

Analytical approximations for the thermodynamics of a planar array of Ginzburg-Landau chains with near neighbor and next near neighbor interactions

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The free energy and the specific heat of a planar arrays of atomic chains, with on-site quartic anharmonicities and inter-chain elastic near neighbor (*nn*) and next near neighbor (*nnn*) interaction, are obtained. A mean-field approximation for the on-site anharmonicities and the Hsue-Chern approach to the quartic oscillators are used. An Ising-type behavior of the specific heat is obtained, both for *nn* and *nnn* cases. The physical relevance of this result, and a careful analysis of other approaches used in literature, are also presented.

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1. Introduction

The low dimensional systems, some of them just tutorial examples in textbooks of solid state physics, attract more and more interest, both for theoretical and practical reasons [1]. The Luttinger model [2,3], considered initially mainly as an academic exactly solvable problem, became an essential tool for the study of quasi-one-dimensional (q1D) conductors, obtained only few years later [4,5]. Application of many body techniques [6-9], of bosonisation and of the renormalization group theory [10] to the Luttinger model, as well as the proposal of many other one-dimensional (1D) models [11] or generalizations of Luttinger's one [12] by several authors, made of this branch of theoretical physics an essential tool for understanding the physical properties of new materials and compounds, like phase transitions [13-15], electrical and optical properties [16-18], etc.

Another important step forward in the physics of low dimensional systems is the development of nanosciences. The dependence of the physical behavior on the size and dimension of the system, the possibility of technological applications at nanoscale are of utmost theoretical and practical interest. The chains of atoms [19-21], of spins [22], of classical or quantum anharmonic oscillators, the polymeric chains [23] are important subjects for nanoelectronics, spintronics, quantum phase transitions, molecular electronics, etc.

However, in spite of intense theoretical investigations, even quite simple systems like chains of anharmonic oscillators, cannot be described exactly [24-25]. In fact, after important theoretical results obtained in the study of low dimensional systems in the '70s and early '80s, the last decades have been dominated by numerical results, mainly Monte Carlo simulations. Although such approaches are essential for the understanding of real systems, the lack -

or scarcity - of analytic results is frustrating. In order to limit this drawback, the effort of several authors was directed in obtaining analytic approximations or even exact results for relevant low dimensional models [26].

In this paper we shall obtain the thermodynamics of the planar array of chains of anharmonic oscillators, using the transfer matrix approach [27-29]. Within this approach, the free energy of a N -dimensional classical system is obtained by evaluating the ground state energy of its dual system - a $N-1$ -dimensional quantum system. In our case, the two systems to be studied are the planar array of chains of classical anharmonic oscillators (a 2D classical system), and the chain of anharmonic quantum oscillators (a 1D quantum system). The ground state energy of the chain of anharmonic oscillators will be obtained with a Green function method.

We shall consider the 2D system as being composed by chains of classical anharmonic oscillators, with quartic anharmonicity (the so-called 2-4 anharmonic oscillator); the on-chain interaction is considerably larger than the inter-chain one, so each chain conserves its individuality. All the chains are disposed in the same plane. We shall treat the chain in the continuum approximation, so the order parameter will be a continuous function $\psi_j(x)$, where j is the chain index. The inter-chain interaction is considered to be elastic; both near neighbor (*nn*) and next near neighbor (*nnn*) inter-chain interactions are taking into account.

The dual system is a chain of anharmonic quartic (2-4) oscillators, interacting through *nn* and *nnn* elastic forces. As it is well known, the quantum quartic oscillator cannot be solved analytically; as much the more, the statistical problem of N such oscillators defies exact solution. As the inter-site interaction seems more important, for the thermodynamic behaviour, than the on-

site one, we shall treat the inter-site interaction exactly and the in-site one - in a mean-field approximation. In this way, we can take advantage of a simple method of evaluation of the ground state energy of bilinear Hamiltonians, which takes into account not only the nn interaction, but also the interaction between arbitrary distant neighbors. However, as beyond the nnn case, the mathematical difficulties are almost insurmountable, we shall restrict ourselves to the nnn case. Finally, the ground state energy of the quantum chain will be identified with the free energy of the array of classical chains.

The main result of this paper is the critical behavior of the planar system, for which a logarithmic singularity of the specific heat is obtained. This is the fingerprint of an Ising-like behavior, and confirms the results obtained for such systems, using other approaches, more or less similar. The logarithmic character of this singularity appears in the both situations: when only the nn interaction is taken into account, as well as when both of them are considered. This fact confirms one of the basic assumptions of the renormalization group approach - that the range of the interaction (if it remains finite) does not affect its critical behavior [30].

