being exactly solvable, at least in what concerns the magnetic degrees of freedom. Pincus^[8] showed that an XY antiferromagnetic linear chain is, at vanishing temperature, unstable, with respect to dimerization. Beni and Pincus^[9] showed next that this instability induces a second order phase transition between the uniform and dimerized phases, *under the assumption that those two phases are the only ones to be considered*. Dubois and Carton^[10] proved that, at the critical temperature T_c and coming from high temperatures, *appears* an order which is indeed a dimerization. The scope of the present work is to study what happens *below* T_c , particularly to test if any other modes (in the immediate neighbourhood of the dimerization mode or not) become unstable as well. We shall exhibit that for all temperatures below T_c , *no other contributions to the structural order appear than the pure dimerization one*. This fact provides "a posteriori" a justification for the calculations of the vanishing external magnetic field free energy, order parameter, specific heat performed in Refs. [9-13] where *pure* dimerization has been assumed for the ordered phase of the XY model (furthermore, in Refs. [14-19] the Heisenberg model becomes, within the framework of certain approximations, equivalent to the XY model; within this restricted context the present justification holds also for the Heisenberg model).

Let us now describe our argument. The magnetic contribution to the Hamiltonian of the $\frac{1}{2}$ - spin XY cyclic linear chain (with unitary crystalline parameter) is given by

$$\mathcal{H}_m = - \sum_{j=1}^N \left\{ J_{2j-1} \left(S_{2j-1}^x S_{2j}^x + S_{2j-1}^y S_{2j}^y \right) + J_{2j} \left(S_{2j}^x S_{2j+1}^x + S_{2j}^y S_{2j+1}^y \right) \right\} \quad (1)$$

where, for future convenience, we have artificially separated the interactions into odd and even ones. Through the Jordan-Wigner transformation^[20] this Hamiltonian can be expressed as follows

$$\mathcal{H}_m = - \frac{1}{2} \sum_{j=1}^N \left\{ J_{2j-1} \left(a_{2j-1}^+ a_{2j} + a_{2j}^+ a_{2j-1} \right) + J_{2j} \left(a_{2j}^+ a_{2j+1} + a_{2j+1}^+ a_{2j} \right) \right\} \quad (2)$$

where the fermionic creation and annihilation operators have been introduced. By using next the Fourier transformed quantities

$$\begin{aligned} b_k &= \frac{1}{\sqrt{N}} \sum_{j=1}^N e^{i(2j-1)k} a_{2j-1} \\ \bar{b}_k &= \frac{1}{\sqrt{N}} \sum_{j=1}^N e^{i 2j k} a_{2j} \end{aligned} \quad (3)$$

$$J_q = \frac{1}{N} \sum_{j=1}^N e^{i(2j-1)q} J_{2j-1}$$

$$\bar{J}_q = \frac{1}{N} \sum_{j=1}^N e^{i 2j q} J_{2j}$$

where $-\pi/2 < k, q \leq \pi/2$, we obtain

$$\begin{aligned} \mathcal{H}_m &= - \frac{1}{2} \sum_q \left\{ J_q \sum_k \left[e^{-i(k-q)} b_k^+ \bar{b}_{k-q} + e^{ik} \bar{b}_k^+ b_{k-q} \right] + \right. \\ &\quad \left. + \bar{J}_q \sum_k \left[e^{-i(k-q)} \bar{b}_k^+ b_{k-q} + e^{ik} b_k^+ \bar{b}_{k-q} \right] \right\} \end{aligned} \quad (4)$$

This expression can conveniently be separated into two terms, namely

$$\mathcal{H}_m = \mathcal{H}_0 + V \quad (4')$$

where \mathcal{H}_0 is the $q=0$ contribution (related to *pure* dimerization) and V the $q \neq 0$ one. Let us finally introduce new quantities through the transformations

$$\begin{aligned} b_k &= \frac{1}{\sqrt{2}} (\alpha_k + \beta_k) \\ \bar{b}_k &= \frac{1}{\sqrt{2}} (\alpha_k - \beta_k) e^{i Q_k} \end{aligned} \quad (5)$$

where

$$\theta_k \equiv \text{arctg} (\eta_0 \text{tg } k) \quad (6)$$

$$\eta_0 \equiv \left| \frac{J_0 - \bar{J}_0}{J_0 + \bar{J}_0} \right| \quad (7)$$

The Hamiltonian (4') will be now given by

$$\mathcal{H}_0 = \sum_k J \left(\epsilon_k^\alpha \alpha_k^+ \alpha_k + \epsilon_k^\beta \beta_k^+ \beta_k \right) \quad (8)$$

and

$$V = \sum_{q \neq 0} \sum_k \left[\Lambda_{kq} \left(\alpha_k^+ \alpha_{k-q} - \beta_k^+ \beta_{k-q} \right) + \Lambda'_{kq} \left(\alpha_k^+ \beta_{k-q} - \beta_k^+ \alpha_{k-q} \right) \right] \quad (9)$$

where

$$J \equiv \frac{|J_0 + \bar{J}_0|}{2} \quad (10)$$

$$- \epsilon_k^\alpha = \epsilon_k^\beta \equiv \epsilon_k \equiv (\cos^2 k + \eta_0^2 \sin^2 k)^{1/2} \quad (11)$$

$$\Lambda_{kq} \equiv \frac{1}{4} \left\{ J_q \left[e^{-i(k-q - \theta_{k-q})} + e^{i(k-\theta_k)} \right] + \bar{J}_q \left[e^{-i(k-q + \theta_k)} + e^{i(k + \theta_{k-q})} \right] \right\} \quad (12)$$

$$\Lambda'_{kq} \equiv \frac{1}{4} \left\{ J_q \left[e^{-i(k-q - \theta_{k-q})} - e^{i(k-\theta_k)} \right] + \bar{J}_q \left[e^{i(k + \theta_{k-q})} - e^{-i(k-q + \theta_k)} \right] \right\} \quad (13)$$

We intend next to calculate the magnetic contribution F_m to the free energy of the system. To perform this we shall work within the temperature dependent Green functions framework^[21], by treating V as a perturbation to \mathcal{H}_0 (we discuss later the implications of this treatment). We obtain, at a given temperature T ,

$$F_m = F_0 + F_1 + F_2 + \dots \quad (14)$$

where F_0 is the magnetic free energy associated to \mathcal{H}_0 and is given by

$$F_0 = -2k_B T \left[N \ln 2 + \sum_k \ln \text{ch} \frac{\epsilon_k}{2k_B T} \right] ; \quad (15)$$

