density and λ_D the electron Debye length. My analysis clearly shows that for a strongly turbulent system, one must consider both these nonlinearities which compete with each other; depending on which of the two nonlinearities is dominant, Q would be positive or negative which in turn will ascertain the possibility of occurrence of Langmuir collapse. For weakly turbulent systems, i.e., for $E^2/nKT \ll k^2\lambda_D^2$, noninertial nonlinearity, considered herein, dominates the nonlinearity arising because of ponderomotive force. It is worth pointing out that in Zakharov's[3] case, $Q<0$ which according to my analysis would correspond to a modulationally unstable case. Hence I assert once again that the collapse takes place only if the system is modulationally unstable and it is wrong to say that in three dimensions Langmuir waves always undergo collapse.

In conclusion, I would like to point out the two basic advantages of my method over the procedure followed by Zakharov.[3] Firstly Zakharov's method is not applicable to low-frequency waves, in particular ion-acoustic waves because of the averaging done on fast time scales. Secondly my method can consistently take care of both the nonlinearities mentioned above without neglecting the electronic nonlinearities and the second derivative in time as done by Zakharov.[3] The extension of present analysis, for strongly turbulent systems, including the ion dynamics, will be reported in a forthcoming paper.

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Ordered Helium Films on Highly Uniform Graphite—Finite-Size Effects, Critical Parameters, and the Three-State Potts Model*

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The heat capacity of the ordering transition of helium films is studied on a new graphite substrate whose microcrystallite allignment and lateral size are ten times that for Grafoil. Transition peaks are considerably sharper than those observed on Grafoil allowing determination of critical parameters $\alpha \cong 0.36$ and $A^+/A^- \cong 0.65$. The α is substantially larger than recent three-state Potts-model calculations in two dimensions (of which the ordering transition is the only example), raising the possibility that the ordered state belongs to a new universality class.

Considerable research has now been conducted with gases adsorbed on the substrate Grafoil, which is graphite exfoliated, and rolled from ground natural crystals.[1] Vapor pressure,[2] heat capacity,[3,4] neutron scattering,[5] NMR,[2,6] spreading pressure,[7] and other techniques have explored the monolayer and multilayer phases for a host of adsorbates on Grafoil. These studies relied on Grafoil's large surface area (20 m^2/g), partial crystallite orientation[5] (basal planes are $\lesssim 30°$ to sheet), and adequate thermal conductivity. However, the crystallites of this substrate appear to be quite small[5,7] ($\lesssim 200$ Å) with a sizable fraction having a random orientation.[5] These features are known to have an appreciable effect on film behavior.[5,8] I report here the results of a heat capacity experiment performed on UCAR oriented graphite grade *ZYX* [1] which is carefully exfoliated from a stress-annealed pyrolytic graphite single crystal of monochronometer grade. Although the surface area is small and mass equilibrium times are long, the expanded *ZYX* is known to have an order of magnitude better crystallite orientation than Grafoil[9] ($\lesssim 3°$). This should be of considerable importance in enhancing signals for orientation-dependent experiments (NMR,[6] Mössbauer,[10] and neutron scattering[5]).

ZYX crystallites are surely far larger and more uniform than Grafoil.[9] To explore size effects I chose to investigate the ordering transi-

tion of He^3 and He^4 partial monolayer films.[3] A sharp heat-capacity anomaly at about 3 K is interpreted as signaling the transition between an imperfect two-dimensional (2D) gas and a regime ordered epitaxially with the hexagonal net of weak potential wells comprising the graphite substrate. Because of hard-core exclusion this ordered state occurs for a coverage of one helium atom per three graphite substrate wells, such that only next-nearest-neighbor wells are occupied. There has been considerable interest in this ordering transition,[11-13] and Alexander[14] has pointed out its analogy with the three-state Potts model[15] in 2D (an extended Ising model with three possible spin orientations). Mukamel, Fisher, and Domany[16] have surveyed the field and infer that the ordering transition is the only 2D three-state Potts-model system now known. Theorists Straley and Fisher[17] estimate the critical exponents for this model to be $\alpha = \alpha' = 0.05 \pm 0.1$, while Kim and Joseph[18] obtain 0.26, and Becker and Wortis estimate $\alpha \cong 0.20$.[19] The helium data on Grafoil indicated $\alpha \cong 0$, but a detailed analysis was not attempted since the peaks start to round quite far from the transition temperature T_c (due to the finite size of the Grafoil crystallites).

