we note that Berker, Ostlund, and Putnam predict $\beta = 0.10$ for their PLG model compared with our $\beta = 0.09 \pm 0.03$.

To conclude, we reiterate the fact that these experiments have illustrated that x rays, in spite of their lack of surface selectivity, may provide extremely detailed information about surface film melting. Our experiments provide strong support for the PLG model of commensurate krypton on graphite, including the crossover from first- to second-order melting. However, primarily because of finite-size effects, we have only been able to demonstrate consistency rather than uniqueness. Critical liquid scattering measurements should be able to resolve the remaining ambiguities. Our initial data suggest that such measurements are indeed within the scope of an x-ray experiment and we anticipate results in the near future.

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Dynamics of an Ising-like Model in Four Space Dimensions

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Using a molecular-dynamics technique, we studied the excitation spectrum of an Ising-like lattice-dynamic model in four space dimensions. We found a central peak close to the transition temperature which does not evolve from the damped soft mode. Our results reveal that the overdamped–soft-mode picture and current interpretations of renormalization-group results for dynamic properties should be revised. They also provide testable predictions for Ising-like dipolar systems.

A recent development has been the application of renormalization-group techniques to dynamic critical phenomena.[1] As in the static case,[2] most calculations have been performed as an expansion in the parameter $\epsilon = 4 - d$, where d is the space dimension. For $d \geq 4$, the exponents for static critical phenomena are expected to be classical in most cases, and the conventional theory of critical slowing down[1] should hold. For an Ising-like system with dipolar forces, however, the static and dynamic critical properties will be classical for $d \geq 3$.[2,3] The renormalization-group analysis also indicates that the critical dynamics of lattice-dynamic systems is equivalent to corresponding stochastic models with conserved energy.[1,4] This has led to the conclusion that for lattice-dynamic systems with short-range forces and $d \geq 4$, the dynamic critical behavior is associated with an overdamped soft mode. In particular, a sharp central peak around zero frequency, additional to a damped soft-mode resonance, should not occur in the spectral density of the order-parameter fluctuations.[1,4] For Ising-like systems with dipolar forces these conclusions are expected to hold for $d \geq 3$.

The purpose of this Letter is to test these predictions by means of a molecular-dynamics technique, the conclusion being that the excitation spectrum of a four-dimensional Ising-like lattice-dynamic model with short-range forces differs, close to T_c, from the overdamped–soft-mode picture. This result does not imply that the critical slowing down is nonconventional. It reveals,

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however, that the current interpretation of renormalization-group results for dynamic properties should be revised. The plan of this Letter is to sketch the model, to discuss some molecular-dynamics results, and to make connection to the renormalization-group predictions and the overdamped-soft-mode picture.

The Hamiltonian of the Ising-like system reads

$$\mathcal{H}=\sum_l \frac{M\dot{X}_l^2}{2}+\frac{A-4dC}{2}\sum_l X_l^2+\frac{B}{4}\sum_l X_l^4 + \frac{C}{2}\sum_{\langle l,l'\rangle}(X_l-X_{l'})^2, \quad (1)$$

where l refers to sites on a d-dimensional simple hypercubic lattice. X_l is the displacement, and $M\dot{X}_l=M\,dX_l/dt$ the momentum, of the particle in the lth unit cell. A, B, C, and M are model parameters, which are chosen here as

$$A=-1,\quad B=\tfrac{1}{3},\quad C=\tfrac{1}{8},\quad M=1,\quad d=4. \quad (2)$$

Here we have adopted the same units as Schneider and Stoll.[5] This choice guarantees that at $T=0$ the order parameter, given by

$$\langle X_l\rangle_{T=0}^2=\frac{16C-A}{B}=9, \quad (3)$$

does not banish. The system undergoes a ferrodistortive phase transition at some $T=T_c>0$.[6] In the so-called Ising limit, where $A\to-\infty$, $B\to+\infty$, but $A/B=1$, the partition function reduces essentially to that of the Ising model.[7] Accordingly, the system defined by Hamiltonian (1) belongs to the Ising universality class. Moreover, the only conserved density for the Hamiltonian (1) is the energy. According to the universality hypothesis for dynamic properties, one would therefore expect the system to exhibit the critical dynamics of a stochastic Ising-like model with conserved energy.[1,4]

