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Crossover between displacive and order/disorder behaviour in the Φ^4 model

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Abstract. The Φ^4 potential is widely employed for modelling systems that undergo displacive phase transition of the soft mode or order/disorder. A molecular dynamic simulation is used to establish where the crossover lies between the two regimes, considering both limits of long range and short range intersite interactions. It appears that with both ranges of interaction the crossover occurs when the on-site potential changes from a single to a double well. The behaviour of the soft mode frequency ω is also investigated, and in particular the ratio of the slopes $d\omega^2/dT$ between $T > T_c$ and $T < T_c$. We find this ratio attains the value -2 given by standard renormalized phonon theory only the double limit of long range coupling and extreme soft mode behaviour. The order parameter varies as $(T_c - T)^{1/2}$ over a wide temperature range, as observed in several materials, in this double limit only.

1. Introduction

The Φ^4 model on a lattice is widely employed for modelling materials that undergo a displacive structural phase transition. The model is able to cover a wide range of phenomena, from the so-called soft mode behaviour (SiO_2, $NaNO_3$, biphenyl,...) to the order/disorder type of transition ($NaNO_2$, NH_4Cl,...). In the soft mode limit, the phase transition can be described in terms of renormalized phonon theory (Bruce 1980). In this, the system possesses a lattice vibration mode which softens as the temperature decreases toward T_c. At the critical temperature, this particular mode freezes completely so a static field of displacement of atom positions is induced within the structure. In the extreme opposite case, the system is described by an order/disorder dynamical behaviour characterized by atoms or clusters fluctuating between two more-or-less discrete states. The length of correlation in time and space grows as the temperature approaches T_c and finally diverges on it (Bruce 1980).

For the following, we consider the Hamiltonian form, the so-called Φ^4 model:

$$H = \sum_i \frac{m}{2}\left(\frac{dU_i}{d\tau}\right)^2 + \sum_i \left(\alpha U_i^2 + \gamma U_i^4 - U_i \sum_{j \neq i} J_{ij} U_j\right) \qquad (1.1)$$

where the parameters α and γ represent the on-site parameter ($\gamma > 0$) and the J_{ij}s represent the pairwise intersite interactions. The variable U_i can stand for the

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rotation of the tetrahedral cluster in quartz or another such generalized coordinate. It can also represent the displacement of the central atom in the unit cell in the perovskite family, such as the titanium in $BaTiO_3$, or the torsion of the benzene rings relative to each other in crystalline biphenyl (Benckert 1987). The parameter m is a corresponding generalized inertia. The coupling terms J_{ij}s determine the wave vector at which the phase transition occurs. By considering strong competition between the J_{ij}s for example, one recovers the Janssen model used for modelling displacive modulated structures (Janssen 1986). For our purpose, we shall only consider a ferro-distortive phase transition by choosing the following exchange interaction:

$$J_{ij} = \begin{cases} J/z > 0 & \text{for certain } (i,j) \text{ neighbours} \\ 0 & \text{otherwise} \end{cases} \qquad (1.2a,b)$$

where (1.2a) applies to the z closest neighbours to a given site and we can vary z from $z = 6$ (extreme short range) to $z = N$ (extreme long range). Here N is the total number of atoms in our computer simulation. Note that the Hamiltonian (1.1) which depends on four parameters (m, α, γ and J_{ij}) can take a simpler form by rescaling U_i, the time τ and the Hamiltonian as follows:

$$U_i \to \sqrt{(\gamma/J)}U_i \qquad (1.3a)$$

$$\tau \to \sqrt{(J/m)}\tau \qquad (1.3b)$$

$$H = \frac{\gamma}{J^2}H = \sum_i \frac{1}{2}\left(\frac{\mathrm{d}U_i}{\mathrm{d}\tau}\right)^2 + \sum_i \left(\frac{\alpha}{J}U_i^2 + U_i^4 - \frac{1}{z}U_i \sum_{j\neq i} U_j\right). \qquad (1.3c)$$

Aside from the range z, the right-hand side of equation (1.3c) depends on the unique combination (α/J) which controls the on-site potential shape, i.e. single or double well as (α/J) is positive or negative, and also the well depth. By considering the total energy when all $U_i = U_0$, it is easy to show that there is no phase transition unless $(\alpha/J) < 1$. We shall refer to

$$\alpha/J \lesssim 1 \qquad (1.4)$$

as the soft mode limit. Note also that the above Hamiltonian describes an optic mode of the variable U_i, but this does not restrict the generality of our discussion as it is always possible to recover an acoustic mode for the variables V_i by the transformation $V_i = U_{i+1} - U_i$. It is not thought that the particular form (1.2) adopted for J_{ij} influences the phase transition materially, except for in the range z.

We conclude from (1.3) that the model is specified by two parameters, namely (α/J) and the range z. Of these, it is well established that the on-site potential is the main factor controlling the behaviour of the system. For a single anharmonic well for the on-site potential, i.e. for (α/J) near the limit (1.4), it is well accepted that the Φ^4 model leads to a soft mode type of behaviour whose displacement on lowering T freezes at T_c. In the opposite case, for a deep double-well on-site potential with $(\alpha/J) \ll 0$, i.e. with large 'hump height' (figure 1) compared to the coupling terms, it is also well established that the Φ^4 describes an order/disorder dynamic system because it approaches an Ising model with U_i near the bottom of one of the wells.