The present copper calorimeter containing five 1-in.$\times$1-in.$\times\frac{3}{16}$-in. *ZYX* wafers with a density $\lesssim 1$ g/cm^3 was outgassed at 800°C for 48 h. An argon isotherm at 77 K gave an adsorption area of 2.5 m^2/g for the 9.0-g sample (vs 20 m^2/g for Grafoil). The capsule with two $\frac{1}{8}$-W Allen-Bradley 330-Ω resistors attached was mounted to a nylon sleeve in the low-vibration cryostat of Magerlein and Sanders.[20] Thermometry was through a programmable ratio-transformer bridge (with 5-μK noise) interfaced for digital data acquisition and ratio incrementation at bridge zero crossings. Before and after temperature drifts due to the 1–3-mK heating pulses were fitted with least-squares exponentials on a PDP-10 computer. Extrapolation adjustments were $\lesssim 0.1\%$[21] and curvature corrections[22] were determined to be insignificant. A sizable, but regular, empty calorimeter signal was subtracted from the film data.

Figure 1 displays the best heat-capacity data for He^4 films on UCAR-*ZYX* and Grafoil substrates.[3] The *ZYX* peak is seen to rise fully twice as high as the Grafoil peak, and there is a significant reduction in peak width. Both curves asymptotically approach the classical 2D gas value $C/Nk = 1$ near 4 K and merge near 2.2 K, the low-temperature limit of the present experiment. A comparison of the film entropies at 4 K reveals a ~12% additional entropy for *ZYX* films. This is evidence that more helium atoms are participating in the ordering transition on the *ZYX* substrate than on Grafoil (probably because crystallite edge effects are less important). He^3 behaves similarly with maximum peak heights being $C/Nk = 10.4$ for *ZYX* and 5.4 for Grafoil.

Figure 2 shows peak temperatures for selected coverages of He^4 and He^3 in the ordering region

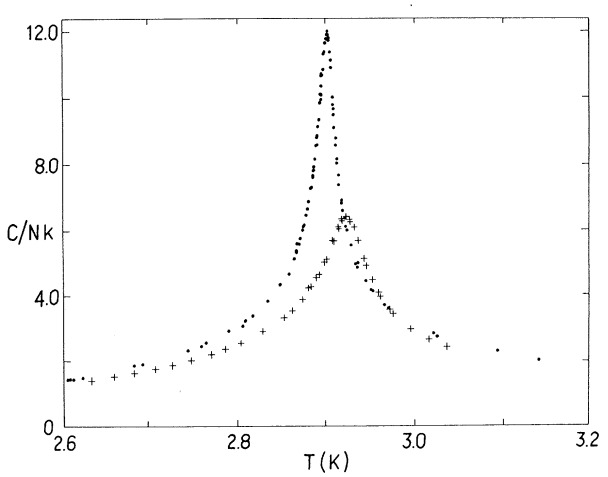

FIG. 1. Heat capacity for ordering peaks of He^4; (•), 3.194 STP cm^3 on UCAR-*ZYX*. (+), 62.45 STP cm^3 on Grafoil.

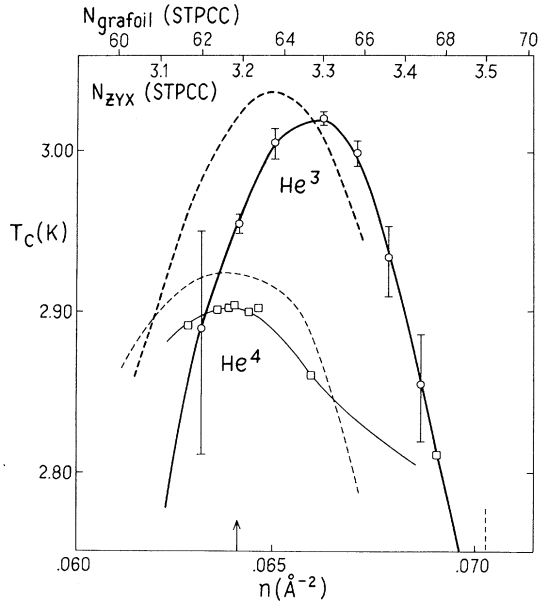

FIG. 2. Peak temperature vs film density n for ordering region on *ZYX*. (Dotted lines are Grafoil. Error bars indicate peak inflection points. Arrow denotes normalization coverage.)

