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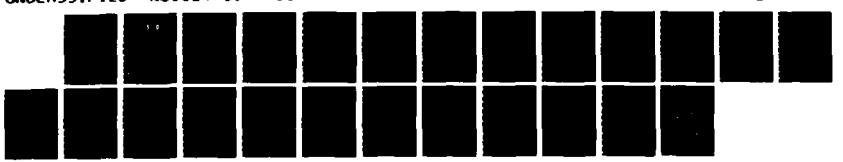

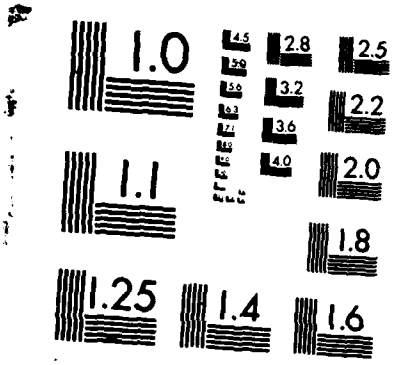

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Heat Capacities of Rare Gases Adsorbed on Graphite

by

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HEAT CAPACITIES OF RARE GASES ADSORBED ON GRAPHITE

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Abstract

The McQuistan-Hock model is used to investigate adsorption for realistic systems. After computing the partition function in the most convenient ensemble for the problem under consideration, i.e., the isothermal-isobaric ensemble, the heat capacity for submonolayer films of Ne, Ar and Xe on graphite is computed for several coverage values. Heat-capacity signatures exhibit maxima at temperature values that are in good agreement with experimental data.

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I. Introduction

The recent discovery that submonolayer films of adsorbates on some substrates have two-dimensional phases has opened a new field: the study of two-dimensional matter, its phases, and the transitions between them. A common way to realize two-dimensional systems is to physisorb gaseous particles (atoms or simple molecules) onto solid surfaces (substrates). Dependent on particle and substrates properties, the adsorbate condenses as a homogeneous film which might reveal two-dimensional properties. Often the only task the substrate is expected to accomplish is to force the adsorbed particles into a plane, so that the substrate disturbs the adsorbed system as little as possible. In order to investigate experimentally the physical properties of two-dimensional systems, one must have substrates of high specific surface areas such as graphite (and its modifications). The essential task in a thermodynamical investigation of a physisorbed two-dimensional system is the construction of a complete group of adsorption isotherms. In practice, for an accurate determination of the entropy, calorimetric experiments to determine the specific-heat signatures are performed. Such experiments offer the possibility of studying low-dimensional order-disorder phenomena such as structural and melting phase transitions.

With respect to the order of a melting transition, the situation depends strongly on whether one considers three- or two-dimensional melting. In the former case, there is no doubt that melting is always a discontinuous event at which the system absorbs latent heat from the surroundings. In the two-dimensional case, the question as to whether melting is a discontinuous or continuous process is still open.

In the present paper we use the McQuistan-Hock model[1] to compute heat-capacity signatures for two-dimensional systems of rare gases adsorbed on graphite. Experimental studies on these systems have demonstrated the existence of ordering and phase transitions[2] and have identified regions of two-dimensional liquid-vapor as well as liquid-solid coexistence, accompanied by a two-dimensional critical point.[3] In the next section, after a brief overview of the McQuistan-Hock model, we present our calculations and results. The third and last section is devoted to our conclusions.

II. Theory and Calculations

In a recent paper,[1] McQuistan and Hock developed an exact solution for the distribution function of q indistinguishable particles on a 2 × N lattice, where N is a positive integer number. The grand-canonical partition function is written as

$$Z_N(x,y,z,t) = \sum_{q=0}^{2N} F_{qN}(x,y,t) z^q \tag{1}$$

where

$$F_{qN}(x,y,t) = t^q \sum_{n_{00}} \sum_{n_{11}} A[N,q,n_{00},n_{11}]\, x^{n_{11}}\, y^{n_{00}} \tag{2}$$

$$t = e^{-V_0\beta}, \tag{3}$$

$$x = e^{-V_{11}\beta}, \tag{4}$$

$$y = e^{-V_{00}\beta} \tag{5}$$

and

$$z = e^{\beta\mu}, \tag{6}$$

in which μ is the chemical potential of the adsorbed particles, $\beta \equiv (k_B T)^{-1}$ with T the absolute temperature and k_B the Boltzmann constant, V_{00} is the interaction between two nearest-neighbor vacant sites, V_{11} is the interaction between two nearest-neighbor adparticles, and V_0 is the interaction between a particle and the surface. The number of unique ways q indistinguishable particles can be arranged on a rectangular 2 × N lattice to form n_{11} occupied nearest-neighbor pairs and n_{00} vacant nearest-neighbor pairs is given by $A[N,q,n_{00},n_{11}]$ in Eq. (2) by making use of a 15-term recursion relation. In a subsequent paper,[4] Hock and McQuistan computed the adsorption isotherms for what we might call the "rigid" lattice in which there is no interaction between two nearest-neighbor vacant sites: $V_{00} \equiv 0$. They showed that the coverage as a function of the spreading pressure does not exhibit a first-order phase transition.

