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Single-particle properties of systems undergoing structural phase transitions

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We study the properties of the single-particle probability density function for a d-dimensional model system exhibiting a structural phase transition. Using renormalization-group methods (in $d = 1$) and exact inequalities (in $d = 3$), we examine, in particular, the extent to which the behavior of this function can reveal the distinctive changeover in dynamical behavior expected for a system exhibiting a displacive phase transition.

I. INTRODUCTION

It is only in the last few years that some of the subtleties in the dynamical properties of systems undergoing structural phase transitions have come to be appreciated. Much of the recent activity in this field can be traced to some inelastic-neutron-scattering experiments[1] performed on the perovskite $SrTiO_3$, widely regarded as the paradigm example of a system exhibiting a displacive phase transition. These experiments revealed a soft-mode spectral function displaying, *in addition to* the expected soft-mode sidebands, a narrow central component which was found to persist over a wide temperature range *above* T_c. While there is some evidence to suggest that at least some (perhaps most) of this scattering is attributable to crystal imperfections (e.g., a surface strain[2]), the occurrence of the phenomenon in molecular-dynamics investigations of one-dimensional[3] and two-dimensional[4] model systems leaves no doubt that the effect is, at least to some extent, intrinsic to the ideal crystal dynamics.

Of the many and varied explanations of the phenomenon,[5] perhaps the most intuitively appealing are those[4,6,7] which correlate it with the existence (even for $T > T_c$) of regions in the crystal in which the local order parameter is nonzero for a time scale long in comparison with typical inverse phonon frequencies. Such clusters of local order are revealed in the computer simulations referred to above and in the explicit analyses of a one-dimensional system performed by Krumhansl and Schrieffer[6] and by Aubry.[3,7] The work of these authors is strongly suggestive of a picture (clearly described by Beck[8]) in which the[9] central peak is attributed to (relatively slow) fluctuations in the local order parameter associated with each cluster, while the soft-mode sidebands reflect the more conventional (and relatively fast) fluctuations about the local equilibrium positions within the clusters.

Unfortunately, despite their conceptual appeal, it has not yet proved possible to set these ideas in a firm theoretical framework. The lack of progress reflects the difficulties characterizing the treatment of the *dynamics* of nonlinear systems by the nonperturbative methods which the one-dimensional analyses[3,6,7] suggest are necessary to capture the intrinsically nonlinear clusterlike solutions. In contrast, the theory of the *static* (equilibrium) properties of such systems has (naturally!) proved somewhat more tractable and has progressed further, particularly with the development of renormalization-group (RG) methods.[10]

In the light of this situation it seems reasonable to examine in some detail those aspects of static crystal properties that can illuminate the nature of the underlying dynamical phenomena. In this paper we consider one such property, namely, the single-particle probability density function $P(X_1)$, where X_1 is a local, continuous variable describing the displacement of an atom from its high-temperature-phase equilibrium position. The motivation for studying this property in particular is twofold.

Firstly, this probability density function is of immediate relevance to experiments which probe the local properties of systems undergoing structural phase transitions. For example, EPR experiments can measure this function directly,[11] while x-ray scattering experiments reflect the behavior of this function implicitly through its formal Fourier transform—the Debye-Waller factor.

Secondly, it is intuitively clear that the physical picture sketched above has immediate implications for the qualitative character of the probability density function. It suggests that this function should exhibit marked deviations from a Gaussian form,[12] even in the high-temperature phase (to which regime this paper is restricted). More specifically, the tendency for each atom to oscillate about positions of local equilibrium which "anticipate" the structure of the low-temperature phase will naturally enhance the value of the probability function around these positions, producing (if the effect is sufficiently strong) a multipeaked structure.

Prompted by the EPR experiments alluded to above, this equation was investigated some time ago with the aid of computer calculations for a two-dimensional model system.[13] That analysis was unable to establish conclusively whether or not (for the Ising-like $d=2$ system considered) the approach to criticality (from above) is invariably accompanied by the formation of a double-peaked structure in the probability density function. It is this question, and related issues, that we examine in this paper, the format of which we now outline.

In Sec. II we define the (continuous-variable Ising-like) model to be analyzed, and formulate the problem of interest in a way which illuminates the connection between the formation of additional peaks in the probability function, and the establishment of *short-range order* in the system.