for *ZYX* (solid line) and Grafoil[3] (dotted line). The top scales give the volumes of gas N (in STP cm^3) adsorbed in the calorimeters and the lower scale gives film density n in atoms per $Å^2$. Normalization of N for *ZYX* was chosen for best agreement with the He^4 Grafoil data. The similarities in the phase diagrams for these films is striking. For both substrates the phase diagrams exhibit a clear isotopic shift in coverage and temperature. These features have been attributed to particle statics and quantum tunneling.[3,12] The *ZYX* films have a small but real temperature depression from the Grafoil data (~20 mK) and the peak heights rise dramatically (Fig. 1 shows these features clearly). This temperature shift and peak behavior can be understood by finite-size considerations. An Ising model calculation of Ferdinand and Fisher,[23] for instance, showed that for a finite 2D rectangular array T_c falls toward the infinite-system transition temperature T_∞ as $\epsilon = (T_c - T_\infty)/T_\infty \sim n^{-1}$ and the heat-capacity maximum C_{max} rises toward a singularity as $\approx \ln(n)$, where n is the number of spins comprising the length of the array. Since spins transcribe to lattice sites, the observed T_c shift and peak sharpening for *ZYX* are evidence for an increased lateral extent of the crystallites.

The present data lend themselves to the careful critical-point heat-capacity analysis of Lederman, Salamon, and Schacklette.[24] I assume a power law $C^{\pm}/Nk = A^{\pm}|t|^{-\alpha} + B^{\pm} + ct$ close to T_c, where $t = (T - T_c)/T_c$, $\pm$ denotes the sign of t, and $B^{\pm}$ and ct are background terms. By eliminating $|t|^{-\alpha}$ at equal values of $|t|$ (the universality hypothesis $\alpha = \alpha'$ is invoked here), one obtains

$$(C^+/Nk - ct) = (A^+/A^-)(C^-/Nk - ct) + (B^+ - B^- A^+/A^-). \quad (1)$$

Plotting the $(C^-/Nk - ct)$ data vs interpolated $(C^+/Nk - ct)$ at equal values of $|t|$, and varying T_c and c one obtains the least-squares straight-line fit which determines T_c, A^+/A^-, and $B^+ - B^- A^+/A^-$. Then $(C/Nk - ct)$ is modified by Eq. (1) and combined with the actual $(C^+/Nk - ct)$ data to give the merged heat capacity C_M. This is plotted vs reduced temperature for determination if α, A, and B from a fit of the form $C_M/Nk = A|t|^{-\alpha} + B + ct$. For interpolation, the C^+/Nk data were approximated to within scatter by a fourth-order othogonal polynomial. Data were fitted over the range $2.5 \leq C^-/Nk \leq 10.75$. The linear plot and fit are given in Fig. 3 inset for $T_c = 2.903$ K. Sensitivity to T_c is apparent from the variation in the resulting parameters: $T_c = 2.9044 \pm 15$, $A^+/A^- = 0.65 \pm 0.13$, $(B^+ - B^- A^+/A^-) = 1.11 \pm 0.55$, $c = -0.5 \pm 2.5$. For T_c's outside of this range a perceptible bow appeared in the plot.