F_1 vanishes because of the quasi-fermions (spinless magnetic excitations associated to the α 's and β 's operators) linear momentum conservation; F_2 is given by

$$F_2 = \sum_{q \neq 0} \sum_k \left\{ |\Lambda_{kq}|^2 G(k,q) + |\Lambda'_{kq}|^2 G'(k,q) \right\} \quad (16)$$

where

$$G(k,q) = \frac{k_B T}{2} \sum_{\omega_n} \left[g_\alpha(k, \omega_n) g_\alpha(k-q, \omega_n) + g_\beta(k-q, \omega_n) g_\beta(k, \omega_n) \right] \quad (17)$$

$$G'(k,q) = \frac{k_B T}{2} \sum_{\omega_n} \left[g_\alpha(k, \omega_n) g_\beta(k-q, \omega_n) + g_\beta(k, \omega_n) g_\alpha(k-q, \omega_n) \right] \quad (18)$$

where

$$g_\alpha(k, \omega_n) = (i \omega_n - J \epsilon_k^\alpha)^{-1} \quad (19)$$

$$g_\beta(k, \omega_n) = (i \omega_n - J \epsilon_k^\beta)^{-1} \quad (20)$$

and $\omega_n = k_B T \pi(2n+1)$ with $n=0, \pm 1, \pm 2, \dots$

The two terms of expression (17) (expression (18)) are diagrammatically represented in Fig. 1.a (Fig. 1.b). Through standard complex plane integration we obtain

$$G(k,q) = -\frac{1}{2J} \frac{\text{th} \frac{\epsilon_k + \frac{q}{2}}{2t} - \text{th} \frac{\epsilon_{k-\frac{q}{2}}}{2t}}{\epsilon_k + \frac{q}{2} - \epsilon_{k-\frac{q}{2}}} \quad (17')$$

and

$$G'(k,q) = -\frac{1}{2J} \frac{\text{th} \frac{\epsilon_k + \frac{q}{2}}{2t} + \text{th} \frac{\epsilon_{k-\frac{q}{2}}}{2t}}{\epsilon_k + \frac{q}{2} + \epsilon_{k-\frac{q}{2}}} \quad (18')$$

where

$$t \equiv k_B T / J \quad (21)$$

We are prepared now to introduce the elastic contribution F_e to the free energy of the system. Contrarily to the magnetic con-

tribution, this one is going to be treated only approximatively , namely within the adiabatic approximation (see Refs. [22,23]) ; by doing this we roughly take into account the *crystalline* three-dimensionality of the real system; i.e. the role played by structural fluctuations is reduced into a minor one. Furthermore, we neglect the anharmonic elastic contributions (we discuss later the small error herein introduced). We have therefore that

$$\begin{aligned}
 F_e &= \sum_{j=1}^{2N} \frac{C}{2} (x_{j+1} - x_j)^2 \\
 &= NC \sum_q \left\{ |x_q|^2 + |\bar{x}_q|^2 - \cos q \left(x_q \bar{x}_q^* + \bar{x}_q x_q^* \right) \right\}
 \end{aligned} \tag{22}$$

where C is the harmonic elastic constant, x_j is the *mean* position of the j-th spin (with respect to its position in the uniform phase) and

$$\begin{aligned}
 x_q &= \frac{1}{N} \sum_{j=1}^N e^{-i 2jq} x_{2j} \\
 \bar{x}_q &= \frac{1}{N} \sum_{j=1}^N e^{-i(2j-1)q} x_{2j-1}
 \end{aligned} \tag{23}$$

Let us now go back to the magnetic contribution. We recall that the Hamiltonian (1) includes interactions only between first-neighbouring spins, characterized by an exchange integral noted $J(u)$, where u denotes their incremental distance with respect to the reference one in the uniform phase. We assume

$$J(u) = J(0) + J'(0)u \tag{24}$$

The truncation of the series associated to $J(u)$ introduces errors comparable to those coming from the non inclusion of anharmonicity in the elastic contribution. It follows from (24) that

$$\begin{aligned}
 J_{2j} &= J(0) + J'(0) [x_{2j+1} - x_{2j}] \\
 J_{2j-1} &= J(0) + J'(0) [x_{2j} - x_{2j-1}]
 \end{aligned} \tag{25}$$

and, by using the two last expressions of (3), we obtain

$$\begin{aligned}
 J_q &= J(0) \delta_{q,0} + J'(0) \left[\bar{x}_q e^{-iq} - x_q \right] \\
 \bar{J}_q &= J(0) \delta_{q,0} + J'(0) \left[x_q e^{-iq} - \bar{x}_q \right]
 \end{aligned}
 \tag{26}$$

The use of relations (26) into definitions (7) and (10) leads to

$$\eta_0 = \left| \frac{J'(0)}{J(0)} \left(\bar{x}_0 - x_0 \right) \right|
 \tag{7'}$$

and

$$J = |J(0)|
 \tag{10'}$$

If we replace now expressions (26) into (12) and (13) and those into (16) we obtain F_2 which added to F_0 (given by relation (15)) and to F_e (given by relation (22)) finally leads to the following diagonalized expression:

$$f = f_0 + \frac{1}{2} \sum_{q \neq 0} \left(\omega_q^2 \eta_q^2 + \omega'_q{}^2 \eta'_q{}^2 \right) + \frac{1}{2} K \eta_0^2
 \tag{27}$$

where we have introduced reduced free energies through

$$\begin{aligned}
 f &\equiv F/NJ \\
 f_0 &\equiv F_0/NJ
 \end{aligned}
 \tag{28}$$

and also

$$\begin{aligned}
 \omega_q &= m_q + |n_q| \\
 \omega'_q &= m_q - |n_q| \\
 m_q &\equiv K + \frac{1}{2\pi} \int_0^{\pi/2} dk \left\{ G(k,q) \left[\cos^2(k - \theta_{k,q}) + \cos^2(k + \theta_{k,q}) \right] \right. \\
 &\quad \left. - 2 \cos q \cos(k - \theta_{k,q}) \cos(k + \theta_{k,q}) \right\} \\
 &\quad + G'(k,q) \left[\sin^2(k - \theta_{k,q}) + \sin^2(k + \theta_{k,q}) \right. \\
 &\quad \left. + 2 \cos q \sin(k - \theta_{k,q}) \sin(k + \theta_{k,q}) \right] \left. \right\}
 \end{aligned}
 \tag{30}$$

$$\begin{aligned} n_q \equiv & K \cos q + \frac{1}{2\pi} \int_0^{\pi/2} dk \left\{ G(k, q) \left[e^{-iq} \cos^2(k - \theta_{k, q}) \right. \right. \\ & \left. \left. + e^{iq} \cos^2(k + \theta_{k, q}) - 2 \cos(k - \theta_{k, q}) \cos(k + \theta_{k, q}) \right] \right. \\ & \left. + G'(k, q) \left[e^{-iq} \sin^2(k - \theta_{k, q}) + e^{iq} \sin^2(k + \theta_{k, q}) \right. \right. \\ & \left. \left. + 2 \sin(k - \theta_{k, q}) \sin(k + \theta_{k, q}) \right] \right\} \end{aligned}$$

where

$$K \equiv CJ(0) / [J'(0)]^2 \quad (31)$$

$$\theta_{k, q} \equiv \frac{1}{2} \left(\theta_{k, +\frac{q}{2}} + \theta_{k, -\frac{q}{2}} \right) \quad (32)$$

and where $G(k, q)$, $G'(k, q)$ and the θ 's are respectively given by relations (17'), (18') and (6). Notice also that in expressions (30) we used the quasi-continuum limit ($N \rightarrow \infty$). Furthermore, we have that

$$\eta_q = \left| \frac{J'(0)}{J(0)} \right| \left| x_q + \frac{n_q}{|n_q|} \bar{x}_q \right| \quad (33)$$

$$\eta'_q = \left| \frac{J'(0)}{J(0)} \right| \left| x_q - \frac{n_q}{|n_q|} \bar{x}_q \right|$$