In Sec. III we analyze the $d=1$ form of our model with the aid of a direct-space renormalization-group argument. The analysis emphasizes the connection between the formation of a double-peaked probability function and the onset of Ising-like behavior.

In Sec. IV we examine the behavior of the probability density for a $d=3$ system, exploiting inequalities recently derived by Fröhlich *et al.*[14] and by Guerra *et al.*[15]

Finally, our results are summarized and discussed in Sec. V.

II. MODEL AND INTRODUCTORY THEORY

We shall be concerned with the (classical) equilibrium properties of the system with configurational energy

$$V(\{X\}) \equiv \sum_l V_S(X_l) + \tfrac{1}{2} C \sum_{l,l'}^{\mathrm{nn}} (X_l - X_{l'})^2 . \tag{1}$$

Here $\{X\}$ denotes a set of local coordinates describing the displacements of atoms about their high-temperature-phase equilibrium positions. The sum over l runs over the sites of a d-dimensional hypercubic lattice, of periodicity a. The scalar (Ising-like) nature of the variables (supposed for simplicity) may be regarded, physically, as expressing a uniaxial anisotropy, while the single-particle potential V_S may be thought of as arising from an underlying sublattice of atoms which do not participate in the phase transition. The second term describes interactions between all nearest-neighbor (nn) pairs of coordinates.

It is appropriate to note at this point an alternative formulation of Eq. (1) favored by certain authors,[16] who write the model potential energy as

$$V(\{X\}) = \sum_l \tilde{V}_S(X_l) - C \sum_{l,l'}^{\mathrm{nn}} X_l X_{l'} \tag{2}$$

with the "single-particle potential" given by

$$\tilde{V}_S(X) = V_S(X) + 2dCX^2 . \tag{3}$$

While this representation has the advantage that it emphasizes the correspondence with the (continuous-spin) Ising model, the translational invariance of the "interaction" term in the former representation is more in keeping with the lattice-dynamical picture underlying our model.

In the notation of Eq. (1) (which we shall follow throughout this paper) a necessary condition for the existence of a phase transition is that[17]

$$V_S''(0) < 0 , \tag{4}$$

which has the implication that the single-particle potential has (at least) two minima.[18] In fact, it has recently been demonstrated that Eq. (4) also constitutes a sufficient condition for a phase transition (at nonzero T_c) provided $d \geq 2$.[19]

We define the single-particle probability density function associated with the atom at some lattice site (labeled by $l=1$) as

$$\begin{aligned} P(X_1) &\equiv \int \prod_l{}' dX_l e^{-\beta v(\{X\})} \Big/ \int \prod_l dX_l e^{-\beta V(\{X\})} \\ &\equiv \frac{1}{Z} \int \prod_l dX_l e^{-\beta V(\{X\})} , \end{aligned} \tag{5}$$

where the prime denotes omission, from the functional integration, of the coordinate X_1, and $\beta \equiv 1/k_BT$. To cast this function in a more transparent form consider its first derivative

$$\begin{aligned} P'(X_1) &= \frac{1}{Z} \int \prod_l{}' dX_l \\ &\quad \times \left(-\beta \frac{\partial V}{\partial X_1} (\{X\}) \right) e^{-\beta V(\{X\})} \\ &\equiv \beta P(X_1) F_a(X_1) , \end{aligned}$$

where

$$F_a(X_1) \equiv \int \prod_l{}' \, dX_l \left(-\frac{\partial V(\{X\})}{\partial X_1}\right) e^{-\beta V(\{X\})} \Big/ \int \prod_l{}' \, dX_l e^{-\beta V(\{X\})} \tag{6}$$

$$\equiv \left\langle -\frac{\partial V(\{X\})}{\partial X_1} \right\rangle_{X_1} \tag{7}$$

may be regarded as the average force experienced by the atom if clamped at displacement X_1. Introducing an "adiabatic" potential function by

$$F_a(X_1) \equiv -\frac{\partial V_a(X_1)}{\partial X_1}, \tag{8}$$

the probability density function can be written [from (6)]

$$P(X_1) = D e^{-\beta V_a(X_1)}, \tag{9}$$

where D is a normalization constant. The structure of the single-particle probability function is thus determined by the "adiabatic" potential function characterizing the average environment seen by an atom (when constrained to move infinitely slowly). In particular, the second derivative of $P(X_1)$ is given by

$$P''(0) = -\beta P(0) V_a''(0) \tag{10}$$

and is positive [indicating a two-peaked structure in $P(X_1)$] if the potential function V_a has a double minimum. Insight into the circumstances in which this will occur is afforded by a consideration of the explicit form