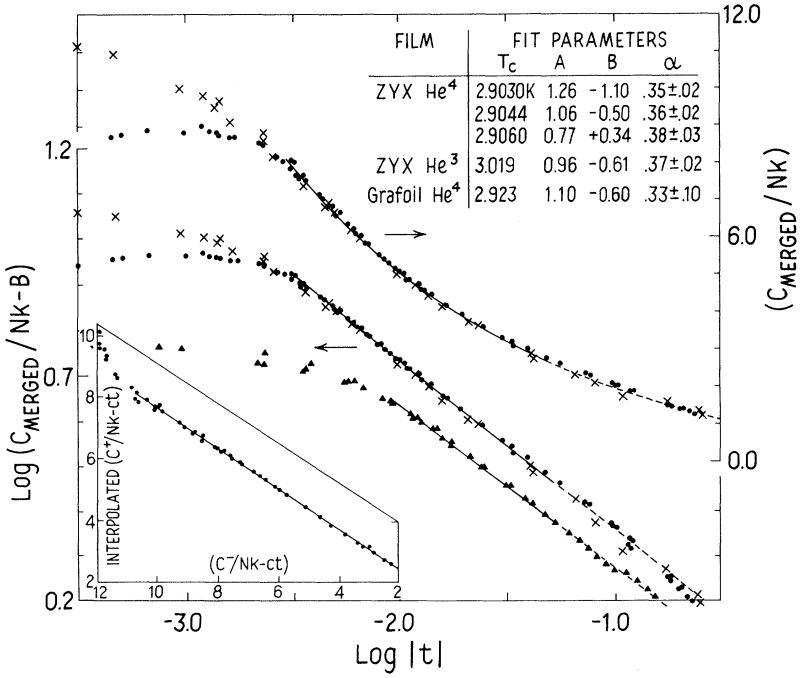

FIG. 3. Merged specific heat C_M for He^4 on *ZYX* with $T_c = 2.9044$ K (●, $t < 0$; ×, $t > 0$). Solid lines are best fits. For clarity the Grafoil data (▲) is offset one division. Inset shows the linear fit discussed in text.

Figure 3 provides log-log and semi-log plots of the *ZYX* He^4 merged data for $T_c = 2.9044$ K. A least-squares power law was fitted to the *ZYX* He^4 merged data over the range $-2.5 \leq \ln|t| \leq -1.3$ (solid line for semi-log plot in Fig. 3). The parameter B was also varied for best linear fit to the data in the log-log plot and the resulting α's agree with those found from the power-law fit. When extended, the curves for both plots follow the data out to $\ln|t| \simeq -0.75$ or over almost two decades in $|t|$. The critical exponent is $\alpha = 0.36 \pm 0.02$, with the uncertainty calculated from[25] $\chi_\pm^2 = \chi_0^2[1 + 1/(p - v - 1)]^{1/2}$, where p is the number of points, v is the number of variables, and χ_0^2 is for the best power-law fit. The results of similar He^4 analyses for other T_c's and for He^3 are included in the Fig. 3 table.

For comparison, the He^4 data were also analyzed with a standard multiparameter nonlinear least-squares program.[26] The results ($T_c = 2.901$, $\alpha = 0.40$, $\alpha' = 0.39$, $A^+/A^- = 0.74$, $B^+ = 0.31$, $B^- = -1.43$, and $c = -5.0$) are in good agreement with the above. When the background was restricted to $B^+ = B^-$, T_c shifted to 2.900 K and A^+/A^- went to 0.99. However, α remained stable at 0.38. An additional term $D^\pm|t|^{-x}$ was added to the analytical power function. The fit yielded $A^+/A^- = 1.06$, $D^+/D^- = 0.94$, $x = 0.13$, $B = -1.95$ with a χ^2 only half as large as in the $B^+ = B^-$ fit. T_c and α remained stable at 2.900 K and 0.37 respectively.

I conclude that α is *insensitive* to variations in T_c and simple modifications of the fitting function. These α's are considerably larger than the Potts-model estimates of 0.05–0.26 and the range usually associated with critical phenomena[23] ($-0.1 \lesssim \alpha \lesssim 0.2$). Furthermore, although the universal quantity A^+/A^- shows some variation it appears to be well below the usual range of 1.0–1.2.[23] This raises the intriguing possibility that the three-state Potts transition in 2D belongs to a new universality class not seen previously.

A similar procedure for the Grafoil data is not satisfactory since most of the peak is subject to background or rounding effects. Nevertheless, an analysis was attempted, the results of which are included in the log-log plot and inset table of Fig. 3. The uncertainty in α is large enough for consistency with the *ZYX* α but is far from 0.0, characteristic of a logarithmic divergence. A quantitative comparison of crystallite areas for the two substrates is possible from a consideration of the points of rounding t_r and C^r/Nk. Since the coherence length at rounding ξ^r is constrained to be no larger than the crystallite length, $\xi^r \sim n$, where n is the number of graphite potential wells along one face of the crystallite. Using $\xi \sim t^{-\nu}$ and $2\nu = 2 - \alpha$ from 2D scaling I obtain at rounding (subscript Z for *ZYX* and G for Grafoil)

$$n_Z/n_G \sim (t_Z{}^r/t_G)^{\alpha/2-1} = (0.0025/0.010)^{0.36/2-1} = 3.1, \quad (2)$$

$$n_Z/n_G \sim [C_Z{}^r/C_G{}^r]^{1/\alpha-0.5} = (10.25/5.3)^{1/0.36-0.5} = 4.5. \quad (3)$$

These estimates are in fair agreement. Thus, the area of the *ZYX* crystallites is 10 to 20 times that for the average Grafoil crystallites, which is presently characterized as $\lesssim 200$ Å^2. A further analysis of the peak maximum involving nonuniversality considerations and rounding exponents probably could provide some information on the distribution of crystallite sizes in *ZYX*.

