

UDC 530.1

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## NON-EQUILIBRIUM GIBBS THERMODYNAMIC POTENTIAL OF A MAGNETIC SYSTEM

The Leontovich definition of the free energy of a non-equilibrium system is discussed. In this approach it is assumed that the system has a definite temperature and is described by some set of reduced description parameters. It is shown that this free energy is obtained from an equilibrium free energy in the presence of an external field by a Legendre transformation that makes the field an internal parameter. The case of a weak non-equilibrium system is considered in details. The obtained result is applied to an isotropic magnetic system, non-equilibrium states of which are described by the phonon subsystem temperature and total magnetic dipole moment. Thereby the motion of magnetic dipoles in the system is taken into account and the temperature meaning is justified. On this basis the Gibbs non-equilibrium thermodynamic potential is calculated up to the fourth order terms in small dipole moment inclusive. The obtained Gibbs potential does not meet the requirements of the Landau theory of phase transitions of the second kind. We suppose that this contradiction is a consequence of the assumption about isotropy of the system which is broken in magnetic systems.

**Keywords:** non-equilibrium Gibbs thermodynamic potential; the Legendre transformation; states in an external field; magnetic system; phase transition.

Обговорюється означення Леонтовича вільної енергії нерівноважної системи. В цьому підході передбачається, що система має певну температуру й описується певною множиною параметрів скороченого опису. Показується, що зазначену вільну енергію можна отримати з рівноважної вільної енергії системи в деякому зовнішньому полі за допомогою перетворення Лежандра, що робить зовнішнє поле внутрішнім параметром. Детально розглядається випадок слабо нерівноважної системи. Отриманий результат застосовується до ізотропної магнітної системи, нерівноважні стани якої описуються температурою підсистеми фононів і повним магнітним дипольним моментом. Тим самим враховується рух магнітних диполів системи та з'ясовується сенс температури. На цій основі розраховується нерівноважний термодинамічний потенціал Гіббса з точністю до внесків четвертого порядку включно за малим дипольним моментом системи. Отриманий потенціал не задовольняє вимогам теорії Ландау фазових переходів другого роду. Вважаємо, що це протиріччя є наслідок припущення про ізотропію системи, яка в магнітних системах порушена.

**Ключові слова:** нерівноважний термодинамічний потенціал Гіббса, перетворення Лежандра, стани у зовнішньому полі, магнітна система, фазовий перехід.

Обсуждается определение Леонтовича свободной энергии неравновесной системы. В этом подходе предполагается, что система имеет определенную температуру и описывается некоторым множеством параметров сокращенного описания. Показывается, что эта свободная энергия получается из равновесной свободной энергии системы в некотором внешнем поле с помощью преобразования Лежандра, которое делает внешнее поле внутренним параметром. Детально рассматривается случай слабо неравновесной системы. Полученный результат применяется к изотропной магнитной системе, неравновесное состояние которой описывается температурой равновесной фононной подсистемы и магнитным дипольным моментом. Тем самым учитывается движение магнитных диполей системы и выясняется смысл температуры. На этой основе вычисляется неравновесный термодинамический потенциал Гиббса с точностью до вкладов четвертого порядка включительно по малому магнитному дипольному моменту системы. Полученный потенциал не удовлетворяет требованиям теории Ландау фазовых переходов второго рода. Полагаем, что это противоречие является следствием предположения об изотропии системы, которая в магнитных системах нарушена.

**Ключевые слова:** неравновесный термодинамический потенциал Гиббса, преобразование Лежандра, состояния во внешнем поле, магнитная система, фазовый переход.

## 1. Introduction

In 1938 Leontovich proposed [1] a definition of the free energy of a non-equilibrium state (see also [2, 3]). It is based on the idea that a non-equilibrium state coincides with an equilibrium state in the presence of a proper external field. In this approach non-equilibrium free energy differs from an equilibrium one by a Legendre transformation. Leontovich shows that this free energy satisfies the necessary conditions: in the equilibrium it coincides with the equilibrium potential, has minimum and defines probability of non-equilibrium states entering the Boltzmann formula (as an effective Hamiltonian).

