

Monolayers of CF_4 adsorbed on graphite, studied by synchrotron x-ray diffraction

K. Kjaer, M. Nielsen, and J. Bohr

Risø National Laboratory, DK-4000 Roskilde, Denmark

H. J. Lauter

Institut Laue-Langevin, 156X, 38042 Grenoble Cedex, France

J. P. McTague

Brookhaven National Laboratory, Upton, New York 11973

(Received 6 May 1982)

With synchrotron x-ray diffraction we have measured the phase diagram of CF_4 monolayers adsorbed on the graphite substrate UCAR-ZYX. We have found four two-dimensional crystalline phases including the 2×2 commensurate structure. Between this and the denser incommensurate hexagonal phase we find an axially compressed phase interpreted to be the stripe domain structure. Our data indicate that the order-disorder transition (melting) of the 2×2 commensurate phase as well as melting of the stripe and of the hexagonal incommensurate phases are continuous.

INTRODUCTION

Physisorbed monolayers on graphite of rare-gas atoms or of simple molecules have been studied extensively in recent years with the main purpose of describing the nature of phase transitions in two-dimensional systems. The transitions studied in most detail are melting of the incommensurate free-floating solid [Ar (Ref. 1) and Xe (Ref. 2) monolayers], order-disorder transition (melting) of the commensurate $\sqrt{3} \times \sqrt{3}$ structure [Kr (Ref. 3) and He (Ref. 4) monolayers], and the commensurate-incommensurate (*C-I*) transition [Kr,⁵ H_2 , and He (Ref. 6) monolayers]. We report here on diffraction studies on monolayers of CF_4 adsorbed on the graphite substrate UCAR-ZYX.

The CF_4 layers on graphite constitute a fundamentally different system from that of the rare-gas monolayers because of two factors. First, the adsorbed particles are larger and thus commensuration occurs at the 2×2 structure instead of at the denser $\sqrt{3} \times \sqrt{3}$ structure. Second, the preferred adsorption site is assumed to be above the vertex points of the graphite honeycomb structure, which have a symmetry different from that of the center positions taken up by the rare-gas commensurate layers, but similar to that of oxygen atoms chemisorbed on Ni(111) surfaces.⁷

Figure 1 shows the honeycomb structure of the (0002) graphite surface and the CF_4 molecules in the commensurate 2×2 structure. They are shown with a three-fluorine plane parallel to the graphite

surface and the projections of the Lennard-Jones spheres of the fluorines are shown shaded. At the temperatures where the 2×2 structure exists, the molecules will have large-amplitude rotational oscillations. In the figure the lowest-energy configura-

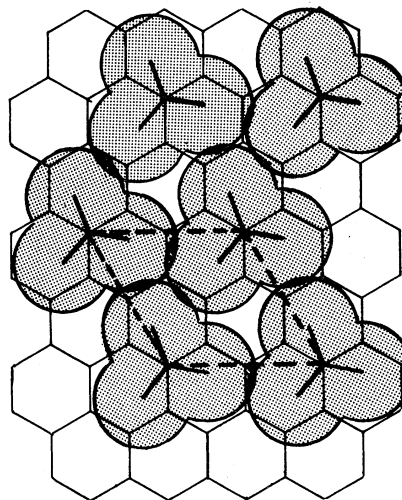


FIG. 1. Schematic arrangement of the CF_4 molecules on the (0002) surface of graphite, in the 2×2 commensurate structure. They are shown by the projection (shaded) of the Lennard-Jones spheres of the fluorines (using the diameter of neon: $2^{1/6}\sigma = 3.17 \text{ \AA}$) assuming that the molecules have a three-fluorine plane parallel to the substrate and are located above the vertex points. Molecules are rotated away from the high-symmetry orientations. Broken line shows the unit cell of the 2×2 structure.