2. The planar array of chains and its dual system

The main goal of this paper is to investigate the critical behavior of a planar array of anharmonic chains. Each chain has the length L and is composed by anharmonic oscillators, interacting with their neighbors through elastic forces. We shall treat each chain in the continuum approximation, so the order parameter will be a continuous function $\psi_j(x)$, where j is the chain index, $1 \leq j \leq M$; cyclic boundary conditions are imposed, so $\psi_j(x) = \psi_{j+M}(x)$. a_s, b, c are the Landau-Ginzburg parameters; their physical meaning is discussed in many references, see for instance [31]. The inter-chain interaction extends to the next and to the near next neighbor and is described by the coefficients c_1, c_2 . In the continuum approximation, this distinction cannot be done, and the interaction between oscillators is melt into the constant c . If l is the distance between oscillators along the chain, the condition:

$$cl \ll c_1, c_2 \quad (1)$$

expresses the fact that the inter-chain interaction is much smaller than the intra-chain one, assuring the individuality of each chain.

So, the free energy functional describing the planar array of classical quartic oscillators is:

$$F_{cl}[\psi] = \sum_{j=1}^M \int_0^L dx \left(a_s \psi_j^2 + b \psi_j^4 + c \left(\frac{d\psi_j}{dx} \right)^2 + c_1 (\psi_{j+1} - \psi_j)^2 + c_2 (\psi_{j+2} - \psi_j)^2 \right) \quad (2)$$

It can be obtained from [32], taking $\xi_0=1$. The dual of the planar array of classical oscillators [26-29] is a chain of quantum oscillators:

$$H = \sum_{j=1}^M \left(\frac{-1}{4\beta^2 c} \frac{d^2}{d\psi_j^2} + a_s \psi_j^2 + b \psi_j^4 + c_1 (\psi_{j+1} - \psi_j)^2 + c_2 (\psi_{j+2} - \psi_j)^2 \right) \quad (3)$$

where periodic boundary conditions are imposed again. According to the transfer matrix theory [27], the free energy of the classical 2D system (per unit length, i.e. divided to the length of a chain)

$$f = \frac{F}{L} = k_B T E_0 \quad (4)$$

is proportional to the ground state energy of the quantum 1D system.

The Hamiltonian (3) can be split in an on-site and an interaction contribution:

$$H = \sum_{j=1}^M (H_{j,s} + H_{j,im}) \\ = \sum_{j=1}^M \left(\frac{-1}{4\beta^2 c} \frac{d^2}{d\psi_j^2} + (a_s + 2c_1 + 2c_2) \psi_j^2 + b \psi_j^4 \right) - 2 \sum_{j=1}^M (c_1 \psi_{j+1} \psi_j + c_2 \psi_{j+2} \psi_j) \quad (5)$$

We shall firstly study the on-site Hamiltonian:

$$H_s = \frac{-1}{4\beta^2 c} \frac{d^2}{d\psi^2} + (a_s + 2c_1 + 2c_2) \psi^2 + b \psi^4 \quad (6)$$

which can be obtained from [32], taking $\xi_0=1$; let us put:

$$a = a_s + 2c_1 + 2c_2 \quad (7)$$

where a_s corresponds to a in [31c]; it is characterizing an one-particle oscillator, so it is an on-site quantity. We shall consider that a_s has a Landau-type temperature dependence (see [26-32]):

$$a_s = a' \left(\frac{T}{T_{MF}} - 1 \right). \quad (8)$$

Even if $a_s < 0$ (we shall consider, in the spirit of Landau theory, that a_s is quite close to 0), a could become positive, due to the contribution of the inter-chain interaction, so (6) can be written as:

$$H_s = \frac{-1}{4\beta^2 c} \frac{d^2}{d\psi^2} + a\psi^2 + b\psi^4, \quad (9)$$

$$a = a' \left(\frac{T}{T_{MF}} - 1 \right) + 2c_1 + 2c_2$$

As already mentioned, the Hamiltonian (9), for $a > 0$, cannot be solved exactly; the case $a < 0$, corresponding to a two well potential, is even more difficult and challenging; consequently, even more analytical approximations are available in literature. One of the most elegant approaches of (9), applicable both for negative and positive coefficient of the quadratic term, is the so-called two-step approach, proposed by Hsue and Chern method and generalized by several authors [31]. In this regime, when $|a|$ is not too large, the Hsue-Chern gives qualitatively correct results. We shall take advantage of this fact later on, while calculating various quantities of physical interest connected to the Hamiltonian (9).

