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# THE ROLE OF HIGH ORDER TERMS IN LANDAU THEORY TOWARD LANDAU-KHALATNIKOV EQUATION OF MOTION

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**Abstract** — We present the numerical analysis of the high order terms in the polynomial series of the Landau density energy. We compare the profiles and hysteresis of three series of Landau equations. First series is up to the only fourth order, the second and the third series are up to the sixth and eighth terms. The polarisation profile curves are obtained by minimizing the Landau equation and solved it using root finding technique while the hysteresis is obtained by solving Landau-Khalatnikov equation. It is found that those three series of Landau model possess similar polarisation profiles and also similar hysteresis when the system is set near Curie temperature, around 40K to 50 K for the  $T_c$  of 50 K. If the temperature of the system is decreased further away from the Curie temperature, we have to consider the high order terms in Landau free energy expression.

**Keywords**— higher order, Landau theory, hysteresis

## I. INTRODUCTION

Landau theory is well known as a suitable approximation to explain the properties of the system in the equilibrium condition near phase transition [1]. The transition between two phases which have different symmetry in their thermodynamic state is described by order parameter, a physical entity which values (non-zero or zero) depend on the symmetry of the state. It has non-zero value in the ordered state, while in disordered phase, the value is zero [2]. This phenomenological theory has been used to study the system with long range interaction, near phase transition such as ferroelectric-paraelectric [3] and ferromagnet-paramagnet [4] using electric polarization  $\mathbf{P}$  and magnetization  $\mathbf{M}$  as order parameters, and also phase transitions in magnetoelectric multiferroics using both polarization and magnetization.

The free energy of the system is approximated by growing the order parameter in power series. This free energy then becomes the starting point to study other parameters of the system. For example, the value of the order parameter in a certain temperature can be obtained by solving the minimum condition of the free energy of the system [2]. The response of the order parameter to the applied disturbance of such fields is also calculated based on the free energy system using Landau Khalatnikov equation yielding the hysteresis curve [5,6]. In the series of order parameter expressing the free energy, we can also add other terms such as the surface effect, the depolarization term, etc. to complete the behaviour of the system [7].

In previous studies, the expansion of order parameters in free energy is up to the sixth order [8,9]. However, there are also theoretical studies which only use fourth order expansion [5,6]. The latter is the simplest expansion in the Landau theory. Motivated by the difference in the expansion of the free energy, in this paper we study the role of the higher order (sixth and eighth order) in the free energy. Hence, we analyze the equilibrium condition to obtain the profile of the order parameter toward the temperature. Then, we also study the effect of that order of expansion to the response of the material to the external disturbance by using Landau-Khalatnikov equation of motion to get the hysteresis curves of the system.

## II. RESEARCH METHOD

We started by firstly defining the free energy. Assuming the system is ferroelectrics, the form of free energy up into the eighth order is

$$F_E = \frac{1}{2}a_0(T - T_c)P^2 + \frac{1}{4}\beta P^4 + \frac{1}{6}\eta P^6 + \frac{1}{8}\kappa P^8 - EP \quad (1)$$

where  $T_c$  represents Curie temperature, while parameters  $a_0$ ,  $\beta$ ,  $\eta$  and  $\kappa$  are dielectric stiffness constants of the ferroelectrics. Parameter  $E$  corresponds the applied electric field. Using  $P = P_s p$  with  $P_s = \sqrt{a_0/\beta}$  as spontaneous polarization, then divide it by  $a_0^2/\beta$ , equation Eq(1) can be brought into

$$\mathcal{F} = \frac{1}{2}(T - T_c)p^2 + \frac{1}{4}p^4 + \frac{1}{6}\tilde{\eta}p^6 + \frac{1}{8}\tilde{\kappa}p^8 - \mathcal{E}p \quad (2)$$

where  $\tilde{\eta} = (a_0/\beta^2)\eta$ ,  $\tilde{\kappa} = (a_0^2/\beta^3)\kappa$  and  $\mathcal{E} = \sqrt{\beta/a_0^3}E$ .