In 1937 Landau elaborated [4] a theory of phase transitions of the second kind. In fact his theory is based on a non-equilibrium thermodynamic potential. However Landau did not discuss in [4] a fundamental definition of this potential. Applying his theory to paramagnetic  $\leftrightarrow$  ferromagnetic phase transition, he constructed [5] a non-equilibrium thermodynamic potential by a Legendre transformation as the Leontovich definition suggests. It was done without any discussion. However, in the last edition of his course [6] he proposed a definition of an effective Hamiltonian. But this definition was not elaborated by him.

In the present paper we discuss in detail the Leontovich definition for weak non-equilibrium states. The obtained results are applied to calculating the Gibbs thermodynamic potential for a magnetic system, which is described by its total magnetic dipole moment and equilibrium phonon subsystem temperature representing the motion of dipoles of the system. The problem is considered with an accuracy that is enough for a comparison with Landau theory of phase transitions of the II kind.

A part of this work will be presented in International Young Scientist Forum on Applied Physics and Engineering (10-14 October 2016, Kharkiv, Ukraine) [7].

The paper is organized as follows: in Sec. 2 the Leontovich definition of the non-equilibrium free energy is discussed and applied for a case of the weak non-equilibrium states, in Sec. 3 the Gibbs non-equilibrium potential for a magnetic system is calculated.

## 2. The Leontovich definition of the non-equilibrium free energy

Let a non-equilibrium state of the considered system is described with a temperature  $T$  and mean value  $\eta_a$  of some microscopic quantities  $\hat{\eta}_a$  ( $a$  is a parameter number; functions  $f(X)$  of the phase variables  $X$  are denoted hereafter as  $\hat{f}$ ).

An example of such system is given by a system of magnetic dipoles in a crystal body. The motion of the dipoles can be described in terms of phonons. We will assume that the phonon subsystem is in an equilibrium state with temperature  $T$ . The total magnetic moment of the system  $m_n$  describes a non-equilibrium state of the system.

Our work is based on the Leontovich idea, according to which it is possible to choose auxiliary field  $\hat{U}(\eta)$  depending on parameters  $\eta_a$  so that the considered non-equilibrium state is equilibrium in this field [1-3]. In other words, it is assumed that the Gibbs distribution gives in this situation a non-equilibrium distribution function that can be compared with one given by the Bogolyubov reduced description method (see, for example, [7]). So, quantities  $T$ ,  $\eta_a$  are reduced description parameters.

The external field is adjusted in such a way that the relation holds

$$\eta_a = \text{Sp } w(\eta) \hat{\eta}_a \quad (\text{Sp} \dots \equiv \int dX \dots) \quad (1)$$

here  $w(\eta)$  is the canonical Gibbs distribution for the system

$$w(\eta) = \exp \frac{F(\eta) - \hat{H} - \hat{U}(\eta)}{T} \quad (2)$$

Here  $\hat{H}$  is the Hamilton function of the system and  $F(\eta)$  is its free energy of the equilibrium state in the presence of the field..

According to Leontovich [1-3] the non-equilibrium free energy of the system is given by the relation

$$F_{\text{neq}}(\eta) = F(\eta) - U(\eta) \quad (U(\eta) \equiv \text{Sp } w(\eta) \hat{U}(\eta)) \quad (3)$$

where  $U(\eta)$  is mean energy of the field.

Expression (3) corresponds to the definition of non-equilibrium system entropy  $S(\eta)$  given in [8] in the framework of the Bogolyubov reduced description method. Really, in [8] the starting point of the entropy definition is the Gibbs formula

$$S(\eta) = -\text{Sp } w(\eta) \ln w(\eta) = (E(\eta) + U(\eta) - F) / T \quad (E(\eta) \equiv \text{Sp } w(\eta) \hat{H}) \quad (4)$$

where  $E(\eta)$  is internal energy of the non-equilibrium system. So, definition (5) and the Leontovich formula (3) give

$$F_{\text{neq}}(\eta) = E(\eta) - TS(\eta) \quad (5)$$

that obviously corresponds to the common idea about relation of non-equilibrium free energy, internal energy and entropy.

