mode phonons is to be expected in the single crystal; therefore, it is not surprising that the temperature dependence of the single T_1 is almost identical to that of T_1^l in the powder.

The ratio of the T_1^s values is unity within the experimental uncertainty. This suggests an isotope-independent (nonresonant) relaxation mechanism[9] such as would result from abrupt changes in the Hamiltonian of a particular spin due to sudden reorientations of the $OsCl_6$ octahedra. A possible explanation for the occurrence of T_1^s and for its observed temperature variation can be given in terms of the formation of dynamic clusters with the impurities acting as nucleation centers for their development. In the region of a cluster the symmetry in the vicinity of a particular nucleus changes abruptly from that of the high-temperature phase to that of the low-temperature phase. The efficiency of the associated relaxation mechanism will be inversely proportional to the average time spent by a nucleus in an environment characteristic of one or other of the phases. Since the difference in energy associated with the two phases decreases as T approaches T_c it is not surprising that the time constant associated with T_1^s is observed to decrease. (Note that the relevant time constant for T_1^s is *not* the cluster lifetime.) According to this picture nuclei near to an impurity are physically distinguishable from those far from an impurity. As the phase transition is approached the coherence length of the correlated fluctuations giving rise to the clusters increases so that the relative number of nuclei contributing to the T_1^s component increases in the manner indicated in Fig. 2. An important question to resolve for K_2OsCl_6 is whether the observed effect is indeed due to impurities or whether it is an intrinsic effect. A related uncertainty exists with respect to the interpretation of the central peak in $SrTiO_3$.[10]

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Observation of Cluster Waves and Their Lifetime

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Using the molecular-dynamics technique we have studied a linear chain with quartic anharmonicity. Particular solutions of the equation of motion, representing clusters (domains), are shown to be relevant for a statistical description of the dynamics. A new excitation branch and the associated central-peak phenomenon are traced back to traveling cluster waves and their lifetime.

Nonlinear wave processes are involved in phenomena occurring in many scientific areas. The state of the art in this exciting field of applied mathematics and theoretical physics has been covered in several reviews.[1-3] Recently it has also been recognized that strong nonlinearity plays a crucial role in lattice-dynamic systems undergoing a continuous phase transition, which takes the system from a high-temperature displacement pattern to a different low-temperature displacement configuration.[4-7] To imitate such systems one may adopt a model, consisting of an infinite set of coupled anharmonic oscillators. The potential energy is chosen in such a way that the mean displacement, representing the order parameter, does not vanish at low temperature. Such models differ from the Fermi-Pasta-Ulam problem[8] in two important respects: (1) The nonlinearity is not small; (2) the system undergoes a phase transition. They also differ from the disordered chain of Lennard-Jones particles considered previously.[9] The common feature is the lack of any general analytic method for discussing the dynamic finite-temperature properties of such nonlinear systems.

Computer simulations have been carried out to shed light on this problem.[5,6] In addition to revealing expected features, such as the static critical phenomena,[5] there are three other interesting observations: the appearance of clusters of locally ordered regions,[5] the occurrence of a "central peak" around $\omega=0$ in the dynamic form factor $S(k_0,\omega)$,[5,6] and the appearance of a new excitation branch close to the displacive limit.[10] The central-peak phenomenon, first observed in $SrTiO_3$, has received a variety of interpretations and remains open to question.[11]

Recently, however, Krumhansl and Schrieffer[7] found, by studying the classical equations of motion of a strongly anharmonic chain, particular solutions, representing clusters, which cannot be described by the conventional phonon perturbation expansions. At low temperatures, they found that the exactly calculated static properties agree with those found from a phenomenological model in which both phonons and clusters are included as elementary excitations.

In this Letter we demonstrate the relevance of these cluster excitations and, in particular, their implications on the dynamic properties. We present direct numerical evidence that cluster waves exist in model systems at least, and close to the transition temperature. A new excitation branch appearing in addition to the phonon resonances, and the central-peak phenomenon are traced back to traveling cluster waves and their lifetime.