Then, the expression of the free energy in Eq.(2) is minimized into

$$\frac{d\mathcal{F}}{dp} = (T - T_c)p + \beta p^3 + \tilde{\eta}p^5 + \tilde{\kappa}p^7 - \mathcal{E} = 0. \quad (3)$$

After that, the profile of the electric polarization toward temperature in equilibrium condition is obtained by excluding the external electric field and solving Eq.(3) with the various values of temperature using root finding technique. Here, we calculated three types of order of the Landau free energy which are up to the fourth order, sixth order and eighth order. Using obtained profile curves, the analysis is performed to study the role of higher order expansion of Landau free energy toward the polarization of the materials.

Next, we studied the response of the electric polarization to the applied electric field by firstly deriving the equation of motion of the system using Landau-Khalatnikov (LKh) as

$$\frac{\partial p}{\partial t} = -\gamma \frac{\partial \mathcal{F}}{\partial p} \quad (4)$$

where  $\gamma$  is phenomenological damping constant. Performing rescaling process as before to get Eq.(2), we can bring Eq.(4) above into the form of

$$\frac{\partial p}{\partial t} = -\tilde{\gamma} \frac{\partial \mathcal{F}}{\partial p} = -\tilde{\gamma}[(T - T_c)p + p^3 + \tilde{\eta}p^5 + \tilde{\kappa}p^7 - \mathcal{E}] \quad (5)$$

where  $\tilde{\gamma} = a_0\gamma$ . Next, using the definition of limit to the derivative of polarization  $p$  to the time  $t$ , the LKh Eq.(5) can be written in the form which is easier to be worked on numerical calculation

$$[1 + \tilde{\gamma}\Delta t(T - T_c)]p_{i+1} - p_i + \tilde{\gamma}\Delta t(p_{i+1}^3 + \tilde{\eta}p_{i+1}^5 + \tilde{\kappa}p_{i+1}^7 - \mathcal{E}) = 0 \quad (6)$$

where  $p_{i+1}$  and  $p_i$  refer to the  $p(t+\Delta t)$  and  $p(t)$  with  $\Delta t$  is the step size of time illustrating the relaxation time. The results are hysteresis curves of the electric polarization  $p$  versus the external electric fields  $\mathcal{E}$ .

## III. RESULT AND DISCUSSION

In the numerical calculation, we set the critical temperature  $T_c = 50$  K. We also set the parameters for dielectric stiffness constants as:  $a_0 = 10^{-6}$  K<sup>-1</sup> and  $\beta = 10^{-15}$  cm<sup>4</sup>/statC<sup>2</sup>, typical for ferroelectrics. The parameters for the sixth and eighth order are fixed at the values  $\eta = 5 \times 10^{-27}$  cm<sup>8</sup>/statC<sup>4</sup> and  $\kappa = 10^{-37}$  cm<sup>12</sup>/statC<sup>6</sup>.

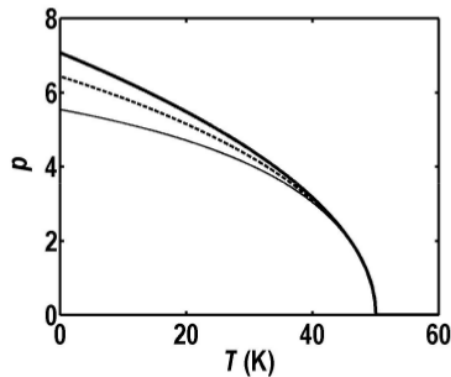


Fig. 1 Profile of the electric polarization toward the temperature with various order of free energy. The thick line represents the 4<sup>th</sup> order.

The dotted-line is for the 6<sup>th</sup> order and the thin line corresponds to the 8<sup>th</sup> order of the Landau free energy, The solutions of Eq.(3) are illustrated in Fig.1. It shows the profiles of the electric polarisation toward the temperature for various order of the polynomial Landau free energy. The thick line in Fig.1 corresponds the fourth order Landau free energy while the polarization profiles of the systems with the sixth and eight order Landau density energy are represented by dotted line and thin line.

