

$(\sqrt{3} \times \sqrt{3})R30^\circ$   $(3 \times 3)$  phase transition in Pb/Ge(111) and Sn/Ge(111): a phenomenological study on the phase transition anomalies and the role of defects

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2005 Nanotechnology 16 325

(<http://iopscience.iop.org/0957-4484/16/2/026>)

[The Table of Contents](#) and [more related content](#) is available

Download details:

IP Address: 150.244.7.82

The article was downloaded on 28/04/2009 at 14:57

Please note that [terms and conditions apply](#).

# $(\sqrt{3} \times \sqrt{3})R30^\circ \leftrightarrow (3 \times 3)$ phase transition in Pb/Ge(111) and Sn/Ge(111): a phenomenological study on the phase transition anomalies and the role of defects

A Cano, A P Levanyuk and E G Michel

Departamento de Física de la Materia Condensada C-III, Universidad Autónoma de Madrid, E-28049 Madrid, Spain

Received 10 September 2004, in final form 5 January 2005

Published 25 January 2005

Online at [stacks.iop.org/Nano/16/325](http://stacks.iop.org/Nano/16/325)

## Abstract

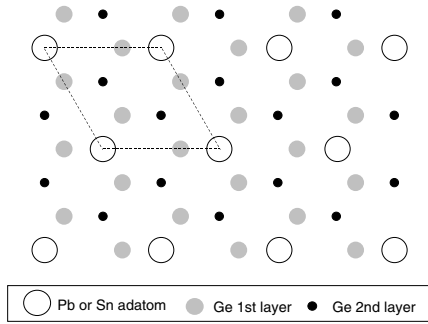
A phenomenological study of the  $(\sqrt{3} \times \sqrt{3})R30^\circ \leftrightarrow (3 \times 3)$  phase transitions occurring in the adsorption systems Pb/Ge(111) and Sn/Ge(111) is presented. The starting point of such a study is the Landau theory. The critical behaviour expected theoretically for the two interfaces, and the corresponding influence of defects, are discussed in detail. Symmetry arguments show that, contrary to general belief, the critical behaviours of Pb/Ge(111) and Sn/Ge(111) are essentially different. The Landau-like approach employed to study the influence of defects provides a consistent and general manner to interpret the existing experimental data. Special attention is paid to the influence of hopping defects in Sn/Ge(111).

## 1. Introduction

The adsorption of Pb and Sn on Ge(111) has been a subject of interest in surface science for many years, in particular because these interfaces were considered model systems to investigate the origin of ordered structures at surfaces and its role in the formation of the metal/semiconductor junction [1, 2]. A large number of ordered phases are observed with varying the adsorbate coverage and the temperature in these systems [3, 4]. Both Sn/Ge(111) and Pb/Ge(111) interfaces exhibit a  $(\sqrt{3} \times \sqrt{3})R30^\circ$  structure (in the following  $\sqrt{3}$ ) at room temperature for a coverage of one-third of a monolayer (ML) of adsorbate [3, 4]. The  $\sqrt{3}$  phase has been characterized using several different surface sensitive techniques [5–9].

The  $\sqrt{3}$  phase reverts to a  $(3 \times 3)$  structure upon lowering the temperature [10, 11]. This phase transition has received a lot of attention (for an updated reference list, we refer the reader to [12]). A crucial question behind this interest is the microscopic origin of the phase transition. This question has been controversial, and there is no consensus on the topic (see e.g. [12]). In this paper we follow a different

approach, attacking the problem from a phenomenological point of view. Our starting point will be the Landau theory of phase transitions and its extensions. As we shall see, such an approach allows one to have a unified view of the subject, clarifying and systematizing existing experimental and theoretical results. On the basis of this view, new results can be obtained. Symmetry arguments, for instance, reveal that the  $\sqrt{3} \leftrightarrow (3 \times 3)$  transition in Pb/Ge(111) is essentially different from the phase transition in Sn/Ge(111), which has been overlooked up to now. One of the extensions of the Landau theory is the continuum-media theory of defects and structural phase transitions (see, e.g., [13] and references therein). This extension is of particular interest when studying the aforementioned transitions in the Pb/Ge(111) and Sn/Ge(111) interfaces. The properties of defects are so accessible experimentally in these interfaces that they can serve as model systems for the analysis of the behaviour of defects in a system close to a phase transition, as well as for the study of the influence the defects on the corresponding transition. Indeed, experimental data already reported in the literature reveal, for the first time, some interesting phenomena related to defects. These phenomena correspond to certain



**Figure 1.** Ball model of the  $\sqrt{3}$  structures of Sn/Ge(111) and Pb/Ge(111).

cases contemplated in the aforementioned continuum-media theory which, however, were not fully developed in the past. The current experimental activity motivates us to develop these cases here.

The paper is organized as follows. In section 2 we summarize the already reported structural data on Pb/Ge(111) and Sn/Ge(111) interfaces, paying special attention to the differences between the corresponding low-temperature structures relevant to our phenomenological analysis. In section 3 we present such an analysis based on a phenomenological Landau-like theory for the  $\sqrt{3} \leftrightarrow (3 \times 3)$  transitions in Pb/Ge(111) and Sn/Ge(111). In section 3.1, considering perfect interfaces, we discuss the corresponding critical behaviours. In section 3.2 we take into account the influence of defects. We first discuss some general aspects of this influence (section 3.2.1), and then we focus our attention on the influence of hopping defects (section 3.2.2). We show that

- (i) the different characteristic temperatures reported for the Sn/Ge(111) interface follow naturally from the presence of hopping defects and
- (ii) the characteristic length of the defect-induced order-parameter modulation may exhibit no anomaly at the transition point.

Some consequences of this latter possibility are further explored in section 3.2.3. Section 4 is devoted to conclusions.

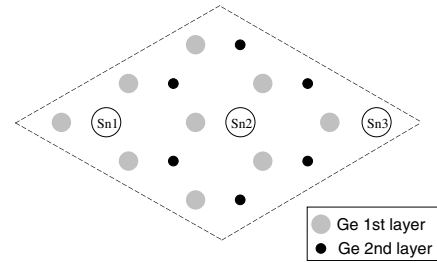
## 2. Summary of structural data

### 2.1. High temperature: $(\sqrt{3} \times \sqrt{3})R30^\circ$ phase

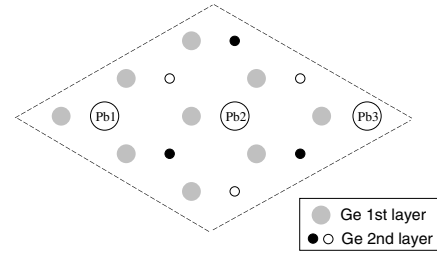
The  $\sqrt{3}$  structure is observed at room temperature for both Pb/Ge(111) and Sn/Ge(111) interfaces, at an adsorbate coverage of  $1/3$  ML. Pb or Sn adatoms occupy substrate  $T_4$  sites [5, 6] (see figure 1). The  $\sqrt{3}$  structure corresponds to the space group symmetry  $p3m1$ . At low temperature, Pb/Ge(111) and Sn/Ge(111) present, however, some important differences.