Further, Leontovich showed [1-3] that it is possible to restrict ourselves by external field of the form

$$\hat{U}(\eta) = \sum_a h_a(\eta) \hat{\eta}_a \quad (6)$$

where functions  $h_a(\eta)$  describe the field intensity. One can say that in  $\hat{U}(\eta)$  external field  $h_a(\eta)$  is switched through microscopic quantities  $\hat{\eta}_a$ . So, non-equilibrium free energy is defined by formula

$$F_{\text{neq}}(\eta) = F(\eta) - \sum_a h_a(\eta) \eta_a \quad (7)$$

The formula shows that  $F_{\text{neq}}(\eta)$  is some thermodynamic potential for equilibrium system in an external field. Non-equilibrium free energy  $F_{\text{neq}}(\eta)$  is obtained from potential  $F(\eta)$  with Legendre transformation. Below we drop arguments  $\eta_a$  in potentials  $F$ ,  $F_{\text{neq}}$  according to thermodynamic habits.

In fact, non-equilibrium free energy construction with a Legendre transformation was proposed by Landau in his theory of the phase transitions paramagnet  $\leftrightarrow$  ferromagnet [5]. However, he avoided using the term "the non-equilibrium free energy" in his theory of the phase transition of the second kind [6] and assumed the presence of its necessary properties without any discussion.

Leontovich established the next properties of the non-equilibrium free energy: it turns into equilibrium free energy  $F_0$  in equilibrium [1-3], it has a minimum in the equilibrium

$$F_{\text{neq}}(\eta_0) = F_0, \quad F_{\text{neq}}(\eta) \geq F_0. \quad (8)$$

Here  $\eta_{a0}$  is an equilibrium value of the parameter  $\eta_a$  and  $F_0$  is equilibrium free energy defined by formulas

$$\eta_{a0} = \text{Sp } w_0 \hat{\eta}_a; \quad w_0 = \exp \frac{F_0 - \hat{H}}{kT}, \quad \text{Sp } w_0 = 1. \quad (9)$$

This is often used in the literature without reference to Leontovich.

The basic thermodynamic relation for the free energy  $F$  has a standard form

$$dF = -SdT - \sum_i A_i da_i + \sum_a \eta_a dh_a + \mu dN. \quad (10)$$

Here  $a_i$  are external parameters which are defined with the system Hamiltonian  $\hat{H}$ ,  $A_i$  are corresponding thermodynamic forces

$$A_i = \text{Sp } w \hat{A}_i, \quad \hat{A}_i \equiv -\frac{\partial \hat{H}}{\partial a_i}, \quad (11)$$

$\mu$  is chemical potential,  $N$  is number of particles in the system (for an one-component system). It follows from (3) that  $-\eta_a$  is the thermodynamic force, which corresponds to the parameter  $h_a$ .

The basic thermodynamic relation for  $F_{\text{neq}}$  can be written as

$$dF_{\text{neq}} = -SdT - \sum_i A_i da_i - \sum_a h_a d\eta_a + \mu dN \quad (12)$$

using relations (4) and (5).

In weak non-equilibrium states the Leontovich definition can be analyzed in detail. In these states deviations  $\delta\eta_a = \eta_a - \eta_{a0}$  of parameters  $\eta_a$  from their equilibrium values  $\eta_{a0}$  and external field  $h_a$  are small and a perturbation theory can be applied for calculating the introduced above quantities. It is convenient to do this using formula

$$\exp \frac{F_0 - F}{T} = \mathbb{F}(h) \quad (13)$$

(following from (2), (6), and (9)) where function  $\mathbb{F}(h)$  is defined by

$$\mathbb{F}(h) = \overline{\exp\left(-\sum_a h_a \hat{\eta}_a / T\right)} \quad (\hat{f} \equiv \text{Sp } w_0 \hat{f}) \quad (14)$$

and is obviously the generating function for mean values  $\overline{\hat{\eta}_{a_1} \dots \hat{\eta}_{a_n}}$ . Instead of averages  $\overline{\hat{\eta}_{a_1} \dots \hat{\eta}_{a_n}}$  of products of quantities  $\hat{\eta}_a$  it is convenient to use the corresponding correlation functions  $\langle \eta_{a_1} \dots \eta_{a_n} \rangle$  which are defined by formulas

