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First-order phase transitions in repulsive rigid *k*-mers on two-dimensional lattices

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In a previous paper [F. Romá, A. J. Ramirez-Pastor, and J. L. Riccardo, Phys. Rev. B **72**, 035444 (2005)], the critical behavior of repulsive rigid rods of length k (k-mers) on a square lattice at half coverage has been studied by using Monte Carlo (MC) simulations. The obtained results indicated that (1) the phase transition occurring in the system is a second-order phase transition for all adsorbate sizes k; and (2) the universality class of the transition changes from 2D Ising-type for monomers ($k = 1$) to an unknown universality class for $k \geq 2$. In the present work, we revisit our previous results together with further numerical evidences, resulting from new extensive MC simulations based on an efficient exchange algorithm and using high-performance *computational capabilities*. In contrast to our previous conclusions (1) and (2), the new numerical calculations clearly support the occurrence of a first-order phase transition for $k \geq 2$. In addition, a similar scenario was found for k-mers adsorbed on the triangular lattice at coverage $k/(2k+1)$. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3678312]

I. INTRODUCTION

Lattice-gas models have been extensively investigated in the last decades because they provide a theoretical framework for the description of many physical, chemical, and biological systems. The adsorption thermodynamics and the understanding of surface phenomena have been greatly benefited from the development of these models.[1–3] Particularly, the two-dimensional (2D) lattice-gas model with repulsive interactions between the adparticles has received considerable theoretical and experimental interest because it provides the theoretical framework for studies of surface phase transitions occurring in many adsorbed monolayer films.[4–12] Most studies have been devoted to adsorption of monoatomic particles. However, if some sort of correlation exists, like particles that occupy several k contiguous lattice sites (k-mers), the statistical problem becomes exceedingly difficult.[13–15] The model of a 2D gas of rigid k-mers is the simplest representation of a strongly adsorbed film of linear molecules in submonolayer or monolayer regime. Examples of this kind of systems are monolayer films of n-alkanes adsorbed on monocrystalline surfaces of metals, such as Pt(111)[16] and Au(111).[17, 18]

From a theoretical point of view, several attempts were made in the past in order to solve the k-mers problem.[13–15, 19–26] However, a rigorous description of equilibrium and dynamic properties of polyatomic species adsorbed on 2D substrates still represents a major challenge in surface science. The inherent difficulty common to processes involving the adsorption of k-mers is to calculate the configurational (entropic) contribution to the thermodynamic potentials properly, which means the degeneracy of the energy spectrum compatible with a given number of particles

and adsorption sites. One way of overcoming these theoretical complications is to use Monte Carlo (MC) simulation method.[27–32] MC technique is a valuable tool for studying surface molecular processes, which has been extensively used to simulate many surface phenomena including adsorption, diffusion, reactions, phase transitions, etc. Here we will try to demonstrate that numerical simulations, combined with a correct theoretical interpretation of the results, can be very useful to obtain a very reasonable description of the critical behavior of polyatomics on 2D surfaces.

By using MC simulations, multiple-histogram reweighting and finite size-scaling techniques, Rżysko and Borówko[33–40] have studied a wide variety of systems in presence of multisite occupancy. Among them, attracting dimers in the presence of energetic heterogeneity,[33] heteronuclear dimers consisting of different segments, A and B, adsorbed on square lattices,[34–38] and trimers with different structures adsorbed on square lattices.[34, 40] In these papers, a rich variety of phase transitions was reported along with a detailed discussion about critical exponents and universality class. The authors found that the majority of systems belong to the 2D Ising class of universality. However, in the case of heteronuclear dimers with repulsive A-A coupling and attractive interactions of B-B and A-B types the fluid exhibits a nonuniversal behavior.[35]

With respect to repulsive couplings, the structural ordering of interacting dimers on a square lattice has been studied by Romá *et al.*[41] By using MC simulation, the authors concluded that there are a finite number of ordered structures for dimers with repulsive nearest-neighbor interaction. The thermodynamic implication of such structural ordering was demonstrated through the analysis of the adsorption isotherm and the collective diffusion coefficient of dimers with nearest-neighbor repulsion.[42] It was shown that two well-defined

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steps appear in the adsorption isotherms; they correspond to a (4×2) ordered phase at $\theta = 1/2$ and a zig-zag order at $\theta = 2/3$, being θ the surface coverage. The analysis of the phase diagram for repulsive nearest-neighbor interactions on a square lattice,[43] as well as the behavior of the entropy with coverage,[41] confirmed the presence of these two well-defined structures.