A standard model Hamiltonian for a system which might undergo a ferrodistortive phase transition reads

$$\mathcal{H}=\tfrac{1}{2}\sum_l \dot{U}_l^2+\tfrac{1}{2}(A-2C)\sum_l U_l^2+\tfrac{1}{4}B\sum_l U_l^4 + \tfrac{1}{2}C\sum_l (U_l-U_{l+1})^2. \tag{1}$$

Here l denotes the lattice sites and U_l, $\dot{U}_l$ are the displacement and velocity of the particle with respect to a rigid reference lattice with lattice constant a and one particle per unit cell. A, B, and C are the model parameters. The time is chosen in such a way that the mass of the particles is equal to one. Here we consider a one-dimensional system only. Neglecting quantum effects, $T=0$, where the isothermal susceptibility

$$\frac{1}{N}\sum_{l,l'}\langle U_l U_{l'}\rangle$$

diverges, corresponds in this case to the transition temperature. We note that $A=-\infty$, $B=+\infty$, and $A/B=-1$ correspond to the Ising limit, and $A=2C$ corresponds to the displacive limit.[12]

Using the molecular-dynamics technique, we investigated a linear chain of 2000 particles, subjected to periodic boundary conditions, as defined by Eq. (1). For technical details on the molecular-dynamics method, we refer to the papers of Rahman[13] and Verlet.[14]

To investigate the excitation spectrum we next consider the dynamic form factor

$$\hat{S}_{\rho\rho}(k,\omega)=\frac{\int_{-\infty}^{\infty}dt\,\exp(-i\omega t)\langle\rho(-k,0)\rho(k,t)\rangle}{\langle\rho(-k,0)\rho(k,t)\rangle}=\frac{S_{\rho\rho}(k,\omega)}{S_{\rho\rho}(k,t=0)}, \tag{2}$$

where

$$\rho(k,t)=N^{-1}\sum_l \exp[ik(al+U_l)], \tag{3}$$

k denotes the wave number, and ω the frequency.

Figure 1(a) shows the calculated frequency dependence of $\hat{S}_{\rho\rho}(k,\omega)$ for model III ($A=\frac{7}{8}$, $B=\frac{1}{3}$, $C=\frac{1}{2}$) for fixed k values and $k_BT=0.0468$. At $k=0$ we observe a central peak and a weak soft-mode resonance. Slightly removed from $k=0$, however, the central peak is seen to split into a double-peak structure, appearing in addition to the soft-mode resonance. Consequently, for finite k a new excitation branch appears in addition to the soft mode and ceases to be well-defined above some cutoff wave vector. In Fig. 1(b) we plot the dispersion curves of the phonon and the new excitation branch, as obtained from the peak maxima. The full curve represents the prediction of the self-consistent phonon approximation (SCPA).

To elucidate the physical origin of the central peak and the associated new excitations first observed in a two-dimensional system,[10] it is suggestive to suppress the high-frequency and short-wavelength phonon excitations. For this purpose we considered only the ω and k values within the rectangle marked in Fig. 1(b). On this basis we

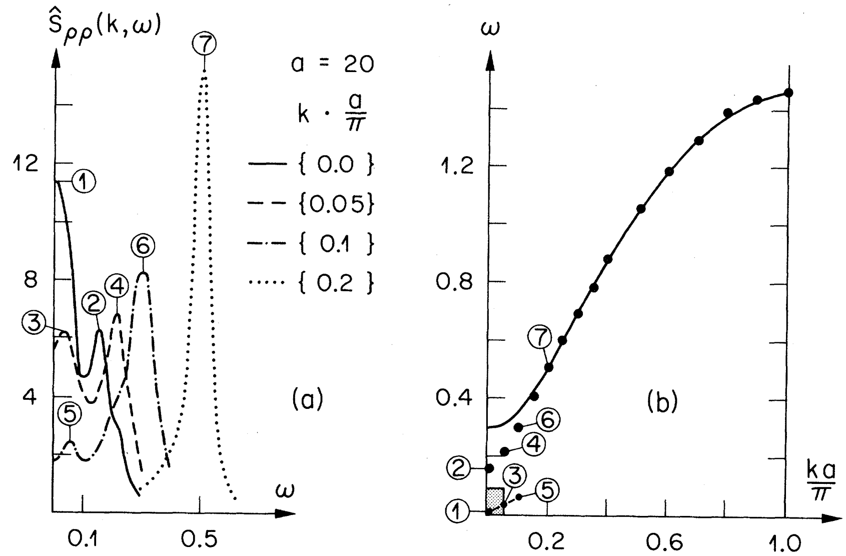

FIG. 1. (a) Frequency dependence of $\hat{S}_{\rho\rho}(k,\omega)$ at some fixed k values in model III ($A=\frac{7}{8}$, $B=\frac{1}{8}$, $C=\frac{1}{2}$) at $k_BT=0.0468$. a is the lattice constant. The peak maxima 2, 4, 6, and 7 are resonances due to the conventional soft mode. Peaks 3 and 5 represent evidence for the new excitation branch. (b) Dispersion curves determined from the peak maxima. The full line was obtained by the SCPA. The numbers label corresponding peak maxima in (a).