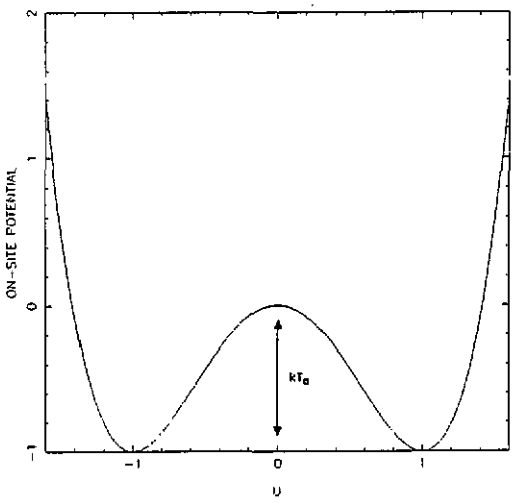

Figure 1. Double-well on-site potential. One argument places the crossover between soft mode limit and order/disorder when $T_c = T_0$ where T_0 is defined in the text.

However the precise location of the crossover between these two behaviours is far from clear in the literature (Blinc and Zeks 1974, Aubry 1975).

In fact, there exist two reasonable arguments centring the crossover between soft mode and order/disorder at two different locations. A trivial argument places the crossover at $\alpha = 0$ which is simply the crossover between a single-well and a double-well on-site potential. In a less obvious argument, the crossover is fixed when the critical temperature has the same energy as the 'hump height' of the double well, for some negative α. By using a mean field estimate T_c (Blinc and Zeks 1974), we deduce that such a crossover occurs at $\alpha = -J$. The argument is as follows. When the kinetic energy of the entities U_i exceed the barrier between the two wells, the system can oscillate across the whole double well, and hence one might suppose that for $T > T_0$ in figure 1 the effect of the central hump can be neglected. In that case one would expect that a system with $T_c > T_0$ would still exhibit soft mode behaviour at $T \geqslant T_c$. When, on the contrary, $T_c < T_0$, the system, on lowering the temperature, would settle into one half of the double well at $T = T_0$ (Normand *et al* 1990) and hence would show order/disorder behaviour at the lower temperature $T_c (< T_0)$. This argument therefore centres the crossover at $T_c = T_0$ which translates approximately into $(\alpha/J) \approx -1$ as already remarked. In this paper, we wish to establish, with the help of computer simulation, the location of the crossover $(\alpha/J)_c$ in model (1.3*c*) between the order/disorder and soft mode behaviours. We also wonder whether the result depends on the range of the coupling between the entities U_i. For this purpose, we use a molecular dynamics simulation (MDS) of the model (1.3*c*) in the two limits of nearest neighbour coupling ($z = 6$) and that of long range coupling ($z = 4095$). In both cases, we consider the power spectrum of the order parameter fluctuations that exhibits the soft mode for different values (α/J) and at different temperatures. In the short range limit, we use a least square fit of the autocorrelation function for various computational reasons discussed in section 2. In section 3 we discuss the criterion that we use for characterizing the soft mode and order/disorder types of transition. In section 4 we discuss the crossover location on the basis of the MDS results in the short range coupling limit, and in section 5, in the long range coupling limit. We find that for both long range coupling and short range coupling

the crossover is close to zero:

$$\alpha/J_c \lesssim 0. \tag{1.5}$$

Below this critical value (1.5), i.e. at $(\alpha/J) = -0.25$, the system already has an order/disorder behaviour and there is still a softish mode, although it ceases to soften completely to $\omega = 0$.

We want to emphasize that the model (1.3) belongs to the same universality class as the Ising model for all values of (α/J) (Bruce 1980). The nature of the critical fluctuations is therefore the same in the soft mode and order/disorder regimes and cannot determine the crossover between them. In both regimes, the system near T_c contains islands of order, with a central peak in the correlation function $S(q,\omega)$ describing their relaxation behaviour (Schneider and Stoll 1975). This is somewhat paradoxical because, in the order/disorder regime, there remains near T_c the additional 'resonant' peak at non-zero ω, while in the soft mode regime this peak has shifted to zero frequency near T_c and has been swallowed up in the critical fluctuations. Our results show how this paradox is resolved. In the order/disorder regime, the weight tends to zero as T tends to T_c so that it 'disappears' in that sense. Thus, to leading order in $|T - T_c|$ the critical fluctuations are the same in the two cases. In any case, the critical fluctuations are a side issue for another reason: we are discussing the behaviour of the system over a broad temperature range, typically from 0.5 T_c to 1.5 or 2 times T_c, not particularly in the narrower Ginzburg interval dominated by critical fluctuations around T_c. Our simulations also clarify another issue as is described below.

In the soft mode regime we have that

$$\begin{aligned} \omega^2(T) &\propto (T - T_c) \qquad \text{for } T > T_c \\ &\propto (T_c - T) \qquad \text{for } T < T_c \end{aligned} \tag{1.6}$$

over quite a wide temperature range $0.5 \leqslant T/T_c \leqslant 2$, and one often considers the ratio slopes in (1.6)

$$R = (\mathrm{d}\omega^2/\mathrm{d}T)_{T<T_c}/(\mathrm{d}\omega^2/\mathrm{d}T)_{T>T_c}. \tag{1.7}$$

Renormalized phonon theory (Bruce 1980) gives the result

$$R = -2 \tag{1.8}$$

which we shall refer to as the standard value. However, experimentally R often differs substantially from the standard value (1.8), as, for example, in bismuth vanadate with $R = -2.7$ (Pinczuk *et al* 1978, 1979). Our simulations will show that the standard value (1.8) is obtained if and only if two conditions are satisfied: namely, that (α/J) is near the soft mode limit (1.4) and that we have long range interaction (z large). This is consistent with (1.8) being given by renormalized phonon theory because the latter effectively makes two approximations, namely replacing the on-site well by an effective potential in the form of a parabola and treating the interactions by mean field theory.