$$\begin{aligned} P''(0) &= \beta P(0) \left(\frac{d}{dX_1} \left\langle -\frac{\partial V(\{X\})}{\partial X_1} \right\rangle_{X_1} \right)_{X_1=0} \\ &= \beta P(0) \left[\left\langle \frac{\partial^2 V(\{X\})}{\partial X_1^2} \right\rangle_{X_1=0} + \beta \left\langle \left(\frac{\partial V(\{X\})}{\partial X_1} \right)^2 \right\rangle_{X_1=0} \right] \\ &= -\beta P(0) \left[V_S''(0) + 4dC - 4\beta C^2 \left\langle \left(\sum_\delta X_\delta \right)^2 \right\rangle_{X_1=0} \right]. \end{aligned} \tag{11}$$

Here the sum on δ extends over the second nearest neighbors of the atom at site $l = 1$. According to this equation, systems characterized by parameters such that $V_S''(0) + 4dC < 0$ will invariably exhibit a two-peaked probability density function. It is natural to expect, therefore, that the dynamical behavior of these systems (which have been called "order disorder"[20]) will invariably reflect both the tunneling and oscillatory modes referred to in the Introduction. In contrast, the systems characterized by parameters satisfying

$$|V_S''(0)|/4dC < 1 \tag{12}$$

(which have been termed "displacive"[20]) might be expected to display a more varied behavior. For these systems, at high enough temperatures the single-particle probability function is singly peaked. This corresponds essentially to the fact that, at high temperatures, the motions even of neighboring atoms become uncorrelated, so that each atom actually "sees" an effective potential $\tilde{V}_S(X)$ [cf. Eq. (3)] which, for a displacive system, has $X = 0$ as a position of stable equilibrium. Thus, at high enough temperature the local dynamical behavior of such systems should be adequately described as quasiharmonic oscillations about the high-temperature-phase equilibrium position. At low enough temperatures, however, it is clear from condition (4) that the local dynamical behavior must be better described in terms of quasiharmonic oscillations about one or other of the potential minima (or, at least, about a position somewhere in one or other of the two wells: cf. Sec. V). A crude (rather too crude, as we shall see later) indication of the onset of this "low-temperature" regime would be afforded by the development of a double-peaked structure in $P(X)$. Qualitatively, Eq. (11) makes it apparent that this will occur if the *short-range order*, reflected in the correlation function in (11), becomes sufficiently strong.

The question of fundamental interest is, then, whether or not the transition from the "high-temperature" regime to the "low-temperature" regime invariably *precedes* the phase transition. If the character of the probability function is accepted as a hallmark of the former "transition," the question is then whether or not the *local* instability, reflected in the vanishing of $V_a''(0)$, associated with the onset of (strong) *short-range* order[21] precedes the *macroscopic* instability associated with the onset of *long-range* order.

This is the question we examine generally in Sec. IV, but for the moment we shall focus attention on a $d = 1$ system where a fuller analysis of $P(X_1)$ proves possible. The price of the ease of analysis is, of course, that this system exhibits a phase transition only at $T = 0\,^\circ\mathrm{K}$, so that the "local" instability must, of necessity, precede the macroscopic instability. Nevertheless, we shall see that the analysis offers some appealing insights into the high-to-low-temperature changeover referred to above.

III. PROBABILITY DENSITY FUNCTION FOR $d = 1$

It is by now well known that the static properties of the one-dimensional model system defined by Eq. (1) can be treated essentially exactly by transfer matrix techniques.[22,23] The behavior of the single-particle probability function may, in fact, be obtained by a simple extension of the arguments of this kind presented by Morf and Thomas.[23] We prefer, however, to present a renormalization-group argument which has the advantage that it renders more transparent certain homogeneity properties that characterize the behavior of the system in the displacive limit $V_S''(0)/C \to 0$.[24]

The calculation of a local property, such as $P(X_1)$, is performed most readily with the aid of a *direct*-space RG transformation which is a generalization (to continuous variables) of the decimation scheme (for a fixed-length spin Ising model) described by Nelson and Fisher.[25] The use of a real-space transformation will, furthermore, enable us to avoid certain worrying features of low-dimensionality momentum-space RG treatments.