In the particular case of a *pure* dimerization ($x_{2j-1} = -x_{2j} \equiv \eta > 0 \forall j$) we have

$$\bar{x}_0 = x_0 = \eta \quad (34)$$

hence

$$\eta_0 = \left| \frac{2 J'(0)}{J(0)} \right| \eta \quad (35)$$

and

$$\eta'_0 = 0 \quad (36)$$

Let us stress that ω_q and ω'_q satisfy $\omega_q \geq \omega'_q \forall q$ and respectively correspond to optic and acoustic structural branches (in particular $q \rightarrow 0$ implies $\omega'_q \propto q$). Furthermore ω_q equals ω'_q for $q = \pm \pi/2$

as long as η_0 vanishes. As it is already known^[9-12], at a critical reduced temperature t_c and coming from above, the dimerized structure appears through a second order phase transition. Coherently with this fact, we expect ω_0 to vanish at t_c ; clearly the critical frontier in the t - K space (see Fig. 2) is given by $\omega_0(t_c, K; \eta_0=0)=0$, which leads, through use of the first of relations (29), to

$$K = \frac{1}{\pi} \int_0^{\pi/2} dk \frac{\sin^2 k}{\cos k} \operatorname{th} \frac{\cos k}{2 t_c} \quad (37)$$

whose asymptotic behaviours are^[10]

$$t_c \sim \begin{cases} 1/8K & \text{if } K \rightarrow 0 \\ e^{-\pi K} & \text{if } K \rightarrow \infty \end{cases} \quad (38)$$

Let us now focalize the dimerization order parameter η_0 . Within the assumption (proved afterwards) that the ordered phase is a *pure* dimerization (and consequently $\eta_q = \eta'_q = 0 \quad \forall q \neq 0$) the free energy given by relation (27) reduces to $f_0 + \frac{1}{2} K \eta_0^2$; therefore the equilibrium value of η_0 is given by

$$\left. \frac{\partial f_0}{\partial \eta_0^2} \right|_{t, K} + \frac{K}{2} = 0 \quad (39)$$

which, through (15) and (28), leads to

$$K = \frac{1}{\pi} \int_0^{\pi/2} dk \frac{\sin^2 k}{\sqrt{\cos^2 k + \eta_0^2 \sin^2 k}} \operatorname{th} \frac{\sqrt{\cos^2 k + \eta_0^2 \sin^2 k}}{2t} \quad (39')$$

Two typical cuts of the surface $\eta_0(t, K)$ are presented in Fig. 3.

Let us now discuss the central point of the present work: what happens with ω_q and ω'_q below t_c , once we have replaced there in the equilibrium value of η_0 (given by Eq. (39))? The answer is presented, for a typical value of K , in Fig. 4 (any other value of K leads to qualitatively the same results). We observe that, below t_c and going towards $t=0$, the whole spectrum ω_q and ω'_q monotonically increases. Therefore it becomes evident that no other thermodynamical structural instability appears other than the one already taken into account: the ordered phase is a *pure* dimerization. The thermal behaviour of ω_0 and of $v \equiv \left. \frac{d\omega'_q}{dq} \right|_{q=0}$ (propor-

tional to the sound velocity) are represented, for a typical value of K , in Fig. 5 (ω_0 and v respectively saturate at the values $\sqrt{2K}$ and $\sqrt{K/2}$ in the limit $t \rightarrow \infty$, as a consequence of the disappearance of the magnetic contribution). At this point, let us go back to the expansion (14), where we neglected terms of the fourth (or higher) order in η_q and η'_q (F_3 vanishes). This is justified only if no structural modes (for any non vanishing value of q) suddenly freeze down, i.e., no new phase transition (necessarily of the first order) occurs besides the one already taken into account. This conjecture is strongly supported by the monotonic increase of the spectrum (for decreasing temperature below t_c) already mentioned.

Let us conclude by saying that the higher order terms we have neglected in both expansions (22) and (24) have no influence at all for $t \geq t_c$, and bring only small quantitative modification for $t < t_c$. For example the inclusion of a fourth order anharmonic elastic constant in the expansion (22), provokes a depression of the order parameter in the region of low temperatures (the smaller the temperature the higher the depression).

One of us (C.T.) acknowledges useful remarks from M.E. Fisher, R.B.Stinchcombe and J.Villain; the other one (R.A.T.L.) was partially supported by a fellowship from CNPq/Brazil.

References

1. Bray J.W., Hart H.R.Jr., Interrante L.V., Jacobs I.S., Kasper J.S., Watkins G.D., Wee S.H. and Bonner J.C., Phys. Rev. Lett. 35, 744 (1975).
2. Smith L.S., Ehrenfreund E., Heeger A.J., Interrante L.V., Bray J.W., Hart H.R.Jr. and Jacobs I.S., Solid State Commun. 19, 377 (1976).
3. Bray J.W., Interrante L.V., Jacobs I.S., Bloch D., Moncton D.E., Shirane G., Bonner J.C., Phys. Rev. B20, 2067 (1979).
4. Huizinga S., Kommandeur J., Sawatzky G.A., Thole B.T., Kopinga K., Jonge W.J.M. de and Roos J., Phys. Rev. B19, 4723 (1979).
5. Mook H.A. et al., Bull. Amer. Phys. Soc. 24, 507 (1979).
6. Bloch D., Voiron J., Bonner J.C., Bray J.W., Jacobs I.S. and Interrante L.V., Phys. Rev. Lett. 44, 294 (1980).
7. Jacobs I.S., Bray J.W., Hart H.R.Jr., Interrante L.V., Kasper J.S., Bloch D., Voiron J., Bonner J.C., Moncton D.E. and Shirane G., J. of Magnetism and Magnetic Mat. 15, 332 (1980).
8. Pincus P., Solid State Commun. 22, 1971 (1971).
9. Beni G. and Pincus P., J. Chem. Phys. 57, 3531, (1972).
10. Dubois J.Y. and Carton J.P., J. Phys. 35, 371 (1973).
11. Tsallis C., "Contribution à l'étude théorique des transitions de phase magnétiques et structurales", Doctoral Thesis, Université Paris-Orsay (1974).
12. Tsallis C. and de Sèze L., Ferroelectrics 14, 661 (1976).
13. Tannous C. and Caillé A., Can. J. Phys. 57, 508 (1979).
14. Beni G., J. Chem. Phys. 58, 3200 (1973).
15. Pytte E., Phys. Rev. B 10, 4637 (1974).
16. Bray J.W., Solid State Commun. 26, 771 (1978).
17. Bulaevskii L.N., Buzdin A.I. and Klomskii D.I., Solid State Commun. 27, 5 (1978).