tion is shown as estimated from the potentials of Ref. 8. The molecules are rotated away from a symmetric orientation by the interaction with the substrate. Within the unit cell of the 2×2 structure (Fig. 1), there are four equivalent adsorption sites per molecule and four more sites connected with the former by a 180° rotation. Further, the above-mentioned rotation may be chosen clockwise or counterclockwise. Therefore, the order-disorder transition of the 2×2 structure is different from the three-state Potts transition⁴ applying for the $\sqrt{3} \times \sqrt{3}$ phase and is to be described by a Heisenberg model with cubic anisotropy or by an Ising model.⁹ For the (*C-I*) transition between the 2×2 structure and the denser incommensurate hexagonal structure, the different symmetry has the following implication: If a hexagonal structure existed with a layer density a little above the commensurate density, this would give a superstructure of domains¹⁰ in which the domain walls form triangles instead of hexagons as for the slightly incommensurate Kr layers.¹¹ A triangular superstructure would in the crossing points of the domain walls have six walls meeting, and this would involve a high wall-crossing energy. According to Ref. 12 this favors the alternative domain configuration, the stripe structure, in which the domain walls are parallel lines with no walls crossing. The latter model is found to describe our results, and this is the first observation of the stripe structure in monolayers adsorbed on graphite. The existence of this phase allows us to study the following transitions: commensurate 2×2 to stripe structure, stripe to incommensurate hexagonal structure, and melting of the stripe phase into an isotropic fluid.

The measurements were done with an x-ray spectrometer at the storage ring DORIS at Deutsches Elektronen-Synchrotron (DESY) in Hamburg. The instrument is described in Ref. 13 and consists of a monochromator with two parallel Ge(111) crystals and a position sensitive detector as analyzer. The graphite substrate is a stack of UCAR-ZYX strips contained in an all metal sample cell with Be windows, and the cell was mounted in a Displex cryostat.

PHASE DIAGRAM

The diffraction from the CF₄ monolayers was measured exclusively in the region near the (10) reflection of the 2×2 structure. At the higher-order reflections the intensity was insufficient to obtain useful groups. Data were taken as function of tem-

perature and CF₄ coverage, and Fig. 2 shows the derived phase diagram. At coverage less than 0.79 (we use as unit of coverage the amount of gas which would give one ideal and completed monolayer with the 2×2 structure), we observe triple-point behavior between dilute gas (*G*), liquid (*L*), and commensurate 2×2 phase (*C*). The gas-liquid coexistence line is drawn in accordance with the critical point taken from Ref. 14. With decreasing temperature the coexisting gas plus the 2×2 phase go into the coexisting gas plus stripe (*S*) phase at 65 K. Below 57 K coexistence between gas and a "three-peak structure" (*3P*) is observed. The nature of this three-peak structure is not interpreted and will not be discussed further here.¹⁵ The stripe structure is observed in a rather broad region around the 2×2 phase, and at higher layer density the hexagonal incommensurate structure (*I*) was found. The nature of the phase transitions between the different phases is indicated by either single lines (continuous transitions) or by shaded coexistence regions (first-order transitions). However, the phase diagram is tentative and several ambiguities remain. In the diffraction data there is no distinct difference between the

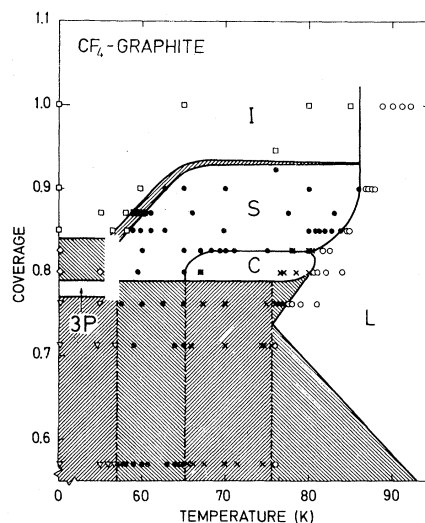


FIG. 2. Phase diagram of CF₄ on (0002) graphite. *C*, *S*, *I*, and *3P* denote the 2×2 commensurate, stripe, hexagonal incommensurate, and three-peak structures, and *L* represents a two-dimensional liquid. Full lines indicate phase boundaries and broken lines show transitions of solid phases in coexistence with the dilute-gas phase (not shown in the figure). Coexistence regions are shaded. Points denote diffraction measurements in the following sense: ○, liquid, ×, single 2×2 peak at 1.475 \AA^{-1} , ●, two peaks with 1:2 intensity ratio, □ one peak at $Q > 1.475 \text{ \AA}^{-1}$, ▽, three peaks, ◇, four peaks (coexistence of *3P* and *I*).

