Local Excitation as Transport Phase Transition in Phonon Systems

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The Peierls-Boltzmann transport equation for phonons, which was re-formulated and modified in a previous paper, is extended to be applicable to arbitrary interactions and phonon processes. As a rule, it turns out that only two types of steady state solutions are possible: hysteresis and threshold. These two solutions reveal the possibility of "transport phase transitions", i. e. a transition from the "thermodynamic" branch to a "nonthermodynamic" one via a cumulative excitation. It is shown that both the threshold and the hysteresis situation exhibit pronounced analogies to phase transitions in thermal equilibrium. The dependence of the steady states from the relevant parameters is discussed.

1. Introduction

Phase transitions in open systems have gained increasing interest in the last years. It turned out that phase transition-like phenomena are not restricted to systems in thermal equilibrium. Furthermore, there are many nonequilibrium systems, not only in physics but also in chemistry, biology, engineering and related topics, where one can find a close similarity in the overall behaviour of the relevant variables.

A common feature of these systems is the fact that they are open for energy and/or matter influx. Thus they can be driven far away from thermal equilibrium and under certain conditions new nonequilibrium states may be stabilized (dissipative structures). Nonlinearities in the evolution equations are responsible for the possible existence of such new states. By gradually increasing the strength of the external stimulation one may carry an open system through a whole sequence of nonequilibrium steady states. In this region, the so-called "domain far from thermal equilibrium", instabilities manifest themselves in a splitting into branches of different behaviour (e. g. stable – unstable, oscillating – nonoscillating, etc.). The respective transition regions behave in many ways analogous to those for equilibrium phase transitions. For details we refer to the excellent review articles of Haken [1, 2], where also the relevant literature is cited.

Recently, the author and M. Wagner have published an example for such a phase transition-like behaviour in an open system [3], henceforth refered to as I. There we have applied a generalized Peierls-Boltzmann equation for phonons in its fully nonlinear form to a simple model system. The aim of the present study lies in the extension of this formalism to arbitrary interactions and phonon processes.

Our starting point is a brief review of the formalism. We discuss in some detail the steady state behaviour and the inherent two types of transport phase transitions: a continuous and a discontinuous one. As a rule it turns out that these two types are the only possible ones. They exhibit the same behaviour as that known from first and second order phase transitions. Furthermore the dependence of the detailed structure of the steady state solutions as a function of some of the relevant parameters is discussed and the usefullness of Thom's catastrophe theory is demonstrated.

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2. Structure of the Modified Peierls-Boltzmann Formalism

The starting point in I was a bilinear phonon density operator $\hat{n}(k\lambda, r, t)$ (Hardy operator) and a Hamilton operator consisting of two parts, a harmonic one, H_0, for the phonons and a second one, H_I, which describes interactions between phonons and between phonons and local modes respectively. A series of conventional transport postulates applied to the commutator $[H_0, \hat{n}]$ in the Heisenberg equation of motion led to the well known linear transport term. The commutator $[H_I, \hat{n}]$ represents the collision term. Further simplifications of the transport and the collision terms, among which we specifically note a spatial averaging procedure, had to be applied. We then were left with the general Boltzmann equation [vid. Eq. (I.29)]

$$\partial_t \langle \hat{N}_k \rangle - \Phi(k,t) = \sum_{m=0}^{\infty} m \left(\sum_{\mu,\nu}^{(1)} - \sum_{\mu,\nu}^{(2)} \right) \varrho_\nu(t)\, \partial_t W_{\mu\nu}(t) \tag{1}$$

with

$$\hat{N}_k \equiv \hat{N}(k\lambda, t) = a^+(k\lambda, t)\, a(k\lambda, t) \tag{2}$$

where a^+ and a are phonon creation and annihilation operators.

The space-independent flux $\Phi(k,t)$ is given by

$$\Phi(k,t) = -V[v(k\lambda)\, \nabla_r \hat{n}(k\lambda, r, t)]_{\mathrm{av}\cdot r}. \tag{3}$$

The r.h.s. of Eq. (1) represents the collision term. $\varrho_\nu(t)$ is the diagonal part of the density matrix and $W_{\mu\nu}(t)$ is the transition probability from state $|\nu>$ to state $|\mu>$:

$$W_{\mu\nu}(t) = -[L_s \exp\{-i(L_0 + Q L_s)t\} L_s]_{\mu\mu\nu\nu} \tag{4}$$

$L_s = [H_{I'}]$ and $L_0 = [H_{0'}]$ are the tetradic Liouville operators and $P = 1 - Q$ is the diagonalization projector (for details vid. I).