For the purpose of this calculation we shall choose for the single-atom potential V_S the familiar explicit form

$$V_S(X) = \tfrac{1}{2}\bar{A}X^2 + \tfrac{1}{4}BX^4 . \tag{13}$$

It is convenient to introduce scaled variables

$$Y_l \equiv (\beta C)^{1/2} X_l \tag{14}$$

and new parameters

$$\Delta \equiv \bar{A}/C, \quad u \equiv B/\beta C^2 . \tag{15}$$

The free energy of the $d = 1$ system then assumes the form

$$F = -kT \ln Z = \tfrac{1}{2} NkT \ln(\beta C) - kT \ln Z_N(\Delta, u) , \tag{16}$$

where

$$Z_N(\Delta, u) \equiv \int \prod_{l=1}^{N} dY_l e^{-\mathcal{H}_N(\{Y\}, \Delta, u)} , \tag{17}$$

with the "effective Hamiltonian" defined by

$$\mathcal{H}_N(\{Y\}, \Delta, u) = \tfrac{1}{2}\Delta \sum_{l=1}^{N} Y_l^2 + \tfrac{1}{4}u \sum_{l=1}^{N} Y_l^4 + \sum_{l=1}^{N} (Y_l - Y_{l+1})^2 . \tag{18}$$

The partition function Z_N can be written

$$Z_N = \int \prod_{l=1}^{N} dY_l K(Y_1, Y_2, \Delta, u) K(Y_2, Y_3, \Delta, u) \times \cdots K(Y_N, Y_1, \Delta, u) , \tag{19}$$

where we suppose periodic boundary conditions and introduce (as in Ref. 23) a transfer function

$$K(Y_i, Y_{i+1}, \Delta, u) \equiv \exp\left[-(1 + \tfrac{1}{4}\Delta)(Y_i^2 + Y_{i+1}^2) - \tfrac{1}{8}u(Y_i^4 + Y_{i+1}^4) + 2Y_i Y_{i+1} \right] . \tag{20}$$

The RG transformation consists of taking a partial trace over the N degrees of freedom—we shall integrate over every second displacement coordinate—followed by a rescaling of the remaining coordinates. Explicitly, we introduce the function

$$H(Y_i, Y_{i+2}, \Delta, u) \equiv \int dy_{i+1} K(Y_i, Y_{i+1}, \Delta, u) K(Y_{i+1}, Y_{i+2}, \Delta, u) \tag{21}$$

and define renormalized parameters $\tilde{\Delta}$ and $\tilde{u}$ by demanding that H be written in the form[26]

$$H(Y_i, Y_{i+2}, \Delta, u) = D_0 K(\tilde{Y}_i, \tilde{Y}_{i+2}, \tilde{\Delta}, \tilde{u}, \ldots) . \tag{22}$$

Here D_0 is a constant (whose value it is necessary to compute only if the free energy is required), and the new variables are related to the original variables by a rescaling

$$\tilde{Y}_i = \zeta Y_i . \tag{23}$$

The coordinate rescaling factor ζ is chosen so that the coefficient of the interaction term in the exponent of the transfer function

$$K(\tilde{Y}_i, \tilde{Y}_{i+2}, \tilde{\Delta}, \tilde{u}, \ldots) \tag{24}$$

remains equal to 2 [cf. Eq. (20)].

Before summarizing the results of this procedure we note first its formal implications for the single-particle probability density function of a coordinate, such as Y_1, which is not integrated out in the RG transformation. We write this function, for the original N-variable system, characterized by parameters Δ and u, as

$$\begin{aligned} P_N(Y_1, \Delta, u) &\equiv \frac{1}{Z_N(\Delta, u)} \int \prod_{l=1}^{N}{}' dY_l e^{-\mathcal{H}_N(\{Y\}, \Delta, u)} \\ &= \frac{\zeta}{Z_{N/2}(\Delta, u, \ldots)} \int \prod_{l=1}^{N/2}{}' d\tilde{Y}_l e^{-\mathcal{H}_{N/2}(\{\tilde{Y}\}, \tilde{\Delta}, \tilde{u}, \ldots)} \\ &= \zeta P_{N/2}(\tilde{Y}_1, \tilde{\Delta}, \tilde{u}, \ldots) . \end{aligned} \tag{25}$$

In the thermodynamic limit we may drop the subscript N and, using (23) once more, obtain

$$P(Y_1, \Delta, u) = \zeta P(\zeta Y_1, \tilde{\Delta}, \tilde{u}, \ldots) . \tag{26}$$