calculated

$$U_l^*(t) = \mathrm{Re} \sum_{|k| \le |k_c|} \sum_{|\omega| \le |\omega_c|} U(k,\omega) \times \exp[-i(kal+\omega t)], \quad (4)$$

describing the slow fluctuations of $U_l(t)$ in space and time.

The continuum version of Eq. (1) suggests that traveling-wave solutions

$$U_l(t) = U(la - vt) \quad (5)$$

of the associated nonlinear equation of motion may dominate $U_l^*(t)$, provided that $v^2 < Ca^2$. In fact, Krumhansl and Schrieffer[7] identified these particular solutions as traveling cluster (domain) waves.

In Fig. 2 we plot the calculated $U_l^*(t)$, where negative values have been suppressed. Increasing blackening corresponds to increasing displacements. This plot demonstrates that clusters of locally ordered regions and cluster walls exist. Another important result is that the clusters propagate with a finite lifetime and a velocity v, where v is distributed around $v_0 \approx 0.25a$. This value agrees quite well with the group velocity of the new excitations [$0.2a$, Fig. 1(b)]. Therefore, from the above results, one will be led naturally to the conclusion that the new excitations appearing close to the displacive limit (model III) and close to $T=0$ correspond to traveling cluster waves. This interesting result is consistent with the existence of traveling-wave solutions [Eq. (5), $v^2 < Ca^2$], but is by no means guaranteed by the existence of these solutions. In fact, for $T \gg 0$, the dynamics is dominated by the high-frequency excitations only.

Let us now turn to the central-peak phenomenon appearing close to and at $k=0$ [Fig. 1(a)]. Recognizing that the clusters propagate with a finite lifetime (Fig. 2), it is obvious that for small k, where $\omega = v_0 k \approx 0$, the damped traveling cluster waves become overdamped, and give rise to the

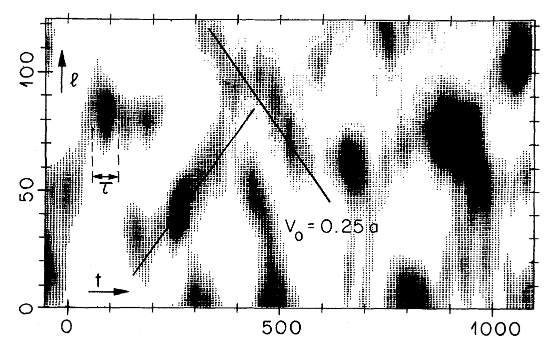

FIG. 2. Hypsometric plot of $U_l^*(t)$ [Eq. (4)] for model III at $k_BT=0.0468$. Negative values of $U_l^*(t)$ have been suppressed. Increasing blackening corresponds to increasing displacements. $v_0=0.25a$ denotes the mean velocity of the cluster waves and τ is the cluster lifetime.

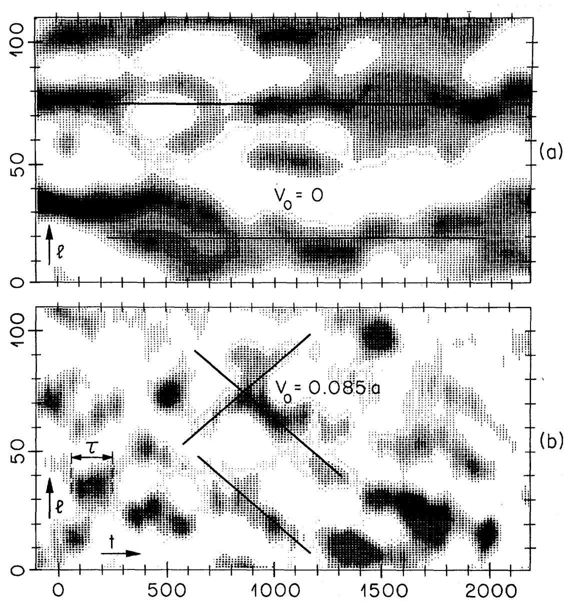

FIG. 3. Hypsometric plot of $U_l^*(t)$ [Eq. (4)] for model I ($A=-1$, $B=\frac{1}{3}$, $C=\frac{1}{2}$) at (a) $k_BT=1.5$ and (b) $k_BT=3.0$. v_0 denotes the mean velocity of the cluster wave.

central-peak phenomenon. Consequently, the central-peak half-width is proportional to the inverse lifetime, and the central-peak height is proportional to the lifetime of the large clusters.

