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The role of bond orientational order and the nature of the melting transition in an incommensurate physisorbed monolayer: argon on graphite at completion

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Abstract

A constant-temperature, constant density molecular-dynamics (MD) method is utilized to study the behavior of bond-orientational, translational and thermodynamic parameters in the solid, melting transition and fluid regimes of argon physisorbed onto graphite (Ar/gr) at complete coverage. For Ar/gr, completion occurs near $\rho = 1.26$ in units of 0.0636 atoms/Å^2, where $\rho = 1$ corresponds to the complete $\sqrt{3} \times \sqrt{3}$ R30° commensurate adlayer. Several bond-orientational and newly introduced pair-distribution order parameters and effective densities prove especially useful in helping evaluate the temperature behavior of the system's structural order. A broad melting transition accompanied by vacancy formation through large vertical fluctuations in the adsorbate atoms is observed at $T_m = 135 \pm 5$ K, spanning a temperature range of roughly 40 K. The melting transition accompanies a rapid formation of a low-density second layer. Substantial bond-orientational order persists into the high-temperature fluid ($T \approx 300$ K), which is not hexatic in nature and which exhibits infrequent desorption from the first layer. The behavior and interaction of the neighbor shells are compared and contrasted to those observed in Kr/gr.

Keywords: Argon; Molecular dynamics; Physical adsorption; Rare gas solids; Two-dimensional melting

1. Introduction

The work presented here is a follow-up study to a similar effort [1] regarding the (commensurate) Kr/gr system at monolayer completion ($\rho = 1$). Since most of the general background of quasi-two-dimensional (2D) melting and justification of interest in the bond-orientational behavior of physisorbed systems is presented there and in many other places, we felt it expeditious to omit it here. The main motivations of our study of Ar/gr are the marked difference in its behavior from Kr/gr, the wealth of experimental and (to a lesser extent) computational studies available about it and the interest in the connection between the bond-orientational behavior of the system and its dynamics at monolayer completion.

The Ar/gr system is perhaps the most interesting rare gas monolayer system. Since the Ar lattice spacing is roughly 8% smaller than that of a registered phase, Ar/gr forms an incommensurate solid, where not all of the adlayer atoms hover above graphite hexagon centers. Hence, in a local sense, $\rho > 1$ for the submonolayer solid patches, which is similar to a floating system. In fact,

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