Molecular-Dynamics Study of a Two-Dimensional Ferrodistortive XY Model with Quartic Anisotropy

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We give numerical evidence that in a two-dimensional lattice-dynamical XY model with quartic anisotropy a conventional continuous phase transition occurs. The formation of clusters in the angular displacement field separated by 90° walls is also demonstrated. Moreover, we give evidence that the critical dynamics of the order-parameter fluctuations is dominated by a central peak which splits at finite wave vectors. This phenomenon is traced back to damped traveling 90° walls.

There has been a number of recent calculations of critical phenomena in two-dimensional XY models.[1] Of particular interest is the lack of a nonvanishing order parameter at any finite temperature in isotropic systems.[2] However, this does not rule out some different phase transitions in the isotropic case,[3] or the existence of a conventional transition in the presence of anisotropy.

In this Letter, we give numerical evidence that a conventional continuous phase transition occurs in a two-dimensional ferrodistortive lattice-dynamical XY model with quartic anisotropy. Moreover, we demonstrate the formation of clusters in the angular displacement field separated by 90° walls. Finally, we give evidence that the excitation spectrum is more complex than the conventional theory of lattice dynamics predicts. Our results reveal, in addition to the expected transverse-soft-phonon branch, a central peak which splits at finite wave vectors. The latter phenomena are traced back to damped traveling cluster walls in the angular displacement field, which dominate the critical slowing down.

Our Hamiltonian reads

$$\mathcal{H}=\sum_{l,\alpha}\frac{P_{l\alpha}^{2}}{2M}+\frac{A}{2}\sum_{l,\alpha}X_{l\alpha}^{2}+\frac{B}{4n}\sum_{l}\left(\sum_{\alpha}X_{l\alpha}^{2}\right)^{2}+\frac{B_{1}}{4}\sum_{l,\alpha}X_{l\alpha}^{4}-C\sum_{\langle l,l'\rangle\alpha}X_{l\alpha}X_{l'\alpha}. \quad (1)$$

Note that a rigid square reference lattice has been assumed; thus the model has no acoustic-phonon branches in the self-consistent phonon approximation.[4] l denotes the lattice sites, $P_{l\alpha}$ and $X_{l\alpha}$ are the αth components ($\alpha=1,2$) of momentum and displacement, respectively, of particles with mass M. M, A, B, C, and B_1 are model parameters. If the kinetic energy is neglected, the Hamiltonian (1) may also be considered as a lattice model for magnetic systems, where at each site l there is a spin variable $\vec{X}_l$ with n components.[5]

Using a molecular-dynamics technique, simulating a canonical ensemble,[4] we calculated some static and dynamic properties of a model I defined by Eq. (1) and the parameters

$$A=-1,\quad B=\tfrac{4}{5},\quad B_{1}=-\tfrac{1}{15},\quad C=\tfrac{1}{4}. \quad (2)$$

The time is chosen in such a way that the mass of the particles is equal to 1. We studied systems of 1600 and 3200 particles, subjected to periodic boundary conditions. For technical details on the molecular-dynamics method, we refer to Ref. 4. From the temperature dependence of the order parameter we estimated $k_{B}T_{c}\approx 2.72$ and $\beta=0.13\pm 0.03$. Similarly, we also estimated the isothermal order-parameter susceptibility and found $\gamma\approx\frac{7}{4}$ for $\epsilon=(T-T_{c})/T_{c}<0.2$ and $\gamma_{\mathrm{eff}}\approx 3$ for $0.2<\epsilon<0.5$. This result may be understood by noting that the model reduces to two decoupled two-dimensional Ising models for $B=0$ and $-B_{1}=B/3$. For $B_{1}=0$, however, it is equivalent to the nonuniversal isotropic XY model.[3] For model parameters associated with an ordered phase one expects, therefore, Ising critical behavior, except for the special case $B_{1}=0$. This picture is well confirmed by our estimates for β and γ and the observed changeover ($\gamma_{\mathrm{eff}}\approx 3$) further away from T_{c}.

To clarify whether or not the formation of clusters, their dynamics, and the associated central-peak phenomena are reminescent of Ising-like

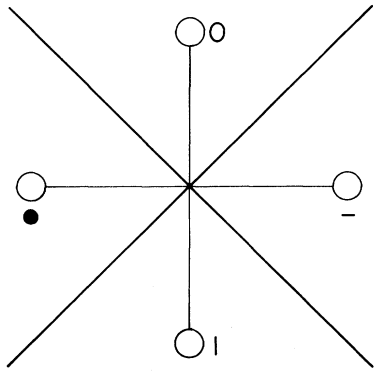

FIG. 1. The four quadrants of the local angular displacement field denoted by −, 0, ●, and |. The instantaneous position of a particle lies in one of the quadrants.

