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ANALYSIS OF MONTE CARLO DATA FOR A MODEL SYSTEM OF ADSORBED DIATOMIC MOLECULES

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1. Introduction

The main object of study of classical statistical mechanics are N-particle systems confined to a finite d-dimensional space domain $\Lambda \subset \mathbb{R}^d$ of volume $V = |\Lambda|$. The mechanical state is described by a set x of canonical variables (momenta and space coordinates of all the particles) and the system is specified by a Hamiltonian $\mathcal{H}_{N,\Lambda}(x)$, defined on the multi-dimensional space $\mathcal{S}_{N,\Lambda}$ of all the allowed values of x. In addition, boundary conditions have to be given to specify the walls $\partial\Lambda$ of the domain Λ and the interaction of the particles with the environment. According to the fundamental postulates of statistical mechanics, if the system is in thermal equilibrium at a fixed absolute temperature T, any extensive macroscopic observable P, associated with a dynamical function $P_{N,\Lambda}(x)$ defined on $\mathcal{S}_{N,\Lambda}$, is given by the canonical average value

$$P_{N,\Lambda}(T) = [Z_{N,\Lambda}(T)]^{-1} \int_{\mathcal{S}_{N,\Lambda}} dx\, P_{N,\Lambda}(x) \exp[-\mathcal{H}_{N,\Lambda}(x)/kT], \tag{1}$$

where k is Boltzmann constant and

$$Z_{N,\Lambda}(T) = \int_{\mathcal{S}_{N,\Lambda}} dx \exp[-\mathcal{H}_{N,\Lambda}(x)/kT] \tag{2}$$

is partition function of the system. In cases when some variables in the set x take discrete values, the corresponding integration symbol in (1) and (2) should be understood as summation. Since the momenta of the particles play a rather trivial role in the classical equilibrium statistical mechanics, one may ignore them and focus the attention on the purely configurational counterparts of equations (1) and (2). A further simplification often used is the discretization of the spatial coordinates of the particles, thus reducing the problem to a lattice one in which the particles occupy the sites of a finite d-dimensional lattice $\mathcal{L} \subset \mathbb{Z}^d$.

Under quite general conditions on the interparticle interactions, observables (1) are analytical functions of temperature and external fields eventually included in $\mathcal{H}_{N,\Lambda}(x)$. Nonanalytical behaviour, however, may occur in the thermodynamic limit (to be denoted by $t - \lim$, which means $N \to \infty$ and $V \to \infty$ at fixed density of the particles $\rho = N/V$) for the intensive observables:

$$t - \lim V^{-1} P_{N,\Lambda}(T) = p(\rho, T). \tag{3}$$

According to the contemporary views, see e.g. [1], every domain in the space of the external thermodynamic parameters in which the limiting intensive observables (3) depend analytically on these parameters corresponds to a macroscopic phase (gaseous, liquid, solid, etc.). The points (lines, surfaces) of nonanalyticity describe phase transitions. The most interesting (and the most difficult) task is the location and description of all the possible singularities for a given model. Unfortunately, there are just a few very simple lattice models which admit an analytical evaluation of the asymptotic behaviour of observables (1) and, hence, an explicit performance of the limit procedure prescribed by the left-hand side of equation (3), see e.g. [2].

About twenty years ago, Fisher [3] suggested that finite systems consisting of many interacting particles obey some universal laws of finite-size scaling near a second order phase transition point. In this case there exists an intrinsic scale of distances determined by the bulk particle-particle correlation length ξ_∞, which diverges at the critical point. According to [3], the variable, determining the finite-size scaling for systems with linear size L, is the ratio L/ξ_∞. For example, let Λ be a hypercube with edge L and let $p_L(K)$ denote some intensive macroscopic observable of the finite system, which in the limit $L \to \infty$ has a singular behaviour of the type:

$$p_\infty(K) \sim |t|^{-\rho}, \quad t = 1 - K/K_c, \tag{4}$$

as $K \to K_c$. Here K is a dimensionless interaction parameter, $\rho > 0$ is critical exponent of quantity p. Then, for finite but large L and K near K_c, the finite-size scaling hypothesis reads [4]

$$p_L(K) \simeq L^{\rho/\nu} Q_p(L/\xi_\infty(K)), \tag{5}$$

where $Q_p(.)$ is a universal function with asymptotic behaviour

$$Q_p(x) \sim x^{-\rho/\nu}, \quad x \to \infty, \tag{6}$$

and ν is the critical exponent for correlation length $\xi_\infty(K)$. Actually, it is more generally valid to write [4].