According to the transfer matrix theory, the free energy (2) can be obtained by calculating the ground state energy of (3). This calculation cannot be done exactly; however, an exact result is available for bilinear Hamiltonians [33]. We shall avoid the difficulties inherent to the anharmonicity applying a mean-field approach and obtain an approximate expression for the ground state energy of (3). The Hsue-Chern method [31] will be used in order to get analytic results.

3. Mean-field approach to the quartic oscillator

We shall treat the anharmonic term of the Hamiltonian

$$H_s = \frac{-1}{2m^*} \frac{d^2}{d\psi^2} + a\psi^2 + b\psi^4 \quad (10)$$

where:

$$\frac{1}{m^*} = \frac{1}{2\beta^2 c} \quad (11)$$

in the mean-field approximation. Let us define:

$$\Delta\psi^2 = \psi^2 - \langle \psi^2 \rangle, \quad \psi^2 = \Delta\psi^2 + \langle \psi^2 \rangle, \quad (12)$$

with $\langle \psi^2 \rangle$ - the quantum average taken with the Hamiltonian (9). Replacing:

$$\psi^4 = (\Delta\psi^2 + \langle \psi^2 \rangle)^2 = (\Delta\psi^2)^2 + 2\Delta\psi^2 \langle \psi^2 \rangle + \langle \psi^2 \rangle^2 \approx 2\Delta\psi^2 \langle \psi^2 \rangle + \langle \psi^2 \rangle^2 \quad (13)$$

in (9), it becomes:

$$H_s \approx \frac{-1}{2m^*} \frac{d^2}{d\psi^2} + (a + 2b\langle \psi^2 \rangle)\psi^2 + b\langle \psi^2 \rangle^2 \quad (14)$$

This theory is meaningful if:

$$a + 2b\langle \psi^2 \rangle > 0. \quad (15)$$

If this condition is not fulfilled, the system is unstable.

The evaluation of $\langle \psi^2 \rangle$ cannot be done exactly, as - already mentioned - the quartic oscillator is not exactly solvable. There are however several analytic approximations which could be used [30]; the most precise refer to one well potentials, or to deep two-wells ones. In our case, when the wells are quite swallow, the Hsue-Chern approach seems to be the most suited.

Using [31c], Eq. (53),

$$\langle \psi^2 \rangle = \frac{1}{4\beta\sqrt{|a|c}} \frac{1}{\omega} \quad (16)$$

where ω is a root of the cubic equation:

$$\omega^3 - \sigma_a \omega - 6\lambda = 0, \quad \sigma_a = \text{sgn}(a), \quad \lambda = \frac{b}{4\beta c^{1/2} |a|^{3/2}}. \quad (17)$$

The precise identification of this root, out of the three ones of (17), is discussed in [31c]. Its explicit form is:

$$\omega = \left(3\lambda + \sqrt{-\frac{\sigma_a}{27} + 9\lambda^2} \right)^{1/3} + \left(3\lambda - \sqrt{-\frac{\sigma_a}{27} + 9\lambda^2} \right)^{1/3}. \quad (18)$$

It is easy to notice that the free term of (17), λ , is proportional to $(m^*)^{-1/2}$:

$$\frac{1}{\sqrt{m^*}} = \frac{1}{\sqrt{2}\beta c^{1/2}}. \quad (19)$$

In the context of the Hsue-Chern approach, the stability condition (15) can be put in a more transparent form. Presuming that $|a|$ is small, or, equivalently, λ is large, we have the following series expansion for $1/\omega$ [31c], Eq. (65):

$$\frac{1}{\omega} = (6\lambda)^{-1/3} \left(1 - \sigma_a (6\lambda)^{-2/3} + \dots \right). \quad (20)$$

Consequently:

$$\langle \psi^2 \rangle = \frac{1}{4\beta\sqrt{|a|c}} (6\lambda)^{-1/3} \left(1 - \sigma_a (6\lambda)^{-2/3} + \dots \right). \quad (21)$$

The stability condition means:

$$a + 2b \langle \psi^2 \rangle = a + 2b \frac{1}{4\beta \sqrt{|a|c}} (6\lambda)^{-1/3} (1 - \sigma_a (6\lambda)^{-2/3} + \dots) > 0. \quad (22)$$