18. Cross M.C. and Fisher D.S., Phys. Rev. B 19, 402 (1979).
19. Takaoka Y. and Motizuki K., J. Phys. Soc. Jap. 47, 1752 (1979).
20. Jordan P. and Wigner E., Z. Phys. 47, 631 (1928).
21. Abrikosov A.A., Gorkov L.P. and Dzialoshinski I.E., "Methods of Quantum Field Theory in Statistical Physics", Dover Publication, Inc, p. 120.
22. Domb C., J. Chem. Phys. 25, 783 (1956).
23. Salinas S.R., J. Phys. C 7, 241 (1974).

CAPTION FOR FIGURES

- Fig. 1 Feynman's diagrams associated to the 2nd order magnetic contribution to the free energy; the full (dashed) lines correspond to α -(β -) propagators. (a) terms of expression (17); (b) terms of expression (18).
- Fig. 2 Reduced critical temperature as a function of the inverse reduced harmonic elastic constant (U and D respectively denote the uniform and dimerized phases).
- Fig. 3 Two typical cuts of the surface which represents the dimerization order parameter as a function of the reduced temperature and harmonic elastic constant: a) for $K=0.4$; b) for $t=0$.
- Fig. 4 Thermal behaviour of the spectrum which characterizes the thermodynamical structural instabilities ($K=0.4$). (a) $t > t_c$; (b) $t = t_c$; (c) $t < t_c$; (d) $t \ll t_c$.
- Fig. 5 Thermal dependence of the $q=0$ optical square reduced frequency (a) and the reduced sound velocity (b) for $K=0.4$.