The McQuistan-Hock model is very interesting because of its simplicity, because it takes into account various interaction terms (V_0,V_{00} and V_{11}), and because it is exactly solvable. We will show here that the model is applicable to realistic systems by comparing the critical temperatures obtained from it with the critical temperatures measured in systems such as rare gases adsorbed on graphite.

In the McQuistan-Hock model, the grand canonical partition function [Eq. (1)], as $N \to \infty$, is given by

$$Z(N,\beta,\mu) \approx k_1 \eta_1^{-N} \quad , \tag{7}$$

where

$$k_1 = - \frac{r(\eta_1)}{s'(\eta_1)} \quad , \tag{8}$$

and η_1 is the smallest root of

$$s(\eta) \equiv \sum_{j=0}^{3} b_j \eta^j = 0 \quad . \tag{9}$$

In Eq. (8),

$$r(\eta) = \sum_{j=0}^{2} a_j \eta^j \tag{10}$$

and

$$s'(\eta) = \left(\frac{\partial s}{\partial \eta}\right)_{x,y,z} \quad . \tag{11}$$

The coefficients $a_j(x,y,z)$ and $b_j(x,y,z)$ in Eqs. (9) and (10) are given in Eqs. (9) and (10) of Ref. 4. The spreading pressure p can be obtained as

$$p = \frac{1}{2\beta} \ln \frac{2\theta s'}{sz} \quad , \tag{12}$$

where θ is the coverage as $N \to \infty$, and

$$\dot{s}(\eta) = \left(\frac{\partial s}{\partial z}\right)_{x,y,\eta} \quad . \tag{13}$$

In order to study the behavior of the heat capacity as a function of the temperature, keeping the spreading pressure p and the number of adparticles q as constant it is most convenient to work within the isothermal-isobaric ensemble, whose partition function will be denoted by $\Delta \equiv \Delta(q,\beta,p)$. The Gibbs free energy, entropy and heat capacity are given by

$$G(q,\beta,p) = -\frac{1}{\beta} \ln \Delta \quad , \tag{14}$$

$$S(q,\beta,p) = k_B[\ln\Delta - \frac{\beta\Delta'}{\Delta}] \quad , \tag{15}$$

$$C(q,\beta,p) = k_B\beta^2 [\frac{\Delta''}{\Delta} - (\frac{\Delta'}{\Delta})^2] \quad , \tag{16}$$

where $C(q,\beta,p)$ has a maximum at the critical temperature ($\beta = \beta_c$), i.e., the solution of the equation

$$2\Delta(\Delta')^2 + 3\beta\Delta\Delta'\Delta'' - 2\beta(\Delta')^3 - \beta\Delta^2\Delta'' - 2\Delta^2\Delta'' = 0 \quad . \tag{17}$$

In Eqs. (15)-(17), the prime denotes the derivative with respect to β. In order to find $\Delta(q,\beta,p)$, we start from its definition

$$\Delta(q,\beta,p) \equiv \Delta_q(\beta,p) \equiv \sum_{N=1}^{\infty} F_q(N,\beta)\eta^N$$

$$= t^q \sum_{N=1}^{\infty} f_q(N,\beta)\eta^N \quad , \tag{18}$$

where $\eta \equiv e^{-2\beta p}$. Multiplying by z^q, summing over q from 0 to 2N and using Eqs. (3) and (4) given in Ref. 4, the following recursion relation is obtained for $\Delta_q(\beta,p)$:

$$d_0\Delta_q = - d_1\Delta_{q-1} - d_2\Delta_{q-2} - d_3\Delta_{q-3} + c_3\delta_{q,3} \quad , \tag{19}$$