### 2.2. Low temperature: Sn/Ge(111)- $(3 \times 3)$

Scanning tunnelling microscopy (STM) and low-energy electron diffraction (LEED) results [11] show that adatom sites in Sn/Ge(111) become inequivalent at low enough temperatures, as a  $(3 \times 3)$  structure is formed. In consequence, there is a translational symmetry breaking in the interface



**Figure 2.** Sn/Ge(111)- $(3 \times 3)$  surface unit cell. Ge atoms in the second layer (●) are equivalent.



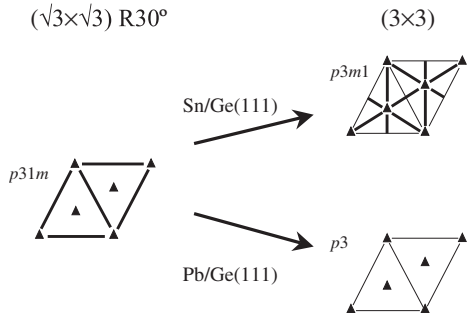
**Figure 3.** Pb/Ge(111)- $(3 \times 3)$  surface unit cell. Ge atoms in the second layer (● and ○) are not equivalent due to different displacements; see [16].

associated with the formation of a  $(3 \times 3)$  structure. Surface x-ray diffraction experiments [14, 15] have shown that the point symmetry of this  $(3 \times 3)$  structure coincides with that of the  $\sqrt{3}$  one (see figure 2). The space group symmetry of Sn/Ge(111)- $(3 \times 3)$  is  $p3m1$ .

### 2.3. Low temperature: Pb/Ge(111)- $(3 \times 3)$

The behaviour of Pb/Ge(111) is somewhat more complicated. In this case, the corresponding  $(3 \times 3)$  structure implies not only the non-equivalence of adatom sites, as revealed by STM [11], but also the non-equivalence of Ge atoms in the second layer of the substrate, as shown by surface x-ray diffraction experiments [16, 17] (see figure 3). In addition to the translational symmetry change, mirror planes are also now eliminated. The resulting symmetry group of Pb/Ge(111)- $(3 \times 3)$  is therefore  $p3$ .

There is, however, a different possibility to explain the behaviour of the Pb/Ge(111) interface. We note that, while the  $(3 \times 3)$  structure appears at  $\sim 250$  K, the data showing  $p3$  symmetry were obtained at 50 K [16, 17]. Therefore, the total lowering of symmetry in Pb/Ge(111) could take place in two steps: a first phase transition would give rise to the appearance of a  $(3 \times 3)$  structure with  $p3m1$  symmetry and a second phase transition would lower this symmetry to  $p3$ , with no increase of the unit cell. If this were the case,  $\sqrt{3}$  to  $(3 \times 3)$  transitions in both Pb/Ge(111) and Sn/Ge(111) systems would be equivalent, but an additional phase transition would exist in Pb/Ge(111) at a lower temperature. We note that nothing prevents a second phase transition from also being observed for Sn/Ge(111) at lower temperatures than so far used; in this case both interfaces would behave in a similar way. To our knowledge Pb/Ge(111) has not been studied enough to confirm any of the above described possible scenarios (one or two different phase transitions). In view of this lack of experimental information, we assume in the following that the



**Figure 4.** Elements of point symmetry in the unit cell of the corresponding structures of the Sn/Ge(111) and Pb/Ge(111) interfaces ( $\blacktriangle$ , three-fold axis;  $\blacksquare$ , mirror plane).

$\sqrt{3}$  to  $(3 \times 3)$  phase transition in Pb/Ge(111) is  $p31m \rightarrow p3$ , i.e., that this phase transition and the one in the Sn/Ge(111) interface are different.

We conclude that the corresponding order parameters of the phase transitions in Pb/Ge(111) and Sn/Ge(111) transform according to different irreducible representations of the  $p31m$  space group of their  $\sqrt{3}$  structures (see figure 4). In other words, there are two different, non-related order parameters. This leads to important consequences, already at the phenomenological level, as shown below.

### 3. Landau theory and its extensions

We expose in the following the phenomenological study of the  $\sqrt{3} \leftrightarrow (3 \times 3)$  phase transitions occurring in Pb/Ge(111) and Sn/Ge(111) interfaces. We consider first ideal interfaces.

#### 3.1. Perfect interfaces

The starting point in this phenomenological study is to construct the corresponding Landau free energy or, in terms of the modern theory, the Landau–Ginzburg–Wilson (LGW) Hamiltonian. To this end, we must take into account the symmetry of the phases involved in the transition.

The problem has been already studied for any structural phase transition taking place in a two-dimensional system [18, 19]. In the case of the  $\sqrt{3}$  to  $(3 \times 3)$  transition in Sn/Ge(111) ( $p31m \rightarrow p3m1$ ), the following Hamiltonian is obtained:

$$H_{\text{LGW}}^{(\text{Sn})} = \frac{A}{2}(\eta_1^2 + \eta_2^2) + \frac{s}{3}(\eta_1^3 - 3\eta_1\eta_2^2) + \frac{B}{4}(\eta_1^2 + \eta_2^2)^2 + \frac{D}{2}[(\nabla\eta_1)^2 + (\nabla\eta_2)^2], \quad (1)$$

where  $\eta_1$  and  $\eta_2$  are the two components of the order parameter. The LGW Hamiltonian for the  $\sqrt{3}$  to  $(3 \times 3)$  transition in Pb/Ge(111) ( $p31m \rightarrow p3$ ) is, however,

$$H_{\text{LGW}}^{(\text{Pb})} = \frac{A}{2}(\eta_1^2 + \eta_2^2) + \frac{B}{4}(\eta_1^2 + \eta_2^2)^2 + \frac{C_1}{6}(\eta_1^2 + \eta_2^2)^3 + \frac{C_2}{6}(\eta_1^3 - 3\eta_1\eta_2^2)^2 + \frac{D}{2}[(\nabla\eta_1)^2 + (\nabla\eta_2)^2]. \quad (2)$$

**3.1.1. Landau theory.** Within the simplest Landau approach, the above Hamiltonians are considered as the non-equilibrium free energies of the systems. A phase transition then takes

place if the coefficient at the quadratic term goes through zero to become negative. In the Landau theory it is assumed that the dependence of this coefficient on a control parameter (temperature in our case) is analytical, and thus it can be represented as a Taylor series. In practice, only the first term of this series is taken into account:  $A \propto T - T_c$ . In the other coefficients, the first term of their Taylor series is a constant.