$$p_L(K) \simeq L^{\rho/\nu} X_p(\tilde{t} L^{1/\nu}), \tag{7}$$

where

$$\tilde{t} = t + (K_{c,L} - K_c)/K_c, \tag{8}$$

and

$$X_p(x) \sim |x|^{-\rho}, \quad x \to \infty. \tag{9}$$

In (8) a shifted critical parameter $K_{c,L}$ has been introduced, which is expected to behave for large L as

$$K_{c,L} \simeq K_c[1 + bL^{-\lambda}], \tag{10}$$

with a shift exponent $\lambda > 0$ and some amplitude b. In the orthodox formulation of the finite-size scaling one accepts $\lambda = 1/\nu$, but generally λ is allowed to be a free parameter [4].

Finite-size scaling gives the possibility to extrapolate the properties of not very large systems to the corresponding properties in the thermodynamic limit, i.e. when $L/\xi_\infty \to \infty$. In case when the model does not have an exact solution, finite-size scaling is the most reliable method for evaluation of the critical exponents, characterizing the corresponding mathematical singularity.

The purpose of the present paper is to present a simple mechanical model of a monolayer of diatomic molecules adsorbed on a solid surface and to study the data, obtained by an appropriate Monte Carlo method in the framework of the existing finite-size scaling hypotheses. This work differs from the previous papers on the subject [5]-[7] in several aspects. First of all, the set of macroscopic observables evaluated by the Monte Carlo technique is enlarged by including an order parameter and the susceptibility associated with it. Second, we impose boundary conditions which favour one of the possible orderings of the system at low temperatures. Finally, we consider the conformity of the numerical data to a new type of critical singularity.

The model and the corresponding stochastic Monte Carlo dynamics are described in Chapter 2 and Chapter 3, respectively. The algorithm used for the numerical simulations is given in Chapter 4. The systematic errors are discussed in Chapter 5.

Preliminary results for the specific heat of lattices 8×8, 12×12 and 16×16 in the light of the three hypothesis of finite-size scaling are discussed in Chapter 6.

2. The model

The dimer problem is a classical problem of the lattice statistics [8]. It may be interpreted as a model of a system of diatomic molecules (dimers), adsorbed on a crystal surface, so that each molecule (dimer) occupies two neighbouring sites of a regular lattice. Due to the short-range repulsive forces, no site is occupied by more than one molecule. The dimer problem, with added interaction potential between the dimers, cannot be solved analytically. That is why Authors' study of such a model is performed by a Monte Carlo method, based on a special algorithm first proposed in [5].

Let $\mathcal{L}$ be a square lattice $L \times L$, close-packed with dimers, see Fig. 1. To complete the definition of the model different boundary conditions may be chosen. Here we have used fixed boundary conditions on two additional rows – the 0-th and $(L+1)$-th. Both of these rows are filled with horizontal dimers, interacting with the ones that cover the lattice. The new dimers are called "boundary", the other ones – "internal" dimers. The vertical boundaries of the system (1-st and L-th row of the lattice) are determined by free boundary conditions. A pair of nearest neighbour parallel dimers (horizontal or vertical) is called internal when both its dimers are internal. If one of the dimers is a boundary one, the pair is boundary too.

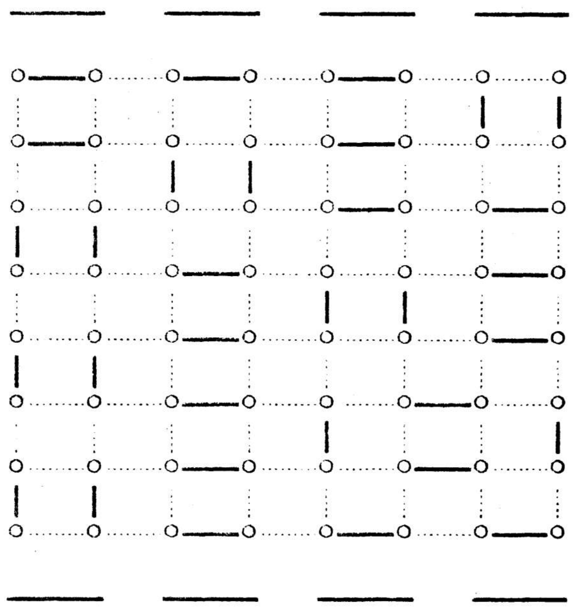

Fig. 1. An example of close-packed lattice 8×8 with fixed boundary conditions.

