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Isothermal melting of near-monolayer xenon on single-crystal graphite

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Abstract. Our study investigates the melting transition of near-monolayer xenon at three temperatures, 121 K, 140 K and 145 K. Xenon on graphite is widely regarded as a model system for the investigation of two-dimensional melting in the presence of a weak orientational field. It has been suggested that it may represent an example in which melting occurs continuously through the unbinding of topological defects in a way analogous to the Kosterlitz–Thouless transition in superfluids. Recent electron diffraction studies are consistent with a two-stage melting process in this system with an intermediate aligned liquid or ‘hexatic’ phase between the solid and quasi-isotropic liquid phases. We report isothermal high-resolution synchrotron x-ray diffraction measurements of the transition from a finite-size limited 2D solid to a well correlated (~ 100 Å) orientationally ordered 2D liquid. Our investigation confirms that at 140 K and 145 K there is a continuous evolution of the length scale of the positional fluctuations up to approximately 1000 Å. At 121 K, despite no discernable evolution of the correlation length, the transition is found to be no sharper in reduced units than at the higher temperatures. The exponent characterizing the decay of the order parameter corresponding to a finite-sized 2D crystal is found to be consistent with values reported for finite-sized two-dimensional magnets undergoing Kosterlitz–Thouless-like (2D-XY) continuous transitions.

1. Introduction

It has been suggested that melting of monolayer xenon on graphite may represent an example in which melting occurs in two successive transitions, each driven by the unbinding of pairs of topological defects. The first transition corresponds to the loss of translational periodicity on passing from the solid to an orientationally aligned liquid similar to the hexatic phase observed in free-standing liquid crystal films. The second corresponds to the loss of orientational order on passing from the aligned liquid into the isotropic liquid state. Importantly, each of these transitions, analogous to the Kosterlitz–Thouless transition in superfluids, may proceed in a continuous manner quite unlike one’s everyday experience of the melting transition in bulk systems. This elegant mechanism for continuous 2D melting originally proposed by Halperin, Nelson and Young takes the acronym ‘KTHNY’ after its proponents [1, 2, 3].

Xenon is a natural system for experimental studies of this type, and there now exists a wealth of literature concerned with physisorbed rare gases [4, 5, 6, 7]. The nature of the melting transition in monolayer adsorbed xenon on graphite has, however,

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remained controversial. Several synchrotron x-ray studies on various polycrystalline graphite substrates have concluded that the associated higher temperature transition is continuous [8, 9, 10, 11].

Despite these elegant studies, the x-ray measurements lacked the clarity and detail, particularly on the orientational order, that only comes from a single-crystal investigation. Specht *et al* [12] performed such an experiment in a closed cell configuration, freezing the xenon adsorbate by decreasing the temperature. They demonstrated the continuous evolution of the critical liquid correlation length up to at least 2000 Å, and also a corresponding transverse ordering that became long ranged at the freezing transition.

In parallel with the synchrotron x-ray investigations, there have been experimental thermodynamic studies and computer simulations. The computer simulations have suggested that the transition at higher temperatures is first order [13]. It is important to note, however, that very large equilibration times have been found in actual xenon on graphite experiments, at least at lower temperatures [14]. This militates against drawing very strong conclusions from computer simulations which probe only the very short time behaviour. Thermodynamic studies by Suter and co-workers have suggested a crossover from first order to continuous behaviour at a temperature of ~ 147 K [15], close to but somewhat higher than has been indicated by the x-ray measurements. Chan and co-workers have reported high-precision heat-capacity and vapour pressure isotherm studies of xenon melting [16]. In contrast to both the synchrotron x-ray conclusions and the results of Suter and co-workers they argue from their data that the phase transition is first order up to at least 156.5 K.

In this paper, we consider the isothermal pressure melting of an atomic monolayer of xenon condensed on a single crystal of graphite at three temperatures: 121 K, 140 K and 145 K. Our experiments have studied only part of the two-stage melting process concentrating on the loss of translational order. We present results obtained by high-resolution synchrotron x-ray scattering at the MIT-IBM beamline X20-C of the National Synchrotron Light Source at Brookhaven National Laboratory.

2. Experiment

Our experimental apparatus has been discussed elsewhere [17]. Incident x-rays were focused by a platinum-coated toroidal focusing mirror and monochromatized to 10.033 keV by a double bounce Si(111) monochromator. The diffracted x-rays transmitted through the substrate were analysed by a Ge(111) crystal mounted on the detector arm. The substrate consisted of a large natural single crystal of graphite measuring (~ 5 mm \times 9 mm \times 0.4 mm). The substrate was prepared by removing a thin upper layer of graphite basal planes on each side of the crystal using adhesive tape. The unusually large size and thickness of the substrate allowed this to be done without damage to the substrate mosaic. In addition, we ensured that at the start of the experiment the graphite substrate was flash heated to approximately 1100 °C in a high vacuum ($\sim 1.0 \times 10^{-7}$ torr at room temperature) environment to desorb any volatile contaminants from the substrate.