The derivation of the recursion relations from Eqs. (21) and (22) is fairly straightforward. We remark only that some care has to be exercised to distinguish between contributions to the operators $\sum_l Y_l^4$ and $\sum_l (Y_l - Y_{l+1})^4$: Although the coefficient of the latter ("anharmonic-bond") term is zero in the starting Hamiltonian, such a term is

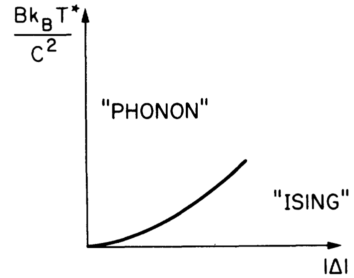

FIG. 1. Dependence upon $|\Delta|$, the deviation from the displacive limit, of the temperature T^*, at which the probability function of the $d=1$ system begins to exhibit two peaks, as given by Eq. (30).

generated by the RG transformation. Near the displacive limit ($\Delta, u \ll 1$) the results may be written as[27]

$$\begin{aligned} \zeta &= b^{-1/2}[1+O(\Delta,u)]\,, \\ \tilde{\Delta} &= b^2[\Delta+O(\Delta^2,u)]\,, \\ \tilde{u} &= b^3[u+O(\Delta^2,u\Delta,u^2)]\,, \end{aligned} \tag{27}$$

where the "decimation factor" $b=2$. The probability density function thus satisfies the homogeneity relation

$$P(Y_1,\Delta,u) \approx b^{-1/2}P(b^{-1/2}Y, b^2\Delta, b^3u, \ldots)\,. \tag{28}$$

Evidently the vanishing of $P^{11}(O)$ will occur at values of the parameters Δ and u satisfying a functional relationship of the form

$$Q(b^2\Delta, b^3u) \approx O\,. \tag{29}$$

Recalling the proportionality of u and T [cf. Eq. (15)], we conclude that the double-peaked structure will appear at a temperature T^* satisfying

$$T^* = Q^*(C^2/Bk_B)\,|\Delta|^{\phi}\,, \tag{30}$$

where $\phi \equiv \frac{3}{2}$ and Q^* is a constant whose value may be determined from the analysis of the Schrödinger-like equation appearing in the transfer-matrix approach of Morf and Thomas, as $Q^* \approx 0.857$.[28] The result (30) is displayed in Fig. 1, and in Table I is compared with the results of explicit molecular-dynamics calculations of the probability function for $d=1$.[29] We shall return to discuss these results in detail in the closing section. For the moment, however, we turn to consider the properties of the probability function in three-dimensional systems.

TABLE I. Temperature T^* (a) as determined by a molecular-dynamics analysis of two models close to the displacive limit (Ref. 29) and (b) compared with the results of Eq. (30).

$\bar{A}$	C	B	$\|\Delta\|$	T^* (a)	T^* (b)
$-\frac{1}{20}$	$\frac{1}{4}$	$\frac{1}{3}$	$\frac{1}{5}$	0.016 ± 0.003	0.014
$-\frac{1}{8}$	$\frac{1}{4}$	$\frac{1}{3}$	$\frac{1}{2}$	0.08 ± 0.02	0.057

IV. PROBABILITY DENSITY FUNCTION FOR $d=3$

The question to which this section is addressed is that raised at the close of Sec. II: Does the existence of a phase transition guarantee a short-range order, at T_c, of sufficient strength to produce a double-peaked probability function? That the answer to this question depends crucially upon the system dimensionality is clear from a qualitative inspection of Eq. (11). For the $d\to\infty$ limit[30] of the model (for which one must then choose a coupling constant C of the order of $1/d$ to obtain a transition at a finite temperature) the correlation term in Eq. (11) is $O(1/d)$, and thus is negligible in comparison with the other terms. Thus, while double peaking of $P(X_1)$ is guaranteed to set in above T_c for $d=1$ (trivially!), it will not do so for "displacive" systems with $d=\infty$ [cf. Eq. (12)].

To analyze the situation in more interesting (and less tractable!) dimensionalities it is convenient first of all to express the correlation function of interest [that appearing in (11)] in a slightly different form.