Let C be a given close-packed dimer configuration over $\mathcal{L}$, $N(C)$ be number of internal and $N_t(C)$ – number of all (boundary and internal) pairs of dimers in C. We consider a simple model in which the interaction energy between the molecules depends on $N_t(C)$ as follows:

$$E(C) = -\epsilon N_t(C), \tag{11}$$

where $\epsilon > 0$ is a parameter.

The canonical Gibbs distribution corresponding to the so defined energy is:

$$\Pi_i^{eq}(K) = \exp[K N_t(C_i)]/Z(K), \tag{12}$$

where $K = \epsilon/kT$, and $Z(K)$ is partition function:

$$Z(K) = \sum_k \exp[K N(C_k)]. \tag{13}$$

Here summation over all possible configurations of the system is carried out.

The Monte Carlo method is based on the generation of such a stochastic chain of configurations $\{C_i\}_{i=1,2,\ldots}$, that allows the canonical average values of every function $O(C)$, defined on the configuration space of the system,

$$O_L(K) = \sum_i \Pi_i^{eq}(K) O(C_i), \tag{14}$$

to be evaluated by arithmetical average values over the stochastic chain $\{C_i\}$

$$O_L^{MC}(K) = N_{MC}^{-1} \sum_{i=1}^{N_{MC}} O(C_i) \to O_L(K), \quad \text{when} \quad N_{MC} \to \infty. \tag{15}$$

3. Stochastic Monte Carlo dynamics

The generation of a stochastic chain of configurations is equivalent to the definition of a discrete stochastic dynamics of the system. If the sequence $\{C_i\}_{i=1,2,\ldots}$ is an irreducible and aperiodic Markov chain, the canonical Gibbs probability distribution is ensured when the corresponding transition probabilities p_{ij} satisfy the detailed balance equation:

$$\Pi_i^{eq}(K) p_{ij} = \Pi_j^{eq}(K) p_{ji}. \tag{16}$$

The algorithm, proposed in [5], is based on the fact that all the possible configurations can be reached from arbitrary initial configuration by a finite sequence of rotations of randomly chosen internal pairs of dimers. Two configurations C_i and C_j are called nearest neighbours, if one of them can be obtained from the other by rotating only one internal pair of dimers. To rotate a pair means, for example, that a pair of horizontal dimers becomes a pair of vertical dimers. The number of nearest neighbours of C_i is obviously equal to the number of internal pairs in C_i. As it was shown in [5], one has a uniform probability distribution when transition probabilities $\tilde{p}_{ij}$ are defined as follows:

$$\tilde{p}_{ij} = \begin{cases} 1/N(C_i) & \text{–if } C_i \text{ and } C_j \text{ are nearest neighbours} \\ 0 & \text{–otherwise.} \end{cases} \tag{17}$$

A solution of the detailed balance equation was found in [5] for the case of completely free boundary conditions. In the case of fixed horizontal boundary conditions considered, a solution of eq. (16) is reported:

$$p_{ij} = \begin{cases} \tilde{p}_{ij} \exp\left\{K\left[N_t(C_j) - N_t(C_i)\right]\right\}, & \text{if } N(C_i) > N(C_j) \text{ or } N(C_i) = N(C_j) \\ & \text{and } N_t(C_i) \geq N_t(C_j) \\ \tilde{p}_{ij} N(C_i)/N(C_j), & \text{if } N(C_i) < N(C_j) \text{ or } N(C_i) = N(C_j) \\ & \text{and } N_t(C_i) < N_t(C_j) \end{cases} \tag{18}$$

One can easily be convinced that these transition probabilities do solve the detailed balance equation. Let C_i and C_j are nearest neighbours, and, for example, $N(C_i) = N(C_j)$ and $N_t(C_i) \geq N_t(C_j)$. Then:

$$\begin{aligned} \Pi_i^{eq} p_{ij} &= \exp[K N_t(C_i)]/Z(K)\tilde{p}_{ij} \exp\{K[N_t(C_j) - N_t(C_i)]\} = \\ &= \exp[K N_t(C_j)]/Z(K)\tilde{p}_{ij} = \exp[K N_t(C_j)]/[Z(K)N(C_i)] = \\ &= \exp[K N_t(C_j)]N(C_j)/[Z(K)N(C_i)N(C_j)] = \\ &= \Pi_j^{eq}\tilde{p}_{ji}N(C_j)/N(C_i) = \Pi_j^{eq} p_{ji}. \end{aligned} \tag{19}$$