For λ large, we can neglect the higher powers of $(6\lambda)^{-1/3}$:

$$a + 2b \frac{1}{4\beta \sqrt{|a|c}} (6\lambda)^{-1/3} > 0, \quad (23)$$

or:

$$a + 2b \langle \psi^2 \rangle = a + \frac{1}{2} \left(\frac{2}{3} \right)^{1/3} \beta^{-2/3} b^{2/3} c^{-1/3} + \dots > 0. \quad (24)$$

This condition is always fulfilled for $a > 0$, and for $a < 0$,

$$|a| < \left(\frac{b^2 k_B T^2}{12c} \right)^{1/3}. \quad (25)$$

It looks not very restrictive, requesting that the product between anharmonicity and temperature, bT , should not be too large. It seems convenient for the study of phase transitions, as it does not limit too much the temperature range. As, for $a < 0$,

$$|a| = a' \left(-\frac{T}{T_{MF}} + 1 \right) - 2c_1 - 2c_2 = \frac{a'}{T_{MF}} (T_{MF} - T) - 2c_1 - 2c_2 \quad (26)$$

the stability condition can be also written as:

$$\frac{a'}{T_{MF}} (T_{MF} - T) - 2c_1 - 2c_2 < \left(\frac{b^2 k_B T^2}{12c} \right)^{1/3}, \quad (27)$$

which can be considered as defining exactly the range of T . The stability condition can be also put in a more rigorous form, using in (24) the exact form of ω given by (18), but for a quite modest benefit, we should get a much more complicated expression.

Presuming that the stability condition is fulfilled, let us put:

$$\frac{1}{2} m^* \Omega_0^2 = a + 2b \langle \psi^2 \rangle \quad (28)$$

and

$$H_s^{MF} = \frac{-1}{2m^*} \frac{d^2}{d\psi^2} + \frac{1}{2} m^* \Omega_0^2 \psi^2 - b \langle \psi^2 \rangle^2. \quad (29)$$

To treat the anharmonicity of (9) in the mean-field approximation, using the Hsue-Chern model for the calculation of $\langle \psi^2 \rangle$, means to put:

$$H_s \approx H_s^{MF}. \quad (30)$$

Replacing in the exact Hamiltonian of the chain of quantum oscillators the on-site term with its mean-field counterpart

$$H_{s,j}^{MF} = \frac{-1}{2m^*} \frac{d^2}{d\psi_j^2} + \frac{1}{2} m^* \Omega_0^2 \psi_j^2 - b \langle \psi^2 \rangle^2 \quad (31)$$

and taking into account that $\langle \psi^2 \rangle$ is site-independent, we get:

$$\begin{aligned} H^{MF} &= \sum_{j=1}^M (H_{j,s}^{MF} + H_{j,int}) \\ &= \sum_{j=1}^M \left(\frac{-1}{2m^*} \frac{d^2}{d\psi_j^2} + \frac{1}{2} m^* \Omega_0^2 \psi_j^2 \right) + \frac{m^*}{2} \sum_{i \neq j} D_{ij} \psi_i \psi_j - M b \langle \psi^2 \rangle^2, \end{aligned} \quad (32)$$

where we have put:

$$D_{ij} = -\frac{4c_1}{m^*} (\delta_{i-j,1} + \delta_{i-j,-1}) - \frac{4c_2}{m^*} (\delta_{i-j,2} + \delta_{i-j,-2}). \quad (33)$$

The Fourier transform of D_{ij} coefficients is:

$$D_k = \sum_j D_{ij} \exp(-ik(R_i - R_j)) = -\frac{8c_1}{m^*} \cos(ka) - \frac{8c_2}{m^*} \cos(2ka). \quad (34)$$

Now, the Hamiltonian of the quantum chain is put in a form suitable for the calculation of its ground state, which is proportional to the free energy of the classical 2D system. In this way, the thermodynamics of the planar array of chains can be obtained.

4. The thermodynamics of the planar array of classical anharmonic oscillators

The formula to be used for the evaluation of the ground state energy of the Hamiltonian (32) is (1.7.2) of [33]:

$$E_G = \frac{\hbar}{2} \sum_{k=1}^M \sqrt{\Omega_0^2 + D_k} - M b \langle \psi^2 \rangle^2. \quad (35)$$

Considering that the length of the quantum chain is L_q

$$\sum_k \dots = \frac{L_q}{2\pi} \int dk \dots \quad (36)$$

so the sum turns into an integral:

$$E_G = \frac{\hbar L_q}{2} \int dk \sqrt{\Omega_0^2 + D_k} - M b \langle \psi^2 \rangle^2 = \frac{\hbar a_q M}{4\pi} \int dk \sqrt{\Omega_0^2 + D_k} - M b \langle \psi^2 \rangle^2. \quad (37)$$