Above we have considered only model III which is close to the displacive limit ($A=2C$). To study the influence of the model parameters we also considered a model I, belonging to the order-disorder regime with $A=-1$, $B=\frac{1}{3}$, $C=\frac{1}{2}$. From Fig. 3(a) it is seen that, in this model, the clusters do not propagate at low temperatures. Moreover, the lifetime of the cluster waves is very long. At higher temperatures [Fig. 3(b)], however, the cluster waves again propagate and their lifetime is seen to be shorter. Therefore, by approaching $T=T_c=0$, we find that the lifetime of the clusters increases, implying that the central-peak half-width vanishes at $T=T_c=0$.

To summarize, we have shown that in a model system for ferrodistortive phase transitions traveling cluster waves exist. Their lifetime was found to increase by approaching $T=T_c=0$ and determines the central-peak height and half-width. From this result it then follows that the central-peak phenomenon is due to the lifetime of the cluster waves. This lifetime also explains the tendency of our systems to approach a thermalized (ergodic) state, in contrast to the system of Fermi, Pasta, and Ulam.[8] Furthermore, we have presented numerical evidence for traveling cluster waves, as suggested by the existence of corresponding particular solutions of the equations of motion.[7] Close to $T=T_c=0$ and close to the displacive limit, these traveling cluster waves give rise to a cluster excitation branch appearing in addition to the phonon resonances. In the order-disorder regime, however, traveling waves are found only at higher temperatures, as shown in Fig. 3(b).

We remark in closing that these results are not reminiscent of one-dimensional systems. In fact, an analysis of two-dimensional ferrodistortive models, similar to those treated in Refs. 5 and 10, leads to the same conclusions close to T_c. Finally, we note that the uncertainties, due to numerical noise, in the results quoted are less than 5% in the region of the central peak. Moreover, our quoted results should not depend on the size of the system, because periodic boundary conditions have been chosen and the temperatures are such that the correlation length is smaller than the linear dimension of the system (see Figs. 2 and 3).

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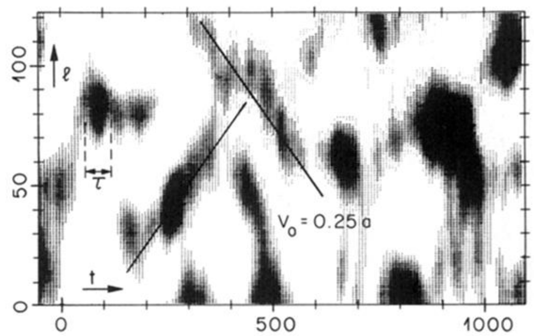

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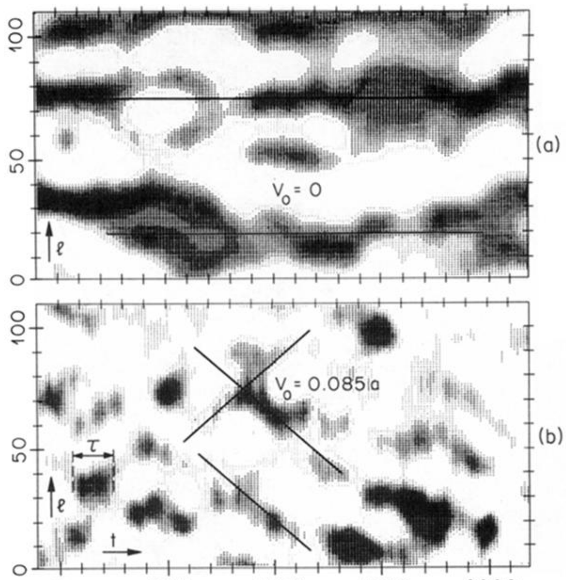

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