Already at this level, some differences can be noticed between the  $\sqrt{3} \leftrightarrow (3 \times 3)$  phase transitions in Sn/Ge(111) and Pb/Ge(111). While the former transition should be discontinuous according to the Landau theory, the latter can be continuous. The discontinuity is due to the third-order invariant present in equation (1) which is absent in equation (2). It is worth mentioning that if this discontinuity is not strong, i.e., if the corresponding first-order phase transition is close to being a second-order one, the Landau approach is, in principle, applicable.

**3.1.2. Small fluctuations.** The assumptions of the Landau theory described above proved to be unjustified close to the transition. The crucial point to understand why is the following. The temperature dependence of any material coefficient playing the role of a ‘generalized rigidity’ (in our case, the coefficient corresponding to the ‘internal deformations’ described by the order parameter) is due to the influence of thermal motion (fluctuations) of other degrees of freedom of the system. Landau tacitly assumed that the only degree of freedom that ‘feels’ the phase transition is the one describing the long-range order, i.e., the mean value of the order parameter over the sample volume. However, this idea is not fully consistent. Let us consider the case of a spatially inhomogeneous order parameter, which indeed would rather describe not the long-range order but rather the short-range one. The fluctuations of the degrees of freedom associated with this inhomogeneous order parameter increase close to the phase transition point, so these degrees of freedom also ‘feel’ the transition.

As long as fluctuations are not too large, their influence can be treated within a first order perturbation theory. The trends revealed in this way are usually confirmed and specified within a much more involved theory valid for large fluctuations, i.e., in the range close to the phase transition (scaling region). An example of such a trend is the case of the first-order transition that, within the Landau theory, is expected in Sn/Ge(111). In this case, and due to this effect, the discontinuity in the order parameter diminishes. Indeed, the coefficient of the third-order invariant in equation (1), being renormalized by fluctuations, becomes temperature dependent and decreases when  $A$  diminishes, i.e. as the stability limit (the spinode point) of the symmetric phase is approached:

$$s \longrightarrow s^* \underset{A \rightarrow 0}{\approx} s[1 - BT/(AD)]. \quad (3)$$

It is impossible to conclude, within this first-order perturbation theory, whether this renormalized value of the coefficient  $s$  in equation (1) goes to zero at the phase transition point itself, i.e. whether the first-order transition becomes a second-order one. However, this conclusion has already been proved exactly, using a theory valid for large fluctuations [20].

The condition of applicability of the first-order perturbation theory is, in fact, the same as that of the Landau

theory. It has been obtained by different reasonings by several authors (see [21–23]) but it can also be deduced from equation (3). For two-dimensional systems it reads

$$TB/(AD) \ll 1, \quad (4)$$

where it is assumed (as well as in equation (3)) that the coefficients of all the terms of fourth order in equations (1) and (2) are of the same order of magnitude. The real meaning of this condition, i.e., whether the region of applicability of the Landau theory exists or does not exist at all, depends on the numerical values for the coefficients.

Two opposite limiting cases are usually considered when analysing structural transitions: displacive and order–disorder transitions (see, e.g., [24]). Assuming that the maximum, i.e. the ‘atomic’, value of the order parameter is equal to unity, the coefficients in the corresponding Landau free energy are estimated as  $A = (T - T_c)/l_{\text{at}}^2$ ,  $B \sim \Theta/l_{\text{at}}^2$  and  $D \sim \Theta$ , where  $l_{\text{at}}$  represents the interatomic distance and  $\Theta \sim T_{\text{at}} = 10^4\text{--}10^5$  K for displacive transitions and  $\Theta \sim T_c$  for order–disorder ones<sup>1</sup>. Substituting in equation (4) one finds  $(T - T_c) \gg T_c$  in both displacive and order–disorder cases<sup>2</sup>. This means that already at  $(T - T_c) \sim T_c$  one may be at the boundary of applicability of the Landau theory (this should not be understood literally: the estimations are rather rough). In other words, the temperature dependences given by the Landau theory are not trustworthy in the region  $(T - T_c) \ll T_c$  where the Landau theory itself is not applicable. This does not mean, however, that the Landau theory is completely useless. Its qualitative conclusions based on symmetry arguments and even order-of-magnitude estimations can still be useful. This is convenient because the Landau theory is simple; but of course, one has to be aware of its limitations.

Let us underline that the non-applicability of the Landau theory does not necessarily mean that the scaling laws can be immediately applied. These laws, having an asymptotic character, are expected to be valid when  $(T - T_c) \ll T_c$ . The corresponding range where the order-parameter fluctuations are very large is conventionally called the ‘scaling’ or ‘fluctuation’ region. This range may be very narrow and may not coincide with the range studied experimentally. But, of course, it is quite useful to know what can be expected in this region (see also below).

**3.1.3. Dynamics.** The dynamics of the corresponding order parameter is expected to be vibrational-like for displacive phase transitions and relaxational-like for order–disorder ones. A naive approximation consists in writing down the equation of motion for the component  $\eta_1$  (and the analogous one for  $\eta_2$ ):

$$m\ddot{\eta}_1 = - \left( \frac{\partial H_{\text{LGW}}}{\partial \eta_1} \right)_{\eta_2} + \nabla \left( \frac{\partial H_{\text{LGW}}}{\partial (\nabla \eta_1)} \right)_{\eta_2} \quad (5)$$

<sup>1</sup> We note that the labels ‘displacive’ and ‘order–disorder’ have been used in a different way in the literature related to Sn/Ge(111) and Pb/Ge(111). In this paper we restrict ourselves to this ‘definition’, commonly employed when dealing with structural phase transitions.

<sup>2</sup> This is a specific feature of the two-dimensional case. The three-dimensional displacive systems have an appreciable region of applicability of the Landau theory, unlike the three-dimensional order–disorder ones (in the absence of special mechanisms of suppression of critical fluctuations, e.g. long-range forces, which frequently occur).

in the displacive case. In the order–disorder case we have

$$\gamma \dot{\eta}_1 = - \left( \frac{\partial H_{\text{LGW}}}{\partial \eta_1} \right)_{\eta_2} + \nabla \left( \frac{\partial H_{\text{LGW}}}{\partial (\nabla \eta_1)} \right)_{\eta_2}, \quad (6)$$

where  $\gamma$  has the meaning of a generalized kinetic coefficient.

Let us assume that the dynamics of the order parameter corresponds to the displacive case. The frequencies of small vibrations can be obtained after linearization of the corresponding equations of motion close to  $\eta_1 = \eta_2 = 0$  in the symmetric phase, and close to spontaneous values of  $\eta_1$  and  $\eta_2$  in the non-symmetric phase. The vibrations obtained by this procedure seem doubly degenerated in the symmetric phase, but in our case [Pb/Ge(111) and Sn/Ge(111)] this simply means that  $\omega(\mathbf{k}) = \omega(-\mathbf{k})$ . In the non-symmetric phase, these vibrations correspond to the centre of the Brillouin zone and the ‘degeneration’ is removed. This result of the Landau theory depends neither on the highest power of the order parameter taken into account nor on the temperature dependence of the coefficients, and is valid beyond the region of its applicability as well.