In order to arrive at Eq. (1), a kind of Markofficity assumption in the Zwanzig Master equation for $\varrho_\mu(t)$ is already incorporated as well as the assumption of vanishing initial correlations [i. e. $Q\,\varrho(t=0) = 0$] or the random phase approximation.

As a next step in our calculations we had to expand the transition probabilities $W_{\mu\nu}(t)$. Restricting ourselves to "irreducible transitions" and a specific time scale (vid. I) we were left with the Golden Rules

$$\begin{aligned}\partial_t W_{\mu\nu}(t) &= 2\pi\lambda^2 V_{\mu\nu}\, \delta(E_\mu - E_\nu) \\ &+ 2\pi\lambda^4 \Big| \sum_\alpha V_{\mu\alpha} V_{\alpha\nu} (E_\alpha - E_\mu)^{-1} \Big|^2 \delta(E_\mu - E_\nu) \\ &+ 2\pi\lambda^6 \Big| \sum_{\alpha,\beta} V_{\mu\alpha} V_{\alpha\beta} V_{\beta\nu} (E_\alpha - E_\mu)^{-1} \\ &\cdot (E_\beta - E_\nu)(E_\beta - E_\alpha)^{-1} \Big|^2 \delta(E_\mu - E_\nu) + \ldots \end{aligned} \tag{5}$$

with $$(H_I)_{\alpha\beta} \equiv \lambda V_{\alpha\beta}.$$

The application of our general Boltzmann equation (1) to arbitrary phonon transporting systems requires further approximations. We choose the same model as in our previous studies: a linear chain, representing the phonon transporting system S_1, and a nontransporting system S_2, e. g. a single additional degree of freedom with excitation energy ω_s.

For the transporting system S_1 we get from Eq. (1) the kinetic equation

$$\partial_t \langle \hat{N}_k \rangle - \Phi(k,t) = -\frac{\langle \hat{N}_k \rangle - \langle \hat{N}_k \rangle_0}{\tau_k} + \sum_{m=0}^{\infty} m \left(\sum_{\mu,\nu}^{(1)} - \sum_{\mu,\nu}^{(2)} \right) \varrho_\nu(t)\, \partial_t W_{\mu\nu}(t) \tag{6}$$

and for the local excitation

$$\partial_t \langle \hat{N}_s \rangle = -\frac{\langle \hat{N}_s \rangle - \langle \hat{N}_s \rangle_0}{\tau_s} + \sum_{m=0}^{\infty} m \left(\sum_{\mu,\nu}^{(1)} - \sum_{\mu,\nu}^{(2)} \right) \varrho_\nu(t)\, \partial_t W_{\mu\nu}(t) \tag{7}$$

with $W_{\mu\nu}(t)$ given by Eq. (5),

$$\sum_{\mu,\nu}^{(1)} \rightarrow N_k^{(\mu)} = N_k^{(\nu)} + m\,; \quad \sum_{\mu,\nu}^{(2)} \rightarrow N_k^{(\mu)} = N_k^{(\nu)} - m\,.$$

The necessity to add relaxation paths to the kinetic equations was stressed in I; they appear as the first terms of the r.h.s. of Equations (6, 7).

3. Arbitrary Processes and Interactions

In contrast to our previous work we do neither restrict ourselves to a bilinear interaction Hamiltonian nor to a 3 phonon process. Irrespective of the order of pertubation theoretical expansion of the transition probability $W_{\mu\nu}(t)$ and of the degree of nonlinearity in the interaction potential H_I the kinetic equations [vid. Eqs. (I.45, 46)]

$$\partial_t \langle \hat{N}_k \rangle - \Phi(k,t) = -\frac{\langle \hat{N}_k \rangle - \langle \hat{N}_k \rangle_0}{\tau_k} + \langle F(N_s, N_k) \rangle \tag{6'}$$

$$\partial t \langle \hat{N}_s \rangle = -\frac{\langle \hat{N}_s \rangle - \langle \hat{N}_s \rangle_0}{\tau_s} - \langle \sum_k F(N_s, N_k) \rangle \tag{7'}$$

can be approximately simplified to a two variable form, whence N_k henceforth is understood to be the single characteristic phonon occupation. The restriction to two variables is for mathematical convenience only.