$$\begin{aligned} \overline{\hat{\eta}_{a_1}} &= \eta_{a_1 0}, & \overline{\hat{\eta}_{a_1} \hat{\eta}_{a_2}} &= \overline{\hat{\eta}_{a_1}} \overline{\hat{\eta}_{a_2}} + \langle \eta_{a_1} \eta_{a_2} \rangle, \\ \overline{\hat{\eta}_{a_1} \hat{\eta}_{a_2} \hat{\eta}_{a_3}} &= \overline{\hat{\eta}_{a_1}} \overline{\hat{\eta}_{a_2}} \overline{\hat{\eta}_{a_3}} + \langle \eta_{a_1} \eta_{a_2} \rangle \overline{\hat{\eta}_{a_3}} + \langle \eta_{a_2} \eta_{a_3} \rangle \overline{\hat{\eta}_{a_1}} + \langle \eta_{a_3} \eta_{a_1} \rangle \overline{\hat{\eta}_{a_2}} + \end{aligned} \quad (15)$$

$$+\langle \eta_{a_1} \eta_{a_2} \eta_{a_3} \rangle$$

and so on with using of cyclic permutations. A generating function  $\mathbb{G}(h)$  for correlations  $\langle \eta_{a_1} \dots \eta_{a_n} \rangle$

$$\mathbb{G}(h) \equiv \sum_{n=2}^{\infty} \frac{(-1)^n}{n!} \sum_{a_1 \dots a_n} h_{a_1} \dots h_{a_n} \frac{1}{T^n} \langle \eta_{a_1} \dots \eta_{a_n} \rangle, \quad (16)$$

is connected with the generating function  $\mathbb{F}(h)$  as

$$\mathbb{F}(h) = \exp\left(-\sum_a h_a \eta_{a0} / T + \mathbb{G}(h)\right) \quad (17)$$

(see, for example, [8, 9]).

In accordance with (7), this relation gives the following expression for the free energy  $F$

$$F = F_0 + \sum_a h_a \eta_{a0} - T \mathbb{G}(h), \quad (18)$$

in the form of an expansion in powers of  $h_a$ . The basic thermodynamic relation (10) gives the formula

$$\eta_a = (\partial F / \partial h_a)_{T, a_i, N}, \quad (19)$$

which allows (together with relations (17) – (19)) to express parameters  $\eta_a$  that describe non-equilibrium state of the system through the auxiliary field

$$\eta_a = \eta_{a0} + \sum_{n=1}^{\infty} \frac{(-1)^n}{n! T^n} \sum_{a_1 \dots a_n} h_{a_1} \dots h_{a_n} \langle \eta_a \eta_{a_1} \dots \eta_{a_n} \rangle \quad (20)$$

This expansion and formulas (7) and (19) give an expression for non-equilibrium free energy through a small field  $h_a$

$$\begin{aligned} F_{\text{neq}} = F_0 + \frac{1}{2T} \sum_{a_1, a_2} h_{a_1} h_{a_2} \langle \eta_{a_1} \eta_{a_2} \rangle - \frac{1}{3T^2} \sum_{a_1, a_2, a_3} h_{a_1} h_{a_2} h_{a_3} \langle \eta_{a_1} \eta_{a_2} \eta_{a_3} \rangle + \\ + \frac{1}{8T^3} \sum_{a_1, a_2, a_3, a_4} h_{a_1} h_{a_2} h_{a_3} h_{a_4} \langle \eta_{a_1} \eta_{a_2} \eta_{a_3} \eta_{a_4} \rangle + O(h^5). \end{aligned} \quad (21)$$

Here and below the non-equilibrium free energy  $F_{\text{neq}}$  is calculated with accuracy up to the fourth order inclusive. Formula (20) can be considered as an equation for the function  $h_a(\eta)$ . This equation for weak non-equilibrium states is solved in a perturbation theory in small deviations  $\delta\eta_a = \eta_a - \eta_{a0}$  and allows transforming the expression for the non-equilibrium free energy (21) into a series in powers of  $\delta\eta_a$ . The result obtained in this way is similar to the basic assumption of the Landau theory of phase transitions of the second kind [4].

### 3. The Gibbs non-equilibrium potential of a magnetic system

Let us consider a magnetic system consisting of magnetic dipoles oscillating in a solid. Motion of the dipoles is described in terms of phonons forming an equilibrium

subsystem. A non-equilibrium state of the system is described by its total dipole moment  $m_n$  as the reduced description parameters  $\eta_a$ . In this case external field  $h_a$  is uniform magnetic field taken with minus  $-H_n$  (because interaction is given by  $\hat{U} = -\hat{m}_n H_n$ ).