According to the above ‘naive’ approximation, ‘soft modes’, i.e., frequencies tending to zero at the phase transition point, should be observed. It is worth noticing that, in fact, no system is known where this ‘naive’ expectation for a displacive transition is fulfilled. Experimentally, the order parameter dynamics is always more complicated than the prediction given by equation (5). The soft mode frequency is always finite at the phase transition point, and it is accompanied by the so-called central peak (see, e.g., [25]).

The nature of this central peak is controversial, and its origin can be different in different systems. The simplest example where the central peak appears is in the case of coupling of the order parameter to a system of hopping defects [13, 26, 27]. This implies a relaxational order-parameter dynamics at low enough frequencies. This seems to be the case of the Sn/Ge(111) interface, as we shall see at some length below.

**3.1.4. Large fluctuations.** In the critical (scaling) region the temperature, wavevector and other dependences are power-like with exponents depending on the form of the LGW Hamiltonian, i.e., on the symmetry properties of the order parameter of the system.

The LGW Hamiltonian equation (1) coincides with the Hamiltonian of the three-state Potts model. The critical behaviour of this model is as follows [28]. The order parameter modulus vanishes at the critical temperature  $T_c$  as  $\sim t^\beta$  with  $\beta = 1/9$ , where  $t = (T - T_c)/T_c$  is the reduced temperature. The order parameter correlation length  $\xi$  diverges as  $\sim |t|^{-\nu}$  with  $\nu = 5/6$ , and the susceptibility  $\chi$  as  $\sim |t|^{-\gamma}$  with  $\gamma = 13/9$ . Critical indices  $\beta$  and  $\nu$  have been confirmed in an He atom scattering experiment for Sn/Ge(111) [29]. This is fairly surprising because it implies that the frozen-in defects do not influence or are practically absent in the system. We recall that for three-dimensional systems these defects seem to overshadow the critical behaviour [13].

The LGW Hamiltonian equation (2) coincides with the Hamiltonian of the XY model with sixth-order anisotropy. The critical behaviour of this model is completely different from the

three-state Potts model. Upon lowering the temperature, first an upper critical temperature is crossed, reaching a Kosterlitz–Thouless region of critical points with continuously varying exponents. Then, a second critical temperature is crossed into an ordered phase [30]. Unfortunately, data on temperature dependence of the order parameter in Pb/Ge(111) are not available at the moment. It would be very interesting to compare the critical behaviour of the two systems.

### 3.2. Defects

It has been assumed up to now that we are dealing with perfect interfaces. However, defects seem to play a ‘special role’ in the phase transitions that we are considering, as seems to be explicitly revealed for the Sn/Ge(111) interface [31]. A theoretical discussion on this ‘special role’ has been made on the basis of numerical calculations [32], which makes it somewhat difficult to extract the relevant information. On the other hand, we note that experimental data [31] also reveal some interesting features not yet discussed in detail (e.g., the absence of anomaly in the characteristic length of the defect-induced order-parameter modulation at the transition point). In the following we summarize the continuum-media approach and further develop it to explain these features.

**3.2.1. Overview.** It is convenient to discuss first some general aspects. Defects can be treated as a perturbation of the ideal crystal matrix whenever the defect concentration is much less than the concentration of the host atoms. The characteristic length of such a perturbation is the correlation length [13]. Its value considerably exceeds the interatomic distance when approaching the phase transition. The defect perturbation can then be treated within a continuum-media approximation. Within this approximation, the system is described by the Landau thermodynamic potential. The defects are modelled as local changes in the corresponding coefficients (non-symmetry-breaking defects) and/or local appearance of new terms (symmetry-breaking defects).

When dealing with the phase transition anomalies, the most important contribution of the non-symmetry-breaking defects is given by

$$A(\mathbf{r})\eta^2 \quad (7)$$

in the corresponding Landau potential (we consider here a one-component order parameter for the sake of illustration). The function  $A(\mathbf{r})$ , describing a defect-induced inhomogeneity, can normally be understood as a local change of the transition temperature. In consequence, this type of defect is known as *random local transition temperature defects* or *T-defects*.

In the case of symmetry breaking defects, the most important term induced is linear in  $\eta$ :

$$h(\mathbf{r})\eta. \quad (8)$$

As a result of this term, a non-zero value of  $\eta$  arises even above

the phase transition. The function  $h(\mathbf{r})$  plays the role of the defect-induced field conjugated to the order parameter. Indeed, in this case, one usually speaks of *random local field defects*. These two types of defects are the only ones relevant for further discussion.

We have so far tacitly assumed that defect parameters do not vary with time, i.e., that they are ‘frozen-in’ defects. However, depending on the relation between the characteristic measurement time and the relaxation time of the defects (i.e. the time needed by a defect to abandon its initial state), it may be necessary to take into account possible changes in the defect state. In this case we are dealing with the so-called *mobile* or *hopping* defects. It is well known, for instance, that hopping symmetry-breaking defects strongly affect the corresponding phase transition temperature [26]. Experimental examples of such an influence are well documented in  $\text{KTaO}_3:\text{Li}$  [33]. This crystal is an incipient ferroelectric in the absence of Li dopants, i.e. its ‘transition temperature’ is negative and close to zero. The Li dopants act as orientable dipoles (hopping random-local-field defects). As a result of these new degrees of freedom, the resulting transition temperature may be high enough to permit that the phase transition takes place. This phenomenon, i.e. the increase of the corresponding phase transition temperature due to the presence of hopping defects, also seems to be observed in the Sn/Ge(111) interface [31]. In the next section we analyse this observation and related phenomena in detail.

**3.2.2. Defects and the  $\sqrt{3} \leftrightarrow (3 \times 3)$  transition in Sn/Ge(111).** The transition temperature of the  $\sqrt{3} \leftrightarrow (3 \times 3)$  phase transition in Sn/Ge(111) is obtained in [31] as the temperature where the  $(3 \times 3)$ -ordered region of the structure extrapolates to infinity. We note that the  $(3 \times 3)$  ordering was assumed to be induced by symmetry-breaking defects. A remarkably different temperature is obtained from He atom scattering experiments [29]. As a result, microscopic models were proposed in order to explain this difference [32]. In this section we discuss possible causes of this apparent discrepancy. In particular, we show that it can be one more example of the above mentioned phenomenon connected to hopping defects (a similar behaviour was theoretically predicted [26] and experimentally observed [33] in several other systems a long time ago). First of all, it is convenient to analyse the specific features of the defect-induced order-parameter modulation expected in this case, irrespective of the hopping of the defects.