It should be noticed that an averaging and a factorizing procedure must be employed to separate a closed subhierarchy from the infinite hierarchy of kinetic equations [(6′) and (7′)], i. e. the nonlinear function $F(N_s, N_k)$ must be replaced by $F(\langle N_s \rangle, \langle N_k \rangle)$ in a rather unknown way. However, the thermal equilibrium $\langle \hat{N}_k \rangle_0$ and $\langle \hat{N}_s \rangle_0$ must remain a solution for vanishing phonon transport. A possible choice was given in Equation (I.50).

We introduce a new set of variables and parameters

$$\begin{aligned} \langle \hat{N}_k \rangle &\equiv N_k \rightarrow y\,, \\ \langle \hat{N}_s \rangle &\equiv N_s \rightarrow x\,, \\ \Phi(k,t) + \tau_k^{-1} \langle N_k \rangle_0 &\rightarrow \Phi\,, \\ \langle F(N_s, N_k) \rangle &\rightarrow -\beta F(x,y)\,, \\ \langle \sum_k F(N_s, N_k) \rangle &\rightarrow -\alpha F(x,y) \end{aligned} \tag{8}$$

[$\alpha, \beta > 0$; they depend on H_I and $W_{\mu\nu}(t)$].
The kinetic equations [Eqs. (6′) and (7′)] then read

$$\partial_t y - \Phi = -y/\tau_k - \beta F(x, y) , \quad (9)$$

$$\partial_t x = -x/\tau_s + \alpha F(x, y) . \quad (10)$$

It should be kept in mind that y represents the excitation of the phonon modes in the transporting system S_1, whereas x stands for the excitation of the local degree of freedom, i. e. the nontransporting system S_2. For moderately low temperatures we may assume that the thermal average $X_0 \equiv \langle \hat{N}_s \rangle_0$ is zero. This will be done in the following, although no principal change arises if we do not use this simplification.

The kinetic equations (9) and (10) have a structure which seems to be typical for most nonlinear evolution equations. They can be solved only, if one specifies the nonlinear function $F(x, y)$. The functional structure of the polynomial $F(x, y)$ on the one hand is determined by the order of pertubation theoretical expansions of the transition probability $W_{\mu\nu}(t)$ and on the other hand by the structure of the unharmonic coupling H_I of systems S_1 and S_2. We first note the existence of an energy conservation for the relevant phonon process, i. e.

$$m\,\omega_s \approx \omega_{k_1} + \omega_{k_2} + \ldots + \omega_{k_n} \quad (11)$$

referred to as a $(n-m)$-process, where n phonons combine to create m local. This process is achievable as a direct process, if a term

$$a_{k_1} a_{k_2} \ldots a_{k_n} (b_s^+)^m + \text{h.c.} \quad (12)$$

is contained in H_I. a_{k_1} and $a_{k_1}^+$ are the annihilation and creation operators for the phonon modes $\hat{N}_{k_1}$; b_s and b_s^+ represent the local modes $\hat{N}_s$. The phonon process (12) requires an expansion of the interaction potential up to high orders. Thus the weight of the direct process will be very small for $m, n > 1$. Alternatively, this process is possible also as a cummulative excitation via energy non conserving intermediate states. Then the operator combination (12) is not contained in H_I, but is built up in $W_{\mu\nu}(t)$ via Golden Rules together in combination with powers of $b_s^+ b_s$ and $a_k^+ a_k$. Instead of (12) we have

$$(a_k^+ a_k)^\nu (b_s^+ b_s)^\mu a_{k_1} \ldots a_{k_n} (b_s^+)^m + \text{h.c.} \quad (13)$$

The underlying process is denoted as $P(\mu, \nu; n, m)$, here n phonons create m local modes and vice versa. In addition ν phonons are created and again annihilated, and the same process occurs with μ local modes. The resulting nonlinear function F reads

$$F(x, y) = \langle (x+m)^{2\mu} y^{2\nu} y^n (x+1)(x+2) \ldots (x+m) - (x-m+1)^{2\mu} (y+1)^{2\nu} (y+1)^n x(x-1) \ldots (x-m+1) \rangle . \quad (14)$$

Having established $F(x, y)$, the factorization and averaging procedure remains to be done.