where $\delta_{q,3}$ is the Kronecker delta function,

$$d_0 = 1 - y^3\eta \quad , \tag{20a}$$

$$d_1 = -\eta(xyd_0 + y^3\eta + 1)t \quad , \tag{20b}$$

$$d_2 = -\eta(x^3d_0 + xy\eta)t^2 \quad , \tag{20c}$$

$$d_3 = x\eta^2(xy-1)[x^2d_0 + y\eta(2xy-1)]t^3 \quad , \tag{20d}$$

and c_3 is given by Eq. (24d) below. From Eq. (18) and from the recursion relation for $f_q(N,\beta)$ as given in Ref. 4, we can compute the initial condition for $\Delta_q(\beta,p)$. We obtain, provided $V_{00} > -2p/3$, that

$$\Delta_0 = \frac{y\eta}{d_0} \quad , \tag{21a}$$

$$\Delta_1 = 2\eta + 4y\eta\Delta_0 + 2y^2\eta\Delta_0^2 \quad , \tag{21b}$$

$$\Delta_2 = x\eta(1+2y\eta) + 2\eta\Delta_0(2xy^2\eta + y^2\eta + x + 1/y) + \eta\Delta_0^2(2xy^3\eta + 4y^3\eta + x + 4) + 2y\eta\Delta_0^3(1 + y^3\eta) \quad . \tag{21c}$$

Let us define

$$\sum_{q=0}^{2N} z^q \Delta_q = \frac{u(z)}{v(z)} \tag{22}$$

where

$$u(z) = \sum_{j=0}^{3} c_j z^j \tag{23a}$$

$$v(z) = \sum_{j=0}^{3} d_j z^j \quad . \tag{23b}$$

Here the d_j's are given by Eqs. (20) and the c_j's are given by

$$c_0 = y\eta \tag{24a}$$

$$c_1 = \eta t\,[2d_0 + y\eta(4y - xy - 1)] \tag{24b}$$

$$c_2 = \eta t^2[xd_0 + xy\eta(2-x^2)] \tag{24c}$$

$$c_3 = x\eta^2t^3\{[x^2y\eta + 4y\eta(1-x)](xy-1) + (4-y-2x)(x-xy^3\eta+y^2\eta) - 1\} \quad . \tag{24d}$$

If we rewrite Eq. (22) as

$$\frac{u(z)}{v(z)} = K + z\,\frac{C_0 + C_1z + C_2z^2}{1 + D_1z + D_2z^2 + D_3z^3} = K + z\,\frac{\gamma(z)}{\delta(z)} \tag{25}$$

with

$$K = c_0/d_0 \quad , \tag{26}$$

$$d_0^2C_j = c_{j+1}\,d_0 - c_0\,d_{j+1} \tag{27}$$

and

$$D_j = d_j/d_0 \quad , \tag{28}$$

we obtain

$$\Delta_q = \sum_{j=1}^{3} \chi(z_j)\, z_j^{-q} \quad , \quad 0 < q \leq 2N \tag{29}$$

where the z_j's are the roots of the equation

$$\delta(z) = 0 \tag{30}$$

and

$$\chi(z) = -\gamma(z)/\delta'(z) \quad . \tag{31}$$

Here the prime denotes the derivative with respect to z.

It is worth noting that for the special case in which $x = y = 1$, $C_2 = D_3 = 0$ and the solution of Eq. (29) gives

$$z_{\pm} = \frac{-\eta \pm \sqrt{\eta}}{\eta t} \quad , \tag{32}$$

and the postive root is always smaller than the absolute value of the negative root, so that for large q,

$$\Delta_q = \frac{(t\sqrt{\eta})^q}{2(1-\sqrt{\eta})^{q+1}} \quad , \tag{33}$$

and the heat capacity per adparticle is

$$C = \frac{k_B \alpha^2}{\sinh^2 \alpha} \quad . \tag{34}$$

where $\alpha \equiv \beta p/2$. We note that this is the same result that would have been obtained for a system of independent, distinguishable particles, each of which has only two accessible states: the particle is on the site or it is not.

For the more general case in which $V_{11} \neq 0$, we have solved Eq. (30) numerically choosing values for the interaction constants that are likely to model rare-gas/graphite systems. In Table I, the chosen values are displayed together with the literature reference from where they have been taken. Several studies of the Ne, Ar, Xe/graphite systems have reported a broad anomaly in the specific heat at temperature values given in Table I, namely at 15.7 K for Ne, 55 K for Ar and 118 K for Xe. In Fig. 1 we present the specific heat as a function of temperature for values of the coverage of 0.1, 0.3 and 0.5. We see that maxima in the specific heat are observed in the range of the experimental critical temperatures.

III. Discussion and Conclusions

From Fig. 1 we can see that maxima in the heat-capacity signatures are found around 10-15 K for Ne, 40-52 K for Ar and 80-102 K for Xe. Let us compare these results with some experimental findings and with some other model calculations.