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Deviations from Dynamic Scaling in Helium and Antiferromagnets

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We compute up to order ϵ^2 all dynamics transients (subleading exponents) for helium and symmetric antiferromagnets. We also discuss the relevance for helium of a fixed point which leads to a "weak" dynamic scaling.

Critical dynamics of systems like helium and symmetric antiferromagnets (AF) involving reversible mode coupling has been widely studied[1,2] using Wilson's[3] recursion method on stochastic Langevin-like equations or techniques of field renormalization which are useful to demonstrate scaling properties and get a relatively easy access to higher-order computations.[4,5] We have taken advantage of these techniques to investigate the two-loop (i.e., second order in $\epsilon = 4 - d$) approximations for the critical dynamic behavior of helium and $O(n)$ symmetric antiferromagnets. In particular we obtain to order ϵ^2 all subleading exponents (transients) which govern corrections to dynamic scaling. This allows us to distinguish in the ϵ-n plane, besides region (I) where standard dynamic scaling[6] holds, a region (II) involving "weak" dynamic scaling due to the occurrence of a "dangerous" irrelevant parameter.[7] In (I) the dynamic exponent is[6] $z = d/2$; in (II) it turns out to be $z = \frac{1}{2}(d + \omega_\lambda)$, where ω_λ is one of the three (He) or two (AF) subleading exponents which are computed here. The possible, but unlikely, physical relevance of region II for He is also discussed.

We write the Langevin stochastic equations for He introduced by Halperin, Hohenberg, and Siggia[1] in the form

$$\dot{\psi}_0{}^\alpha(t) = -\Gamma_0(1 + ib_0)\frac{\delta\mathcal{H}}{\delta\psi_0{}^{\alpha *}(t)} + ig_0\psi_0{}^\alpha(t)\frac{\delta\mathcal{H}}{\delta E_0(t)} + \eta^\alpha(t), \tag{1}$$

$$\dot{E}_0(t) = \Lambda_0\nabla^2\frac{\delta\mathcal{H}}{\delta E_0(t)} + ig_0\sum_\alpha\left(\psi_0{}^{\alpha *}(t)\frac{\delta\mathcal{H}}{\delta\psi_0{}^{\alpha *}(t)} - \psi_0{}^\alpha(t)\frac{\delta\mathcal{H}}{\delta\psi_0{}^\alpha(t)}\right) + \zeta(t), \tag{2}$$

where the space argument is understood and

$$\mathcal{H} = \int d^dx\{\textstyle\sum_\alpha(|\nabla\psi_0{}^\alpha|^2 + r_0|\psi_0{}^\alpha|^2) + \frac{1}{2}E_0{}^2 + \gamma_0 E_0\sum_\alpha|\psi_0{}^\alpha|^2 + (u_0/3!)(\textstyle\sum_\alpha|\psi_0{}^\alpha|^2)^2\}. \tag{3}$$

Here $\psi_0{}^\alpha$ is the (complex) order parameter field ($\alpha = 1$ for He; we use $\alpha = 1, \ldots, n/2$), E_0 is the conserved "energy," and η^α and ζ are the associated Gaussian noises whose correlations are related to the kinetic coefficients Γ_0 and $-\Lambda_0\nabla^2$ by Einstein relations. γ_0 and u_0 are the static couplings and g_0 is the reversible mode coupling constant. Equations for the AF case, where the order parameter ψ_0 is a real n-component vector ($n = 3$) and $E_0{}^{\alpha\beta}$ represent the generators of the $O(n)$ symmetry, are simply related[1,2] to (1)–(3).

These equations are cast in a form tractable by field theoretic methods by using the Martin-Siggia-Rose Lagrangian[8] which involves conjugate variables $\hat{\psi}_0{}^\alpha$ and $\hat{E}_0$. The *statics* as contained in