We briefly illustrate the described way to Eq. (14) for the example [vid. I, Eq. (41)]

$$H_I \equiv \lambda V = \lambda \sum_k \gamma(k, s) (a_k + a_{-k}^+) (b_s + b_s^+) \quad (15)$$

$$\omega_s \approx \omega_{k_1} + \omega_{k_2} + \omega_{k_3} . \quad (16)$$

In our notation this process reads P (1,0; 3,1) with the relevant operator combination

$$b_s^+ b_s a_{k_1} a_{k_2} a_{k_3} b_s^+ + \text{h.c.}$$

This results from the combination of λV from Eq. (15) and the second Higher Golden Rule which is necessary for a energy conserving process. From Eq. (14) one then deduces

$$F(x, y) = \langle (x+1)^3 y^3 - x^3 (y+1)^3 \rangle \quad (17)$$

[vid. I, Equation (43)].

4. Steady State Solutions

We want to solve Eqs. (9) and (10) for the time independent case. First we eliminate y in Eq. (10) with the relation

$$y_s = \Phi \cdot \tau_k - (\beta \tau_k / \alpha \tau_s) x_s \quad (18)$$

and we arrive at

$$F(x, y_s) - x_s/\alpha \tau_s = 0 . \quad (19)$$

The latter equation is a polynomial of order

$$r = 2\mu + 2\nu + n + m - 1 \quad (20)$$

for the steady state excitation X_s of the local degree of freedom as a function of phonon flux Φ. We have investigated about 35 different processes $P(\mu, \nu; n, m)$ and quite surprisingly we have found only two types of steady state behaviour [4]. Some of the processes are listed in Table 1. The steady state solutions are drawn in Figs. 1 and 2.

In addition, from our results we deduce for $S \equiv \beta \tau_k (\alpha \tau_s)^{-1}$ that the solutions remain unchanged in the whole region $0 \leqq S < 1$. But the case $S < 1$ is only of physical relevance since we

Table 1. Processes $P(\mu, \nu; n, m)$ and their possible realizations. H_{ij} is the interaction Hamiltonian:
$H_{ij} = \sum \Lambda (k_1, \ldots k_j; s)\,(a_{k1} + a_{k1}^+) \ldots (a_{kj} + a_{-k}^+)\,(b_s + b_s^+)$.
FGR and HGRn (i. e. Fermi and Higher Golden Rules) are the transition probabilities which are necessary to allow for the processes $P(\mu, \nu; n, m)$.

P (0.0; 2.1)	H_{21}	+FGR
P (1.0; 2.1)	$H_{11}+H_{12}$	+HGR 1
P (0.0; 3.2)	$H_{11}+H_{21}$	+HGR 1
P (1.0; 3.2)	$H_{11}+H_{12}$	+HGR 2
P (0.0; 3.1)	H_{31}	+FGR
P (1.0; 3.1)	H_{11}	+HGR 2
P (1.1; 3.1)	$H_{11}+H_{21}$	+HGR 2
P (0.0; 4.2)	H_{21}	+HGR 1
P (1.0; 4.2)	H_{22}	+HGR 1
	H_{11}	+HGR 3
	$H_{11}+H_{21}+H_{12}$	+HGR 2

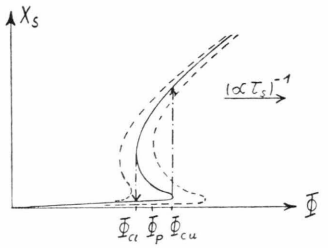

Fig. 1. Excitation of a local mode coupled to a one-dimensional phonon system for arbitrary processes $P(\mu, \nu; n, m)$ with $\tilde{r} \geqq 3$, i. e. hysteresis case, for three different values $\alpha \tau_s$. Φ_{cl} and Φ_{cu} are the critical values for decreasing and increasing flux, respectively. Φ_p is the most probable transition, x_s is the occupation deviation from thermal equilibrium.