*(a). Defect-induced order-parameter modulation.* Let us consider a defect inducing an order-parameter modulation such that  $\eta_1 \neq 0$ ,  $\eta_2 = 0$ . In the linear approximation (see, e.g., [13, 27]), the equation for this defect-induced order parameter  $\eta_{\text{def}}$  reads

$$(A + 2s\eta_{1,\infty} + 3B\eta_{1,\infty}^2)\eta_{\text{def}} - D\nabla^2\eta_{\text{def}} = h\delta(\mathbf{r}) \quad (9)$$

in accordance with equation (1), where  $h$  is the defect strength and  $\eta_{\infty}$  represents the (constant) order parameter far enough from the defect. Let us assume that far enough from the defect  $\eta_2 = 0$  also (which implies  $s < 0$ ). Then  $\eta_{1,\infty}$  can be taken as

$$\eta_{1,\infty} = \begin{cases} 0 & \text{(symmetric phase),} \\ \frac{|s|}{2B} \left( 1 + \sqrt{1 - 4\frac{AB}{s^2}} \right) & \text{(non-symmetric phase),} \end{cases} \quad (10)$$

and the (first-order) phase transition takes place at  $A_c = \frac{2}{9} \frac{s^2}{B}$ . The latter condition, which can be obtained from the continuity of the free energy at the transition point, defines the corresponding transition temperature  $T_c^0$ .

Within this linear approximation,  $\eta_{\text{def}}$  is given by

$$\eta_{\text{def}}(\rho) = \frac{h}{2\pi D} K_0(\rho/\rho_c), \quad (11)$$

where  $K_0$  is the MacDonal function,  $\rho$  is the distance to the defect and

$$\rho_c = \begin{cases} (D/A)^{1/2} & (A > A_c), \\ [D/(A + 2s\eta_{1,\infty} + 3B\eta_{1,\infty}^2)]^{1/2} & (A < A_c). \end{cases} \quad (12)$$

In the absence of defects, the magnitude  $\rho_c$  coincides with the characteristic length of the thermal fluctuations of the order parameter, so it represents the ‘bare’ correlation length. It is worth mentioning that the defect-induced order parameter amplitude (11) decays as  $\sim(\rho_c/\rho)^{1/2} \exp(-\rho/\rho_c)$  for  $\rho \gg \rho_c$ . In [31, 32], however, a different form  $\sim \exp(-\rho/\rho_c)$  was used to fit the experiments.

In equation (11) we see that, as long as the transition is of first order ( $s \neq 0$ ), the width of the defect-induced modulation does not become infinite at the transition point. Such a divergence is possible only if the transition is continuous ( $s = 0$ ) and takes place at  $A_c = 0$ , and a similar situation takes place if the defects are hopping defects, as they are considered in [31, 32]. In this case one has to bear in mind that, even if the phase transition is continuous, it takes place at a point where the coefficient  $A$  is not zero [13, 27]. Consequently, although the magnitude  $\rho_c$  can be quite large, it should remain finite at the transition point. Let us examine this question in more detail.

(b). *Free energy contribution due to the system of hopping defects: shift of the transition temperature.* The Sn/Ge(111) interface presents a significant concentration ( $\sim 1\%$ ) of Ge substitutional atoms and vacancies [32]. These substitutionals and vacancies may act as hopping random-local-field defects: depending on the position where one such a defect was located, one of the three possible ‘domains’ of the  $(3 \times 3)$  phase is induced [31, 32]. It is known that hopping defects result in new variables of the system that, being linearly coupled to the order parameter [13, 27], give rise to an increase of the corresponding transition temperature. Let us estimate such an increase for the case of Sn/Ge(111).

Following e.g. [13, 27], the contribution to the free energy of the system due to the hopping defects can be written as

$$\Phi_{\text{def}} = \sum_i N_i \Phi_i + \Delta \Phi_{\text{def}}, \quad (13)$$

where  $N_i$  is the number of defects inducing the  $i$  ‘domain’ ( $i = 1, 2, 3$  in Sn/Ge(111)),  $\Phi_i$  are the corresponding

contributions to the energy due to isolated defects and  $\Delta \Phi_{\text{def}}$  is the contribution associated with the fluctuations in  $N_i$  (i.e. the entropy of the system of (independent) defects). To further progress, it can be assumed that far enough from the defects the order parameter takes the value  $(\eta_1, \eta_2) = (\eta_\infty, 0)$ . Thus, the above energies can be taken as

$$\Phi_1 \simeq \Phi_d(\eta_\infty - \eta_d)^2, \quad (14a)$$

$$\Phi_2 = \Phi_3 \simeq \Phi_d[(\eta_\infty + \frac{1}{2}\eta_d)^2 + \frac{3}{4}\eta_d^2], \quad (14b)$$

where  $\Phi_d \simeq \pi D \ln^{-1}[\pi \rho_c/(2d)]$  and  $\eta_d \simeq \eta_{\text{def}}(d)$ , with  $d$  indicating the dimensions of the defect core (which can be taken as an atomic distance) [27].

Close to the transition point, the contribution  $\Delta \Phi$  due to fluctuations can be calculated as follows. Let  $w$  be the probability of finding the system in a given configuration of defects,  $N$  the total number of defects and  $P_i$  the probabilities that a given defect is in the corresponding  $i$  state. We then have

$$w = \frac{N!}{\prod_{i=1}^3 N_i!} \prod_{i=1}^3 P_i^{N_i}. \quad (15)$$

By putting  $N_i = (N/3)(1 + \xi_i)$ , with  $\xi_i \ll 1$  and  $\sum_i \xi_i = 0$ , and assuming that  $N \gg 1$ , we further get

$$\Delta \Phi_{\text{def}} = -T \ln w \approx \frac{NT}{6} (\xi_1^2 + \xi_2^2 + \xi_3^2). \quad (16)$$

The free energy per unit volume can then be written as

$$\Phi_{\text{tot}} \simeq \Phi_0 + \frac{A_{\eta\eta}}{2} \eta_\infty^2 + \frac{s}{3} \eta_\infty^3 + \frac{B}{4} \eta_\infty^4 + A_{\eta\xi} \eta_\infty \xi_1 + \frac{nT}{6} (\xi_1^2 + \xi_2^2 + \xi_3^2), \quad (17)$$

where  $A_{\eta\eta} = A + n\Phi_d$  and  $A_{\eta\xi} = -n\Phi_d\eta_d$ , with  $n$  being the concentration of defects. As we see, the presence of hopping defects results in both a renormalization of the coefficient  $A$ , and the appearance of the new variables  $\xi_i$  (accounting for the influence of the new degrees of freedom associated with the hopping of the defects), linearly coupled with  $\eta_\infty$ . The former is due to the energy associated with the presence of defects itself (it comes from the first sum in equation (13)). As a result of the linear coupling between  $\xi_1$  and  $\eta_\infty$ , the phase transition temperature is increased (see below).