In addition, it is assumed below that the system is isotropic in absence of the magnetic field. In this case, according to the rotational invariance, the equilibrium value of parameters  $m_n$  equals to zero

$$m_{n0} = \overline{\hat{m}_n} = 0 \quad (22)$$

and the first correlation functions have a structure

$$\begin{aligned} \langle m_n m_l \rangle &= \frac{1}{3} \langle m^2 \rangle \delta_{nl} \quad (\langle m^2 \rangle = \langle m_n m_n \rangle), \quad \langle m_n m_l m_m \rangle = 0, \\ \langle m_n m_l m_m m_s \rangle &= \frac{1}{15} \langle m^4 \rangle (\delta_{nl} \delta_{ms} + \delta_{nm} \delta_{ls} + \delta_{ns} \delta_{lm}) \quad (\langle m^4 \rangle \equiv \langle (m_n m_n)^2 \rangle). \end{aligned} \quad (23)$$

It allows simplifying the previous results. In particular, formula (20) gives such an expression for the total dipole moment  $m_n$  via the magnetic field  $H_n$

$$m_n = \frac{1}{3T} H_n \langle m^2 \rangle + \frac{1}{30T^3} H_n H^2 \langle m^4 \rangle + O(H^5). \quad (24)$$

Therefore the free energy (21) takes the form

$$F_{\text{neq}} = F_0 + \frac{1}{6T} H^2 \langle m^2 \rangle + \frac{1}{40T^3} H^4 \langle m^4 \rangle + O(H^6) \quad (25)$$

( $H^2 \equiv H_n H_n$ ,  $H^4 \equiv (H^2)^2$ ). The non-equilibrium free energy has to be expressed through the total magnetic moment  $m_n$ . To do this, (24) is solved as an equation for the function  $H_n(m)$

$$H_n = \frac{3T}{\langle m^2 \rangle} m_n - \frac{27 T \langle m^4 \rangle}{10 \langle m^2 \rangle^4} m_n m^2 + O(m^5) \quad (26)$$

that with formula (25) gives the necessary expression for

$$F_{\text{neq}} = F_0 + \frac{3}{2} \frac{T}{\langle m^2 \rangle} m^2 - \frac{27 T \langle m^4 \rangle}{40 \langle m^2 \rangle^4} m^4 + O(m^6). \quad (27)$$

The obtained non-equilibrium free energy (27) is a function  $F_{\text{neq}} = F_{\text{neq}}(T, V, N, m)$  of the temperature  $T$ , volume  $V$ , number of magnetic dipoles,  $N$  and total magnetic moment  $m_n$ . Theory of phase transitions is usually formulated at fixed temperature  $T$  and pressure  $p$  because of connection with common experimental conditions. With this and in view of the use of Gibbs thermodynamic potential that is introduced by the Legendre transformation

$$\Phi_{\text{neq}} = F_{\text{neq}} + pV, \quad (28)$$

it should be calculated as a function of variables  $T$ ,  $p$ ,  $N$ ,  $m_n$ . For its calculation it is necessary to calculate the pressure with using the basic thermodynamic relation (12) that takes the form

$$dF_{\text{neq}} = -SdT - pdV + H_n dm_n + \mu dN \quad (29)$$

and gives

$$p = -\left(\frac{\partial F_{\text{neq}}}{\partial V}\right)_{T,m,N}. \quad (30)$$

Then one has to find function  $V(T, p, N, m)$  that is a reverse function for the pressure  $p(T, V, N, m)$ .