gas plus homogenous solid (say 2×2 phase) and the homogenous solid phases, and thus the corresponding phase lines are uncertain. A very noticeable effect is that the homogenous 2×2 structure is not found at the coverage 1 but rather near 0.8. This must imply that the 2×2 phase contains a high concentration of vacancies or a nonideal behavior of the substrate. The total substrate surface area was determined before and after the measurements by measuring Ar vapor-pressure isotherms at $T = 65$ K, and reference to diffraction measurements on Kr monolayers using the same sample cell.¹⁶ The Ar isotherms give a practical check on the area of adsorption on an uncalibrated scale. The absolute scale of the area was determined in more complicated measurements where a series of diffraction scans are measured at low temperature for adsorbed monolayers of Kr with increasing coverage. The sharp onset of the commensurate-to-incommensurate phase transition defines the coverage giving the $\sqrt{3} \times \sqrt{3}$ monolayer.

In the region where three of the phases in Fig. 2, I , S , and $3P$, come together, the phase diagram is not known, but a pronounced first-order transition is observed from $3P$ to I and from I to S .

Figure 3 shows the position of the observed Bragg peaks as function of temperature for three different coverages. The signature of the C phase is a single peak at the position $Q = 1.475 \text{ \AA}^{-1}$ which is half the (10) reciprocal-lattice vector of the graphite honeycomb structure. The stripe structure is characterized by two peaks, and the distance between them is a measure of the axial compression. Our conjecture that the S phase of Fig. 2 has the stripe structure¹² rests on the following observations: When we move out of the 2×2 region either by decreasing the temperature or by increasing the coverage, we observe that the single (10) reflection of the 2×2 structure splits into a double peak such that the low- Q component remains at the commensurate position $Q = 1.475 \text{ \AA}^{-1}$, and the other one moves to slightly higher Q values. There is a constant ratio of 1:2 between the intensity of the two peaks, the low- Q component being the smallest. The distance between the peaks is a continuous function of temperature and coverage. This is explained by an axial compression of the structure or a corresponding expansion of the reciprocal lattice as is shown in the upper part of Fig. 3. The diffraction from an ideal stripe structure comprises extra peaks (satellites) on both sides of the doublet described above. These are due to the periodic density modulation associated with the difference of layer density in the middle of the domains and in

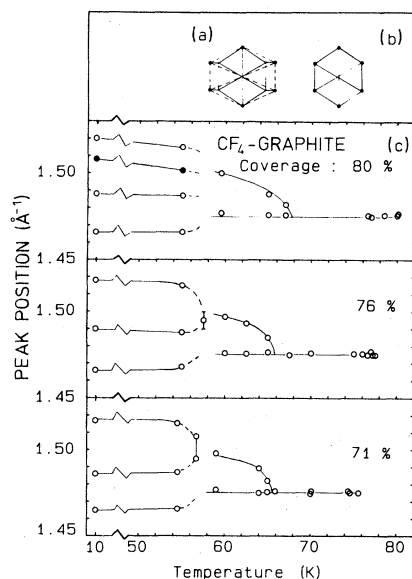


FIG. 3. (a) Points and dashed lines: reciprocal lattice of the uniaxially compressed (stripe) structure. (No satellite reflection points are shown.) Full lines: reciprocal lattice of the 2×2 structure. (b) Reciprocal lattice of the 2×2 structure. (c) Peak positions vs temperature for three different coverages. In the upper panel, the filled circles represent the signal from the I phase coexisting with the three-peak structure. Lines are guides to the eye.

the domain walls.¹⁶ However, if the walls are very broad, the satellite intensity is weak. We have not observed any such satellites.