Given the form of the free energy (17), and taking into account that  $\sum_i \xi_i = 0$ , we then have  $\xi_2 = \xi_3 = -\xi_1/2$ . Consequently,

$$\Phi_{\text{tot}} = \Phi_0 + \frac{A_{\eta\eta}}{2} \eta_\infty^2 + \frac{s}{3} \eta_\infty^3 + \frac{B}{4} \eta_\infty^4 + A_{\eta\xi} \eta_\infty \xi_1 + \frac{A_{\xi\xi}}{2} \xi_1^2, \quad (18)$$

where  $A_{\xi\xi} = nT/2$ . Further minimization over  $\xi_1$  yields

$$\Phi_{\text{tot}} = \Phi_0 + \frac{A^*}{2} \eta_\infty^2 + \frac{s}{3} \eta_\infty^3 + \frac{B}{4} \eta_\infty^4, \quad (19)$$

where  $A^* = A_{\eta\eta} - A_{\eta\xi}^2/A_{\xi\xi}$ . This implies that the actual transition temperature is

$$T_c \sim T_c^0 + \frac{n}{n_{\text{at}}} \left( \eta_d^2 \frac{\Theta}{T_c} - 1 \right) \Theta, \quad (20)$$

where  $T_c^0$  is the transition temperature in the absence of defects and  $n_{\text{at}} = l_{\text{at}}^{-2}$ .

In order–disorder systems, this transition temperature almost coincides with  $T_c^\circ$ , as long as  $\Theta \sim T_c$  for these systems and the defect parameters are such that  $\eta_d^2 < 1$ ,  $n/n_{\text{at}} \ll 1$ . In displacive systems, however, this temperature may be quite different from  $T_c^\circ$ : with  $\eta_d^2 \sim 1$  and  $n/n_{\text{at}} \sim 10^{-4}$ , for instance, we obtain  $T_c - T_c^\circ \sim 100$  K (recall that  $\Theta \sim T_{\text{at}} \sim 10^4\text{--}10^5$  K in displacive systems). This latter possibility can explain the two different temperatures found in [31] and [29].

(c). *Characteristic length of the defect-induced modulations: possibility of no anomaly at the phase transition point.* It is worthwhile to explore further consequences of this separation of the characteristic temperatures  $T_c^\circ$  and  $T_c$ . As exposed above, the two temperatures can be very different in displacive systems. Let us focus our attention on the characteristic length of the defect-induced order-parameter modulation. The temperature dependence of this length has been studied in [31] for the Sn/Ge(111) interface. The slope of the squared inverse of this length does not change when crossing the real transition temperature. This is quite surprising. In a pure sample and within the Landau theory, this slope is expected to be  $A'_{\eta\eta}/D$  in the symmetric phase, where primes denote the derivative with respect to temperature (see equation (12)). In the non-symmetric phase, however, it is expected to be  $-2A'_{\eta\eta}/D$  if the transition is continuous. Similarly, some anomaly should also be observed if the transition is discontinuous. As we shall see, the explanation of what is observed in [31] could be connected to the above mentioned separation of characteristic temperatures due to hopping defects.

To analyse the particular case of Sn/Ge(111) is somewhat cumbersome, and not very instructive. The relevant effects can be more easily revealed by analysing the case of a continuous transition with a one-component order parameter (two domains). In this case, the equation for the defect-induced order-parameter modulation can be written as

$$(A + 3B\eta_\infty^2)\eta_{\text{def}} - D\nabla^2\eta_{\text{def}} = \pm h\delta(\mathbf{r}), \quad (21)$$

where the different signs account for the two possible different states of the defect. The solution to this equation is of the form equation (11), with

$$\rho_c = \begin{cases} (D/A)^{1/2} & \text{(symmetric phase),} \\ [D/(A + 3B\eta_\infty^2)]^{1/2} & \text{(non-symmetric phase).} \end{cases} \quad (22)$$

To find the phase transition point and the value  $\eta_\infty$  taking into account the influence of hopping defects, we can proceed as in section 3.2.2b. In order to reveal the effects we are interested in, it is necessary to compute the free energy of the system taking into account fourth-order terms in the variables  $\xi_i$  associated with the hopping defects. As a result, we find that

$$\eta_\infty^2 = \begin{cases} 0, & \text{if } A_{\eta\eta} > A_{\eta\xi}^2/A_{\xi\xi}, \\ -\frac{A_{\eta\eta} - A_{\eta\xi}^2/A_{\xi\xi}}{\tilde{B}}, & \text{if } A_{\eta\eta} < A_{\eta\xi}^2/A_{\xi\xi}, \end{cases} \quad (23)$$

where  $A_{\eta\eta} = A + 2n\Phi_d$ ,  $A_{\eta\xi} = -2n\Phi_d\eta_d$ ,  $A_{\xi\xi} = nT$  and  $\tilde{B} = B + B_\xi A_{\eta\eta}^3/(A_{\xi\xi} A_{\eta\xi}^2)$ ; with  $\Phi_d \simeq \pi D \ln^{-1}[\pi\rho_c/(2d)]$ ,  $\eta_d = \eta_{\text{def}}(d)$  and  $B_\xi = nT/3$  ( $n$  is the concentration of defects and  $d$  indicates the dimensions of the defect core). As

we see, the phase transition temperature  $T_c$  is now defined by the condition  $A_{\eta\eta} = A_{\eta\xi}^2/A_{\xi\xi}$ . It can be estimated as  $T_c \simeq T_c^\circ + \frac{n}{n_{\text{at}}}(\eta_d^2 \frac{\Theta}{T_c} - 1)\Theta$ , where  $n_{\text{at}} = I_{\text{at}}^{-2}$  and  $T_c^\circ$  would be the transition temperature in the absence of defects. As in the case previously discussed for the Sn/Ge(111) interface, this shift in the transition temperature is negligible in the order–disorder case but it may not be in the displacive case.