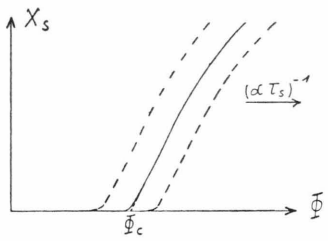

Fig. 2. Excitation of a local mode coupled to a one-dimensional phonon system for arbitrary processes $P(\mu, \nu; n, m)$ with $\tilde{r} < 3$, i. e. threshold case, for three different values $\alpha \tau_s$. Φ_c is the averaged critical phonon flux.

have to restrict ourselves to $\beta < \alpha$ and to $\tau_k < \tau_s$. $\beta < \alpha$ follows from the fact that α contains an additional phonon summation beyond that of β. The same argument holds for $\tau_k < \tau_s$, since in a transporting system the possibility of additional losses aside from the direct interaction is more probable than for a local excitation. This follows from a rather simplified estimate employing Callaway's rule.

$\tau_k \ll \tau_s$ (i. e. $\varkappa_k \gg \varkappa_s$, $\varkappa =$ damping) may be used as a starting point for an adiabatic elimination of the stable modes N_k. This procedure corresponds to a neglection of the time dependence of N_k. Then the N_k plays the role of smooth parameters (pool variables).

The above cited restriction of the parameter S leads to a rather interesting situation. Without creating relevant changes in the steady state behaviour of the nontransporting system we may neglect the function $F(x, y)$ in Equation (9). This may be achieved in the following way. Due to the coupling of the transporting system S_1 to an external stimulus phonon modes are excited. Through $\hat{N}_k - \hat{N}_s$ interaction local modes are excited. The total neglection of βF is tantamount to the assumption that the influence which the energy transition from S_1 to S_2 has onto the phonon flux Φ is vanishing. This approximation is consistent with the assumption we had to introduce to derive the simplified transport term.

Furthermore it is interesting to have a look on the behaviour for $S > 1$. In this unphysical case we have found several different solutions for Eqs. (9) and (10), e. g. a double hysteresis or an additional branch of solutions. A detailed analysis shows us that these additional steady states are either unstable or negative or that they are positive and stable but the phonon modes having negative occupations.

5. Discussion of the Results

With $S < 1$ and the neglection of βF in Eq. (9) the order of the polynomial [Eq. (19)] is reduced to

$$r \to \tilde{r} = 2\mu + m\,. \qquad (20')$$

As a rule we have found that for $\tilde{r} < 3$ only the threshold type of solutions exists, whereas in the opposite case only a hysteretic behaviour is displayed. The nonexistence of a hysteresis in the first case is plausible, since a quadratic polynomial has at most two real solutions. But for $\tilde{r} \geqq 3$ it is not obvious why there exists only the typical hysteresis as a solution up to polynomials of very high order. In a forthcoming paper [5] we will analyse this rule and we will give a mathematical prove for it.

We want to discuss now the solutions in some detail for the threshold and the hysteresis situation.

a) Threshold Behaviour

For $\Phi < \Phi_c$ the excitation of the local mode is vanishingly small and for $\Phi > \Phi_c$ the excitation

increase very strong with increasing flux. This is the typical behaviour which one knows from the laser [6] and from chemical reactions with a bimolecular step (auto- or crosscatalytic reaction) [7].

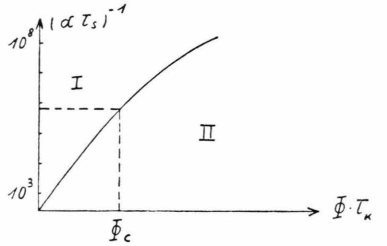

Fig. 3. Dependence of the steady state solution x_s from the parameters of the system in the threshold case. Region I is the stable unexcited solution, region II the stable and highly excited one. For $\alpha\tau_s$ given, a unique value Φ_c results.

Beyond the threshold value Φ_c the additional degree of freedom x_s is extremely excited above its thermal equilibrium as defined by the phonons in the chain. A continuous transition takes place to a state far from thermal equilibrium. This transition may be viewed – with some restrictions – as a second order phase transition, since it shows the typical behaviour of this type of transition in thermal equilibrium systems.