The ratio R may not be of great importance in itself, but the issue of the standard value (1.8) is linked to another matter of some interest. Quite a number of materials

are known to undergo a structural phase transition where the order parameter $U(T)$ for $T < T_c$ follows the simplest Landau theory

$$U(T) \propto (T_c - T)^{\beta} \tag{1.9}$$

with $\beta = 1/2$ over a surprisingly wide range of temperature e.g. $LaAlO_3$, As_2O_5 and others (Salje *et al* 1991, Salje 1990). These contrast with other materials, and with computer simulations where the exponent in (1.9) lies anywhere in the range 0.3 to 0.5 (outside the Ginzburg region of critical fluctuations) (Giddy *et al* 1989, 1990). If $\omega^2(T)$ varies linearly (1.6) below T_c, as it does in our simulations in the case studied, then the standard value $R = -2$ (1.9) is equivalent to the simple Landau result $\beta = 1/2$, and the pair of conditions that applies to the former is also necessary for the latter. As already remarked, one of the conditions is that the interaction J_{ij} should be long ranged, and it is interesting to note that interactions of infinite range occur in ferroelastic phase transitions where the coupling is mediated by elastic strain (Marais *et al* 1991, 1992). The present work therefore contributes to an understanding of materials obeying (1.9) with $\beta = 1/2$ over a wide temperature interval, and this spin-off may be more significant than the original purpose of locating the crossover.

2. Molecular dynamics simulation

The molecular dynamics simulation (MDS) of the Hamiltonian (1.3*c*) is performed on a parallel processor array (AMT-DAP, Cambridge) and runs on a lattice of 16X16X16 elements with periodic boundary conditions. We consider the two extreme ranges of interaction already introduced below (1.2), namely a coupling to the $z = 6$ cubic nearest neighbours for only short range, and equal coupling to all other elements in the simulation for long range ($z = 4095$). In both cases, the simulations are performed within the framework of a microcanonical ensemble (see Dove 1988 for details). In the self-consistent renormalized phonon theory (Bruce 1980), the soft mode belongs to the normal coordinate representing the wave vector q at which the transition occurs. For our case of a ferro-type transition, we have $q = 0$ (Γ mode) with normal coordinate given by the mean displacement

$$U(\tau) = U_{q=0}(\tau) = \frac{1}{N}\sum_{i=1}^{N} U_i(\tau) \tag{2.1}$$

and the order parameter is the time average of the above quantity $U(\tau)$. As usual, the frequency of the mode is determined from the power spectrum of the fluctuation correlation function given here by

$$S(\omega, q = 0) = \int \mathrm{d}\tau\ \mathrm{e}^{-i\omega\tau}\langle U(\tau = 0)U(\tau)\rangle. \tag{2.2}$$

The above expression is known to carry two important features (Schneider and Stoll 1973, 1975, 1978), namely, a resonant peak centred around a temperature dependent frequency ω_R, i.e. the phonon mode in which we are interested, and a 'central peak' around $\omega = 0$. Thus, the power spectrum can be written in the form:

$$S(\omega, q = 0) = 2AB/(B^2 + \omega^2) + CD/[D^2 + (\omega - \omega_R)^2] \tag{2.3}$$

where A, B, C, D and ω_R are the parameters to be determined. This corresponds to a correlation function in time:

$$S(\tau) = Ae^{-B\tau} + Ce^{-D\tau}\cos(\omega_R\tau). \tag{2.4}$$

Experimentally one measures $S(\omega, q = 0)$, but this is not a satisfactory quantity in a finite system as small as in our computer simulation. The important point to note is that the form (2.4) would apply to an infinite system. In our case, it only applies for times τ less than the circulation time τ_C for a signal to pass around the computer sample, i.e. for a fluctuation to propagate to the edge of the system and to reappear by the periodic boundary conditions on the opposite side. Such a circulation dominates $S(\tau)$ for τ greater than τ_C causing erratic fluctuations in it. Our procedure is therefore to determine ω_R, A, B, C and D by fitting the form (2.4) to the correlation function $S(\tau)$ for $\tau < \tau_C$ only. We then have from (2.3) the power spectrum $S(\omega)$ of an effective infinite system. This corresponds to removing the tail of the computational $S(\tau)$ and replacing it by the tail appropriate for an infinite system. However, that procedure is not applicable for infinitely long range coupling of $z = 4095$ in our case because the velocity of sound c is then infinite. In that case, the power spectrum that we consider is given by the square of the Fourier transform of the order parameter (2.1) (Dove 1988).

In both cases, we report the soft mode frequency ω_R, its width and the power spectrum for different values of (α/J) and different temperatures (figures 3 to 7). In the short range coupling limit, we also report the height A/B of the central peak (figure 6).