It is simple to show that

$$\left\langle \left(\sum_{\delta}\right) X_{\delta} \right\rangle^{2}_{X_1=0} \equiv \left\langle \left(\sum_{\delta} X_{\delta}\right)^{2} \right\rangle_{r}\,, \tag{31}$$

where $\langle\cdots\rangle_r$ denotes an average in the ensemble associated with the potential

$$V_r(\{X\}) = \sum_{l} \tilde{V}_S(X_l) - C \sum_{l,l'}^{\mathrm{nn}}{}' X_l X_{l'}\,, \tag{32}$$

where the prime denotes omission, from the sum, of those terms involving the atom $l=1$.[31] Now, according to a theorem originally proved for fixed-length Ising spins by Griffiths[32] and generalized to the classical systems of interest here by Guerra, Rosen, and Simon,[15] reducing the strength of one or more bonds in a system characterized by ferromagnetic interactions cannot result in an increase of any correlation function. It follows that

$$\left\langle \left(\sum_{\delta} X_{\delta}\right)^{2} \right\rangle_{r} \leqslant \left\langle \left(\sum_{\delta} X_{\delta}\right)^{2} \right\rangle. \tag{33}$$

It is now convenient to introduce Fourier coordinates by

$$X_l = \frac{1}{N} \sum_{\vec{q}} X(\vec{q})\, e^{i\vec{q}\cdot\vec{R}_l}\,. \tag{34}$$

One then finds that

$$\left\langle\left(\sum_{\delta} X_{\delta}\right)^{2}\right\rangle = \frac{1}{N}\sum_{\vec{q}}\langle|X(\vec{q})|^{2}\rangle F^{2}(\vec{q}), \qquad (35)$$

where we have introduced

$$F(\vec{q}) \equiv \sum_{\delta} e^{i\vec{q}\cdot\vec{\delta}}. \qquad (36)$$

In the thermodynamic limit we then have

$$\left\langle\left(\sum_{\delta} X_{\delta}\right)^{2}\right\rangle = \left(\frac{a}{2\pi}\right)^{d}\int\langle|X(\vec{q})|^{2}\rangle F^{2}(\vec{q})\,d^{d}q, \qquad (37)$$

where the integral is over the Brillouin zone.

Now, according to the theory of infrared bounds developed recently by Fröhlich, Simon, and Spencer[14] the propagator $\langle|X(\vec{q})|^{2}\rangle$ satisfies the inequality

$$\langle|X(\vec{q})|^{2}\rangle \leqslant \frac{k_{B}T}{2C}\,\frac{1}{2d-F(\vec{q})}. \qquad (38)$$

Combining this result with Eqs. (11), (33), and (37) gives for $T>T_c$

$$P''(0) \leqslant -\beta P(0)\left[V_{S}''(0)+4dC - 2C\left(\frac{a}{2\pi}\right)^{d}\int\frac{F^{2}(\vec{q})\,d^{d}q}{2d-F(\vec{q})}\right]. \qquad (39)$$

This result may be reexpressed in terms of the (generalized) Watson integral[33]

$$q(d)=\frac{1}{(2\pi)^{d}}\int\prod_{i=1}^{d}d\theta_{i}\left(d-\sum_{i=1}^{d}\cos\theta_{i}\right)^{-1} \qquad (40)$$

as

$$P''(0) \leqslant -\beta P(0)[V_{S}''(0)+8dC-4d^{2}Cq(d)]. \qquad (41)$$

In $d=3$ we have $q(3)=0.5054\ldots$, so that

$$P''(0)|_{d=3} \leqslant -\beta P(0)[V_{S}''(0)+5.805C]. \qquad (42)$$

Thus, in $d=3$, systems characterized by parameters such that

$$|V_{S}''(0)|/4dC<0.484 \qquad (43)$$

will be *sufficiently* displacive in character that their single-particle distribution function does *not* develop a two-peaked structure above T_c.

V. DISCUSSION

We begin with a discussion of our results for the $d=1$ system examined in Sec. III. While the essential result of this analysis [Eq. (30)] is implied directly by simple homogeneity arguments based upon the continuum analog of Eq. (1), the RG approach yields rather more insight into its significance. In particular, it makes it clear that the exponent ϕ appearing in (30) is to be interpreted as the exponent characterizing the crossover from the Gaussian behavior associated with the "displacive-limit" fixed point ($\Delta^*=u^*=0$) to one-dimensional Ising critical behavior (characterized by a fixed point that does not lie within the Δ-u parameter space). The temperature T^* may thus be thought of as locating a crossover, which it is natural to regard as a particularly simple (perhaps misleadingly so) realization of the "phonon-Ising" crossover referred to by Murata.[34]

This interpretation is certainly in intuitive accord with the definition of T^*: it is also corroborated by a result of Krumhansl and Schrieffer.[6] These authors show that the contribution to the free energy (of the $d=1$ system) made by cluster-type excitations is characterized by an activation energy which, in the notation employed here, may be written as $2k_BT^*/Q^*$. Thus T^* does indeed approximately locate a crossover to a regime in which the thermodynamics is dominated by cluster-type rather than phonon-type excitations.