When the temperature is lowered through 57 K at coverages less than 0.76 the double peak of the stripe phase changes into three peaks. Very close to the transition the diffraction response gets broad, and we cannot conclude whether this transition is of first or second order. The positions of the three peaks are $Q = 1.465, 1.486, \text{ and } 1.518 \text{ \AA}^{-1}$, and these as well as the transition temperature are independent of coverage in the solid plus gas coexistence regime. At coverage 0.8 the $3P$ structure coexists with the incommensurate hexagonal phase observed at higher density. The latter component gives a single peak, in the upper panel of Fig. 3 at $Q = 1.502 - 1.508 \text{ \AA}^{-1}$. The nature of the $3P$ phase has recently been discussed on the basis of neutron scattering results,¹⁵ however, more information is needed to conclude on the structure of this phase.

Figure 4 shows the intensity of the (10) Bragg peak of the 2×2 phase at four different coverages. Below the liquid point at the triple-point line, at a coverage 0.74, the intensity of the (10) peak disap-

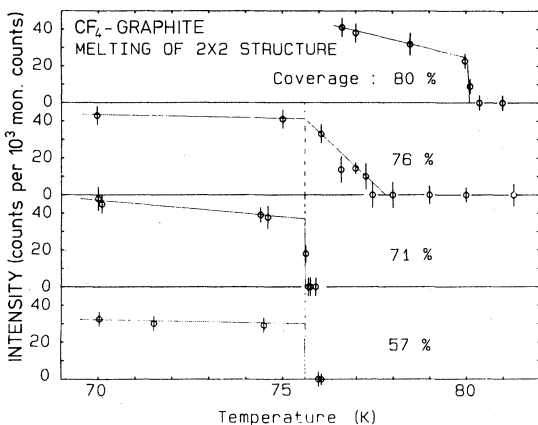


FIG. 4. Peak intensity of the 2×2 reflection at 1.475 \AA^{-1} vs temperature at four different coverages. All the diffraction groups represented in the figure had the resolution width of the instrument, corresponding to a coherence length of $L > 1100 \text{ \AA}$. From above, the panels show melting of the homogenous 2×2 phase, "pressure-broadened" decrease of 2×2 intensity due to coexistence with a dense liquid, and (lower two panels) sharp triple-point transition from 2×2 plus dilute-gas coexistence to liquid plus gas coexistence.

pears discontinuously at the triple point. For coverage 0.76 the intensity decreases gradually as we go through the liquid plus solid coexistence region. However, at the triple point there should be an abrupt decrease of intensity as we go from solid plus gas coexistence to solid plus liquid coexistence. Thus the liquid plus solid region of Fig. 2 may be incorrectly indicated, but further measurements are needed to solve this problem. The upper panel of Fig. 4 shows that at coverage 0.80 the 2×2 phase has a very sharp melting transition: The intensity disappears within 0.1 K. This transition is analogous to the melting of the commensurate $\sqrt{3} \times \sqrt{3}$ phase of Kr on graphite described by the three-state Potts model with a continuous transition. For CF₄ monolayers the symmetry is different as discussed above, and it would be of particular interest to study the melting by diffraction in the same detail as has been done for Kr.³ It should be noted that the phase diagram of Fig. 2 shows that the transition changes over from first order to second order with increasing coverage, and this is analogous to what was found for Kr on graphite³ and discussed theoretically in Ref. 17.

At higher coverages first the C phase changes into the stripe phase, the melting of which is discussed in the next section, and then the dense incommensurate hexagonal structure is formed.

Melting of the latter was studied in a constant coverage scan at $\rho = 1.0$. A set of (10) diffraction groups typical for a continuous melting transition was observed. Above the melting temperature T_M the groups broaden. They can be fitted by powder-averaged Lorentzian functions^{1,2} with correlation lengths ξ which ideally should diverge at T_M but in practice are limited by finite-size effects. We find that ξ varies from $\xi > 1200 \text{ \AA}$ below T_M , a limit set by our instrumental resolution, to $\xi = 170 \text{ \AA}$ at 6 K above T_M . This is a much slower variation than observed for Xe and Ar films^{2,1} on the same substrate, but more studies are necessary before this can be interpreted.