3. The crossover criterion

The main issue is to establish a sharp criterion as to whether the system is behaving in an order/disorder or soft mode manner. Clearly we have order/disorder behaviour if ω_R is non-zero at all temperatures T around T_c, i.e. if the resonant peak of $S(\omega)$ remains identifiably separate from the central peak. Conversely, if ω_R tends to zero at T_c we have soft mode behaviour, but there would appear to be a problem: how can we tell the behaviour of ω_R near T_c when ω_R is so small that the two peaks overlap and merge? This problem is not as severe as it sounds because the expected behaviour of ω_R is shown in figure 2 due to damping. Remember that a simple harmonic oscillator with inherent undamped frequency Ω and damping γ oscillates with frequency

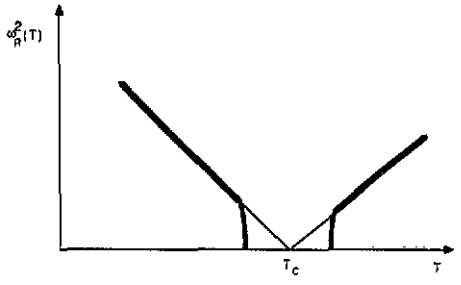

Figure 2. Expected behaviour of $\omega_R(T)$ near T_c in the soft mode regime (schematic and enlarged). Bold line: frequency as observed; light line: ideal undamped frequency.

$$\omega = (\Omega^2 - \gamma^2)^{1/2} \tag{3.1}$$

and that the response is diffusive in the overdamped regime $\Omega < \gamma$. Therefore, if we expect the undamped $[\omega_{RU}(T)]^2$ to extrapolate linearly to zero at T_c, the observed $\omega_R(T)$ actually disappears over a region of temperature around T_c (figure 2). This is seen for example in figure 3(*a*), (*b*). Presumably critical fluctuations will also affect the behaviour near T_c in another way so that the two straight lines (1.6) in figure 2 need not intersect zero exactly at T_c. The situation is further illuminated by considering the behaviour of the central peak. In an order/disorder situation one has a diffusive (non-oscillatory) local flipping between two rather distinct states, and indeed this can be taken as an alternative definition of order/disorder character. In consequence the weight of the central peak is large over a wide temperature range above T_c (and presumably quite widely spread in q-space), as seen in figure 4(*d*) where the individual atoms flip between the two sides of the local double well. In the soft mode regime there is also a central peak, but of slightly different origin. This occurs only near T_c (figure 4(*a*)). Even when the on-site well is a single well, the total energy of the system below T_c has two possible minima due to the addition of the interaction J_{ij}; otherwise there would be no phase transition. Thus near T_c there are clusters of sufficient size to show the two possible minima of the total energy and hence diffusive flipping between them, or rather diffusive motion of cluster boundaries. These are fluctuations of the order parameter largely confined to temperature around T_c (figure 4(*b*)) and presumably to a region near $q = 0$ in reciprocal space. The fluctuations of the order parameter and oscillations of the soft mode are motions of the same character. Thus the soft mode resonance peak is in a sense swallowed up into the central peak near T_c in the soft mode regime when the soft mode becomes overdamped (figure 2).

4. Behaviour with short range coupling

In the short range coupling limit i.e. nearest neighbour interaction, we first consider the case $(\alpha/J) = 0.5$ corresponding to a single-harmonic-well potential anharmonically perturbed. The power spectrum at different temperatures above and below T_c is presented in figure 5. The phonon peak frequency softens when the temperature is decreased, and increases again below T_c. In figure 3(*a*), the frequency squared is plotted versus the temperature. The 'square' in figure 3(*a*) represents the region of difficulty in fitting form (2.3) of the correlation function to the computational data: no ω_R could be fitted. It corresponds to the central region in figure 2 where $S(\tau)$ is expected to be the sum of two real exponentials and not of the form (2.3). We conclude that $(\alpha/J) = 0.5$ is definitely in the soft mode regime according to the criterion of section 3. Figure 4(*a*) depicts the integral of the central peak versus temperature. Note that the maximum occurs at T_c due to the critical fluctuations (Schneider and Stoll 1978). For $(\alpha/J) = 0$, the same discussion can be applied to the data of figures 3(*b*) and 4(*b*): they also characterize a soft mode behaviour. However, for both $(\alpha/J) = -0.25$ and -2, the situation is radically different. In figures 6 $(\alpha/J = -2)$ one obtains a wide phonon peak, i.e. strongly damped, which nevertheless persist throughout the critical temperature, as shown in figures 3(*c*) and (*d*). This is the signature of order/disorder behaviour. Note however that its weight