In a later work,[44] from the (4×2) phase appearing in dimers at half coverage, it was possible (1) to predict the existence of a ($2k \times 2$) structure for k-mers at half coverage and (2) to obtain the critical temperature $T_c(k)$ characterizing the transition from the disordered state to the ($2k \times 2$) phase as a function of the size k of the adsorbed molecules. Ref. 45 was a step further, analyzing the universality class of the phase transition. By using MC simulations and finite-size scaling analysis, the complete set of static critical exponents (α, β, γ, and ν) was determined for adsorbate sizes ranging between $k = 2$ and 4. The obtained values of the critical exponents revealed that the universality class of the transition changes from 2D Ising-type for monomers to an unknown universality class (according to the current classification of order-disorder transitions on surfaces given by Schick[7–9]) for $k \geq 2$.

Further studies,[46,47] motivated by this apparent change of universality, led us to revise our previous conclusion of a second order behavior with exponents of an unknown universality class in favor of an interpretation in terms of a first-order phase transition. The *primary objective* of this paper is to provide a detailed description *of this new scenario*. In order to do so, extensive MC simulations, based on an efficient exchange algorithm[48–50] and using high-performance *computational capabilities*,[51] have been carried out. All the quantities measured in Ref. 45 were recalculated. In addition, the study was complemented with new measurements such as energy cumulant, energy distribution and order parameter cumulant over a wide range of temperatures. The obtained results indicated that (1) the nature of the phase transition occurring at half coverage in a system of repulsive rigid k-mers on a square lattice changes from second order for $k = 1$ to first order for $k \geq 2$; and (2) the critical exponents calculated in Ref. 45 for $k \geq 2$ should be reinterpreted in terms of effective exponents.[52] Finally, *we have also reviewed our* past work that explored the critical behavior of repulsive linear k-mers on the triangular lattice at coverage $k/(2k+1)$.[53] As in the square lattice case, we found a scenario with a change from second-order (for $k = 1$) to first-order phase transition when $k \geq 2$.

The paper is organized as follows. In Sec. II, we describe the lattice-gas model and the simulation scheme is presented in Sec. III. Finally, the results and the general conclusions are given Sec. IV.

II. LATTICE-GAS MODEL

We address the general case of adsorption of homonuclear k-mers on 2D surfaces. The adsorbate molecules are assumed to be linear rigid particles containing k identical units, with each one occupying a lattice site. Small adsorbates would correspond to the monomer limit ($k = 1$). The distance between k-mer units is assumed to be equal to the lattice constant; hence, exactly k sites are occupied by a k-mer when

adsorbed. The surface is represented as an array of $M = L \times L$ adsorptive sites in a square lattice arrangement, where L denotes the linear size of the array. In order to describe the system of N k-mers adsorbed on M sites at a given temperature T, let us introduce the occupation variable c_i which can take the values $c_i = 0$ if the corresponding site is empty and $c_i = 1$ if the site is occupied. On the other hand, molecules adsorb or desorb as one unit, neglecting any possible dissociation. Under these considerations, the Hamiltonian of the system is given by

$$H = w \sum_{\langle i,j \rangle} c_i c_j - N(k-1)w + \varepsilon_0 \sum_i c_i, \qquad (1)$$

where w is the nearest-neighbor (NN) interaction constant which is assumed to be repulsive (positive), $\langle i, j \rangle$ represents pairs of NN sites, and ε_0 is the energy of adsorption of one given surface site. The term $N(k-1)w$ is subtracted in Eq. (1) since the summation over all the pairs of NN sites overestimates the total energy by including $N(k-1)$ bonds belonging to the N adsorbed k-mers. Because the surface was assumed to be homogeneous, the interaction energy between the adsorbed k-mers and the atoms of the substrate ε_0 was neglected for the sake of simplicity.

In order to characterize the phase transition on the square lattice, we use the order parameter defined in Ref. 45, which can be written as

$$\delta = \left| \frac{N_v - N_h}{N} \right|, \qquad (2)$$

where N_v (N_h) represents the number of k-mers aligned along the vertical (horizontal) axis and $N = N_v + N_h$. When the system is disordered ($T > T_c$), the two orientations (vertical or horizontal) are equivalent and δ is zero. As the temperature is decreased below T_c, the k-mers align along one direction and δ is different from zero. Thus, δ appears as a proper and computationally convenient order parameter to elucidate the phase transition.