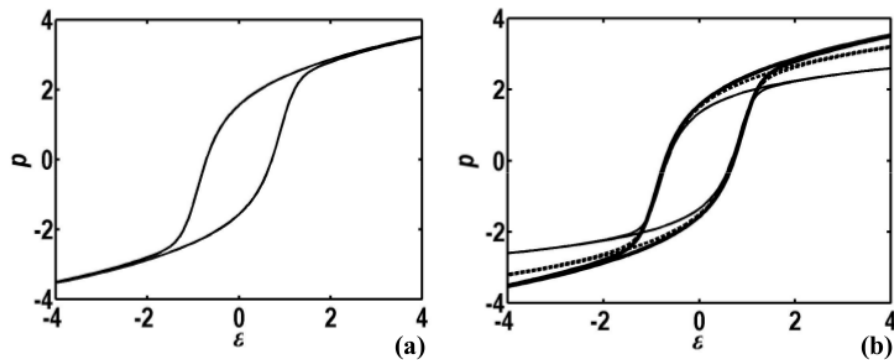


Fig. 2 Hysteresis of ferroelectrics system based on the 4<sup>th</sup> order, 6<sup>th</sup> order and 8<sup>th</sup> order Landau free energy. Picture (a) illustrates hysteresis near  $T_c$  while graph (b) shows the hysteresis at 20 K below  $T_c$ .

The thick line represents the 4<sup>th</sup> order the dotted-line is for the 6<sup>th</sup> order and the thin line corresponds to the 8<sup>th</sup> order of the Landau free energy, in the temperature less than 30 K, figure.1 shows the significantly different values of the electric polarization for the three orders of Landau free energy. It means that in this range of temperature, if we approximate the system using only the fourth order Landau free energy, the results will be different from the approximation based on the higher (6<sup>th</sup> or 8<sup>th</sup>) order of Landau free energy. The result will certainly be precise if it is involving the higher order Landau density energy. It can also be seen from Fig.1, that the polarizations from the systems with the different order of Landau density energy have the same values in the range around 40 K to 50 K ( $T_c$ ). Hence, near critical temperature, the system can be approached using only the fourth order Landau free energy without losing precision.

The effect of the order of the Landau free density energy to the shape of the hysteresis is drawn in Fig.2. The hysteresis is obtained by solving the Eq.(6) numerically. The results at temperature near critical temperature  $T_c$  for the 4<sup>th</sup> order, 6<sup>th</sup> order and 8<sup>th</sup> order of Landau expression are the same, as it is illustrated in Fig.2a. These results are consistent to the electric polarization profile in Fig.1. Where the polarization from the three types of Landau density energy are similar near  $T_c$ . The results of the hysteresis for the temperature further away from the critical point are presented in Fig.2b. It can be seen that the saturated polarization decreases (from around 3.5 to 2.8) when the order of Landau equations are increased from the 4<sup>th</sup> order to the 8<sup>th</sup> order. These results are correlated to the behaviour of the polarization at the temperature far from Curie temperature which shows the similar tendency.



It is assumed that the accurate result is produced by including the higher order term of Landau equation in the calculation. Excluding the higher terms in the calculation, it will result in the bigger values with less accuracy. However, analysing the system by including the higher terms of Landau equation is not easy, especially when the other effects are also considered: inhomogeneity, surface effect, depolarisation field, etc. The easier calculation with accurate results can be performed by setting the system in the condition near Curie temperature. It is illustrated by numerical results of polarization profile and the hysteresis in Fig.1 and Fig.2. It is shown that the simplest and easiest form of Landau free Energy (the polynomial is up only to the fourth order) can be used properly near critical temperature. In this temperature, less than 10% of  $T_c$  ( $T_c$  is around 50 K), the result is the same when it is using higher order terms (6<sup>th</sup> order and 8<sup>th</sup> order). Then, the good accuracy can be maintained.

#### IV. CONCLUSIONS

The higher order terms (6<sup>th</sup> and 8<sup>th</sup> order) in Landau density energy form are important when we are conducting the analysis of the ferroelectric system far from critical temperature. The difficulties of involving the higher terms in the calculation can be avoided by considering that temperature of the system is near Curie temperature since in this condition the higher terms can be excluded.

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