The phase diagram of CF₄ on graphite has earlier been studied by neutron diffraction.¹⁵ The result was different from ours reported here, due to the lower resolution of the neutron scattering technique. In particular the stripe phase was not seen. Also low-energy electron diffraction measurements have been performed on this system.¹⁸ The 2×2 structure was identified and around 60 K a rotated slightly incommensurate structure was observed. The latter observation is difficult to combine with our interpretation.

PHASE TRANSITIONS INVOLVING THE STRIPE PHASE

Two transitions involving the stripe phase, S in Fig. 2, were studied in detail, namely C to S and S to L. Figure 5 shows some observed diffraction groups near the (10) reflection of the 2×2 structure when we go from C to S by changing temperature at constant coverage near the top of the C-phase region in the phase diagram. In the upper panel, at $T = 80.01 \text{ K}$, we have the typical single group of the 2×2 structure at $Q = 1.475 \text{ \AA}^{-1}$ with the Warren line shape.⁶ The full line is the fitted diffraction profile with a Gaussian coherence length⁶ of $L = 1100 \text{ \AA}$, which is determined by the spectrometer resolution. Presumably the intrinsic coherence length of the 2×2 structure is longer, and given by the ZYX substrate to be $L \sim 1600 \text{ \AA}$.¹³ When we go through the C-to-S transition, the double peak of the S phase develops in a continuous way. Clearly the groups in Fig. 5 at 74.96 or 70 K cannot be constructed as coexistence groups between the upper-panel group and one of the others. The full lines are fitted curves assuming a uniaxial compression of the monolayers in the direction shown in the upper panel of Fig. 3. The integrated intensities of the two peaks have the ratio 1:2. The position of

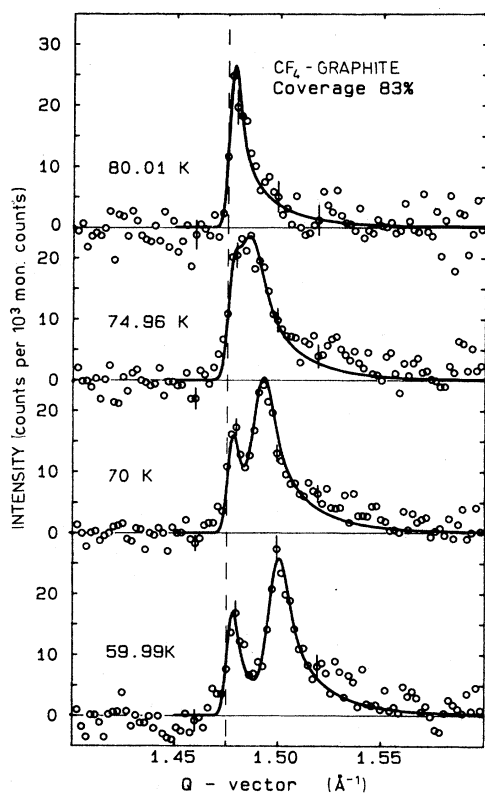


FIG. 5. Measured diffraction intensities (circles) and fitted line shapes (full lines) at constant coverage for four different temperatures. Upper panel: single 2×2 peak of resolution limited coherence length $L = 1100 \text{ \AA}$. Lower three panels: double peaks in 1:2 integrated intensity ratio, characteristic of the stripe phase. Left peak has $L_1 = 1100 \text{ \AA}$ and right peak has L_2 (fitted) = $500\text{--}600 \text{ \AA}$. Gaussian cross sections have been used in the Warren (Ref. 6) formula for both peaks for simplicity.

the low- Q component is fixed at $Q = 1.475 \text{ \AA}^{-1}$, and the high- Q component is shifted outwards according to the increasing compression as T is lowered. For the group at $T = 74.96 \text{ K}$, which is very close to the transition, this shift is $\Delta Q = 0.0073 \text{ \AA}^{-1}$ corresponding to an axial strain of $\epsilon = 0.0066$ or a domain wall separation of 40 lattice distances which is $\sim 200 \text{ \AA}$. In the calculation of the curves fitted to the double groups of Fig. 5 we have assumed that the low- Q component has the same Warren line shape as the group of the pure 2×2 structure with $L = 1100 \text{ \AA}$. For the high- Q component we also used, for simplicity, the Warren line shape, but let the Gaussian coherence length vary, and best fits were obtained with $L = 600 \text{ \AA}$. In fact, different types of line shapes are expected for the two components if the uniaxial compression does not alter the range of coherence of the two-

dimensional (2D) crystal in the direction perpendicular to the strain while fluctuations of the local-strain amplitude limit the correlation length in the parallel direction. The statistical accuracy of our data points does not allow a detailed analysis of line shapes, but in principle the groups describe the type of fluctuation existing in the S phase.^{19,20}