($n=1$) systems,[4] we analyzed the time evolution of the system in more detail. An analysis of the angular displacements revealed that clusters are formed, built up of particles connected by nearest-neighbor bonds. Figure 1 illustrates the four cluster patterns, separated by 90° walls, which may be distinguished. Figure 2 shows snapshots of cluster configurations in model I at various temperatures. The system has been prepared so that $(\langle X_{l1}\rangle^2)_{T=0}=(8C-A)(B/2+B_1)$ and $\langle X_{l2}\rangle=0$ at $T=0$, so that the system is in a single-domain state initially. It is seen that with increasing temperature clusters denoted by open circles and vertical bars are formed at first, while closer to T_c, clusters denoted by filled circles also appear. Moreover, the cluster size increases when T_c is approached from above or below. Finally, clusters are preferably separated by 90° walls. Above T_c, the number of large clusters is seen to decrease with increasing temperature.

It is clear that the complicated motions associated with the formation of such clusters will also affect the excitation spectrum. To explore this expectation, we calculated the following dynamic structure factors:

$$\left.\begin{matrix}\hat{S}_L(\vec{k},\omega)=\hat{S}_{11}(\vec{k},\omega)\\ \hat{S}_T(\vec{k},\omega)=\hat{S}_{22}(\vec{k},\omega)\end{matrix}\right\}=\frac{\int_{-\infty}^{+\infty}dt\,e^{-i\omega t}S_{\alpha\alpha}(\vec{k},t)}{S_{\alpha\alpha}(\vec{k},t=0)},\qquad(3)$$

where

$$S_{\alpha\alpha}(\vec{k},t)=\langle X_\alpha(-\vec{k},0)X_\alpha(\vec{k},t)\rangle,\quad\alpha=1,2,\qquad(4)$$

$$X_\alpha(\vec{k},t)=(1/\sqrt{N})\textstyle\sum_l\exp(i\vec{k}\cdot\vec{R}_l)\times[X_{l\alpha}(t)-\langle X_{l\alpha}\rangle].\qquad(5)$$

Figure 3(a) shows the frequency dependence of

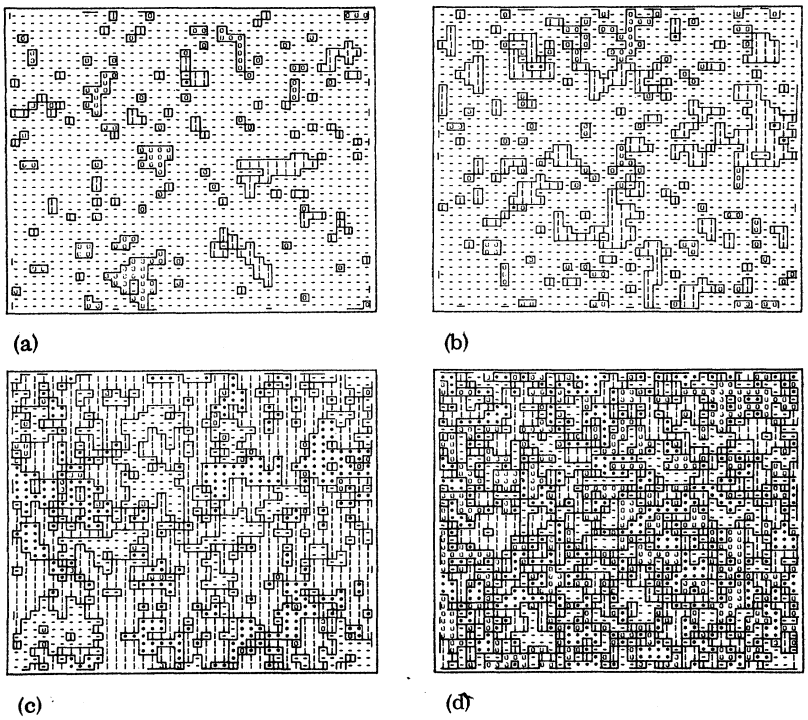

FIG. 2. Snapshots of cluster patterns in model I at various temperatures: k_BT equals (a) 2.0, (b) 2.5, (c) 3.0, and (d) 6.0. According to Fig. 1, we distinguish four cluster patterns; clusters are particles connected by a nearest-neighbor bond with positions lying in the same quadrant.