To investigate the static and dynamic properties of the system, we shall consider the variables

$$X(\vec{q})=N^{-1/2}\sum_l(X_l-\langle X_l\rangle)\exp(i\vec{q}\cdot\vec{R}_l), \quad (4)$$

$$\mathcal{H}(\vec{q})=N^{-1/2}\sum_l(\mathcal{H}_l-\langle\mathcal{H}_l\rangle)\exp(i\vec{q}\cdot\vec{R}_l), \quad (5)$$

where

$$\mathcal{H}_l=\frac{M\dot{X}_l}{2}+\frac{A}{2}X_l^2+\frac{B}{4}X_l^4-C\sum_{l'}X_lX_{l'}. \quad (6)$$

These variables describe displacement and energy fluctuations of wave vector $\vec{q}$. The vectors $\vec{R}_l$ define the rigid reference lattice. The excitation spectrum might be analyzed with the aid of the spectral densities

$$\hat{S}_{XX}(\vec{q},\omega)=\int_{-\infty}^{+\infty}dt\,e^{-i\omega t}\frac{\langle X(-\vec{q},t)X(\vec{q},0)\rangle}{\langle X(-\vec{q},0)X(\vec{q},0)\rangle}, \quad (7)$$

$$\hat{S}_{\mathcal{H}\mathcal{H}}(\vec{q},\omega)=\int_{-\infty}^{+\infty}dt\,e^{-i\omega t}\frac{\langle \mathcal{H}(-\vec{q},t)\mathcal{H}(\vec{q},0)\rangle}{\langle \mathcal{H}(-\vec{q},0)\mathcal{H}(\vec{q},0)\rangle}. \quad (8)$$

At low temperatures ($T\ll T_c$), where anharmonic perturbation theory is adequate, $\hat{S}_{XX}(\vec{q},\omega)$ will be dominated by the phonon resonance at[8]

$$M\omega^2(\vec{q})=(A-16C)+3B\langle X_l^2\rangle + 4C[F(\vec{0})-F(\vec{q})], \quad (9)$$

where

$$F(\vec{q})=\sum_{i=1}^{4}\cos aq_i; \quad (10)$$

here a is the lattice constant and q_i the ith component of $\vec{q}$. Because of energy conservation and the linear coupling between order parameter and energy fluctuations, both $\hat{S}_{XX}(\vec{q},\omega)$ and $\hat{S}_{\mathcal{H}\mathcal{H}}(\vec{q},\omega)$ will also exhibit a heat-diffusion peak or second-sound resonances for small $\vec{q}$ and ω.

To elucidate the temperature dependence of the excitation spectrum, we used a molecular-dynamics technique simulating a canonical ensemble with nearly conserved energy. For a detailed description of this technique, we refer to Ref. 8. Here we considered a system of 10^4 particles, subjected to periodic boundary conditions. The transition temperature T_c was estimated from the temperature dependence of the order parameter and of the static susceptibility, by taking the logarithmic corrections into account. On this basis we found $k_BT_c\approx7.8$, which can be compared with the mean-field result[9]

$$k_BT_c=\alpha\frac{16C}{3B}(16C-A)=8.92,\quad \alpha\approx1.49. \quad (11)$$

α is a monotonic function of $A/16C$, where $\alpha=3$ in the Ising limit and $\alpha=1$ for $A/16C=1$.