Experimental data were obtained in successive experiments at temperatures of 140 K, 145 K and 121 K. In each case the melting experiments were carried out at fixed temperature. The transition was driven by varying the chemical potential, μ_A . In thermodynamic equilibrium, the chemical potential of the ambient xenon vapour in the cell, μ_V , equals that of the condensed adsorbate, $\mu_V = \mu_A$. Simple ideal gas considerations relate the vapour chemical potential μ_V to the pressure of the gas at a given temperature. $\mu_A = \mu_V = kT \ln[Ph^3/(kT(2\pi mkT)^{3/2})]$. Experimentally, the isothermal melting of the xenon monolayer was achieved through a variation of the xenon vapour pressure in

equilibrium with the adsorbate. Each datum point was allowed to equilibrate for at least 30 minutes as very slow kinetics had been seen in our experiments at lower temperatures [14].

3. Results

3.1. The ordered phase

The experiments reported in this paper extend the observations of isothermal pressure melting [17] to include data obtained at several temperatures, 121 K, 140 K and 145 K. In each case the data we have obtained span the transition from a finite-sized crystalline solid (FSCS) at high over-pressures to an aligned, well-correlated 2D liquid. Given these two distinct phases at the extremes of the pressure range of the experiment, differing x-ray lineshapes must be expected at the highest and lowest vapour pressures measured. For the high vapour pressure FSCS phase a detailed analysis of the x-ray lineshape is required in order to obtain a measure of the underlying physical properties. This requires a careful convolution of the model lineshape, the sample mosaic and the measured instrumental resolution. The instrumental resolution was measured from a Si(111) reflection. Detailed discussion of the resolution function in a similar configuration is given by Noh *et al* [18].

The monolayer xenon FSCS phase is incommensurate with the underlying ($\sqrt{3} \times \sqrt{3}R30^\circ$) sites on the graphite substrate. The commensurate sites which are spaced by 4.254 Å are given by $(\frac{1}{3}, \frac{1}{3}, 0)$ reciprocal lattice units (rlu) in terms of the graphite substrate's reciprocal lattice. These same sites may equivalently be expressed as $(1.7055, 0) \text{ \AA}^{-1}$ in terms of the xenon adsorbate 2D triangular reciprocal lattice. ($2\pi/(4.254 \cos 30^\circ) = 1.7055$.) We shall adopt a crystallographic coordinate system in this paper defined with respect to the xenon adsorbate lattice and scaled in inverse Ångströms. The FSCS and aligned liquid scattering discussed in this paper gives rise to diffraction peaks aligned with the graphite substrate but with an incommensurate 2D reciprocal lattice vector $\mathbf{G} \sim (1.6, 0) \text{ \AA}^{-1}$. The xenon-graphite system is not an example of a true floating monolayer completely free from the effects of substrate corrugation [19, 20]. The clearest evidence of the role of the substrate is necessarily the orientational alignment of the FSCS and aligned liquid phases. It is this role of the substrate that is the principal difference between the experimental system and ideal KTHNY behaviour. Given the significant incommensurability of the xenon adsorbate at these temperatures, the role of the substrate on translational periodicity appears to be minimal. It is evident, therefore, that of the two distinct transitions observed in the melting process [21] it is the FSCS to aligned liquid transition that should exhibit the features of simple KTHNY behaviour.

The scattering profile from the sample in the FSCS phase was extremely anisotropic, very sharp in the longitudinal direction while broad in the transverse direction due to the sample mosaicity. The sample mosaic was found to consist of two crystallites of width (Gaussian sigma) $\sim 0.011 \text{ \AA}^{-1}$ (0.4°) separated by $\sim 0.027 \text{ \AA}^{-1}$ (0.95°). It was important to take proper account of the combined effect of the transverse mosaic and the instrumental resolution in the analysis of the scattering profile in the longitudinal direction [18]. Full two-dimensional convolutions were performed to describe the longitudinal scans.

Previous work has demonstrated that the high-temperature incommensurate phase of monolayer xenon on graphite consists of an hexagonal domain wall phase [22]. For such a solid with an infinite size, the expected lineshape would be a power-law profile of the type $S(q) \sim 1/q^{2-\eta_G}$ [23, 24]. In the case of a finite-sized adsorbate, however, the appropriate lineshape is not a simple $q^{-2+\eta_G}$ power law but rather is given by the hyper-geometric Kummer function [25], which, reflecting the hexagonal nature of the xenon domain structure,

is taken to be isotropic:

$$S(Q) = S_0 \cdot \Phi \left(1 - \frac{\eta_G}{2}, 1; -\frac{(Q - G)^2 L^2}{4\pi} \right). \quad (1)$$

We therefore utilized an isotropic Kummer function as the model lineshape within our data analysis of the FSCS phase. We followed the methodology developed by Dimon [10, 26] in regard to the appropriate form of the Kummer function. For η_G non-varying, the integrated intensity of the Kummer function lineshape is directly proportional to the term S_0 .