III. MONTE CARLO SIMULATION

The lattices were generated fulfilling the following conditions: (1) the sites were arranged in a square lattice of side L, with conventional periodic boundary conditions, (2) because the surface was assumed to be homogeneous, the interaction energy between the adsorbed k-mers and the atoms of the substrate ε_0 was neglected for the sake of simplicity, (3) in order to maintain the lattice at half coverage, $\theta = kN/M = 1/2$, the number of k-mers on the lattice was fixed as $N = M/2k$, and (4) for each value of k, appropriate values of L were used in such a way that the ($2k \times 2$) ordered structure is not altered by boundary conditions (see Fig. 1).

In order to study the critical behavior of the system, an efficient exchange MC method[48–50] has been used. As in Ref. 48, a compound system of m noninteracting replicas of the system concerned has been built. The ith replica is associated with a heat bath at temperature T_i (or $\beta_i = 1/k_B T_i$, where k_B is the Boltzmann constant). To determine the set of temperatures $\{T_i\}$, the highest temperature T_1 is set in the high-temperature phase where relaxation (correlation) time is
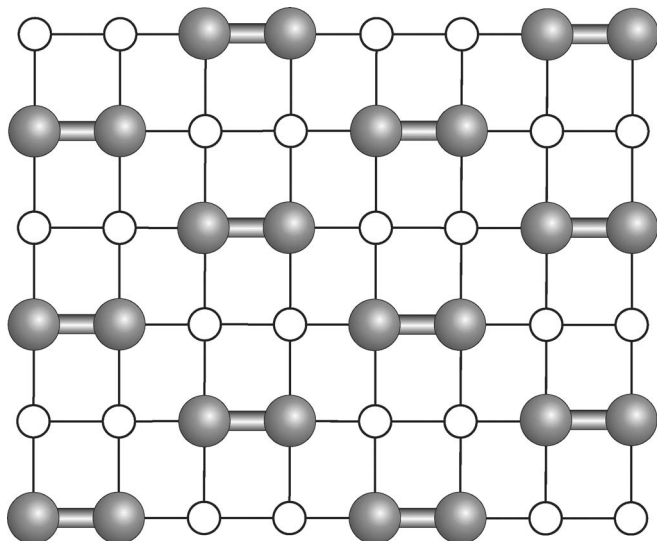
FIG. 1. Snapshot of the ordered phase for dimers at half coverage. Solid circles (joined by thick lines) and open circles represent dimers and empty sites, respectively.

expected to be very short and there exists only one minimum in the free energy space. On the other hand, the lowest temperature T_m is set in the low-temperature phase whose properties we are interested in. Finally, the difference between two consecutive temperatures, T_i and T_{i+1} with $T_i > T_{i+1}$, is set as $\Delta T = (T_1 - T_m)/(m-1)$ (equally spaced temperatures).

Under these conditions, the algorithm to carry out the simulation process is built on the basis of two major subroutines:

(i)   Replica update. Interchange vacancy particle and diffusional relaxation. The procedure, which is repeated M times, is as follows. (a) One out of the m replicas is randomly selected (for example, the ith replica). (b) A k-mer and a k-upla of adjacent empty sites, both belonging to the replica chosen in (a), are randomly selected and their coordinates are established. Then, an attempt is made to interchange its occupancy state with probability given by the Metropolis rule,[54]

$$P = \min\left\{1, \exp(-\beta_i \Delta H)\right\}, \qquad (3)$$

where ΔH is the difference between the Hamiltonians of the final and initial states. (c) A k-mer is randomly selected. Then, a displacement is attempted (following the Metropolis scheme) by either jumps along the k-mer axis or reptation through a 90° rotation of the k-mer axis, where one of the k-mer centers remains in its position (interested readers are referred to Fig. 1 of Ref. 55 for a more complete description of the reptation mechanism). This procedure (diffusional relaxation) must be allowed in order to reach equilibrium in a reasonable time.

(ii) Exchange. Exchange of the two configurations $\mathbf{X}_i$ and $\mathbf{X}_{i'}$, corresponding to the randomly chosen ith and i'th replicas, respectively, is tried and accepted with probability $W(\mathbf{X}_i, \beta_i | \mathbf{X}_{i'}, \beta_{i'})$. In general, the probability of exchanging configurations of the ith and i'th replicas is

given by[48]

$$W(\mathbf{X}_i, \beta_i | \mathbf{X}_{i'}, \beta_{i'}) = \begin{cases} 1, & \text{for}\, \Delta \leq 0 \\ \exp(-\Delta), & \text{for}\, \Delta > 0 \end{cases}, \qquad (4)$$

where $\Delta = (\beta_i - \beta_{i'})[H(\mathbf{X}_{i'}) - H(\mathbf{X}_i)]$. As in Ref. 48, we restrict the replica exchange to the case $i' = i + 1$.