4. The Monte Carlo algorithm

1. *Generation of initial configuration.* The lattice is close-packed with horizontal dimers. A generator for pseudo-random numbers is used to choose a pair of dimers to be rotated. The initial configuration is obtained after 10^4 rotations.

2. *Configuration representation.* The position and the type (horizontal or vertical) of every dimer are written in an array $A(n, m)$. To a horizontal dimer with left end at the site (k_1, k_2) of the lattice we ascribe the values 1 and -1 in the (k_1, k_2)-th and $(k_1, k_2 + 1)$-th elements, respectively, of the array A. To a vertical dimer with

upper end at site (k_1, k_2) of the lattice correspond values 2 and -2 in the (k_1, k_2)-th and $(k_1 + 1, k_2)$-th elements of A.

A second array $B(n, m)$ is created to count the number of pairs in the dimer system. The value 1 of the (k_1, k_2)-th element of B corresponds to a horizontal pair with left end of the upper dimer at site (k_1, k_2) of the lattice; by analogy, the value 2 corresponds to a vertical pair with the same position in the lattice. For every configuration $C_i, N(C_i)$ and $N_t(C_i)$ are calculated. All pairs in C_i are numbered successively.

3. A random Monte Carlo step per site (RMCS) consists of L_{MC} attempted rotations with $L_{MC} = L^2 + r$, where r is 0 or 1 with equal probability. (There are configurations, which cannot be obtained from the initial one through an even number of rotations.)

4. The rotation of a pair of dimers is done according to the transition probabilities (18). A random number between 1 and $N(C_i)$ is generated and the chosen pair is rotated if the corresponding Metropolis criterion is satisfied. To rotate a pair with left upper end at site (k_1, k_2) of the lattice means to change the corresponding values in arrays A and B. If a rotation is accepted, arrays A and B are updated and $N(C_i), N_t(C_i)$ are calculated for the new configuration.

5. When a certain number, N_{MC}, of random Monte Carlo steps is realized, the specific heat $c_L(K)$ per dimer is calculated by:

$$c_L(K) = 2K^2L^{-2}\left\{\frac{1}{N_{MC}}\sum_{i=1}^{N_{MC}} N^2(C_i) - \left[\frac{1}{N_{MC}}\sum_{i=1}^{N_{MC}} N(C_i)\right]^2\right\} \tag{20}$$

Let N_{hod} be the number of horizontal dimers with left ends lying in an odd column, N_{vod} – the number of the vertical dimers with upper ends in an odd row. We define here an order parameter $P_L(K)$ as:

$$P_L(K) = \sum_i \Pi_i^{eq}(K)P(C_i), \tag{21}$$

where

$$P(C) - 2L^{-2}[N_{hod}(C) - N_{vod}(C)]. \tag{22}$$

After some N_{MC} of RMCS the variance of the quantity $P(C)$, which has the meaning of a susceptibility, is calculated:

$$\chi_L(K) = 2L^{-2}\left\{N_{NC}^{-1}\sum_{i=1}^{N_{MC}} P^2(C_i) - \left[N_{MC}^{-1}\sum_{i=1}^{N_{MC}} P(C_i)\right]^2\right\}. \tag{23}$$

5. Error Analysis

The values of c_L and χ_L, obtained by Monte–Carlo sampling, depend not only on the lattice size L and the temperature K, but on the number of RMCS, too. There are primarily two reasons for systematic errors in Monte–Carlo sampling: due to the finite size of lattices studied and due to the finite number of simulations made [8]. In this paper we use the method of evaluating the systematic error suggested in [11]. Values, necessary to calculate c_L^M and χ_L^M, are recorded in a file on every 10 MCS. Let Monte Carlo sampling have M MCS. This sampling is divided into parts of length n MCS each. For every subsampling estimates c_L^n and χ_L^n for c_L and χ_L, respectively, are obtained and then their average values and variances are calculated for different values of n in the interval [20, 20000], see Fig. 2.