The most important conceptual difference between the power-law lineshape appropriate to the infinite solid and the Kummer function is the latter's intrinsic width associated with the finite size of the ordered phase. This finite size is parametrized in the Kummer function lineshape by L . The finite size L can be best estimated from the data at the highest pressure, where the central width of the peak is most pronounced from the diffuse tails. The diffuse scattering is described by the power-law tail with exponent η_G . In fitting the experimental data some care must be taken to minimize the coupling between these two independent parameters. We assume the origin of the finite-size L to be associated with fixed defects on the substrate surface and, as such, to be independent of the varying vapour pressure above the adsorbate. The data presented in this paper have been analysed using initial parameters that were established by allowing the value of L to float in a fit to the highest pressure data, at each temperature. For these initial fits the value of η_G was fixed at 0.3, a value slightly less than the value $\eta_G = \frac{1}{3}$ predicted by theory to be the maximum applicable at the melting transition. In the case of the FSCS fits to the high vapour pressure data, the parameter L is principally associated with the behaviour of the scattering around the centre of the peak while the parameter η_G describes the extended tails of the scattering. None of the data obtained in our experiments is found to be consistent with the very small values of η_G that would be expected for an ordered 2D harmonic solid at the relevant temperature [23]. We find consistently that η_G is always of a significant magnitude ($\eta_G \gtrsim 0.3$) implying that even for the data obtained at the highest vapour pressures studied, the system is still in the critical region and not deep in the ordered (FSCS) phase.

In this paper we discuss data obtained at three temperatures 121 K, 140 K and 145 K. In figure 1 we illustrate the evolution of the longitudinal scattering profile with vapour pressure at each of the three temperatures studied. The three scans illustrated to the left of the figure, although obtained at different temperatures, correspond to approximately the same reduced chemical potential. The background subtracted datum points are shown on a semi-logarithmic scale to illustrate clearly the power-law tails of the Kummer function lineshape parametrized by η_G . The solid lines through the data correspond to fits to the expected FSCS Kummer function lineshape convolved with the resolution and the mosaic. The data illustrated on the right hand side of figure 1 correspond to the observed scattering lineshapes in the vicinity of the melting transition. At this reduced chemical potential the Kummer function FSCS lineshape alone cannot be expected to describe the data fully. Unfortunately, the data are not of sufficiently high statistical quality to permit a fit to a composite lineshape including FSCS and aligned liquid contributions.

In figure 2 we illustrate the decrease in the x-ray peak intensity obtained from fits to our FSCS lineshape as a function of the ambient vapour pressure at each of the three temperatures studied. It is important to note that at 121 K the range of pressure over which the ordered phase is seen to melt into the aligned liquid phase is extremely small. The entire transition is seen to occur over approximately 1 torr. At higher temperatures the corresponding range of pressures becomes more extended until at 145 K the transition

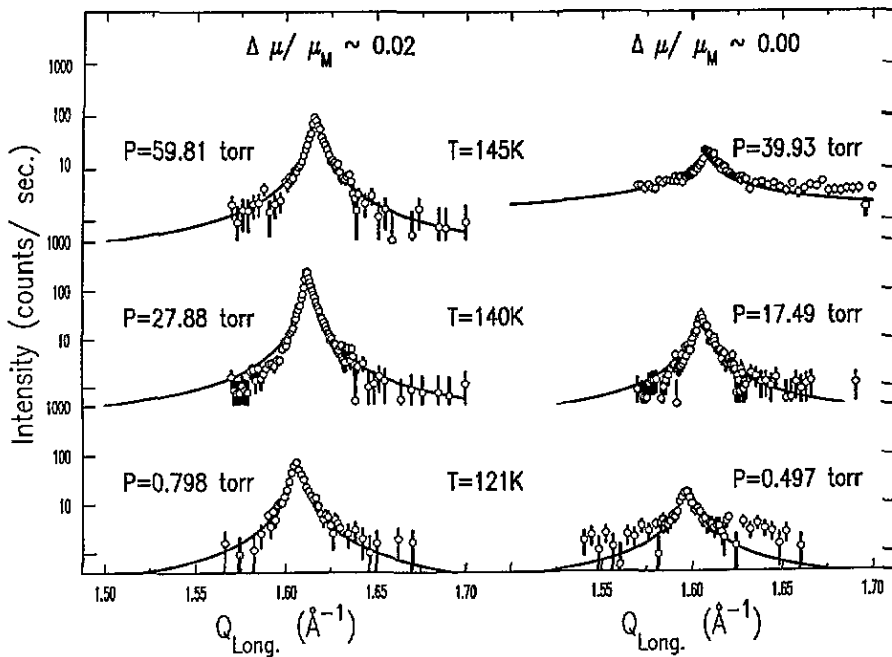


Figure 1. Isotropic Kummer function-based fits to longitudinal scans with the background subtracted at each of the temperatures studied. High- and low-pressure scans are shown for each temperature. The scans represent data obtained at equivalent reduced chemical potentials of magnitude $\Delta\mu/\mu_M = |(\mu - \mu_M)/\mu_M|$ for each of the three temperatures. In each case, the low-pressure data are no longer best described in terms of the solid-phase lineshape alone.

region is seen to span approximately 30 torr. It is important to note that despite the larger pressure ranges at higher temperatures, the transition may generally be described as being very sharp. This is particularly the case if the transition is driven by varying the sample temperature. In that case the whole transition is seen to occur over a very narrow range of temperature. It is partly as a consequence of this very sharp behaviour with temperature that our measurements have been made isothermally by varying the ambient vapour pressure.