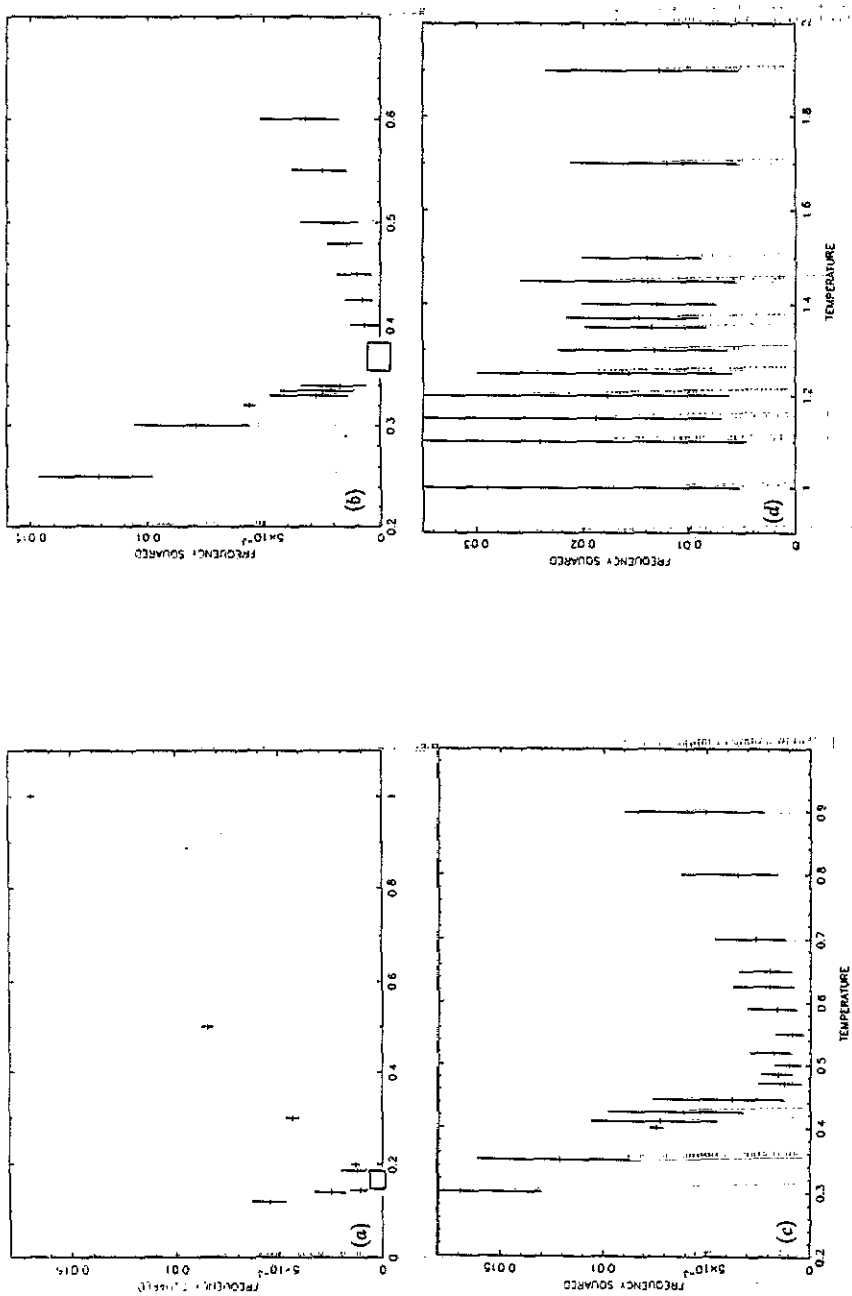

Figure 3. Soft mode frequency squared versus temperature behaviour for the four parameter values considered in the short range interaction limit. The vertical bars denote the lifetime broadening of the phonon mode as given by D in equations (2.3) and (2.4) and the phonon peak width in figures 5 and 6. (*a*) $(\alpha/J) = 0.5$. Note the complete softening of the phonon mode around $T_c \approx 0.16$. The box denotes the region of overdamped oscillations where no frequency could be found. (*b*) $(\alpha/J) = 0$. The soft mode softens completely around $T_c \approx 0.36$. (*c*) $(\alpha/J) = -0.25$. The phonon mode frequency does not soften completely ($T_c \approx 0.50$). (*d*) $(\alpha/J) = -2$. Here, the phonon mode does not show any signs of softening at $T_c \approx 1.27$.