$S_L(\vec{k},\omega)$ and $S_T(\vec{k},\omega)$ for fixed $\{k_X,0\}$ values and $k_BT=2.5$. At $\vec{k}=0$, $S_L(\vec{k},\omega)$, being associated with the order-parameter fluctuations, only exhibits a central peak, which splits into a double-peak structure at finite k_X values. The $S_T(\vec{k},\omega)$ spectrum is dominated by a phonon resonance and no central peak appears. In Fig. 3(b) we plotted the dispersion curves as obtained from the peak maxima. There are two low-frequency branches: a transverse phonon branch with a gap, and a new excitation branch arising from the splitting of the central peak.

To elucidate the physical origin of these results, it is illustrative to consider the continuum limit of the Hamiltonian (1). The corresponding equation of motion for the angular component of the displacement field is

$$\Delta\varphi(X,Y)-\frac{M}{Ca^2}\ddot{\varphi}(X,Y) = -\frac{B_1\rho^2(X,Y)}{4Ca^2}\sin 4\varphi(X,Y), \qquad (6)$$

where ρ is the radial component of the field, and a denotes the lattice constant of the square reference lattice. Equation (6) is just the sine-Gordon equation which is invariant under a Lorentz transformation.[6] From the model parameter chosen and the cluster patterns shown in Fig. 2, it follows that the essential features of the critical dynamics may be extracted by keeping the radial component of the field constant. Consequently, we expect small-amplitude solutions, corresponding to the transverse phonon branch, and 90°-cluster-wall solutions. The small-amplitude solutions have frequencies

$$M\omega_T^2(\vec{k}) = -B_1\rho^2 + Ca^2k^2, \qquad (7)$$

and the traveling 90°-wall solutions are given by[7]

$$\varphi(X, Y=0, t) = \arctan[\pm\exp(\pm\alpha\zeta)], \qquad (8)$$

where

$$\alpha^2 = \frac{-B_1\rho^2}{Ca^2}\left(1-\frac{V_X^2M}{Ca^2}\right)^{-1}, \quad \zeta = X - V_Xt. \qquad (9)$$

V_X is the velocity of the 90° wall and enters here as a parameter. To our knowledge Eq. (8) is the only stable traveling-wave solution, $\varphi(X,t) = \varphi(X-V_Xt)$, of Eq. (6).[6] A reasonable value for the magnitude of the radial field is $\rho^2 = \langle X_{I1}^2 + X_{I2}^2\rangle$.

It should be emphasized that the 90°-wall solution is consistent with the observed cluster patterns and the associated boundaries (Fig. 2).

Noting that the small-amplitude solution leads, below and not too close to T_c, to transverse fluctuations only, one is naturally led to the conclusion that the peaks in $S_T(k_X,\omega)$ represent the transverse phonon resonance with frequency ω_T [Eq. (7)]. The 90°-wall solution, however, obviously leads to longitudinal fluctuations. The central peak and its splitting must then be attributed to overdamped and underdamped traveling 90° walls. This conclusion is also consistent with our results on one-component systems.[4,7]

Above T_c, the longitudinal and transverse dynamic form factors become identical. As shown in Fig. 4(a), the $\vec{k}=\vec{0}$ spectrum is then dominated by a central peak, which at finite wave vectors $\{k_X,0\}$ splits into a symmetric four-peak structure. In accordance with the above analysis, we identify the resulting lower branch [see Fig. 4(b)] as due to overdamped or underdamped traveling cluster walls. The higher branch corresponds to the transverse phonon branch. At higher temperatures, the central peak and the associated cluster-wall excitation branch disappear. Moreover, the phonon branches develop a gap which increases with temperature.

We also note that the central-peak half-width is found to decrease, and its height to increase, when T_c is approached from above or below.

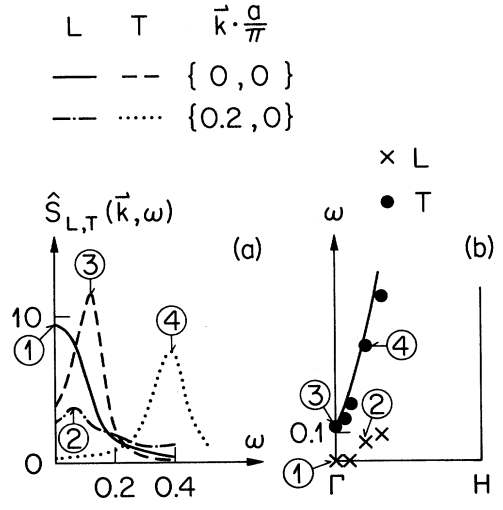

FIG. 3. (a) Frequency dependence of the longitudinal $[S_L(\vec{k},\omega)]$ and transverse $[S_T(\vec{k},\omega)]$ dynamic form factor for various $\{k_X,0\}$ at $k_BT=2.5$. Peaks 3 and 4 are resonances associated with the transverse phonon branch. Peaks 1 and 3 are due to the traveling cluster walls. (b) Dispersion curves determined from the peak maxima; the numbers label corresponding peak maxima.