The sharpness of these transitions has been the subject of some comment in the literature [16, 21]. We concur with these observations, and point out that the behaviour observed here in the xenon-graphite system is very different from the extremely gradual re-entrant freezing behaviour seen in the case of krypton on graphite [27].

3.2. The critical 2D-aligned liquid

In figure 3 we present experimental data obtained at 145 K for pressures in the vicinity of the melting transition. On decreasing the pressure below approximately 40 torr, we observe a continuous broadening of the longitudinal scattering data consistent with critical scattering from an aligned liquid phase. The critical aligned liquid is confirmed to retain orientational order through the observation of a clear diffraction peak in the in-plane transverse direction, as shown in figure 4. Similar effects were also observed at 140 K and data obtained at that temperature have been published previously [17]. The solid lines through the data correspond to fits assuming a model lineshape for the aligned liquid phase. The lineshape consists of an anisotropic 2D Lorentzian convolved longitudinally with the experimental resolution and transversely with the sample mosaic. The poor mosaic of the adsorbate

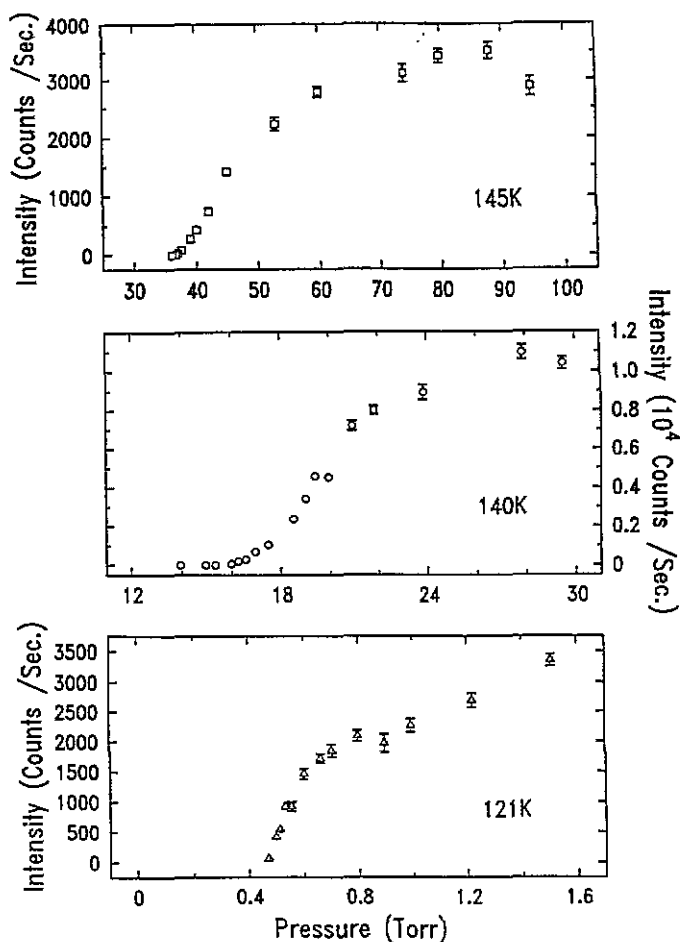


Figure 2. Behaviour of the peak intensity as determined from our FSCS analysis. The decay of the ordered phase is illustrated as function of the ambient vapour pressure.

sample unfortunately prohibits a detailed assessment of anisotropies in the critical scattering close to the melting transition. The mosaic limited feature is, however, quite consistent with the existence of an aligned liquid phase and previous observations [12]. The form of the diffraction lineshape appropriate to aligned liquids and hexatic phases is a subject of active theoretical interest. For instance, Peterson and Kaganer have recently calculated the form of the azimuthal lineshape, particularly appropriate for studies of the hexatic–isotropic transition [28]. The radial lineshape of particular importance to our experiments has been discussed by Aeppli and Bruinsma who conclude that in the vicinity of a continuous hexatic–isotropic transition the radial lineshape should be square-root-Lorentzian while otherwise it should be Lorentzian [29]. As our experiments consider the FSCS–aligned liquid transition a Lorentzian has been judged appropriate.

In figure 5 we summarize the results from analysing our data in terms of an aligned liquid model for each of the three temperatures studied. At 145 K and 140 K, we observe a continuous evolution of the longitudinal positional correlation length, $\xi = 1/\kappa$, consistent with a continuous melting transition. In both the 145 K raw data published here (figure 3)