The elementary step of the simulation process, or Monte Carlo step (MCS), consists of the following: (i) replica update, (ii) exchange, and (iii) repeat from step (i) m times. The complete simulation procedure is the following: (1) initialization, (2) elementary step, and (3) repeat from step (2), $n_2 + n_{MCS}$ times. The initialization of the compound system of m replicas, step (1), is as follows. By starting with a random initial condition, the configuration of replica 1 is obtained after n_1 MCS* at T_1 (one MCS* consists of M realizations of the replica update subroutine). Second, for $i = \{2, \ldots, m\}$, the configuration of the ith replica is obtained after n_1 MCS*s at T_i, taking as initial condition the configuration of the replica to T_{i-1}. This method results more efficient than a random initialization of each replica. Procedures (1)–(3) are repeated for all lattice sizes. For each lattice, the equilibrium state can be well reproduced after discarding the first n_2 MCSs. Then, averages are taken over n_{MCS} successive MCSs. As was mentioned above, a set of equally spaced temperatures is chosen in order to accurately calculate the physical observables in the close vicinity of T_c.

The thermal average $\langle \ldots \rangle$ of a physical quantity A is obtained through simple averages,

$$\langle A \rangle = \frac{1}{n_{MCS}} \sum_{t=1}^{n_{MCS}} A[\mathbf{X}_i(t)]. \qquad (5)$$

In the last equation, $\mathbf{X}_i$ stands for the state of the ith replica (at temperature T). Thus, the specific heat C (in k_B units) is sampled from energy fluctuations,

$$\frac{C}{k_B} = \frac{1}{(L k_B T)^2}[\langle H^2 \rangle - \langle H \rangle^2]. \qquad (6)$$

The quantities related to the order parameter, such as the susceptibility χ and the reduced fourth-order cumulant U introduced by Binder and Landau,[56] are calculated as

$$\chi = \frac{L^2}{k_B T}[\langle \delta^2 \rangle - \langle \delta \rangle^2] \qquad (7)$$

and

$$U_L = 1 - \frac{\langle \delta^4 \rangle}{3 \langle \delta^2 \rangle^2}. \qquad (8)$$

Finally, in order to discuss the nature of the phase transition, the fourth-order energy cumulant V_L was obtained as

$$V_L = 1 - \frac{\langle H^4 \rangle}{3 \langle H^2 \rangle^2}. \qquad (9)$$

IV. RESULTS AND CONCLUSIONS

The critical behavior of the present model has been investigated by means of the computational scheme described in Sec. III and finite-size scaling theory.[28, 32, 57–61] Because
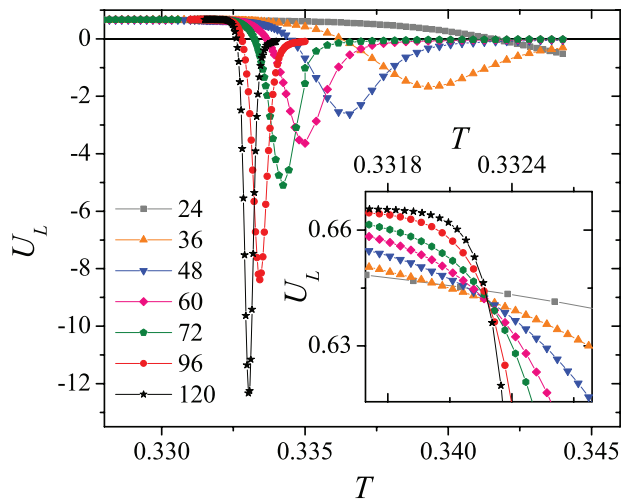
FIG. 2. The order parameter cumulant, U_L, versus T for different sizes, for adsorption of dimers ($k = 2$) on the square lattice. In the inset, a detail of the same curves near the intersection point.
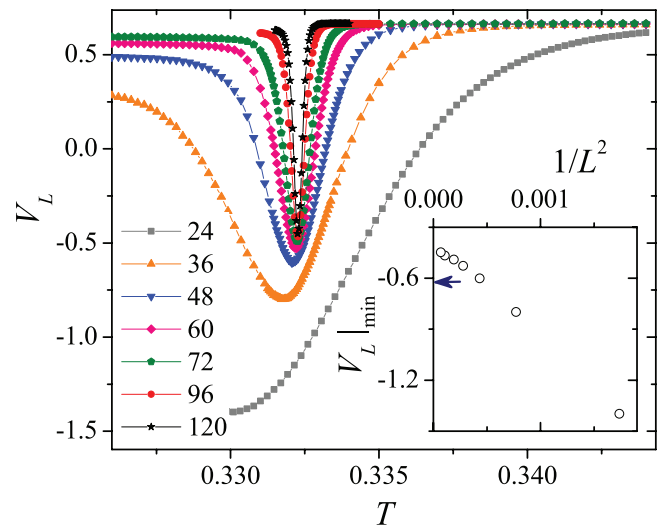