We are now in a position to discuss the temperature dependence of the characteristic length of the defect-induced order-parameter modulation. By putting  $A_{\eta\eta} = (A_{\eta\xi}^2/A_{\xi\xi})(1+a)$  we find that, close to the transition point, the characteristic length (22) of the defect-induced order parameter in the non-symmetric phase ( $a \rightarrow 0^-$ ) is actually such that

$$\rho_c^{-2} = \rho_{c,0}^{-2} \left( 1 + a - \frac{3a}{1 + \kappa(1+a)^3} \right), \quad (24)$$

where  $\rho_{c,0}^{-2} = A_{\eta\xi}^2/(A_{\xi\xi} D) \sim \eta_d^2(n/n_{\text{at}})(\Theta/T_c)I_{\text{at}}^2$  and  $\kappa = (B_\xi/B)(A_{\eta\xi}/A_{\xi\xi})^4 \sim \eta_d^4(n/n_{\text{at}})(\Theta/T_c)^3$ . The case where  $\kappa \ll 1$  may occur for weak enough defects and/or for a small enough concentration of them. Indeed, the occurrence of this case is unavoidable in order–disorder systems, where  $\Theta \sim T_c$ . We then have

$$(\rho_c^{-2} - \rho_{c,0}^{-2})/\rho_{c,0}^{-2} \underset{a \rightarrow 0^-}{\approx} -2a, \quad (25)$$

and the above expectation is confirmed: in the non-symmetric phase,  $(\rho_c^{-2})' \simeq -2\rho_{c,0}^{-2}a' \sim -2A'_{\eta\eta}/D$ . But one has to realize that, in a displacive transition, the case where the defect parameters are such that  $\kappa \gg 1$  is also well possible. This implies a fairly high concentration of relatively strong defects:  $\eta_d^4(n/n_{\text{at}}) \gg 10^{-6}\text{--}10^{-9}$ . In this case:

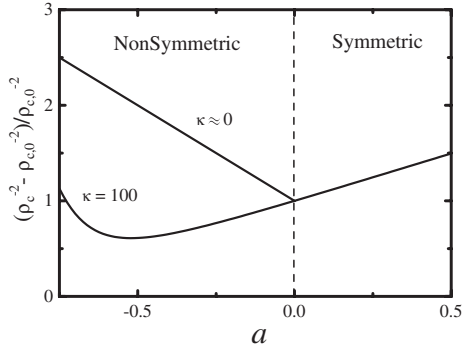
$$(\rho_c^{-2} - \rho_{c,0}^{-2})/\rho_{c,0}^{-2} \underset{a \rightarrow 0^-}{\approx} a + (9/\kappa)a^2 \quad (26)$$

and, consequently, by crossing the transition point no change in the slope of  $\rho_c^{-2}$  will be observed. Indeed no change is expected up to temperatures such that  $9|a|/\kappa \sim 1$ . This could imply temperatures of the order of the phase transition temperature itself [34]<sup>3</sup>.

It is worth noticing that in these estimates no ‘special’ values of the coefficients are chosen, but a high enough concentration of defects and/or a high enough strength of them (within the applicability of the theory). Therefore, this effect may occur in another displacive phase transition provided that hopping defects are present. Indeed it can be seen that it is just the case for the (displacive) transition in the Sn/Ge(111) interface. To reproduce the corresponding increment in the phase transition temperature here, the range of defect parameters is such that  $\eta_d^2(n/n_{\text{at}}) \sim 10^{-4}$ . This falls into the latter case where  $\kappa \gg 1$  (it is possible that, e.g.,  $\eta_d^2 \sim 1$  and  $n/n_{\text{at}} \sim 10^{-4}$  and, consequently,  $\kappa \sim 100$ ). Thus, this could explain the observation reported in [31] of the lack of anomaly associated with the crossing of the transition temperature in the defect-induced modulation of the order parameter (see figure 5).

<sup>3</sup> At this point, one should notice that, depending on the defect parameters, our formalism to treat the influence of defects is justified close to the real transition temperature ( $T \rightarrow T_c$ ), but it might not be in the immediate vicinity of the bare transition point ( $T \rightarrow T_c^\circ$ ). The corresponding criterion of applicability can be obtained following Levanyuk and Sigov [34].





**Figure 5.** Possible behaviours of the characteristic length of the defect-induced modulation of the order parameter.

### 3.2.3. Defects and phase transition anomalies.

(a). *Frozen-in defects.* Frozen-in defects give rise to deviations in phase transition anomalies from the behaviour expected for perfect systems. Elementary considerations indicate that these deviations should be similar to those due to thermal fluctuations. Random-local-field defects can be regarded as something like ‘frozen fluctuations’ of the order parameter, so their influence can be even stronger. In fact, it can be proved that, in the region of applicability of the first-order corrections to the Landau theory, there exists a crossover between a region where the anomalies are driven by thermal fluctuations (far from the transition point) to a region where they are driven by defects (not so far from the transition point) [13]. For the sake of illustration, let us consider the x-ray scattering intensity in the vicinity of a Bragg peak (diffuse scattering):  $I(\mathbf{k}) \propto \langle \eta(\mathbf{k})\eta(-\mathbf{k}) \rangle$ . This intensity contains the contributions due to both thermal and defect-driven fluctuations. The two contributions are additive in the region of applicability of the first-order corrections to the Landau theory [13]. Thermal fluctuations of the order parameter are such that

$$\langle \eta(\mathbf{k})\eta(-\mathbf{k}) \rangle_T = \frac{T}{A + 3B\eta_0^2 + Dk^2} \quad (27)$$

while the defect-driven fluctuations are

$$\langle \eta(\mathbf{k})\eta(-\mathbf{k}) \rangle_{\text{def}} = \frac{nh^2}{(A + 3B\eta_0^2 + Dk^2)^2}. \quad (28)$$

(Hereafter we restrict ourselves to continuous transitions.) Here  $n$  is the defect concentration,  $h$  is the corresponding defect strength and  $\eta_0$  is the value of equilibrium of the modulus of the corresponding order-parameter ( $\eta_0 = 0$  in the symmetric phase). In this case, the phase transition point is defined by the condition  $A = 0$ . The different dependences on  $A + 3B\eta_0^2 + Dk^2$  in these expressions permit the above mentioned crossover. For small wavevectors, it takes place at  $A_{\text{cross}} \sim nh^2/T_c$  in the symmetric phase. That is  $(T_{\text{cross}} - T_c)/T_c \sim (n/n_{\text{at}})(T_{\text{at}}/T_c)^2$  for strong defects.

To date there is no theory taking into account the presence of defects, analogous to the theory developed for the scaling region in perfect systems [35]. However, there is no reason to expect that in this region the anomalies are driven by thermal fluctuations only. Indeed, there is no experimental example

in 3D systems where the thermal-fluctuation-driven critical behaviour is observed unambiguously [36]. There are also examples in 2D of this difficulty [37, 38]. However, in other interfaces which are far from being perfect, this behaviour seems to have been observed [29, 39].