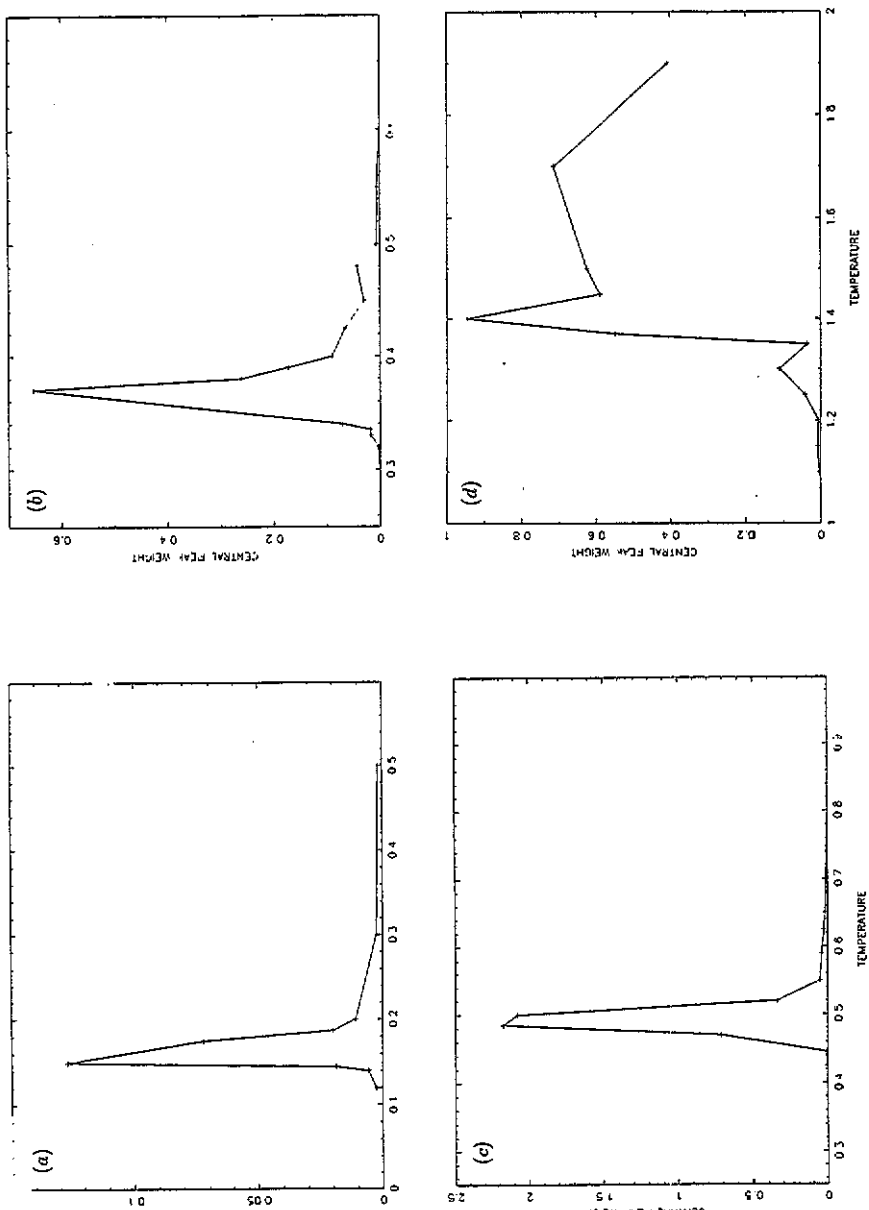

Figure 4. Height of the central peak (equal to A/B in equations (2.3) and (2.4)) in the short range coupling limit. (*a*) $(\alpha/J) = 0.5$. Note a strong peak at T_c resulting from the critical clustering effect. (*b*) $(\alpha/J) = 0$ and (*c*) $(\alpha/J) = -0.25$: both cases present a slight persistence of the central peak at high temperatures. (*d*) $(\alpha/J) = -2$. The central peak is present at all temperatures and results from the order/disorder nature of the system, i.e. the double well at each site.

relative to the central peak tends to zero at T_c in accordance with the discussion of section 1. We note that for $(\alpha/J) = -2$ the central peak persists over a wide range of temperature (figure 4(*d*)). Near $T_c \approx 1.27$ this is, once more, due to clusters, as in the soft mode regime, i.e. to critical fluctuations. However, the central peak cannot be due to critical fluctuations because at high temperature the correlation length is very short: it is, rather, due to local excitation from one side of the double well to the other (Schneider and Stoll 1978).

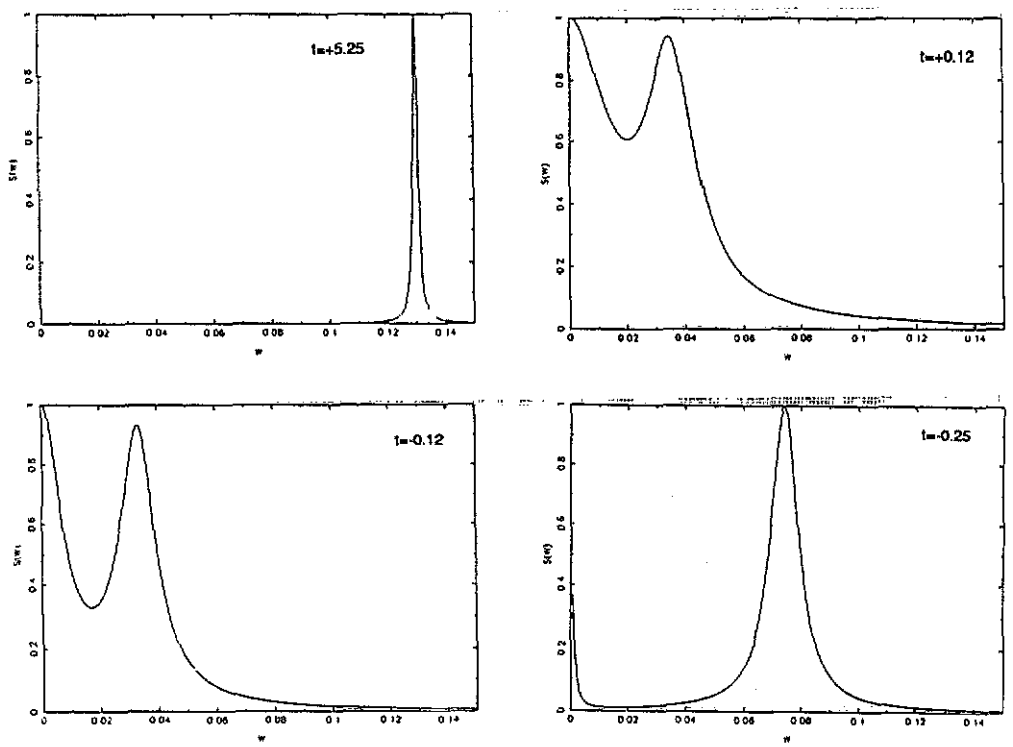

Figure 5. Power spectrum at different temperatures resulting from fitting (2.3) in the nearest neighbour coupling limit for $(\alpha/J) = 0.5$. The reduced temperature $t = (T - T_c)/T_c$ is indicated in the figures. A central peak occurs near T_c where critical clustering dominates.