Essentially, we have to evaluate the integral:

$$\begin{aligned} \int dk \sqrt{\Omega_0^2 + D_k} &= \int dk \sqrt{\Omega_0^2 - \frac{8c_1}{m^*} \cos(ka) - \frac{8c_2}{m^*} \cos(2ka)} \\ &= \Omega_0 \int dk \sqrt{1 - \frac{8c_1}{m^* \Omega_0^2} \cos(ka) - \frac{8c_2}{m^* \Omega_0^2} \cos(2ka)} \end{aligned} \quad (38)$$

which is equivalent to

$$I(A, B) = \int dx \sqrt{1 - A \cos(x) - B \cos(2x)}, \quad A = \frac{8c_1}{m^* \Omega_0^2}, \quad B = \frac{8c_2}{m^* \Omega_0^2}. \quad (39)$$

If $c_2 = 0$, i.e. when the mn interaction is neglected, it becomes the complete integral of second kind:

$$I(A, B) \Big|_{B=0} = \int_0^\pi \sqrt{1 - A \cos(x)} = \frac{2}{\sqrt{1+A}} E(k_1), \quad (40)$$

with

$$k_1^2 = \frac{2A}{1+A}, \quad (41)$$

which coincides with a result obtained by several authors, using different methods. However, it is not an exact result, as claimed sometimes [31], as it has been obtained dealing with the on-site anharmonicity in the mean-field approximation, and only with the interaction - exactly.

The second derivative of the free energy of the 2D system, which is in fact, according to (4), (40), proportional to the second derivative of $E(k_1)$, has a logarithmic singularity if:

$$k_1^2 = 1 \text{ or } A = 1. \quad (42)$$

According to (39), it is equivalent with:

$$m^* \Omega_0^2 = 2a + 4b \langle \psi^2 \rangle = 2a + \left(\frac{2}{3}\right)^{1/3} \beta^{-2/3} b^{2/3} c^{-1/3} = 8c_1. \quad (43)$$

This is an equation which allows the determination of the critical temperature.

The logarithmic singularity is the fingerprint of an Ising-type phase transition; this result is not surprising, as, in the limit of very deep wells, the anharmonic oscillator has an Ising-type behaviour, in the sense that the atom can

occupy only two positions, the "left" or the "right" well. In this limit, the partition sums of the two systems become identical [34]. Also, the fact that a bilinear Hamiltonian might have an Ising-type critical behaviour has been demonstrated almost half a century ago. In their seminal paper on the 2D Ising model as a soluble problem of many fermions, Schultz, Mattis and Lieb notice that "it becomes apparent that the 2D Ising model, rather than being entirely different from the trivially soluble many-body problems, reduces in some ways to one of them, being just the diagonalization of a quadratic form" [35].

If the nnn interaction is also taken into account, i.e. $c_2 \neq 0$ or, equivalently, $B \neq 0$, the integral $I(A, B)$ can be calculated exactly [32]; even if its expression is more complicated, the critical behaviour is equally given by the logarithmic divergence of the elliptic integral $E(k)$. Again, this is not an exact result, as claimed in [31], as only the interaction, and not the on-site anharmonicity, is treated exactly. It is interesting to note that, taking into account, besides the nn interaction, the nnn one, the singularity of the phase transition does not change its character. This confirms one of the postulates of the renormalization group theory, namely the fact that the range of the interaction (as long as it remains finite) is not significant for the critical behaviour [30]. It is also interesting to note that, even an approximate treatment of the on-site interaction, can conduct to the correct critical behavior.

5. Conclusions

An analytical approximation for the thermodynamics of a 2D solid, consisting in a planar array of Ginzburg-Landau atomic chains, is obtained. Each chain is treated in the continuous approximation, but the inter-chain approximation remains discrete; it is described by elastic forces, between next neighbors (nn) and near next neighbors (nnn). The transfer matrix approach is used in order to replace the 2D classical system with its 1D quantum dual. A Green function techniques allows the calculation of the ground state of the 1D system, which is in the same time the free energy of its 2D dual. The on-site anharmonicities are treated in a mean-field approximation, applied to the Hsue-Chern approach to the quartic oscillator. A logarithmic behavior of the specific heat is obtained, representing the fingerprint of an Ising-type phase transition. The physical significance of this results, as well as the limitations of the approximation used in this paper and in other approaches, are carefully discussed.

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