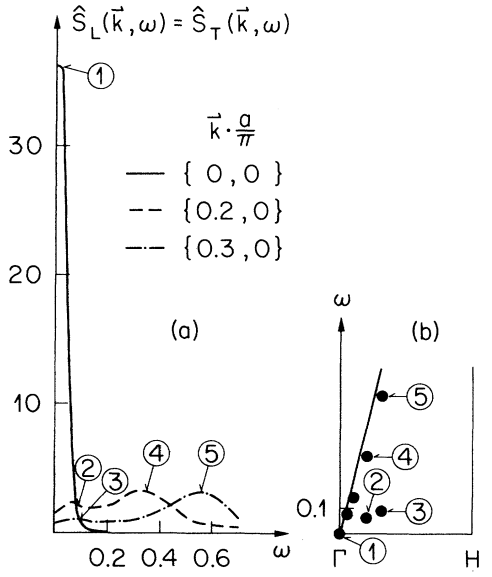

FIG. 4. (a) Frequency dependence of $S_L(\vec{k},\omega) = S_T(\vec{k},\omega)$ for various $\{k_x,0\}$ at $k_BT = 3 > k_BT_c \approx 2.72$. Peaks 4 and 5 are due to a transverse phonon, and peaks 2 and 3 arise from underdamped traveling cluster walls. (b) Dispersion curves determined from the peak maxima; the numbers label corresponding peak maxima.

Hence, the critical slowing down of the order-parameter fluctuation is intimately reflected in the central-peak phenomenon.

In conclusion, we have presented numerical evidence that the critical behavior of a classical two-dimensional lattice-dynamical XY model with quartic anisotropy is consistent with the exponents of the two-dimensional Ising model.

Moreover, we have shown that the formation of clusters, their dynamics, and the associated central-peak phenomena are not artifacts of one-component systems.[4] In fact, we have demonstrated that clusters are formed in the angular displacement field which are separated by 90° walls. The excitation spectrum was found to be more complex than the conventional theory of lattice dynamics would predict. In fact our results reveal that, sufficiently close to T_c, the dynamic form factor of the order-parameter fluctuations is dominated by a central peak which splits at finite wave vectors and dominates the critical slowing down. The central peak and the associated excitation branch were traced back to traveling 90° walls.

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[1]For a review see D. D. Betts, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1974), p. 569.

[2]N. D. Mermin and H. Wagner, Phys. Rev. Lett. 17, 1133 (1966).

[3]A. Luther and D. J. Scalapino, unpublished.

[4]T. Schneider and E. Stoll, Phys. Rev. B 13, 1216 (1976).

[5]M. E. Fisher, Rev. Mod. Phys. 46, 597 (1974).

[6]A. Barone, F. Esposito, C. J. Magee, and A. C. Scott, Riv. Nuovo Cimento 1, 227 (1971).

[7]T. Schneider and E. Stoll, Phys. Rev. Lett. 31, 1254 (1973), and 35, 296 (1975).

Slow Ferroelectric Cluster Dynamics in KH_2AsO_4 and KH_2PO_4 from Cr^{5+} Paramagnetic Resonance

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We succeeded in substituting the paramagnetic Cr^{5+} ($S=\frac{1}{2}$) ion with the same size as As^{5+} in KH_2AsO_4, for P^{5+} in KH_2PO_4, and in their deuterated isomorphs. In the crystals the high-temperature resonance spectrum showing a local tetragonal symmetry is broken on the time scale of the EPR experiment below $T^* > T_C$, so that orthorhombic spectra appear in the paraelectric tetragonal phase. T^* shifts proportionally to the Curie temperature T_C upon deuteration. We deduce the existence of ferroelectric clusters with lifetimes τ longer than 1.5×10^{-8} sec below T^*.

In this Letter, an EPR investigation on Cr^{5+} is presented which shows in the paraelectric phase the existence of slow local dynamic fluctuations in potassium dihydrogen arsenate and phosphate (KDA and KDP) as well as their deuterated isomorphs. These fluctuations are of ferroelectric