145K

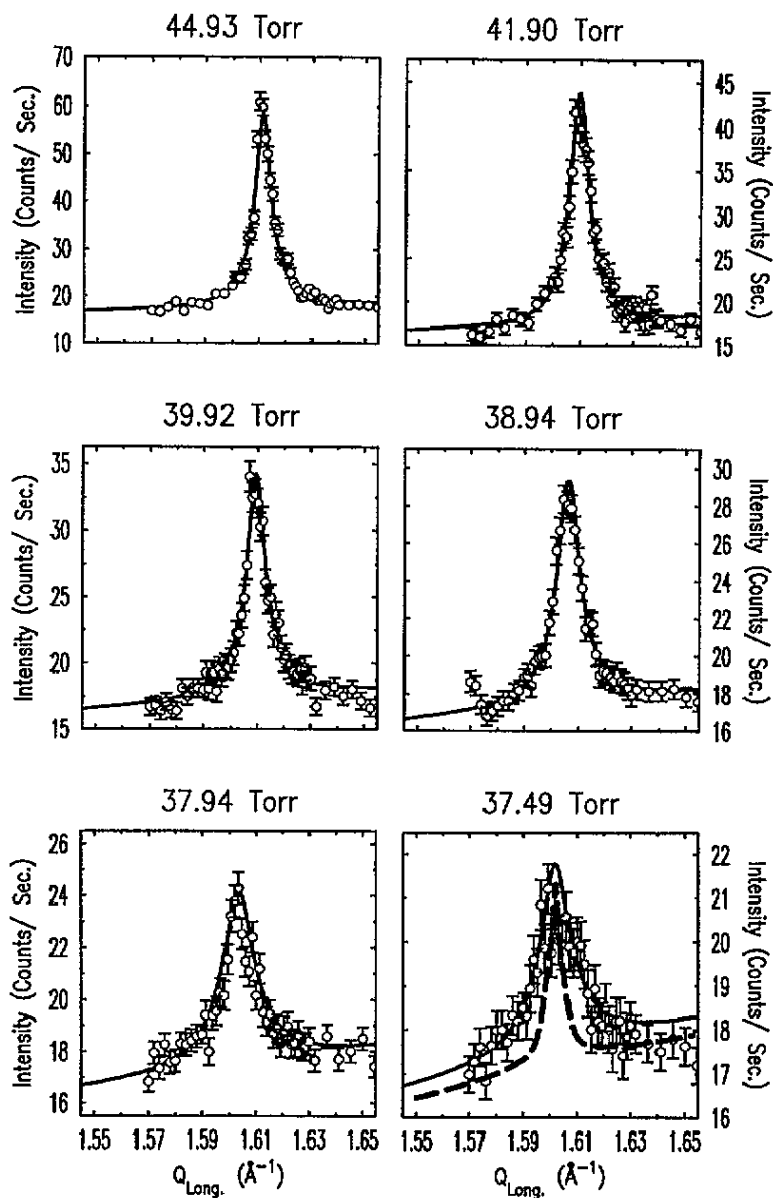


Figure 3. Longitudinal scans obtained at 145 K close to the melting transition, as analysed in terms of our aligned liquid model. Note that the peak half-width at half-maximum evolves continuously as the finite-sized solid melts into the aligned liquid phase. The dashed lines shown in the panel at 37.49 torr correspond to the lineshape predicted from a finite-sized solid analysis with the exponent η_G constrained to be $1/3$, its theoretical maximum value. It is clear that the data at lower vapour pressures can no longer be described by the FSCS-ordered phase line shape.