The threshold flux Φ_c depends on the parameters α, τ_k and τ_s. Φ_c increases with decreasing coupling strength and thus with decreasing α. High damping $\varkappa_k$ (i. e. small relaxation τ_k) requests a large flux Φ to reach the threshold. In Fig. 3 were have drawn the dependence of the threshold flux Φ_c as a function of α, τ_k and τ_s.

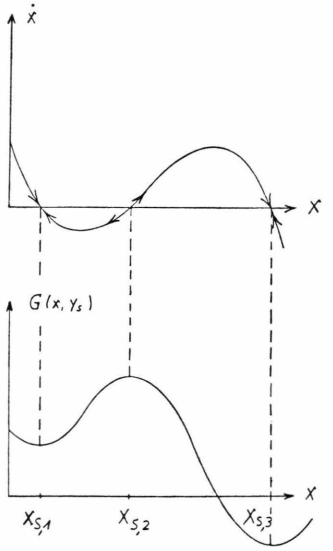

Fig. 4. Phase plane diagram ($\dot{x}$ as a function of x) and plot of the "energy-function" $G(x, y_s)$ for the hysteresis type of solution. x is the averaged occupation of the local mode, $x_{s,i}$ $(i = 1, 2, 3)$ are the corresponding steady state solutions. The arrows indicate the time behaviour of the system. $G(x=0, y_s)$ is arbitrarily chosen.

b) Hysteresis

The relevant feature of this structure of solution is the bistability. For $\Phi_{cl} < \Phi < \Phi_{cu}$ we have two stable steady state solutions. The phase transition is a discontinuous one and we have two marginal transition points. Formally we can define an "energy" or "potential" function $G(x, y_s)$ by writing

$$\partial_t x = -x/\tau_s + \alpha F(x, y_s) = -\partial_x G(x, y_s) \,. \quad (21)$$

In Fig. 4 we have drawn $\partial_t x$ and $G(x, y_s)$ as a function of x for the processes P (1.0; 3.1). In analogy to the Van der Waals gas we can formally define the coexistence of both stable steady states. In this way we can derive the most probable transition from one branch to the other (vid. Φ_p in diagram 1). A more accurate consideration would have to include fluctuations.

Through comparison with a Van der Waals gas one may assume that the hysteresis type of solutions might be viewed as a first order phase transition. There exist many examples for systems exhibiting hysteretic behaviour, e. g. chemical reactions with a trimolecular step [7] or an enzymatic inhibition [8].

The position and the width of the hysteresis (i. e. Φ_{cl}, Φ_{cu} and $\Delta = \Phi_{cu} - \Phi_{cl}$) depends remarkably on the relevant system parameters. As in the threshold case, the parameters determine the hysteresis only in the combinations $\alpha\cdot\tau_s$ and $\Phi\cdot\tau_k$. With increasing $\alpha\cdot\tau_s$, Φ_{cl} and Φ_{cu} are shifted to lower values and Δ decreases. An increasing damping $\varkappa_k$ (i. e. decreasing τ_k) corresponds to an increasing flux. Thus great losses in the transporting system must be overcompensated by increasing phonon flux in order to get an excitation of the additional degree of freedom. Figure 5 shows this behaviour for arbitrary processes $P(\mu, \nu; n, m)$. In addition one can show that increasing values for μ and m increase Δ, whereas increasing values for ν and n decrease Δ. Thus high nonlinearities with respect to x lead to very broad hysteresises and high nonlinearities with respect to y act against this tendency and can lead to extremely narrow hysteretic extensions, i. e. $\Delta \to 0$.

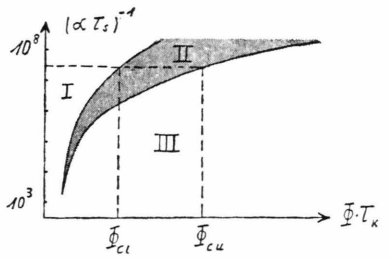

Fig. 5. Dependence of the steady state solutions x_s from the parameters of the system in the hysteresis case. In the regions I and III only one stable steady state exists. In region II one has an unstable steady state and two stable ones. For $\alpha\tau_s$ given, unique values for the critical fluxes Φ_{cl} and Φ_{cu} are determined.