Steele and Karl[8] reported peaks in the specific-heat measurements of Ne adsorbed on graphitized carbon black powder, where at half coverage they found a peak at 16.1 K. Antoniou[9] in turn found a peak at 12 K. Huff and Dash[10] made the first set of measurement of Ne adsorbed on Grafoil, for submonolayer coverages in the temperature range of 2-20 K and found peaks between 12 and 15 K. The anomalies they obtained have been confirmed by Rapp et al,[11] who found, besides a first peak at 13.6 K, a second one at

around 16 K which moves up in temperature as the coverage increases, and then disappears at higher coverages.

Recent experimental studies of neutron scattering,[12,13] heat capacities[14] and synchrotron x-ray scattering[15] on submonolayers of Ar on graphite were all interpreted as being consistent with a continuous melting process. Migone et al[16] reported heat-capacity measurements of submonolayer Ar on Graphite foam, with anomalies at 47.2, 49.5 and 55 K, and interpreted the first peak as a "weak" first-order transition. For Xe the situation is similar: melting of Xe physisorbed on graphite has been interpreted both as continuous[17,18] and first-order melting.[19]

The interesting aspect of the McQuistan-Hock model is that it does not allow for any first-order transition. Yet, when applied to "realistic" systems, it predicts maxima in the heat-capacity signatures at temperatures very close to the experimental ones. A single model that forbids a first-order transition but displays maxima in the heat-capacity curve is the one-dimensional lattice with nearest-neighbor interactions. Unfortunately, this model predicts those maxima at temperature values well below the experimental data. A two-dimensional model such as the square lattice predicts second-order critical temperatures above the experimental data, while the hexagonal lattice gives better agreement with experiments.

In Table II we report the ratio $R \equiv |V_{11}/T_c|$ as predicted by various models, as well as the experimental average value $\bar{R}$ of this ratio as obtained from Table I ($|\frac{R-\bar{R}}{R}| < 10\%$). The predictions from the McQuistan-Hock model are also reported as averages because, as can be seen from Fig. 1, R is not exactly constant. From this table it is interesting to see that the McQuistan-Hock model predicts values of T_c that are in good agreement

with the experimental measurements. The model does not allow for first-order transitions, nor does the linear-lattice model with nearest-neighbor interactions. On the other hand, the linear-lattice model predicts maxima in disagreement with experiments. In good agreement with experiments are also the anomalies predicted by the square-lattice and by the hexagonal models, the difference between the two being what we call the coordination number (CN in Table II), i.e., the number of nearest neighbors attached to each site: the experimental T_c's lie in between the predictions from the models with CN = 3 and CN = 4.

Our conclusions are the following: (i) The McQuistan-Hock model has the advantage of being exactly solvable and of giving good agreement between computed and experimental temperatures at which the maxima in the heat-capacity signatures occur. (ii) The model contains very few parameters, usually V_0, V_{00} and V_{11}; yet those parameters are sufficient to determine the "gross" features of the statistics of adsorption. This implies that the details of the various interactions occurring in real systems do not seem to play an important role. (iii) The model contains a parameter, V_{00}, which accounts for distortions of the substrate, an effect that has been invoked by various authors[10,11] to explain some details of the experimental heat-capacity signatures. It would seem worthwhile to explore the dependence of the adsorption thermodynamics on V_{00}. (iv) The impossibility of showing a first-order transition, on one hand, and the good agreement between the computed and measured critical temperatures suggests that the order of the transition seen at temperatures around the T_c-values of Table I might not necessarily be of first order. (v) An important role seems to be played by the number of nearest neighbors attached to each site: the McQuistan-Hock model (with CN = 3) represents a logical extension from a one-dimensional to

the complete two-dimensional system. It would therefore be interesting to examine the influence of the average number of nearest neighbors on the heat capacity as a function of temperature.

Acknowledgments

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Table I. Adparticle-surface (V_0) and adparticle-adparticle (V_{11}) interactions in Kelvin. T_c is the critical temperature.