145K

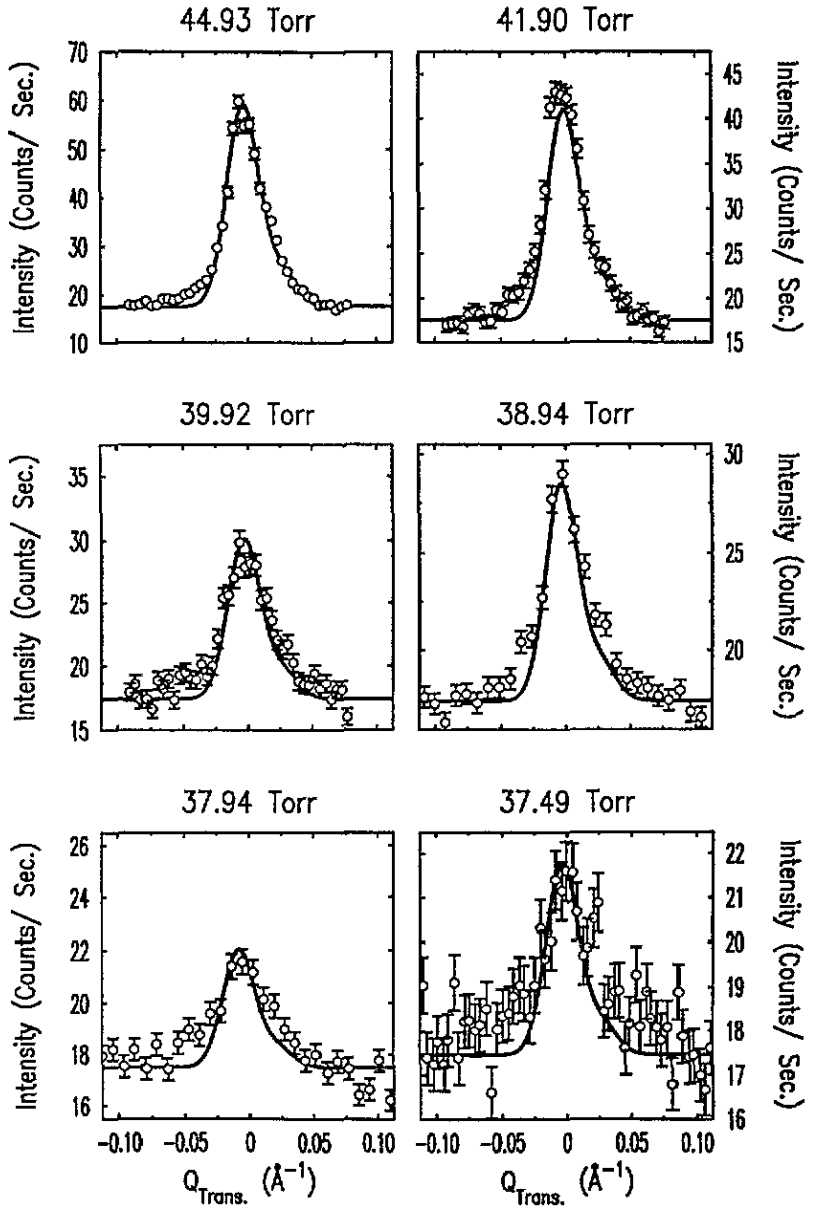


Figure 4. Transverse scans obtained at 145 K complementary to the data illustrated in figure 3. The solid lines correspond to the transverse fit of the aligned liquid model. At each vapour pressure the transverse width is determined by the sample mosaic.

and the 140 K data published previously [17] it is possible to see directly the smooth evolution of the aligned liquid peak width. This corresponds to the evolving length scale

of the real-space critical fluctuations. As is illustrated in the bottom panel of figure 5 there is no evidence for an equivalent evolution of the correlation length at 121 K. It should be emphasized that this does not exclude the possibility of there being critical scattering at 121 K over a very narrow range of pressure. As we shall see in the next section, the decay of the order parameter at all three temperatures appears to behave in the same way with respect to the fundamental quantity, i.e. the reduced chemical potential.

Our observations of a continuous phase transition at the higher temperatures between a FSCS state and an aligned liquid phase and the possibility of a first-order transition at lower temperatures complement recent studies performed using transmission high energy electron diffraction [21]. Venables and co-workers have reported an investigation of monolayer xenon melting in the temperature range $110 \text{ K} < T < 120 \text{ K}$ [21, 30] and they conclude that both the solid-hexatic and the hexatic-isotropic liquid transition are, at most, weakly first order. Although the experiments reported here do not allow us to comment on the hexatic-isotropic transition we find that our observations at 121 K lead us to conclude that at that temperature the melting transition may be weakly first order while at the higher temperatures it appears more clearly continuous.

3.3. Universality

One of the most compelling aspects of experimental studies of critical phenomena is the degree of consistency between measurements made on a given system under different conditions and perhaps, even more intriguingly, from extremely different physical systems whose fundamental physical properties may be described in terms of equivalent symmetries.

Detailed considerations of critical phenomena in two-dimensional systems at finite temperatures are, however, somewhat unusual given the constraint that for systems where the order parameter has continuous symmetry there is strictly no phase with true long-range order. The observation, however, that our xenon adsorbate is not a perfect 2D solid but is limited by finite size effects has a fortuitous consequence. The finite-sized nature of the adsorbate ordering means that the order parameter has a non-zero value when it is truncated at the margins of the ordered regions. We may therefore extract an effective order parameter exponent β which may be compared with values obtained in studies of finite sized 2D-XY magnets.

Recently, Bramwell and Holdsworth have considered the behaviour of finite-sized 2D-XY magnets by means of a modified renormalization group technique [31, 32]. They conclude that an effective value of $\beta = 0.23$ is a universal signature of 2D-XY behaviour in finite-sized systems. In figure 6(a) we illustrate with a solid line a power-law lineshape parametrized by $\beta = 0.23$ as suggested by Bramwell and Holdsworth. The power law has been rounded by convolution with a Gaussian distribution of melting chemical potentials of magnitude $\Delta\mu/\mu_M = 0.0043$ to allow for small inhomogeneities across the sample and finite-size effects. The data points correspond to the measured intensities of the FSCS Kummer function lineshape discussed in section 3.1.

The values of the melting pressures from the FSCS to aligned liquid phase, P_M , are given in table 1 and have been determined from power-law fits to the experimental data. These individual power-law fits allow us to estimate the values of the critical exponent β most appropriate for the data in each case. At 145 K $\beta = 0.226 \pm 0.022$, at 140 K $\beta = 0.232 \pm 0.012$ and at 121 K $\beta = 0.234 \pm 0.031$. The data presented in figure 6(a) have been matched in each case to the proposed $\beta = 0.23$ lineshape through the adjustment of two independent variables, the melting point P_M and an intensity normalization factor.