The phase transition $C \rightarrow S$ was studied in a series of scans for coverages between $\rho = 0.57$ and 0.83 , and the same behavior as that of Fig. 5 was observed. This also applies for $\rho \leq 0.79$, where the phase transition occurs with the solid coexisting with a dilute gas. Thus the driving mechanism for this transition is of a different kind from that operative in the commensurate-incommensurate transition of Kr, where a considerable spreading pressure is needed.¹³ However, CD_4 monolayers on graphite undergo the transition from $\sqrt{3} \times \sqrt{3}$ to a more dilute incommensurate hexagonal structure at $T \approx 50 \text{ K}$, also in a phase coexisting with its dilute gas.²¹ This transition is thus analogous to the C -to- S transition of the CF_4 layers in the sense that it is the change of the amplitudes of the thermal oscillations of the molecules which drives the change of phase. An important factor must be the molecular rotations, and it would be of particular interest to study this by nuclear magnetic resonance. It should also be noted that the relatively high concentration of vacancies in the solid phases may be an important factor for the transition.

Although the observed diffraction groups near the C -to- S transition rather clearly indicate a continuous phase transition, there can of course be a small not resolved discontinuity of the order parameter in the transition. The separation ΔQ of the two groups of the S phase varies in our data from $\Delta Q = 0.03 \text{ \AA}^{-1}$ to $\Delta Q = 0.006 \text{ \AA}^{-1}$, the smallest value we can resolve. An indication that there may be a small first-order jump is that we saw a small hysteresis when a set of groups like those of Fig. 5 was measured with increasing and decreasing temperature. This could be as large as 1 K but many groups would be needed for a precise determination of the hysteresis. Hysteresis of the C -to- S transition has been discussed theoretically in Ref. 10 and may in fact not require a first-order transition.

As shown in Fig. 2 the stripe phase for a range of densities melts into a liquid. This raises the following question: How can the uniaxially compressed S phase transform into an isotropic fluid? Figure 6 shows a constant coverage scan through the melting transition. The general feature seems to be that the double peak of the S phase gradually becomes a single, broad, liquidlike group by both the high- and

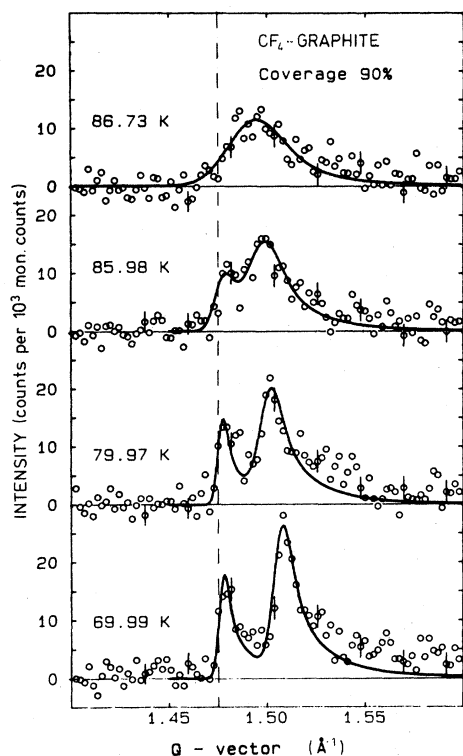


FIG. 6. Constant coverage scan through the melting transition of the stripe phase. Circles: measured intensities. Lines: fitted curves. Upper panel: single broad peak fitted by a Gaussian Warren line shape ($L=200$ Å). (Alternative line shape discussed in text.) Second panel: double peak fitted by two Warren line shapes of coherence lengths $L_1=600$ and $L_2=350$ Å, in 1:2 intensity ratio. Lower two panels: double peak fitted by Gaussian Warren line shapes in 1:2 intensity ratio. Left peak has $L_1=1100$ Å (resolution limited), and right peak has L_2 (fitted) = 500–600 Å.