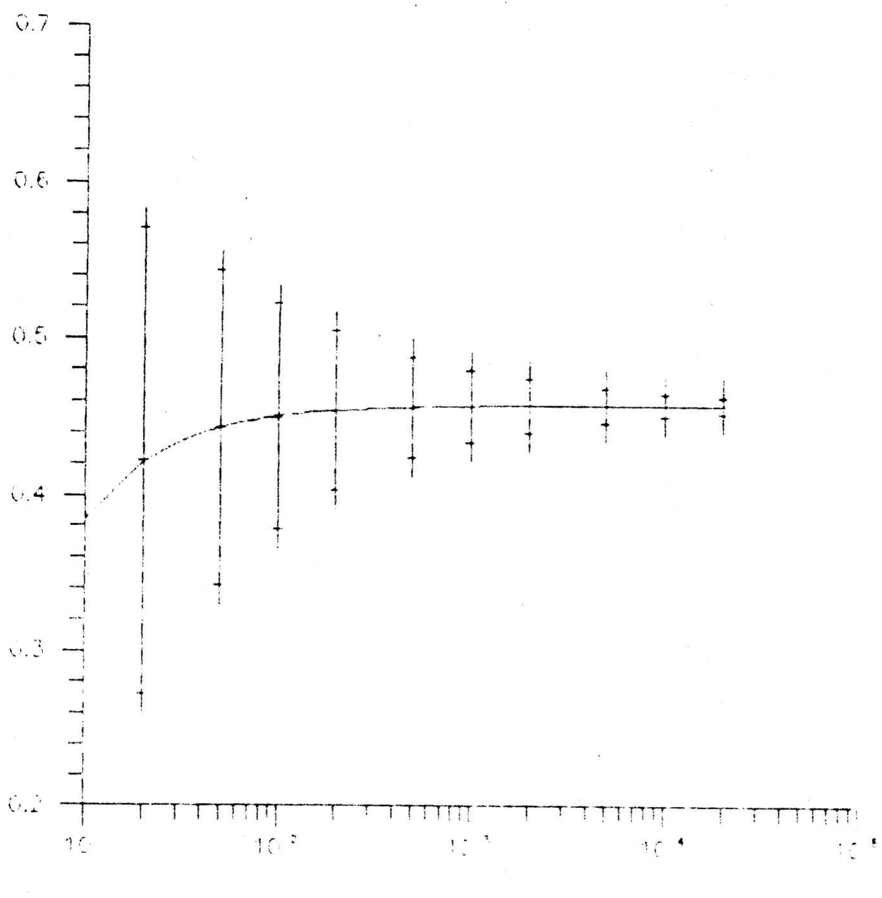

Fig. 2. Estimates of the specific heat and its statistical error as a function of the bins lenght for a lattice 8x8, $K = 1.6$

Then extrapolations to $n = \infty$, namely c_L^∞ and χ_L^∞, are made using the expression:

$$c_L^n = c_L^\infty[1 - (2a_0 + 1)/n], \tag{24}$$

and similarly for χ_L.

6. Numerical results and finite-size scaling

The specific heat is given as a function of inverse temperature K for lattices 8×8, 12×12 and 16×16 (Fig. 3). As one can see, the peak of specific heat narrows, the height of its maximum grows and K_L^{max} moves to lower temperatures (larger K values) with the increase of L. To obtain estimates of K_L^{max} and c_L^{max} we use a parabolic approximation of the numerical results in the neighbourhood of the maximum. Fig. 3 shows how the results conform with this idea.