The location of the crossover according to the results outlined above is formally between the values -0.25 and 0. The argument placing it at -1 is definitively inconsistent with the data; the other one i.e. $(\alpha/J)_c = 0$, however, is still just within the range and we can only place it within that interval, i.e. between -0.25 and 0.

5. Behaviour with long range coupling

We wish to check whether the above conclusion about the crossover depends on the range of interaction between the U_is, and we now use the extreme long range coupling version of (1.3*c*) to test this as described in section 2. In figure 7(*a*), the squared frequency versus T for $(\alpha/J) = -0.25$ is shown for the long range coupling

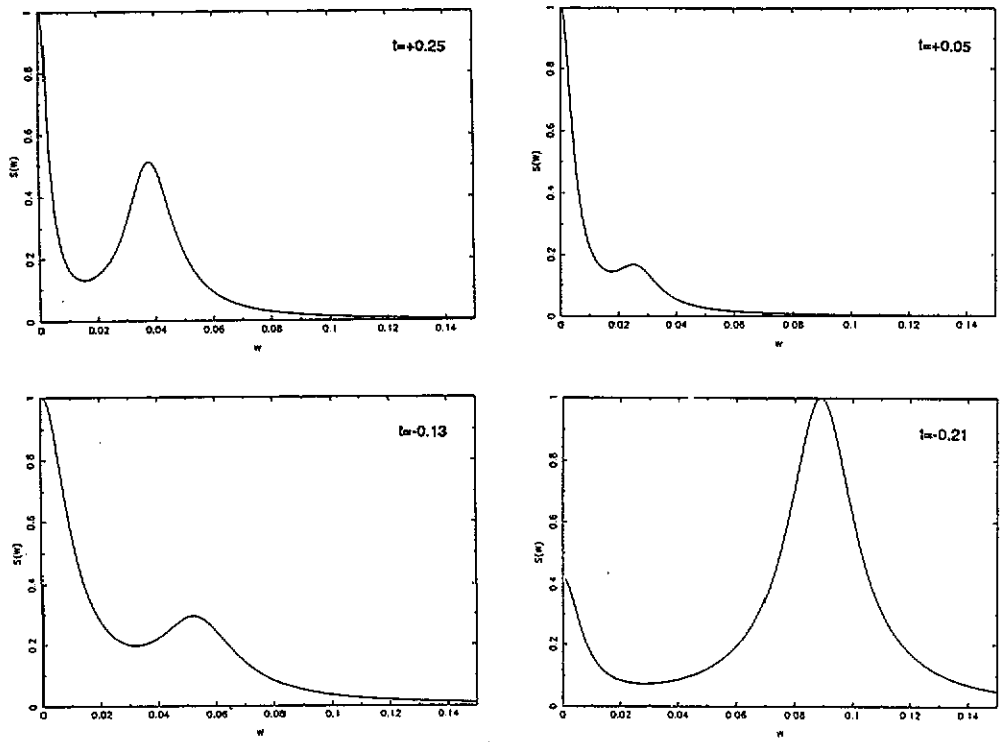

Figure 6. Power spectrum for $(\alpha/J) = -2$ in the nearest neighbour coupling limit. Note the strong persistent central peak at all T related to the order/disorder dynamical process. t is the reduced temperature as in figure 5.

limit. The phonon mode frequency can be followed all through T_c which, according to section 3, corresponds to an order/disorder behaviour. In figure 7(*b*) the squared frequency is presented for $(\alpha/J) = 0$. Note that the phonon mode can be followed until it almost vanishes, even at $t = 0.04$. We therefore have no region of overdamped fluctuations, such as was denoted by the square box in figure 3(*a*), (*b*). In the long range coupling limit, we expect from theory that critical fluctuations are absent, so the central peak does not overshadow the phonon peak. For $(\alpha/J) = 0$ the latter can be followed until $\omega \approx 0$ and we have soft mode limit behaviour again. We therefore conclude that the range of coupling does not change the nature of the transition and hence the location of the crossover.

6. Variation of soft mode frequency

We have already discussed in section 1 the importance of the variation of $\omega^2(T)$ with temperature and the ratio R (1.7) of the slopes below and above T_c. In particular, R being equal to the standard value -2 given by (1.8) together with $\omega^2(T)$ varying linearly with T below T_c, indicates that the order parameter has the simplest Landau form (1.9) with $\beta = 1/2$ for its temperature variation.

An obvious question arises within the framework of the present study: what is the necessary and sufficient condition for model (1.3*c*) to give the standard ratio $R = -2$ (1.8)? Our MDS results enable us to establish this. In table 1 we present the values