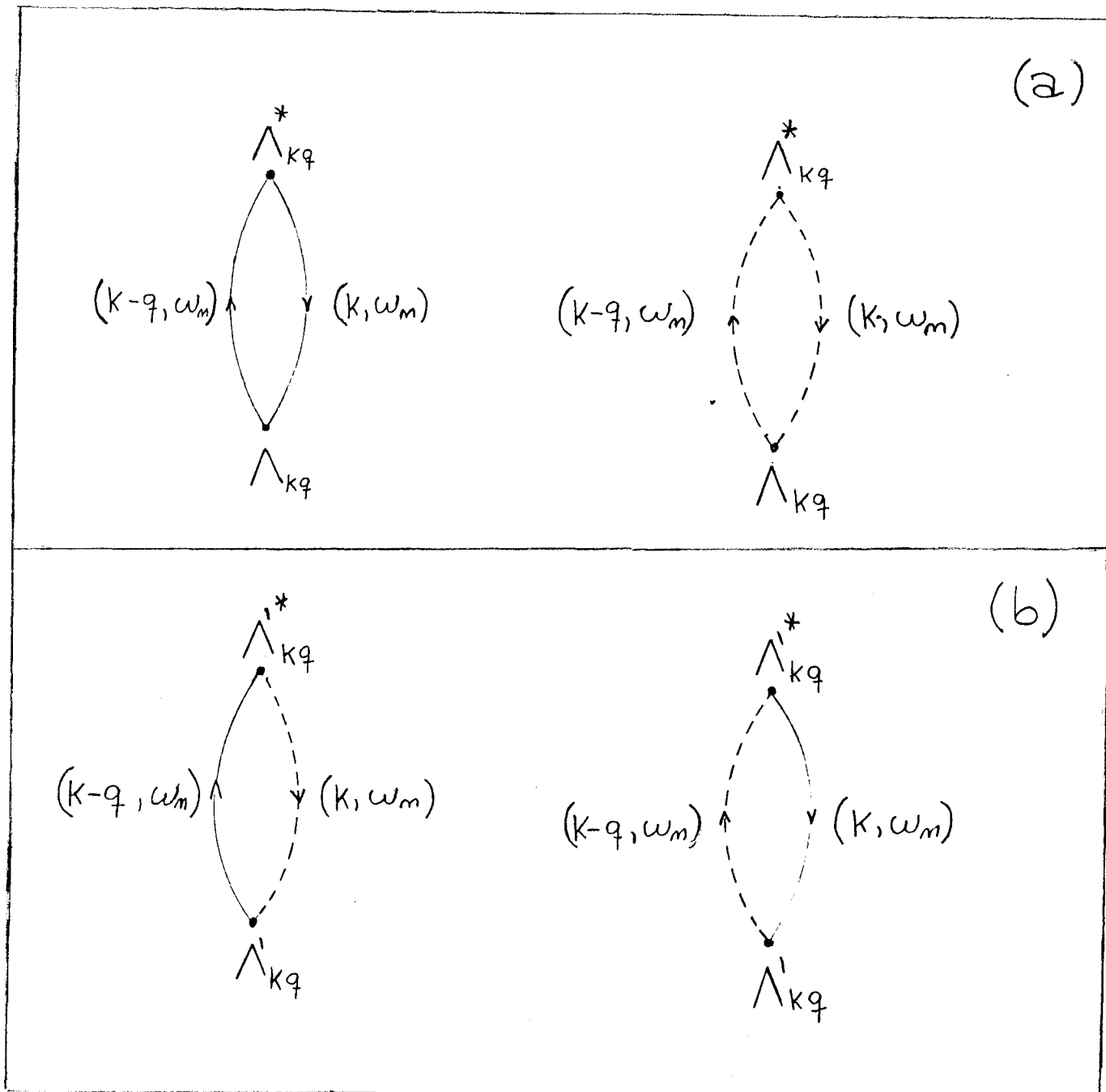


FIG. 1

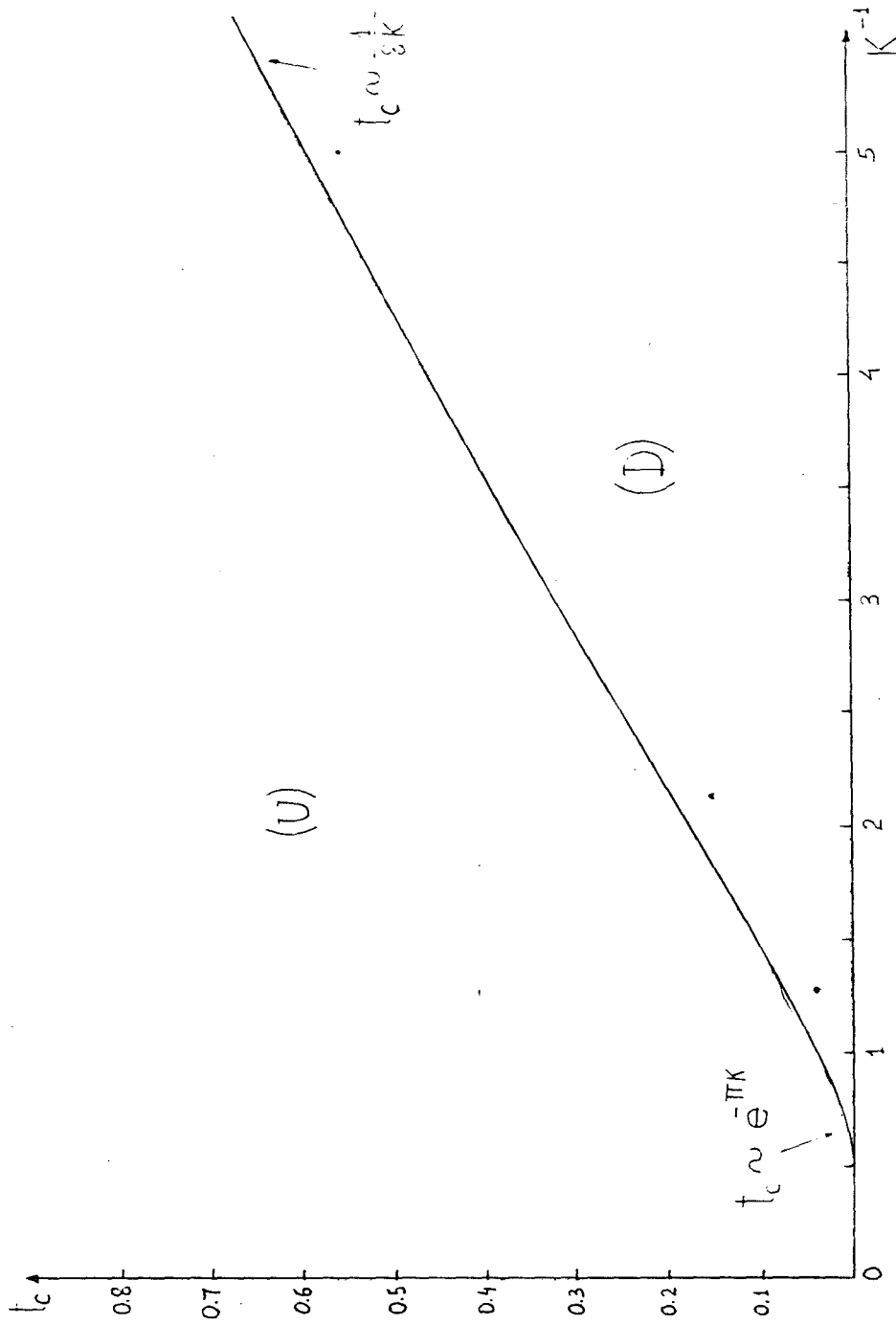