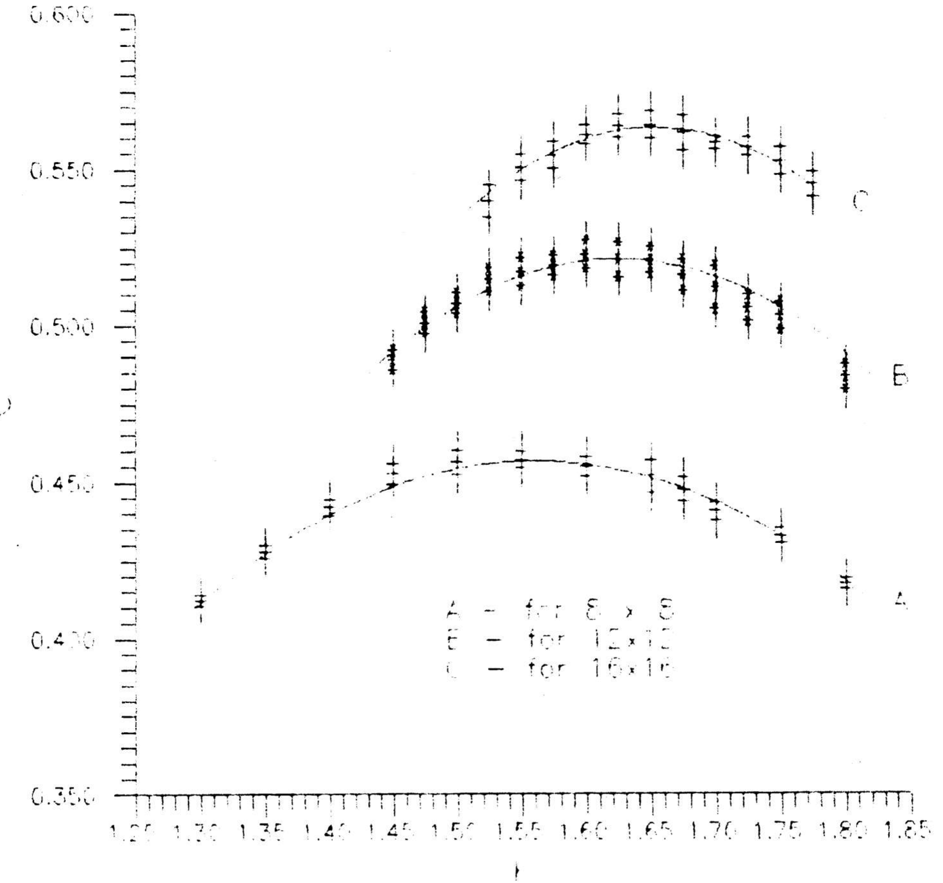

Fig. 3

Trying to explain the observed behaviour by the existing finite–size scaling theory, we must point out that in this case the type of specific heat singularity in the infinite system limit, $c_\infty(K)$, is unknown. That is why, we investigate here three main possibilities: power law, logarithmic and higher order singularities. If $c_\infty(K)$ has an algebraic singularity $|K - K_c|^{-\alpha}, \alpha > 0$, then according to the finite–size scaling hypothesis for two–dimensional systems with free boundaries, the asymptotic form of $c_L(K)$ for large enough L, is [12]:

$$C_L(K = L^{\alpha/\nu} X_\alpha(\tilde{t}L^{1/\nu}) + \sum_{k=0}^{2} L^{-k}\psi_k(t) + O(L^{-2}), \tag{25}$$

where α and ν are the critical exponents of specific heat and correlation length, respectively; $\psi_k(t)$, $k = 0, 1, 2$ are some regular at $t = 0$ functions. If (25) is to reproduce the thermodynamic behaviour in the limit $L \to \infty$, $t \neq 0$, the universal scaling function $X_\alpha(\tau)$ must have the following asymptotic form as $\tau \to \infty$:

$$X_\alpha(\tau) = Y_2|\tau|^{-\alpha} + Y_1|\tau|^{-(\alpha+\nu)} + Y_0\tau^{-2} + \cdots,$$

where Y_0, Y_1, Y_2 are universal coefficients.

It follows from (25) that the height of the maximum, $c_L(K_L^{max})$, has the following asymptotic behaviour as $L \to \infty$:

$$c_L(K_L^{max}) = L^{\alpha/\nu} X_\alpha(0) + \mathcal{O}(1) \tag{26}$$

By differentiating (25) twice with respect to K and setting $\tilde{t} = 0$, for the curvature of the parabolic approximation we obtain

$$c''_L(K_L^{max})/2 = K_c^{-2} X''(0) L^{(2+\alpha)/\nu} + \mathcal{O}(1). \tag{27}$$

In the case of logarithmic singularity we assume the following [13]:

$$c_L(K) = 2q_0 \ln(L^{1/\nu}) + X_0(tL^{1/\nu}) + \sum_{k=0}^{2} L^{-k}\psi_k(t) + \mathcal{O}(L^{-2}), \tag{28}$$

where $q_0 > 0$ is a universal constant and scaling function $X_0(\tau)$ has asymptotic form as $\tau \to \pm\infty$:

$$X_0(\tau) \cong -2q_0 \ln|\tau| + C_\pm,$$

where $C_\pm$ are universal amplitudes.