(b). *Hopping defects.* As we have seen, defects can be understood as additional degrees of freedom of the system (see, e.g., [40, 41]). Then, if a system contains hopping defects only, the corresponding critical behaviour is obtained from the theory for a perfect crystal. The presence of both frozen-in and hopping defects, being more natural, is more intriguing. When computing the critical scattering, for instance, one must bear in mind that, as we have seen, there can be a significant increase of the transition temperature, which is not observed in the defect-induced order-parameter modulations. This implies some dependence of the coefficients of the corresponding Landau potential on the distance or, equivalently, some wavevector dispersion of these coefficients:

$$A \longrightarrow A(\mathbf{k}) = \begin{cases} \tilde{A} & (k \ll l_d^{-1}), \\ A & (k \gg l_d^{-1}), \end{cases} \quad (29)$$

where  $\tilde{A}$  results from the above mentioned renormalization due to the hopping of the defects ( $\tilde{A} = 0$  at the transition point), and  $l_d$  is the characteristic distance between defects. This dispersion must be taken into account when computing the thermal fluctuations of the order parameter, i.e.,  $A$  must be replaced by  $\tilde{A}$  in equation (27) for small enough wavevectors. The fluctuations due to frozen-in-defects, however, are not affected by this dispersion as long as they are connected to the properties of the system at distances smaller than the distance between defects, i.e., equation (28) remains valid. In consequence, the defect contribution to the diffuse scattering, unlike to the thermal one, will show no anomaly at the real transition point, but it will do close to the minimum of  $A + 3B\eta_0^2$ .

## 4. Conclusions

The  $\sqrt{3} \leftrightarrow (3 \times 3)$  phase transitions occurring in Pb/Ge(111) and Sn/Ge(111) interfaces are studied within the phenomenological Landau theory and its extensions. Symmetry arguments reveal that the critical behaviours theoretically expected for these transitions are essentially different. The influence of defects in these transitions is discussed in detail. We propose that the experimental observation of no anomaly of the ‘bulk’ correlation length at the phase transition temperature in Sn/Ge(111) is explained as a result of the presence of hopping defects. This phenomenon is shown to be of general character, and could also be observed in other systems.

## Acknowledgments

We thank I Brihuela and J M Gómez-Rodríguez for useful discussions. EGM is funded by Comunidad de Madrid (grant GR/MAT/0022/2004).

## References

- [1] LeLay G and Metois J J 1983 *Appl. Surf. Sci.* **17** 131
- [2] Metois J J and LeLay G 1983 *Surf. Sci.* **133** 422
- [3] Ichikawa T 1983 *Solid State Commun.* **46** 827
- [4] Ichikawa T 1984 *Surf. Sci.* **140** 37
- [5] Gothelid M, Hammar M, Tornevik C, Karlsson U O, Nilsson N G and Flodstrom S A 1992 *Surf. Sci. Lett.* **271** L357
- [6] Pedersen J S, Feidenhans'l R, Nielsen M, Kjaer K, Grey F and Johnson R L 1987 *Surf. Sci.* **189** 1047
- [7] DiCenzo S B, Bennett P A, Tribula D, Thiry P, Wertheim G K and Rowe J E 1985 *Phys. Rev. B* **31** 2330
- [8] Carlisle J A, Miller T and Chiang T C 1993 *Phys. Rev. B* **47** 3790
- [9] Carlisle J A, Miller T and Chiang T C 1993 *Phys. Rev. B* **47** 10342
- [10] Carpinelli J M, Weitering H H, Plummer E W and Stumpf R 1996 *Nature* **381** 398
- [11] Carpinelli J M, Weitering H H, Bartkowiak M, Stumpf R and Plummer E W 1997 *Phys. Rev. Lett.* **79** 2859
- [12] Ortega J, Pérez R and Flores F 2002 *J. Phys.: Condens. Matter* **14** 5979
- [13] Levanyuk A P and Sigov A S 1987 *Defects and Structural Phase Transitions* (New York: Gordon and Breach)
- [14] Bunk O, Zeysing J H, Falkenberg G, Johnson R L, Nielsen M, Nielsen M M and Feidenhans'l R 1999 *Phys. Rev. Lett.* **83** 2226
- [15] Zhang J, Ismail, Rous P J, Baddorf A P and Plummer E W 1999 *Phys. Rev. B* **60** 2860
- [16] Mascaraque A, Avila J, Alvarez J, Asensio M C, Ferrer S and Michel E G 1999 *Phys. Rev. Lett.* **82** 2524
- [17] Mascaraque A, Alvarez J, Avila J, Asensio M C, Ferrer S and Michel E G 2000 *Surf. Sci.* **454–456** 191
- [18] Rottman C 1981 *Phys. Rev. B* **24** 1482
- [19] Schick M 1981 *Prog. Surf. Sci.* **11** 245
- [20] Alexander S 1975 *Phys. Lett. A* **54** 353
- [21] Pippard B A 1950 *Proc. R. Soc. A* **203** 210
- [22] Levanyuk A P 1959 *Sov. Phys.—JETP* **9** 571
- [23] Ginzburg V L 1960 *Sov. Phys.—Solid State* **2** 1824
- [24] Strukov B A and Levanyuk A P 1998 *Ferroelectric Phenomena in Crystals* (Berlin: Springer)
- [25] Ginzburg V, Levanyuk A and Sobyenin A 1980 *Phys. Rep.* **57** 151
- [26] Halperin B I and Varma C M 1976 *Phys. Rev. B* **14** 4030
- [27] Levanyuk A P 1979 *Zh. Eksp. Teor. Fiz.* **76** 345  
Levanyuk A P 1979 *JETP* **49** 176 (Engl. Transl.)
- [28] Baxter R J 1982 *Exactly Solved Models in Statistical Mechanics* (London: Academic)
- [29] Floreano L, Cvetko D, Bavdek G, Benes M and Morgante A 2001 *Phys. Rev. B* **64** 075405
- [30] José J V, Kadanoff L P, Kirkpatrick S and Nelson D R 1977 *Phys. Rev. B* **16** 1217
- [31] Melechko A V, Braun J, Weitering H H and Plummer E W 1999 *Phys. Rev. Lett.* **83** 999
- [32] Melechko A V, Simkin M V, Samatova N F, Braun J and Plummer E W 2001 *Phys. Rev. B* **64** 235424
- [33] Vugmeister B E and Glinchuk M D 1990 *Rev. Mod. Phys.* **62** 993
- [34] Levanyuk A P and Sigov A S 1988 *Phase Transit.* **11** 91
- [35] Dotsenko V S 1995 *Phys. Usp.* **38** 457
- [36] Ginzburg V, Levanyuk A and Sobyenin A 1987 *Ferroelectrics* **73** 171
- [37] Roelofs L D, Kortan A R, Einstein T L and Park R L 1981 *Phys. Rev. Lett.* **46** 1465
- [38] Family F 1982 *Phys. Rev. Lett.* **48** 367
- [39] Nakayima Y, Voges C, Nagao T, Hasegawa S, Klos G and Pfnür H 1997 *Phys. Rev. B* **55** 8129
- [40] Ma S 1976 *Modern Theory of Critical Phenomena* (London: Benjamin)
- [41] Patashinsky A Z and Pokrovsky V L 1979 *Fluctuation Theory of Phase Transitions* (Oxford: Pergamon)