FIG. 2

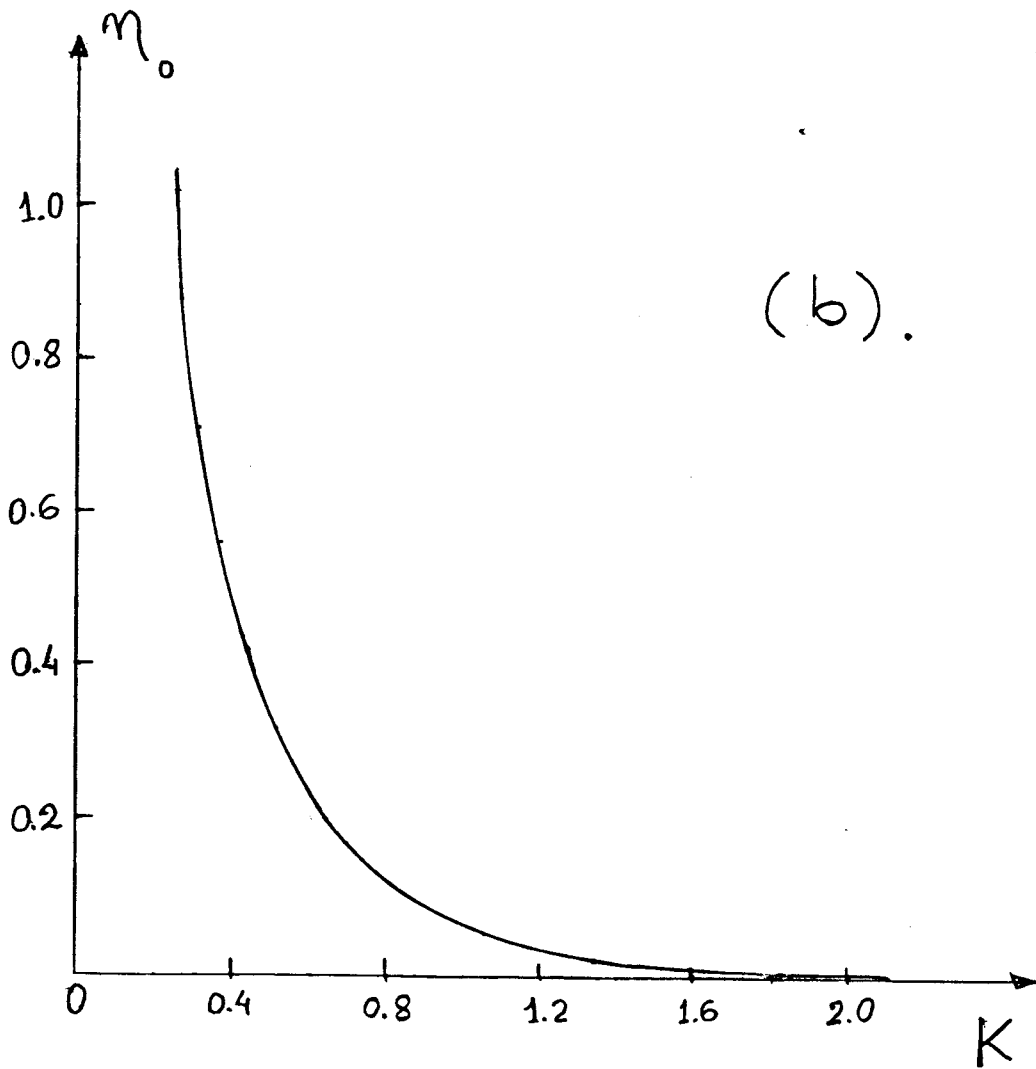
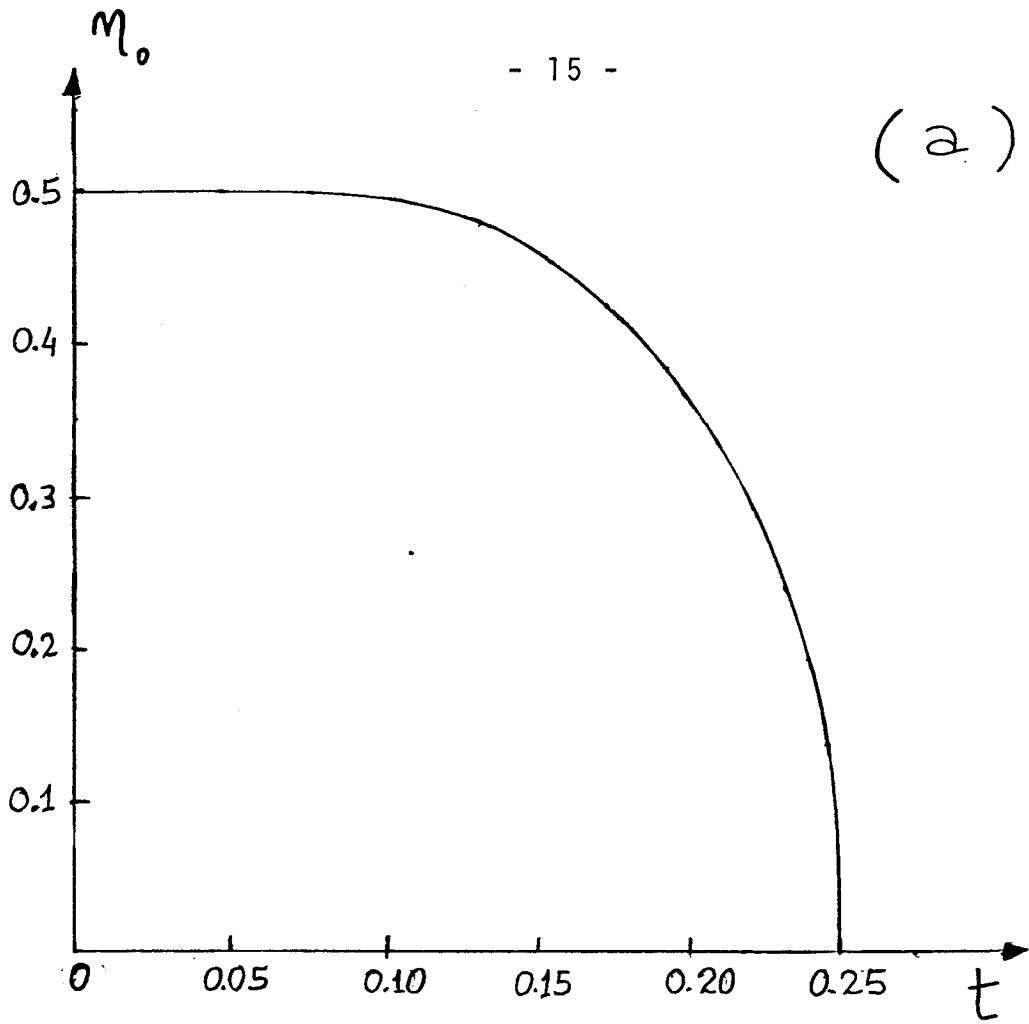