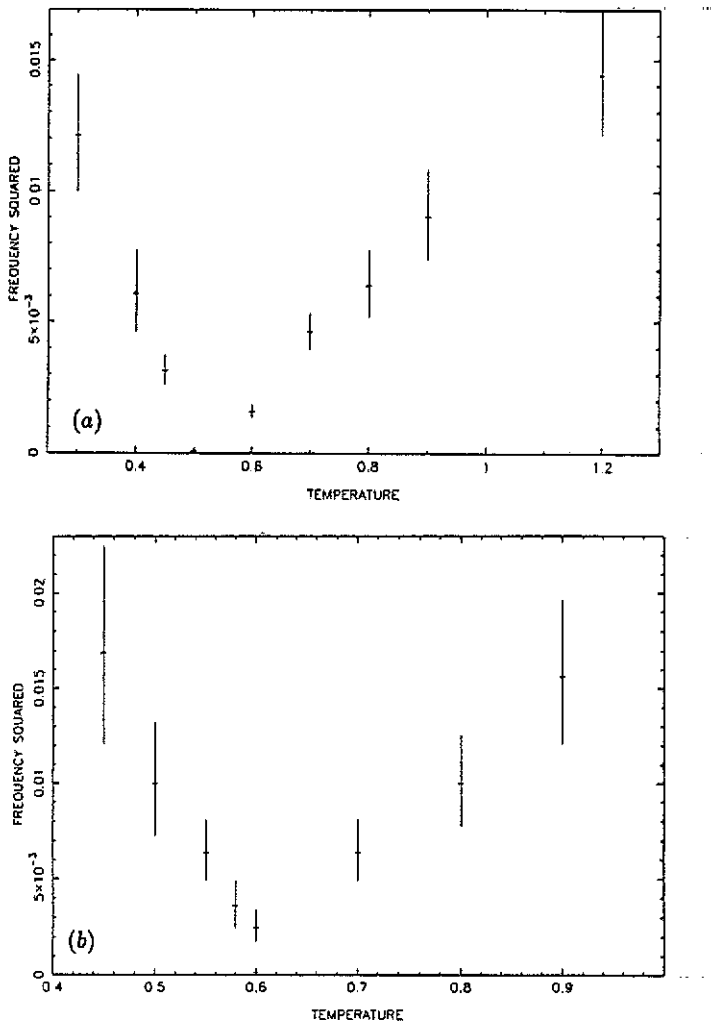

Figure 7. Soft mode squared versus temperature in the long range coupling limit. The vertical bars denote the broadening as in figure 3. (*a*) $(\alpha/J) = 0$; the soft mode softens completely at $T_c \approx 0.52$. (*b*) $(\alpha/J) = -0.25$; the soft mode does not soften completely. Note that both results are in agreement with the short range coupling results. The range of interaction has no effect on the type of transition (soft mode or order/disorder regime).

Table 1. Slope ratio R of the soft mode frequency squared between $T < T_c$ and $T > T_c$ for different values of the potential parameter (α/J) in the long range and short range coupling limits. Note that the standard ratio -2, which is found in self-consistent renormalized phonon theory, is closely approached in the extreme long range coupling limit combined with the displacive limit.

	α/J	−0.5	−0.25	0.0	0.3	0.5
R	Long range	−5.5	−3.0	−2.7	—	−2.3
	Short range	−10.5	−8.0	−9.5	−6.0	−7.0

of the ratio R (1.7) for various values of (α/J) in the soft mode regime for both

short and long range coupling. For the short range case, the ratio R differs strongly from the standard -2 and does not, even in the soft mode limit $(\alpha/J) \to 0$, improve significantly. In the opposite case, the long range interaction already gives $R = -2.6$ at $(\alpha/J) = 0$, close to the crossover location. The ratio at $(\alpha/J) = 0.5$ improves to $R = -2.3$. This suggests that from MDS, the necessary and sufficient condition to obtain the standard ratio $R = -2$ as given by renormalized phonon theory is to be in the soft mode limit $(\alpha/J) \to 1$ with long range coupling. Both conditions must be satisfied.

This result could have been predicted if we had considered carefully the approximations used in the renormalized phonon theory. Firstly, the method approximates the anharmonic on-site potential by a renormalized harmonic potential. The fit is certainly better in the soft mode limit, i.e. $(\alpha/J) \approx 1$, where one already has a single well. Secondly, the use of the random phase approximation picks up the soft mode and places it in the mean squared fluctuation of all the other modes. This is typically a mean field approximation. In the long range coupling used here, one site is equally coupled to all others so there is only one dominant mode in the system: the soft mode itself. In that limit the decoupling is natural because one can neglect all the other modes which are pushed to a much higher frequency.

7. Summary

We have determined the crossover between the soft mode limit and the order/disorder behaviour in the Φ^4 model. Firstly, we have shown that the behaviour is largely controlled by the shape of the on-site potential i.e. whether it is a single or double well. Secondly, we have used a molecular dynamic study of the system which has enabled us to determine its behaviour for a varying shape of the on-site potential. Also, we have performed the simulations in the two extremes of long range and short range coupling, and in both cases have found that the crossover appears very close to the point where the on-site potential is turned from a single well to a double well. In particular, we have found that for a double well, even for a shallow double well in the on-site potential, the soft mode frequency does not tend completely to zero at T_c as it does in the soft mode regime.

We have also discussed the variation of the soft mode frequency with temperature, in particular the ratio of the slopes $d\omega^2(T)/dT$ below and above T_c. We have found that the standard ratio $R = -2$, suggested by the self-consistent renormalized phonon treatment of the Φ^4 model is also given by the simulations, but only in the limit of long range coupling and with the soft mode limit $(\alpha/J) \to 1$. Therefore, this pair of conditions also applies to the order parameter having a temperature variation given by the simplest Landau form (1.9) with $\beta = 1/2$. The fact that such materials exist, with order parameters of simple forms over a wide range of T, suggests they have long range coupling mediated by strain (Marais *et al* 1991, 1992).

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