The degree of consistency between the three measurements obtained at different

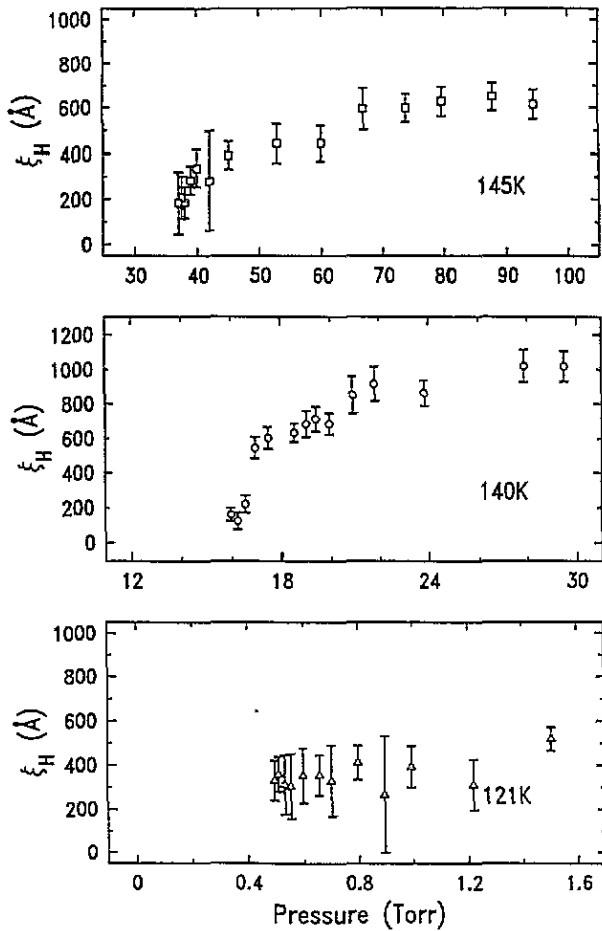


Figure 5. Summary of the behaviour of the longitudinal Lorentzian correlation length ($\xi_H = 1/\kappa$) as a function of ambient vapour pressure. In this case all of the data have been fitted with the 2D anisotropic Lorentzian lineshape expected for an aligned liquid. Note that at 145 K and 140 K there is a continuous evolution in the positional correlation length as the xenon monolayer is melted. The 121 K data show no such continuous diminution.

Table 1. Tabulated are the adsorbate temperature, the finite-size parameter from our Kummer function analysis, the largest correlation length observed from our aligned liquid analysis, the melting pressure that was obtained from a power-law fit to the order parameter and the monolayer coverage at melting, from Jin *et al* [16].

Temperature (K)	L (Å)	ξ_{MAX} (Å)	P_M (Torr)	f_M (ML)
121	770	400 ± 150	0.492	0.84–0.91
140	1230	1030 ± 190	18.067	0.88–0.93
145	900	650 ± 130	40.383	0.92–0.95

temperatures suggests that the melting process may be no different at 121 K than at the higher temperatures. The consistency of the measurements with recent observations from finite-sized 2D magnetism lead us to conclude that, at least in the case of the FSCS-aligned

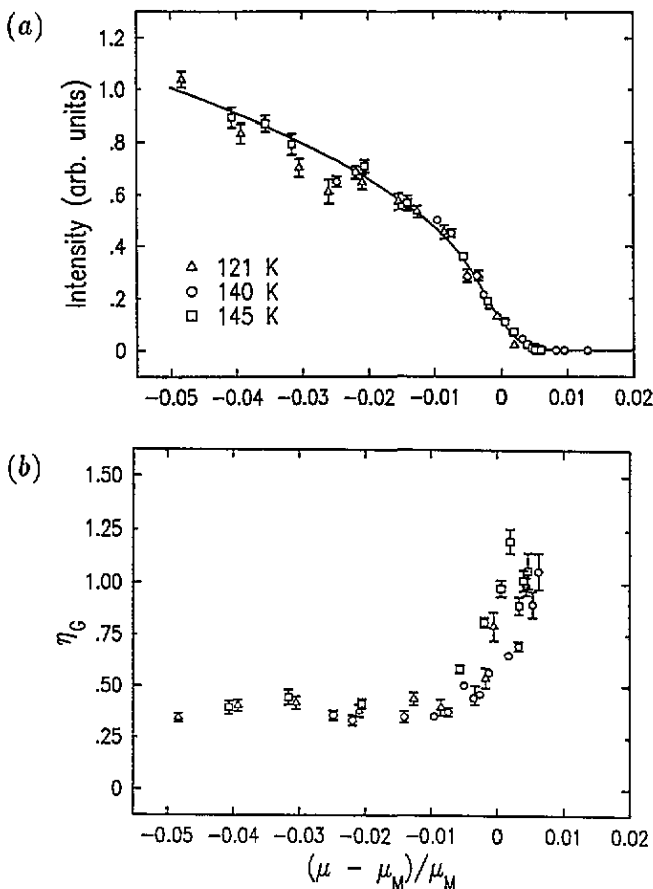


Figure 6. (a) Intensity, S_0 , obtained from our finite-sized solid model versus reduced chemical potential. The solid line represents the power-law form $|\mu/\mu_M - 1|^{2\beta}$ with $\beta = 0.23$ as has recently been suggested for finite-sized systems in the 2D-XY universality class [31, 32]. (b) Behaviour of the exponent η_G as a function of reduced chemical potential for all data as analysed by our isotropic Kummer function model.

liquid transition, the system may indeed fall into the 2D-XY universality class consistent with the predictions of KTHNY theory.

