Spreading pressure study of the commensurate solid to domain-wall fluid transition of monolayer ^{4}He on graphite

Thomas A. Rabedeau*
Department of Physics, University of Washington, Seattle, Washington 98195
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Results of a spreading pressure study of the commensurate solid to domain-wall fluid (CS-DWF) transition of monolayer ^{4}He on graphite are reported. The CS-DWF phase boundary between 1.2 and 2.8 K is determined from the location of pronounced maxima in isothermal compressibility data constructed from the spreading pressure data. The scaling of the compressibility data at this transition is consistent with a chiral crossover exponent smaller than $\frac{2}{3}$.

Present understanding of the commensurate-incommensurate (C-IC) transition in physisorbed films relies upon the domain-wall description of the weakly incommensurate regime. Specifically, the weakly incommensurate phase is described in terms of domains of the commensurate structure separated by domain walls where the domain-wall free energies alone determine the thermodynamic stability of a given domain-wall structure.[1] An important aspect of the domain-wall description of the C-IC transition is the recognition that domain walls can possess a definite helicity and that the dissimilar energetics of domain walls with different helicities reduces the symmetry of the commensurate solid Hamiltonian.[2] This introduction of chirality is expected to alter the universality class of the C-IC transition. In this Brief Report we discuss a spreading pressure investigation of domain-wall thermodynamics at the commensurate solid to domain-wall fluid (CS-DWF) transition in monolayer ^{4}He on graphite.

Consider the monolayer ^{4}He-graphite phase diagram presented in Fig. 1.[3-5] The phase denoted $\sqrt{3}\times\sqrt{3}$ is a commensurate structure with a threefold-degenerate ground state.[6] The disordering of the commensurate solid at the critical density (i.e., $n=1$ in Fig. 1) falls in the same universality class as the two-dimensional, three-state Potts model.[7,8] At densities slightly in excess of the critical density, the commensurate solid is thermodynamically unstable while at considerably higher densities a triangular, incommensurate solid is obtained.[6,9] In the density regime intermediate between the commensurate solid and the triangular solid two phases are evident. Direct measurements of the structure of these two intermediate phases are not available; however, theoretical calculations[10] and computer simulations[11] suggest that the low-temperature phase is an incommensurate, uniaxial domain-wall solid which melts into a disordered domain-wall network (i.e., domain-wall fluid) at approximately 1 K. This interpretation is consistent with low-energy electron diffraction[12] (LEED) and neutron-diffraction[13] results for para-H_2/graphite which has a phase diagram similar to that of ^{4}He-graphite.[14] The data collected in this investigation illuminate several aspects of the ^{4}He-graphite phase diagram. Isothermal compressibilities constructed from spreading pressure data are used to locate the CS-DWF phase boundary from 1.2 to 2.8 K. Heretofore the location of this phase boundary has been indeterminate as prior investigations have not been sensitive to the CS-DWF phase boundary. Additionally, the scaling properties of the compressibility data are examined for the influence of chirality upon this transition.

The strain gauge employed in this study has been described in detail elsewhere.[15] Briefly, the strain gauge is composed of a cylindrical capacitor whose inner conductor is constructed of MAT, a semiordered exfoliated graphite, while the outer conductor is dimensionally inert. The two conductors are separated by a small gap. When a film is adsorbed onto the MAT substrate, the radius of the inner conductor dilates in response to the film-induced surface stress. This dilation is measured capacitively. To lowest order the film spreading pressure ϕ is proportional to $1/C_{\text{empty}} - 1/C_{\text{film}}$. The proportionality constant was

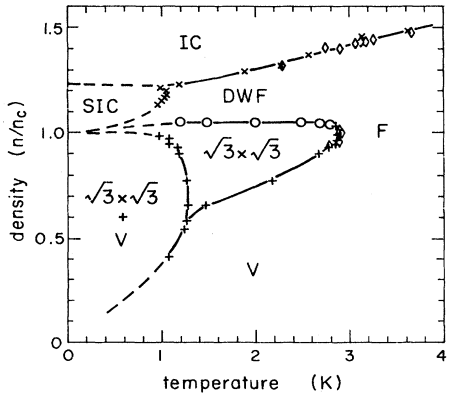

FIG. 1. Phase diagram of monolayer ^{4}He physisorbed on graphite. The data depicted are taken from the heat-capacity studies of Ref. 3 (×), Ref. 4 (◇), and Ref. 5 (+) and the isothermal compressibility results of this study (○). The dashed lines indicate approximate locations of phase boundaries. The abbreviations denote incommensurate solid (IC), striped incommensurate solid (SIC), $\sqrt{3}\times\sqrt{3}R30°$ commensurate solid ($\sqrt{3}\times\sqrt{3}$), domain-wall fluid (DWF), and fluid (F) and vapor (V) phases.

determined at an elevated temperature where the isothermal differential of the Gibbs free energy was employed to relate the film spreading pressure to the vapor pressure of coexisting bulk vapor. The adsorption area of the strain cell was calibrated at the order-disorder transition where the isosteric temperature derivative of the spreading pressure provides a clear signature of the critical density.[16]

Spreading pressure data collected along seven isosteric trajectories with densities ranging from $1.004n_c$ to $1.087n_c$ are presented in Fig. 2. The most compelling feature of these data, the negative slope of ϕ for $T < 3$K, is a consequence of positive entropy variation with coverage as seen by invoking the Maxwell relation[17]

$$-A(\partial\phi/\partial T)\,|_{A,N} = n(\partial S/\partial n)\,|_{T,N}\,.$$

Phenomenologically, this result is a reflection of the low entropy of the commensurate structure at the critical coverage.