the low- Q component being broader and broader through the transition. However, the scatter of the data points of the $T=85.98$ K group does not allow a definitive conclusion in this regard. The change of the splitting ΔQ is only a minor effect. The full lines in Fig. 6 show the fitted groups where we again have used Gaussian structure factors in the Warren-line-shape formula for simplicity. As discussed above, the low- Q component of the double group is insensitive to fluctuations in the lattice parameter in the direction of the uniaxial compression, and a correct line-shape calculation should involve an anisotropic and non-Gaussian structure factor around the reciprocal-lattice points of Fig. 3(a). Although the quality of our data does not allow such detailed analysis, they do indicate that when approaching the melting transition, there exist

fluctuations in the S phase which change the structure factor in both directions.

We have also fitted the liquidlike scattering profile of the upper panel of Fig. 6, at $T=86.73$ K, to a powder-averaged Lorentzian structure factor.² We find that the correlation length is $\xi \approx 300$ Å, a value similar to what was found in liquid Xe (Ref. 2) and liquid Ar (Ref. 1) near the melting transition between the liquid and an isotropic incommensurate solid with hexagonal structure. As Xe and Ar layers have continuous melting transitions, the intrinsic correlation lengths of the liquids diverge at the melting points, but finite-size effects limit the value and give rise to a “rounding” effect very close to the transition. A value of $\xi=300$ Å is within this rounding regime of the transition. Thus the liquid phase just above the S -to- L melting transition is, within the accuracy of our data, equivalent to the liquid phase of adsorbed Xe or Ar, but it would be of interest to study this with much improved intensity.

Contrary to the two phase transitions described above the transition from stripe to the hexagonal incommensurate phase appeared to be distinctly of first order. In constant coverage scans at $\rho=0.85$ and 0.87 we measured groups in the shaded coexistence region of Fig. 2, which could be constructed as weighted sums of the S - and I -phase signal. We do not have sufficient data to decide whether the coexistence region extends all the way to the melting line as indicated in the tentative phase diagram of Fig. 2. The first order character of the S -to- I transition is theoretically expected.¹²

CONCLUSION

The study of the CF₄ films has shown that they have a rather complicated phase diagram with at least five different phases in the monolayer regime. At present our experimental information about the system is very limited; in particular no measurement or calculation of the rotational state or the position of the adsorbed molecules has been performed. Despite this, diffraction measurements can identify phases already discussed theoretically^{12,19} and give important information about the nature of the phase transition between structures with different symmetries. Most significant in our data is the clear evidence that the commensurate-to-incommensurate transition between the 2×2 structure and the denser hexagonal structure takes place through the intervening uniaxially compressed phase, the stripe domain structure. The C -to- S , the

S-to-*L*, and the *I*-to-*L* transitions are continuous, or at least very weakly of first order if so, whereas *S* to *I* and *3P* to *I* are of first order. Our diffraction data also give some information about the fluctuations near the continuous phase transitions: It is clear, e.g., that the structure factor near the reciprocal-lattice points of the stripe phase are anisotropic near the *C*-to-*S* transition but become more isotropic very near the melting line of the phase diagram. However, a detailed analysis of the functional form of the structure factors requires much higher scattering intensity than was available in our measurements.

ACKNOWLEDGMENTS

The participation of Dr. J. Als-Nielsen in part of the experiment, as well as the invaluable help of our colleagues in the HASYLAB, DESY, Hamburg, is gratefully acknowledged. It is also a pleasure to thank Dr. P. Bak and Dr. E. Riedel for many very fruitful discussions about the interpretation of our data. This work was supported in part by the Danish Natural Science Foundation and by the American National Science Foundation.