FIG. 3

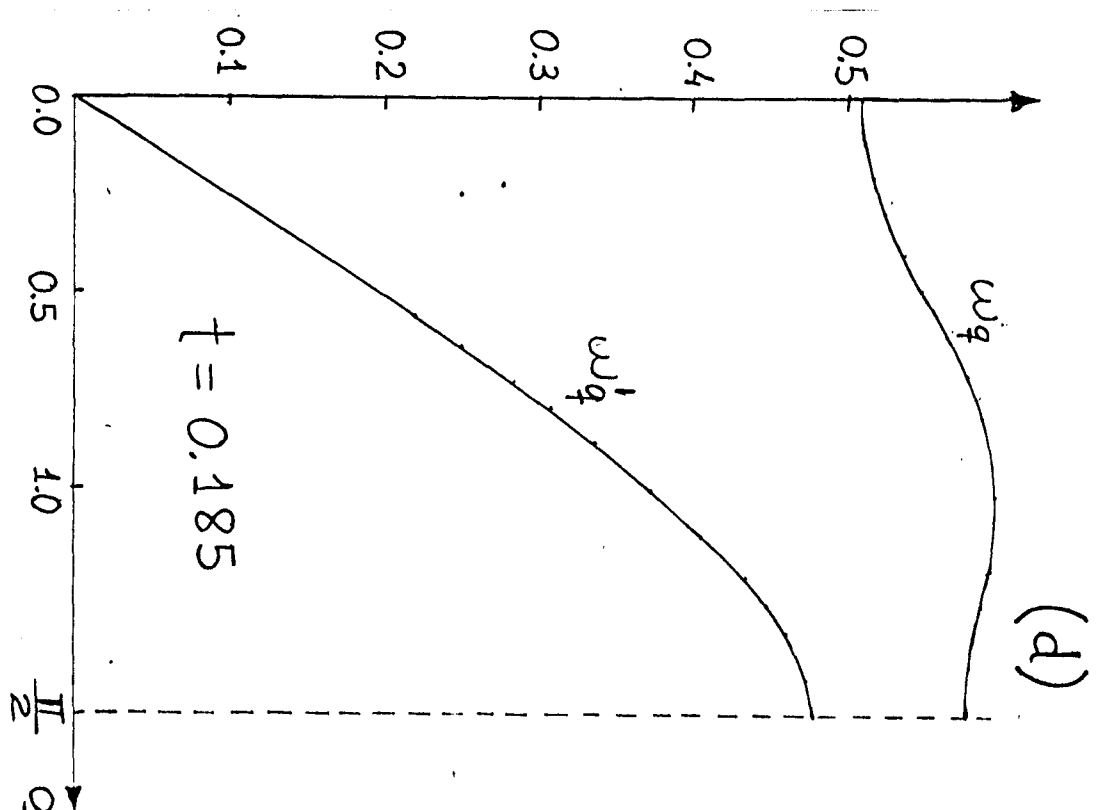
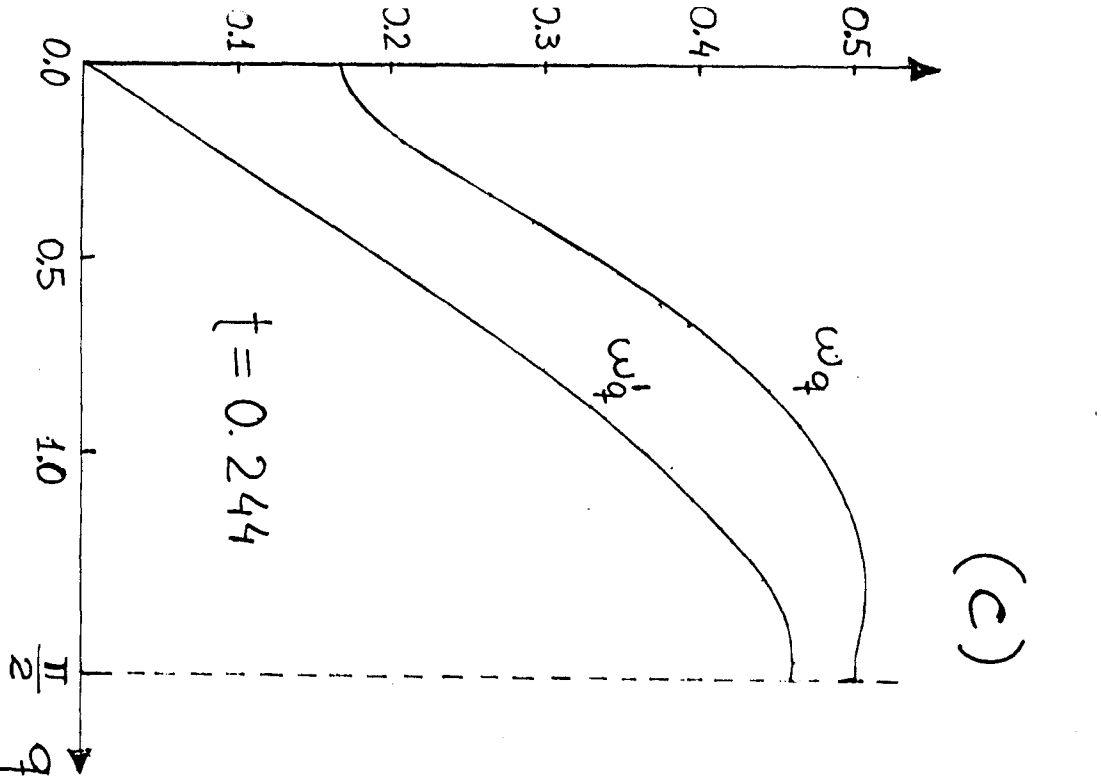
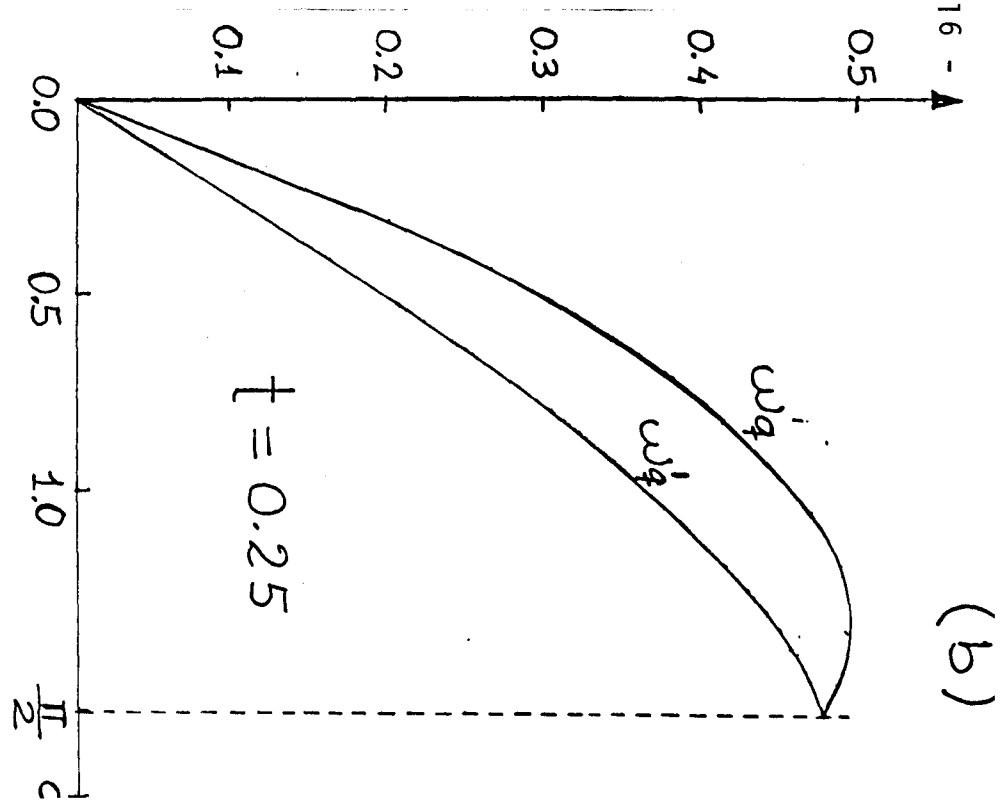
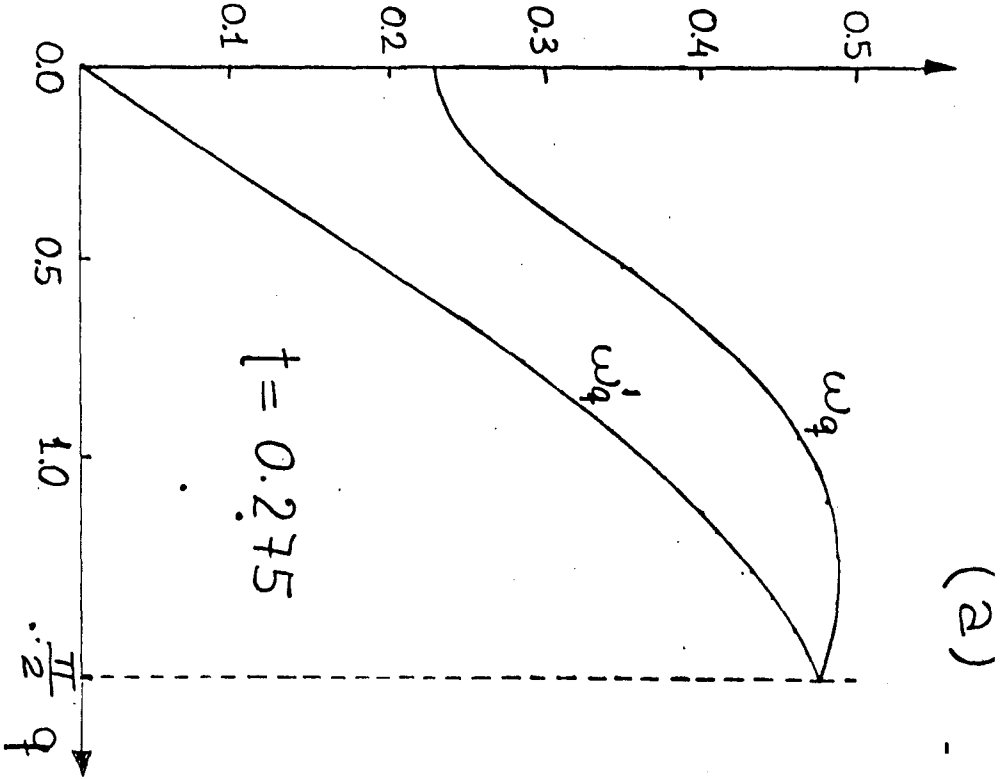