Let us then turn to the excitation spectrum close to T_c. Figure 1 shows the calculated ω dependence of $\hat{S}_{XX}(\vec{q},\omega)$ for some $\vec{q}$ values at $k_BT=7$. A crucial feature is the occurrence of a central peak (CP) in addition to the damped soft-mode resonance. This structure is not expected for $\vec{q}\neq0$. In fact, below T_c there is a direct coupling between energy and displacement fluctuations. The heat-diffusion CP will appear, therefore, in both $\hat{S}_{\mathcal{H}\mathcal{H}}(\vec{q},\omega)$ and $\hat{S}_{XX}(\vec{q},\omega)$; its half-width tends to zero, however, as q^2. Consequently, the CP at $\vec{q}=0$ (Fig. 1) cannot be traced back

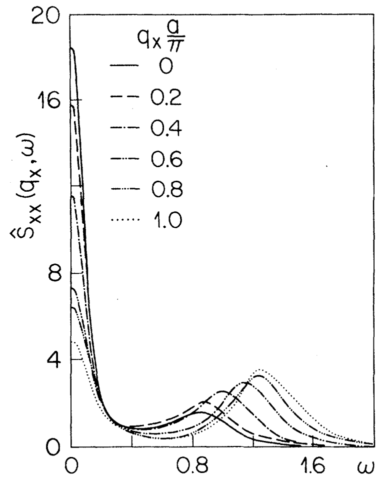

FIG. 1. $\hat{S}_{xx}(\vec{q},\omega)$ [Eq. (7)] at $k_B T=7$ for $\vec{q}=(q_x,0,0,0)$.

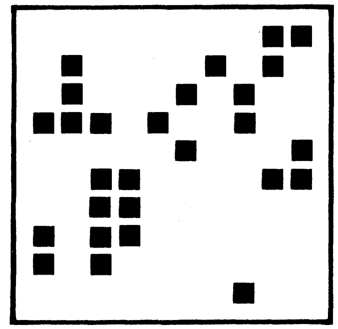

FIG. 2. Snapshot of the displacement pattern in an $(X_1, X_2, 0, 0)$ plane at $k_B T=7$ demonstrating the formation of clusters separated by walls. The squares designate particles whose local displacements X_l have sign opposite to $\langle X_l \rangle_{T=0}$. The displacements of the missing particles have the same sign.

to heat diffusion. Moreover, its occurrence is inconsistent with the conclusions drawn from renormalization-group considerations, according to which the CP at $\vec{q}=0$ should evolve from the gradual overdamping of the soft mode, with frequency approximately given by Eq. (9).

The essential features of $\hat{S}_{XX}(\vec{q},\omega)$ (Fig. 1) are consistent, however, with those of the corresponding three-dimensional system, which we studied recently.[8] There the CP at $\vec{q}=0$ has been traced back to large-amplitude motions associated with the dynamics of clusters. The clustering phenomenon is equivalent to the appearance of d-dimensional microdomain structures in displacement pattern. As Fig. 2 demonstrates, such clustering also occurs for $d=4$. Our results for $k_B T=5$, 10, and 12 reveal that the equivalence of the excitation spectrum between the three- and four-dimensional systems is similar to $k_B T=7$, and seems to hold at all temperatures. In particular, we also found for $d=4$ that the strength of the soft-mode resonance decreased on approaching T_c.

To summarize, we have shown that the CP appearing in $\hat{S}_{XX}(\vec{q}=0,\omega)$ does not evolve from a damped or overdamped soft mode. In analogy to the three-dimensional system, this CP is traced back to the formation and the dynamics of clusters separated by walls. The appearance of the CP additional to the soft-mode resonance is inconsistent with the conclusions drawn from renormalization-group considerations. This inconsistency does not, however, invalidate the estimates for the dynamic critical exponents; it merely reveals that the CP and, in turn, the critical slowing down must be attributed to the cluster dynamics and not to an overdamped soft mode, which becomes irrelevant for small $\vec{q}$ values on approaching T_c. The removal of the damped soft mode as the driving mechanism for the critical slowing down offers then a pictorial illustration of universality in the Ising class in terms of clusters and their dynamics. Clearly, the cluster dynamics will also be affected by energy conservation. Moreover, the demonstrated irrelevance of the soft mode in the critical dynamics calls for reliable experiments, in particular light- and neutron-scattering ones in dipolar Ising-like systems, where the present results should hold in three dimensions.

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