- ¹J. P. McTague, J. Als-Nielsen, J. Bohr, and M. Nielsen, *Phys. Rev. B* **25**, 7765 (1982).
- ²P. A. Heiney, R. J. Birgeneau, G. S. Brown, P. M. Horn, D. E. Moncton, and P. W. Stephens, *Phys. Rev. Lett.* **48**, 104 (1982); E. M. Hammonds, P. Heiney, P. W. Stephens, R. J. Birgeneau, and P. M. Horn, *J. Phys. C* **13**, L301 (1980).
- ³P. M. Horn, R. J. Birgeneau, P. Heiney, and E. M. Hammonds, *Phys. Rev. Lett.* **41**, 961, (1981); R. J. Birgeneau, G. S. Brown, P. M. Horn, D. E. Moncton, and P. W. Stephens, *J. Phys. C* **14**, L49, (1981).
- ⁴M. Bretz, *Phys. Rev. Lett.* **38**, 501 (1977).
- ⁵D. E. Moncton, P. W. Stephens, R. J. Birgeneau, P. M. Horn, and G. S. Brown, *Phys. Rev. Lett.* **46**, 1533 (1981).
- ⁶M. Nielsen, J. M. McTague, and L. Passell, in *Proceedings of the NATO Advanced Study Institute on Phase Transitions in Surface Films*, edited by J. G. Dash and J. Ruvalds (Plenum, New York, 1980), p. 127.
- ⁷L. D. Roelofs, A. R. Kortan, T. L. Einstein, and R. L. Park, *Phys. Rev. Lett.* **46**, 1465 (1981); M. Schick, *ibid.* **47**, 1347 (1981).
- ⁸Takako Shinoda, *Mol. Cryst. Liq. Cryst.* **55**, 101 (1979).
- ⁹E. Riedel and P. Bak (private communication).
- ¹⁰P. Bak, in *Solitons and Condensed Matter Physics*, edited by A. R. Bishop and T. Schneider (Springer, Berlin, 1980), p. 216.
- ¹¹The triangular structure of the domain walls follows if the 2×2 structure is slightly compressed isotropically and the adsorbed particles subsequently moved towards the nearest ideal adsorption site. The same procedure applied to the $\sqrt{3} \times \sqrt{3}$ structure results in a honeycomb structure of domain walls.
- ¹²P. Bak, D. Mukamel, J. Villain, and K. Wentowska, *Phys. Rev. B* **19**, 1610 (1979).
- ¹³M. Nielsen, J. Als-Nielsen, J. Bohr, and J. P. McTague, *Phys. Rev. Lett.* **47**, 582 (1981).
- ¹⁴P. Dolle, M. Matecki, and A. Thomy, *Surf. Sci.* **91**, 271 (1979).
- ¹⁵B. Croset, C. Marti, P. Thorel, and H. Lauter (unpublished); H. J. Lauter, B. Croset, C. Marti, P. Thorel, in *Ordering in Two Dimensions*, edited by S. K. Sinha (North-Holland, Amsterdam, 1980), pp. 211–214.
- ¹⁶M. Nielsen, J. Als-Nielsen, and J. P. McTague, in *Ordering in Two Dimensions*, edited by S. K. Sinha (North-Holland, Amsterdam, 1980), pp. 135–141.
- ¹⁷S. Ostlund and A. N. Berker, *Phys. Rev. Lett.* **42**, 843 (1979).
- ¹⁸S. Calisti, Ph.D. thesis, l'Université d'Aix-Marseille II, 1981 (unpublished).
- ¹⁹J. Villain, in *Ordering in Strongly Fluctuating Condensed Matter Systems*, edited by T. Riste (Plenum, New York, 1980), p. 221.
- ²⁰V. L. Pokrovsky and A. L. Tapalov, *Phys. Rev. Lett.* **42**, 65 (1979); S. N. Coppersmith, D. S. Fisher, B. I. Halperin, P. A. Lee, and W. F. Brinkman, *Phys. Rev. B* **25**, 349 (1982).
- ²¹P. Dutta, S. K. Sinha, P. Vora, M. Nielsen, L. Passell, and M. Bretz, in *Ordering in Two Dimensions*, edited by S. K. Sinha (North-Holland, Amsterdam, 1980), pp. 169–174.