The isothermal compressibility near the CS-DWF phase boundary was constructed by interpolating the data of Fig. 2 to yield $\phi(n)$ for temperatures between 1.2 and 2.8 K. The interpolated data were then used to calculate

$$(-1/n^2)(\delta n/\delta\phi)\,|_{T,A} \approx (\partial a/\partial\phi)\,|_{T,N}\,.$$

The computed compressibility data along five isotherms are plotted in Fig. 3 as functions of ϕ. Anticipating the scaling theory results presented below, the open circles of Fig. 1 denote the CS-DWF phase boundary in the n-T plane as determined from the location of the isothermal compressibility maxima. Below 2.5 K the phase boundary follows the $1.050n_c$ isostere to the precision of the data ($\pm 0.007n_c$).

Now consider the scaling of the isothermal compressibility data presented in Fig. 3. The low-temperature compressibility isotherms are strongly peaked. With increasing temperature the compressibility peaks broaden and weaken until a rounded anomaly is obtained as evidenced by the 2.75-K isotherm. While such an evolution can be attributed solely to path renormalization effects for a second-order transition with a phase boundary of the form depicted in Fig. 1,[18] it is appropriate to examine these data for the influence of a chiral field. The low-temperature data are inadequate to determine scaling exponents and as such are consistent with arbitrary values of the chiral crossover exponent y_c.

In the vicinity of the disordering transition at n_c the data are less ambiguous. Consider the effect of a relevant chiral field which vanishes at n_c such that the singular part of the free energy is a homogeneous function of two relevant fields $t(\phi,T)$ and $u_c(\phi,T)$ with $u_c(\phi_c(n_c), T_c(n_c)) = 0$. Scaling theory indicates that the compressibility scales to leading order as

$$\left.\frac{\partial a}{\partial\phi}\right|_{T,N} \sim t^{(d-y_t)/y_t} \sim \Delta\phi^{2(d-y_t)/y_t}\,, \tag{1}$$

provided $y_t > 2y_c$ where y_t is the Potts scaling exponent. The right-hand expression follows upon expanding t as a power series in $\Delta\phi = |\phi(T) - \phi_c(T)|$ and recognizing that the quadratic term is the lowest term in the expansion since $\partial t/\partial\phi = 0$ at n_c. Inserting $y_t = \frac{6}{5}$ (Ref. 19) yields $\partial a/\partial\phi \sim \Delta\phi^{4/3}$. This power law is consistent with the high-temperature compressibility data of Fig. 3. For $y_t < 2y_c$, one finds to leading order

$$\left.\frac{\partial a}{\partial\phi}\right|_{T,N} \sim u_c^{(d-2y_c)/y_c} \sim \Delta\phi^{(d-2y_c)/y_c}\,, \tag{2}$$

where the expression on the right-hand side of Eq. (2) employs the first term in a power-series expansion for u_c in terms of $\Delta\phi$. Inspection of the higher-temperature compressibility data of Fig. 3 reveals that $\partial K_T/\partial\phi \to 0$ as $\Delta\phi \to 0$. Consistency of these data with Eq. (2) requires $(d-2y_c)/y_c > 1$ or $y_c < \frac{2}{3}$. This constraint upon y_c agrees with the extended scaling relation for y_c derived by

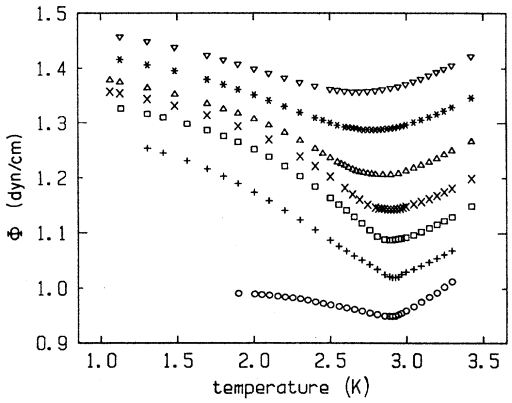

FIG. 2. Spreading pressure data along seven isosteres. Coverages measured relative to the critical density (n_c) are 1.004 (○), 1.018 (+), 1.032 (□), 1.043 (×), 1.057 (△), 1.073 (*), and 1.087 (▽).

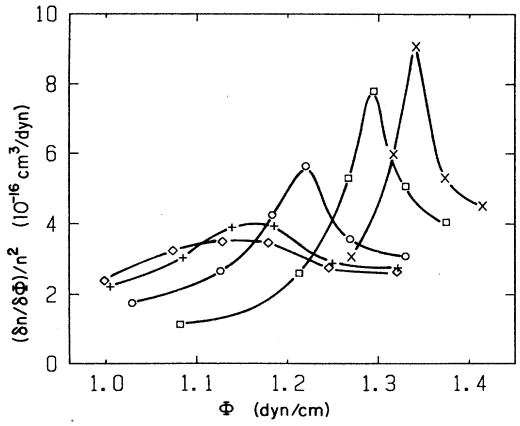

FIG. 3. Computed isothermal compressibility data along five isotherms: 1.50 K (×), 2.00 K (□), 2.50 K (○), 2.70 K (+), and 2.75 K (◇). The lines are guides to the eye.

den Nijs which predicts that $y_c = y_t - 1 = \frac{1}{5}$.[20] Finally, for the more general case of a relevant chiral field which does not vanish at the disordering transition at the critical coverage, heat-capacity data[8] require that the scaling exponents at the critical density be identical to those of three-state Potts model in the absence of chirality. The compressibility scales as shown in Eq. (1).

To recapitulate, the results of this study provide the first, accurate determination of the CS-DWF phase boundary in ^{4}He on graphite. The scaling of isothermal compressibility data at this transition is consistent with a chiral crossover exponent smaller than $\frac{2}{3}$.

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*Present address: Gordon McKay Laboratory, Harvard University, Cambridge, MA 02138.

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