FIG. 4

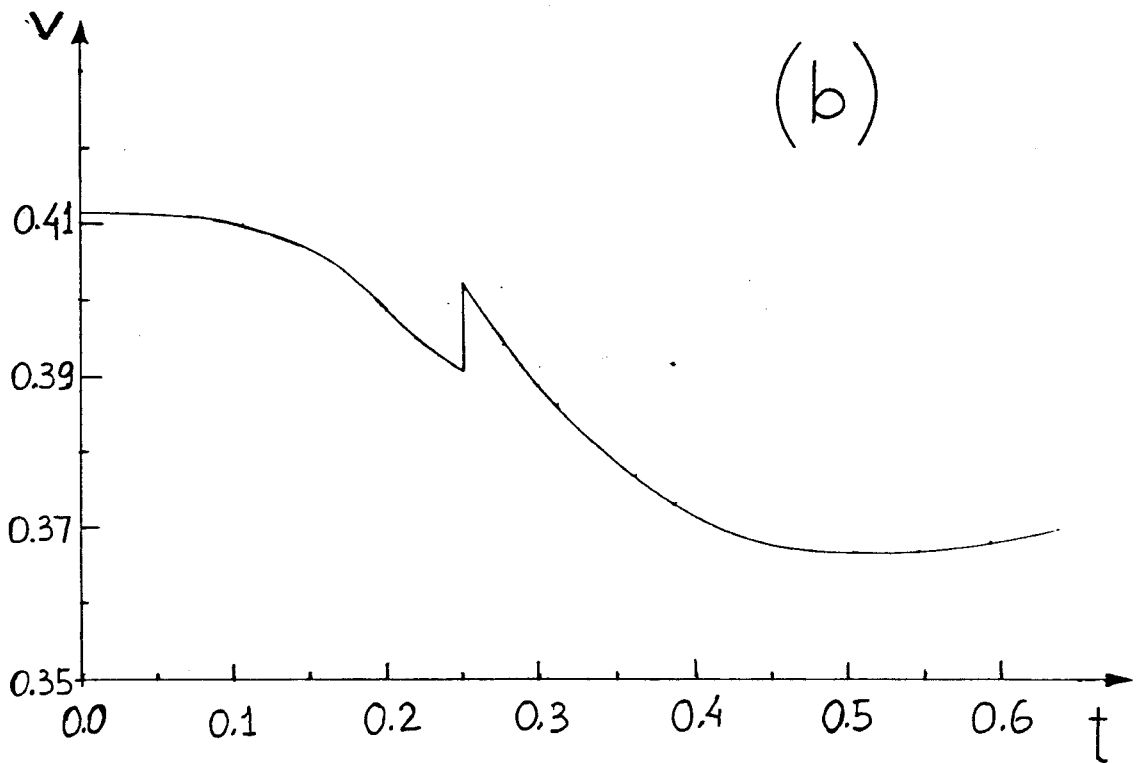
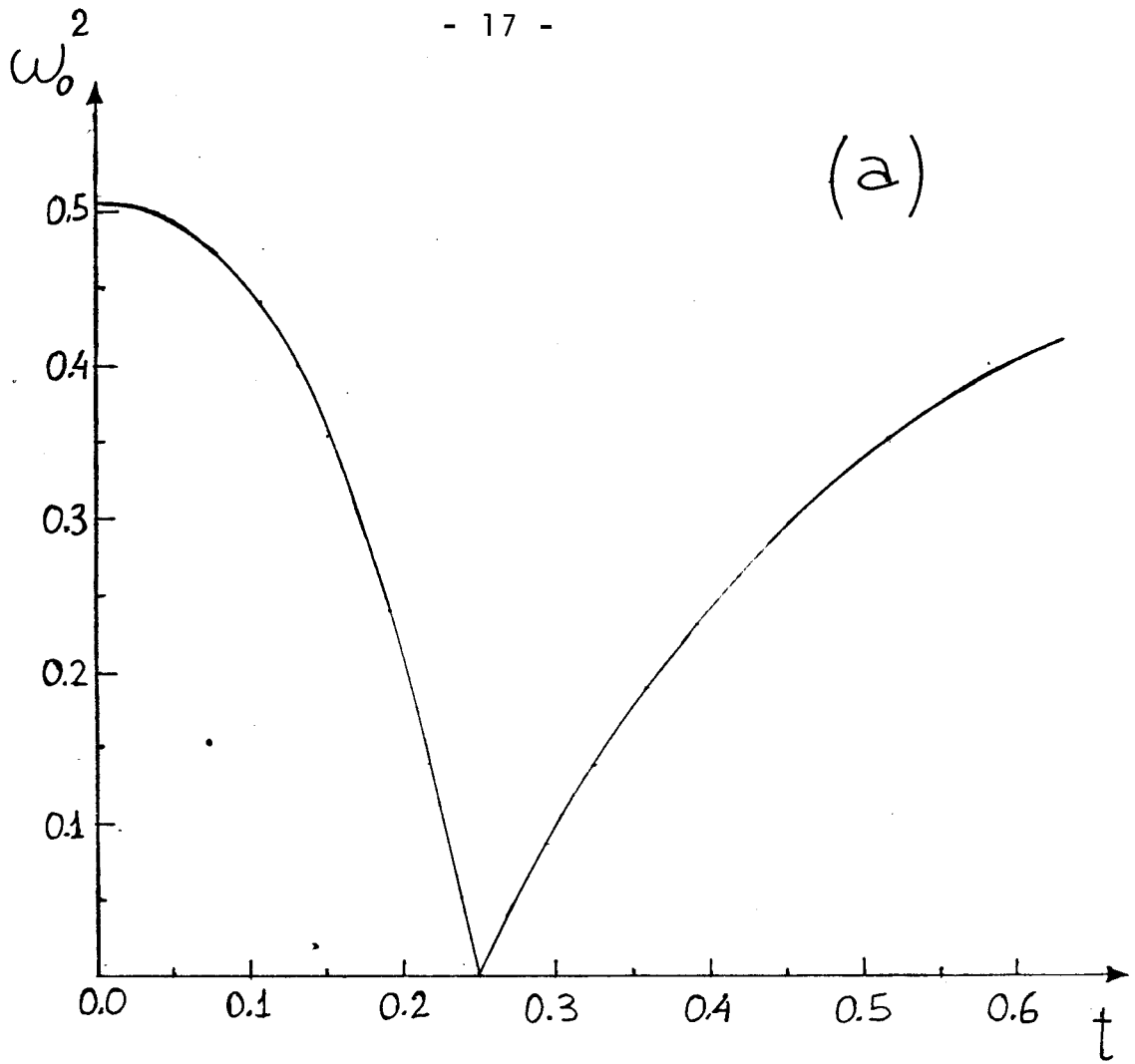


FIG. 5