The analysis of the $d=3$ system, however, reveals that the phonon-Ising crossover is somewhat more subtle than the appealing simplicity of the $d=1$ system would suggest. While, in *some* respects at least, a crossover to a "universal" Ising behavior is expected (sufficiently close to T_c) for the *complete* range of parameters giving rise to a phase transition, the result (43) shows that the spectrum of universal features does not include the formation of a double-peaked probability function (for $d\geqslant 3$).[35] Thus, while there certainly exist "displacive" systems satisfying Eq. (12) which *do* exhibit this phenomenon, there exist also "sufficiently displacive" systems [satisfying (43)] which do not. A prominent example of the latter case is $SrTiO_3$.[11]

One might, of course, regard the result as an indication that the central-peak phenomenon, in so far as it is intrinsic to the ideal crystal behavior, is itself not a universal feature of a structural transition. It is, however, rather more probable that the double peaking of the probability function offers rather too crude a criterion of the validity of the physical situation conjectured in Sec. I, which may manifest itself in rather less dramatic distortions of the probability density function.[36]

More specifically, in the spirit of the picture outlined in Sec. I, suppose that, at a temperature $T>T_c$, the single-particle behavior were to consist effectively of quasiharmonic oscillations of mean-square amplitude δX_L^2 about two positions of local equilibrium displaced by amounts $\pm X_L$ from the true high-temperature-phase equilibrium position. The associated probability function (a superposition of two Gaussians) would *fail* to display a two-peaked structure if $\delta X_L > X_L$.

The implication of these considerations is that the vanishing of $P''(0)$ may not be a sufficiently refined criterion for locating satisfactorily the temperature of the high-to-low-temperature crossover: the question of whether or not such a changeover invariably precedes a phase transition and (correspondingly?) whether or not the central-peak phenomenon is universal in character are therefore left open.

As regards the development of a more refined criterion to locate this changeover, there are two remarks to be made. Firstly, our analysis has by no means exhausted the insights into the dynamical behavior implicit in purely static quantities. In particular, the probability distributions for *clusters* of neighboring atoms may be more sensitive to this changeover.

Secondly, developing an idea of Aubry,[37] it may be helpful—it is certainly suggestive—to pose the question of the location of this changeover thus: Below what temperature is the local dynamical behavior better described in terms of quasiharmonic oscillations about one or other of *two* positions, rather than quasiharmonic oscillations about the *single* high-temperature-phase equilibrium position? The answer might be expected to correspond closely with the temperature at which a self-consistent phonon theory[38] (which *imposes* a quasiharmonic description on the system) predicts a phase transition.[39]

Finally, we remark that, although our analysis (particularly of the $d=1$ system) has made use of the methods and language of critical phenomena, it is rather misleading to regard the high-to-low-temperature "crossover" that we have discussed as a true critical effect. It is probably more appropriate to regard it simply as a necessary consequence of the existence of a double-well single-particle potential, a consequence whose occurrence is bound up with the onset of short-range rather than long-range order.

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formation generates scaling fields that are not present in the initial Hamiltonian (18). Although certain of these—e.g., the local operator Y_l^6—are actually more strongly "relevant" than the local operator Y_l^4, it is not hard to show that they do not invalidate the final (asymptotic) result (30).

[27] We neglect a term of order u in the equation for $\tilde{\Delta}$, which again does not invalidate the result (30).

[28] We are grateful to R. Morf for communicating the result to us.

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[30] The critical temperature in the $d=\infty$ limit is believed to coincide with the mean-field theory result. See R. Oppermann and H. Thomas, Z. Phys. 22, 387 (1975).

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[37] S. Aubry (private communication). Aubry conjectures a close correspondence between the onset temperature for the central peak and the occurrence of a phase transition within self-consistent phonon theory—a conjecture substantiated by numerical results for $d=1$.

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