If we combine Fig. 1 and Fig. 5 to a threedimensional folded surface (see Fig. 6), one has a behaviour which René Thom has called a catastrophe of cusp type [9]. For the processes P(1.0; 3.1) one has indeed a forth order "energy function", but for other processes with $\tau \geqq 3$ this is not true. However, the higher order "energy functions" look like a forth order function since there are only two minima and one maximum in the $G(x, y_s)$-x-diagram (vid. Figure 4). One can eliminate the third order term in $G(x, y)$ and then one gets

$$G(\tilde{x}, y_s) = \tilde{x}^4 + a\,\tilde{x}^2 + b\,\tilde{x} + c \qquad (22)$$

with

$$a = (\alpha\tau_s)^{-1} - (\Phi\cdot\tau_k)^3 \leqq 0\,,$$
$$b = \text{function}\,(\Phi\cdot\tau_k) < 0\,.$$

$\tilde{X}$ depends linearly on x. The control variable a is not independent of the control variable b. Therefore the shaded region A B C in Fig. 6 is not the same as that in Figure 5. In the latter one should replace $(\alpha\tau_s)^{-1}$ by $(\alpha\tau_s)^{-1} - (\Phi\cdot\tau_k)^3$. Diagram 6 represents one of the seven elementary catastrophes. As already noted, the hysteresis corresponds to a cusp type of catastrophe, i. e. one of codimension two, which long time ago has been known as a Riemann-Hugoniot catastrophe. The elementary catastrophes have the important property of structural stability. This means that the geometry is not changed by small pertubations of the "related energy function". Structural stability is not connected with our notation of stability of the steady states. It turns out that the lower and the upper branch between the two is absolutely unstable. In [5] we will show that this situation remains true for all processes $P(\mu, \nu; n, m)$.

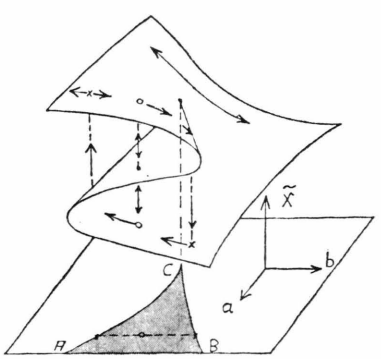

Fig. 6. Geometrical interpretation of the hysteresis type of solutions. The local excitation x is the behaviour variable, a and b are the control variables. The folded surface represents the steady state solutions x_s, in the shaded region A B C three steady states exist.

6. Summary and Outlook

We have extended the results of our previous work to include arbitrary phonon processes. Again we conceive the local excitation as an incoherent cumulative phenomenon, but we have neither restricted ourselves to a process where 3 phonons combine in succession to create the high-energetic local mode nor to bilinear coupling between the transporting and the nontransporting system.

The most important fact is the existence of only two types of steady state behaviour, a hysteresis type of solution and a threshold one. This rule was supported by about 35 different processes $P(\mu, \nu; n, m)$, indicating that these two solutions are the only relevant ones for our generalized Peierls-Boltzmann equations for phonons when applied to the model system of I, i. e. a phonon transporting system coupled to a nontransporting degree of freedom. The threshold solution corresponds to a continuous transition from the nonexcited to the excited state and might be viewed as a second order phase transition. The hysteresis type of solution exhibits discontinuous transitions from one stable state to the other one. It displays a behaviour which one knows from first order phase transitions in systems which are in thermal equilibrium.

Both types of steady state solutions show the possibility to stabilize by flux a singular degree of freedom far away from thermal equilibrium. Thus, a local energy storage in molecular chains seems to be possible [10]. Also, the order of magnitude for the phonon flux near the transition points (critical region) seems to be not too high for realistic values of the chain parameters.

In a forthcoming paper [5] we will show that the threshold and hysteresis types of solutions are the only possible ones. In addition, we will demonstrate that this behaviour remains valid for different averaging and factorization procedures in Equations (9) and (10). Furthermore, the stability conditions will be studied in more detail and without the restriction to a certain process $P(\mu, \nu; n, m)$. We will also prove that oscillating solutions (limit cycles) do only exist if we take into account a certain feedback mechanism between the phonon flux Φ and the local excitation.

So far these calculations have little to do with real physical phonon transporting systems. But there is hope that in the near future one may find phonon transporting systems that might be considered as one dimensional with respect to phonon propagation. In addition an extension of the above method to three dimensions seems possible and our results should hold also in three dimensions.

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