Now the height of the maximum, $c_L(K_L^{max})$, has the following asymptotic behaviour as $L \to \infty$:

$$c_L(K_L^{max}) = (2q_0/\nu)\ln L + \mathcal{O}(1) \tag{29}$$

and the curvature of the parabolic approximation is

$$c''_L(K_L^{max})/2 = K_c^{-2}X''(0)L^{2/\nu} + \mathcal{O}(1). \tag{30}$$

The third finite–size scaling hypothesis treats the case, when $c_\infty(K_c)$ is finite, but $c_\infty(K)$ as a function of K has a cusp at $K = K_c$. In this case the finite–size scaling hypothesis has to be formulated for the first derivative of $c_L(K)$, which in the limit $L \to \infty$ becomes singular at $K = K_c$:

$$\frac{\partial}{\partial K} c_\infty(K) \to A_\pm, \; t \to 0_\pm, \tag{31}$$

Thus we obtain

$$c_L(K) \cong c_\infty(K_c) + L^{-1/\nu}X(\tilde{t}L^{1/\nu}), \tag{32}$$

where $X(\tau)$ has an asymptotic form as $\tau \to \pm\infty$:

$$X(x) \cong A_\pm \tau + B_\pm. \tag{33}$$

From (32) it follows that

$$c_L(K^{max}) \cong c_\infty(K_c) + L^{-1/\nu}X(0), \tag{34}$$

and

$$c''_L(K_L^{max})/2 \cong K_c^{-2}X''(0)L^{1/\nu}. \tag{35}$$

Similarly these three possibilities may apply for the susceptibility χ_L and its critical exponent γ.

The comparison of the above consequences of the finite–size scaling hypotheses with the available numerical data from Monte Carlo simulations could allow us, in principle, to assess the type of singularity which occurs in the thermodynamic limit.

Table 1.
Comparison of Monte Carlo data for $C_L(K_L^{max})$, $L = 8, 12, 16$, with the best approximations according to the three different hypotheses, see Eqs. (26), (29) and (34), respectively.

L	$C_L(K_L^{\max})$	I	II	III
8	0.4577	0.4581	0.4589	0.4575
12	0.5219	0.5176	0.5197	0.5270
16	0.5633	0.5644	0.5629	0.5612

Table 2.
Comparison of Monte Carlo data for $C''_L(K_L^{max})$, $L = 8, 12, 16$, with the best approximations by a power low, see Eqs. (27), (30) and (35).

L	$C''_L(K_L^{\max})$	I - III
8	-1.3688	-1.3440
12	-2.2652	-2.0572
16	-2.5298	-2.7826

From the above results we obtain the following predictions for the critical exponents α and ν:

Hypothesis	Predicted values of α and ν	
I.	$\left.\begin{array}{l}\alpha/\nu \cong 0.3 \\ (2+\alpha)/\nu \cong 1.05\end{array}\right\}$	$\Longrightarrow \nu \cong 2.67, \quad \alpha \cong 0.8.$
II.	$2/\nu \cong 1.05$	$\Longrightarrow \nu \cong 2.1, \quad \alpha = 0$
III.	$1/\nu \cong 1.05$	$\Longrightarrow \nu \cong 0.952, \quad \alpha = -1.$

According to the hyperscaling relation [3, 4], one should have

$$d\nu = 2 - \alpha. \tag{36}$$

Obviously, in our case $d = 2$, and relation (36) is violated for all the three hypotheses investigated. However, the smallest discrepancy is observed under hypothesis III, when $d\nu \cong 1.9$ and $2 - \alpha = 3$.

In conclusion we shall note the following. Obviously, the ideas of finite-size scaling are indispensable in the anlysis of Monte Carlo data for statistical mechanical models which describe a phase transition in the limit of an infinite number of degrees of freedom. However, in order to discriminate between the different possible hypotheses about the type of the critical singularity, one needs Monte Carlo data for a rather large series of lattice sizes. The results, available so far, for our model of adsorbed diatomic molecules are insufficient for definite conclusions to be drawn. The analysis presented here provides the guidelines for further Monte Carlo simulations. Our tentative gradation of the three hypotheses considered, in order of decreasing likelihood is: III, II, I.

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