In figure 6(b) we see a clear continuous increase in the extracted value of η_G through the melting transition at each temperature. Theoretically, as P is varied through P_M the true theoretical lineshape should evolve from (2) to the 2D anisotropic Lorentzian characterizing the aligned liquid phase. Naturally, each lineshape would be expected to be inappropriate deep in the other phase and also in the vicinity of the transition where a mixed lineshape might be expected. The smooth increase in the extracted values of η_G in the vicinity of the transition and in the aligned liquid phase are therefore a sign of the breakdown of applicability of the FSCS lineshape. The power-law tails to the scattering parameterized by η_G are adjusting in an attempt to fit what is really an increasing Lorentzian width associated with the critical aligned liquid. From both the smooth evolution of the correlation length illustrated at 140 K and 145 K in figure 5, the behaviour of the order parameter and the smooth increase in η_G illustrated in figure 6 we see evidence for continuous behaviour

across the FSCS-aligned liquid phase transition at these higher temperatures.

In table 1 we bring together some of the observations emerging from our analyses in terms of the FSCS lineshape and a Lorentzian associated with a critical aligned liquid. In particular, we note that although the maximum correlation length ξ_{MAX} emerging from the aligned liquid analysis was defined quite independently of the finite-size parameter L , both imply that the ordered phase had a spatial extent $\gtrsim 1000$ Å at 140 K. However, for both measures, ξ_{MAX} and L , the ordered state was seen to be somewhat less well correlated at 145 K. The solid phase correlations were diminished still further by the time of the third set of measurements, made at 121 K. This reduction in the ordered phase quality in the successive measurements presumably reflects an erosion in the cleanliness or physical quality of the substrate surface during the course of the experiments.

4. Discussion

We have studied the isothermal melting of near-monolayer xenon on single-crystal graphite at three temperatures 121 K, 140 K and 145 K. At 140 K and 145 K we observe a continuous evolution of the positional correlation length on melting consistent with previous x-ray studies. In addition, a smooth increase is seen in the measured value of the exponent η_G as the transition is approached. We conclude, therefore, that at 140 K and 145 K, near-monolayer xenon on graphite exhibits a continuous melting transition from a finite-size limited 2D solid to an orientationally ordered fluid. Jin *et al* [16], however, have reported that their thermodynamic measurements demonstrate that the melting of monolayer xenon on graphite is always first order. In addressing the differing conclusions drawn by ourselves and other groups using x-ray diffraction and the thermodynamic studies of Jin *et al* it is appropriate to point out that although we can never exclude an arbitrarily small first-order jump right at the melting point, the very high-quality single-crystal substrate allows us to see critical scattering diverging to length scales of approximately 1000 Å, which is presumably better than is obtainable from the substrates used in the thermodynamic studies.

Our result appears to be consistent with the vapour pressure measurements of Gangwar *et al* [15] who conclude that a tricritical point occurs at ~ 147 K; however, their data certainly permit continuous melting down to 140 K [33]. Venables and co-workers have recently highlighted a possible explanation for the divergent conclusions emerging from the thermodynamic and the x-ray diffraction measurements [21]. They comment that much of the thermodynamic work has concentrated in describing the melting process as being a single transition. Venables and co-workers [21, 30] have recently confirmed that the melting of monolayer xenon at these temperatures consists of two distinct transitions as discussed in section 3.1. It is suggested that the thermodynamic studies are more sensitive to the aligned-isotropic liquid transition while the single-crystal x-ray diffraction experiments are more sensitive to the FSCS-aligned liquid transition. Venables and co-workers propose therefore that the continuous behaviour observed in our x-ray diffraction experiments [17] might be consistent with the FSCS-aligned liquid transition while the loss of orientational order would be a first-order discontinuous process.

Elegant heat capacity studies of quasi-2D liquid crystal systems such as 80SI, have reported observing two-step melting transitions with, in particular, a much stronger change in entropy associated with the smectic C (isotropic) to smectic I (hexatic) transition compared with that at the smectic I to crystalline J phase transition [34]. This latter transition is analogous to the transition studied in our experiments. As discussed previously, however, the xenon monolayer experiments are far from ideal realisations of KTHNY behaviour in regard to the loss of orientational order. It is the presence of an ordering substrate which

implies that two-dimensional melting in adsorbed xenon on graphite may not be simply equivalent to the behaviour seen in crystalline J-hexatic I-smectic C liquid crystal systems. Nevertheless, the possibility of two-dimensional melting proceeding through two distinct, possibly continuous, transitions is now well established.

Finally, we have analysed our data in terms of an effective order parameter for a finite-sized 2D solid and compared the value of β obtained with recently published reports for the finite-sized 2D-XY magnet [31, 32]. We observe a satisfying degree of self-consistency between the measurements made at all three temperatures. The data are also found to be consistent with the recent predictions made for systems in the 2D-XY universality class, as predicted by KTHNY theory. The width of the transition has been reported [21] to be somewhat sharper than that predicted by KTHNY theory. While we agree that the transition is indeed very sharp, particularly as a function of temperature, our isothermal diffraction measurements reveal a rate of decrease for the order parameter at the FSCS-aligned liquid transition consistent with 2D-XY theory.

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