	[a] V_0	[b] V_{11}	[c] T_c	[d] T_c
Ne	-378	-34.6	15.7	14.5
Ar	-1103	-120	55	51.0
Xe	-1912	-236	118	102.0

[a] From Ref. 5

[b] From Ref. 6

[c] From Ref. 7

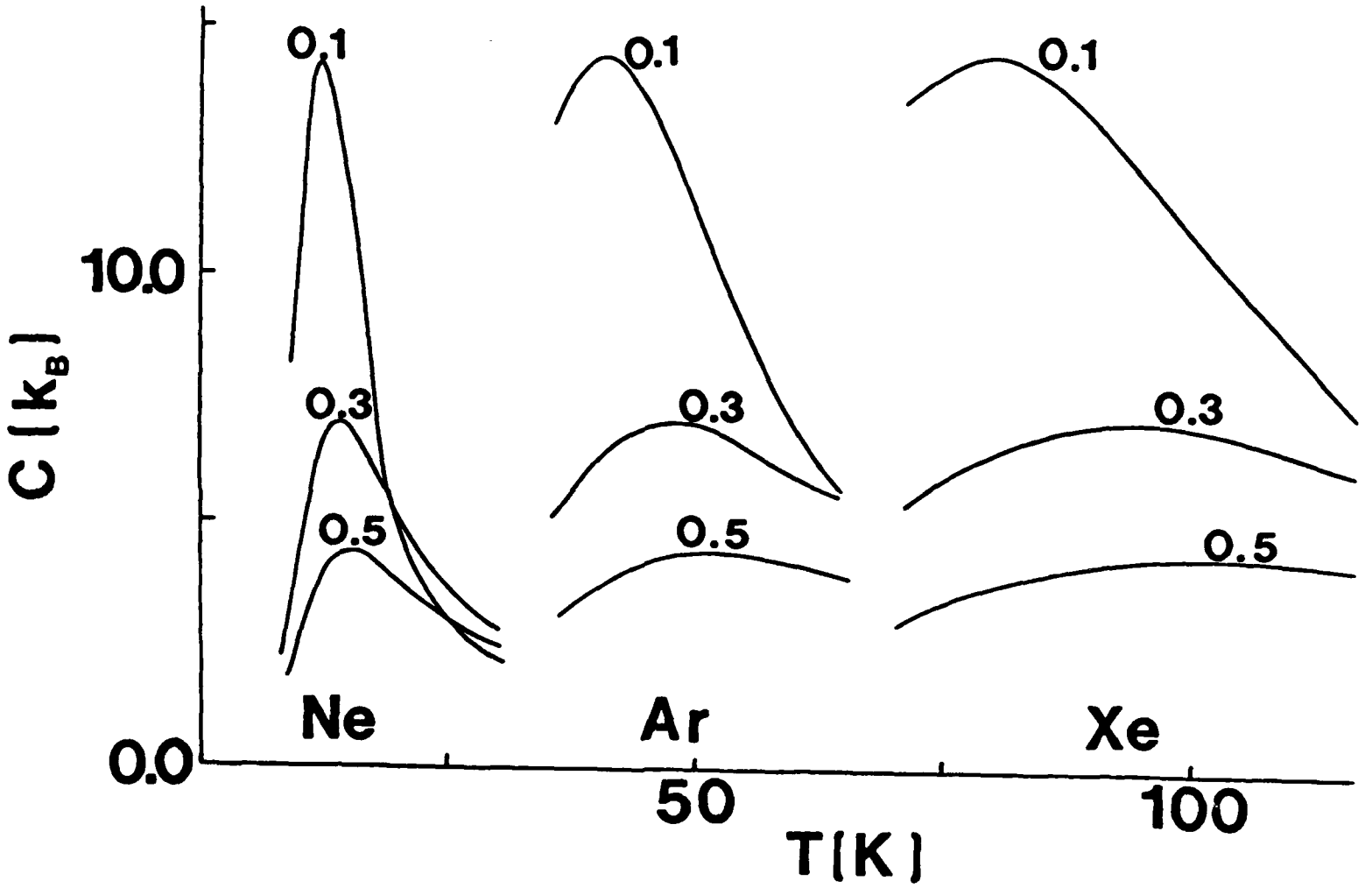
[d] From this study at coverage value of 0.5

Table II. Values of the ratio $R \equiv |V_{11}/T_c|$ from various models and from experiments (EXP); 1D = linear lattice, HL = hexagonal lattice, MQH = McQuistan-Hock lattice, SL = square lattice. For EXP and MQH the average values are reported. The coordination number is indicated in parentheses.

Systems	R
1D (CN = 2)	4.80
HL (CN = 3)	2.63
MQH (CN = 3)	2.35
EXP	2.13
SL (CN =4)	1.76

Figure Caption

1. Specific heat for Ne, Ar and Xe adsorbed on graphite using the interaction parameters given in Table I and the coverage values of 0.1, 0.3 and 0.5.


0.1
0.1
0.1
10.0
C [k_B]
0.3
0.3
0.3
0.5
0.5
0.5
Ne
Ar
Xe
0.0
50
100
T[K]

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