The structure of the free energy as a function  $F_{\text{neq}}(T, V, N, m)$  is given by the formula

$$F_{\text{neq}} = F_0 + a(T, \tilde{V}) \frac{m^2}{N} + b(T, \tilde{V}) \frac{m^4}{N^3} + O(m^6) \quad (\tilde{V} \equiv V / N) \quad (31)$$

where

$$a(T, \tilde{V}) \equiv \frac{3T}{2\psi_1(T, \tilde{V})}, \quad b(T, \tilde{V}) \equiv -\frac{27T\psi_1(T, \tilde{V})}{40\psi_2(T, \tilde{V})^4}, \quad \langle m^{2s} \rangle \equiv N\psi_s(T, \tilde{V}) \quad (32)$$

that can be proved on the basis of the principle of spatial correlation weakening (see, for example, [8]). Formulas (30) and (31) allow calculating the function  $V(T, p, N, m)$

$$\tilde{V} = \tilde{V}_0 + c(T, p) \frac{m^2}{N^2} + d(T, p) \frac{m^4}{N^4} + O(m^6) \quad (33)$$

where  $\tilde{V}_0$  is an equilibrium volume and

$$c \equiv \left(\frac{\partial a}{\partial \tilde{V}}\right)_T \left(\frac{\partial p_0}{\partial \tilde{V}}\right)_T^{-1}, \quad d \equiv \left[\left(\frac{\partial b}{\partial \tilde{V}}\right)_T + c \left(\frac{\partial^2 a}{\partial \tilde{V}^2}\right)_T - \frac{1}{2} c^2 \left(\frac{\partial^2 p_0}{\partial \tilde{V}^2}\right)_T\right] \left(\frac{\partial p_0}{\partial \tilde{V}}\right)_T^{-1}. \quad (34)$$

In this expression  $p_0(T, \tilde{V})$  is an equilibrium pressure and a substitution  $\tilde{V} \rightarrow \tilde{V}_0(T, p)$  is assumed.

Simple calculations based on formulas (28), (31), and (33) give the final expression for the non-equilibrium Gibbs potential

$$\Phi_{\text{neq}}(T, p, m, N) \equiv \Phi_0 + \alpha(T, p) \frac{m^2}{N} + \beta(T, p) \frac{m^4}{N^3} + O(m^6) \quad (35)$$

where it is denoted

$$\alpha \equiv a(T, \tilde{V}_0(T, p)) \quad (\Phi_0 \equiv \Phi_0 / N);$$

$$\beta \equiv d \left(\frac{\partial \Phi_0}{\partial \tilde{V}}\right)_T + \frac{1}{2} c^2 \left(\frac{\partial^2 \Phi_0}{\partial \tilde{V}^2}\right)_T - c \left(\frac{\partial^2 a}{\partial \tilde{V}^2}\right)_T - \tilde{V}_0 \left(\frac{\partial b}{\partial \tilde{V}}\right)_T + b. \quad (36)$$

Here  $\Phi_0$  is an equilibrium Gibbs potential and a substitution  $\tilde{V} \rightarrow \tilde{V}_0(T, p)$  in  $\beta$  is assumed. Note that corrections of the second order in  $m_n$  for the free energy (31) and the Gibbs potential (35) are equal. This is a consequence of the general theorem proved in [6].

This expression is similar to one which is a basis of the Landau theory of phase transitions of the second kind paramagnet  $\leftrightarrow$  ferromagnet (see, for example, [5, 6]). However the coefficient in (35) at  $m^2$  is positive in contradiction with the Landau assumption. We suppose that this contradiction is a consequence of our assumption about the system isotropy that is broken in magnetic systems at some temperatures.

#### 4. Conclusions

In this paper on the basis of the Leontovich definition of the non-equilibrium free energy the Gibbs non-equilibrium thermodynamic potential is built for weak non-equilibrium states of a magnetic system. In understanding of this definition, it is very important to explain the meaning of the temperature. We consider a system of moving magnetic dipoles, so the subsystem that describes their motion (for example, phonon subsystem) is assumed to be in equilibrium with the mentioned temperature. In calculations we restrict ourselves by non-equilibrium potentials obtained with accuracy up to the fourth order terms in the small total magnetic moment of the system inclusive. In this way we try to justify the phenomenological Landau assumption concerning the thermodynamic potential used in his theory of the phase transitions of the II kind. In the present paper the thermodynamic Gibbs potential, which does not meet the Landau requirements, is constructed. Our explanation of this contradiction connects it with our assumption about anisotropy of the considered system that is false below the Curie point. In this situation equilibrium averages (in the absence of the magnetic field) should be considered as the Bobolyubov quasi-averages (see, for example, [10]). In a subsequent paper we will discuss this idea in details.

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*Received 20.05.2016*