FIG. 3. Energy cumulant, V_L, versus T for different sizes, for adsorption of dimers ($k = 2$) on the square lattice. In the inset, variation of $V_L|_{min}$ with $1/L^2$. The arrow indicates the value of $V_L|_{min}$ calculated from Eq. (10).

the replica temperatures were chosen equally spaced, the acceptance probability of the replica exchange decreases in the transition region, reaching a minimum whose value is always greater than 50%. The equilibration has been tested by studying how the results vary when the simulation times n_2 and n_{MCS} are successively increased by factors of 2. We require that the last three results for all observables agree within the error bars. This simple method has shown to be useful to test equilibration.[56]

We start the analysis considering the case of dimers ($k = 2$). The parameters used for simulation were as follows: $m = 80$, $n_1 = 10^5$ MCS*s and $n_2 = n_{MCS} = 2.5 \times 10^7$ MCSs for lattice sizes, L, ranging from 24 to 120. From now on, for simplicity, temperatures will be given in units of w/k_B. In addition, the transition temperature will be denoted as T_c. As in Ref. 45, order parameter, specific heat, susceptibility and order parameter cumulant were calculated as a function of temperature for several lattice sizes. The results obtained for C and χ (not shown here for brevity) confirm previous work,[45] validating the current numerical scheme. In the case of the order parameter cumulant (Fig. 2), this quantity requires a more detailed analysis. If one observes around the transition temperature (see inset of Fig. 2), the crossing point obtained in Ref. 45 is perfectly reproduced. However, when the order parameter cumulant is calculated in a wider range of temperatures, the curves exhibit the typical behavior of U_L in the presence of a first-order phase transition. Namely, the order parameter cumulant shows a characteristic deep (negative) minimum, instead of a smooth drop from 2/3 to 0 as in a continuous transition.[52,59–61] This finding represents the first evidence that contradicts the second order behavior informed in Ref. 45.

As is well known,[59] the finite-size analysis of V_L is a simple and direct way to determine the order of a transition. In the case of a first-order phase transition, V_L shows a peak as a function of T that becomes sharper as the lattice size increases (in contrast with a constant value of 2/3 expected for

a continuous or second-order phase transition). The results in Fig. 3 confirm the first order behavior observed in Fig. 2.

The scaling properties during the transition have been also considered. In a first-order phase transition, it is expected that all finite-size effects depend on the volume of the system (L^2 in this case), instead of a dependence with the system size through different critical exponents related with the divergence of the correlation length, as occurs in a second-order transition.[52,59–61] In other words, there are no critical exponents associated with a first-order transition and divergences are a consequence of the phase coexistence. In this framework, the transition temperature can be calculated from the location of the maximum of the specific heat [$T_c^C(L)$], the susceptibility [$T_c^\chi(L)$], the logarithmic derivative of δ [$T_c^\delta(L)$], as well as the minimum of fourth-order cumulant of the energy [$T_c^V(L)$]. In fact, all these quantities versus the lattice size are expected to scale as $T_c + bL^{-2}$, where T_c is the asymptotic value ($L \to \infty$) of the transition temperature (see Fig. 4). The determination of T_c in this way results in a good agreement with the value obtained in Ref. 45 from the cumulants intersection (indicated by an arrow in the figure). In the same reference, the value found for the correlation exponent ν (= 0.53(1)), is very close to $1/d$ (= 1/2), where d is the lattice dimension. This fact explains, in part, the misinterpretation on the nature of the transition. It is worth mentioning that the mere crossing of cumulants in a nearly unique point is not an indication of a second-order phase transition. In fact, in the presence of a first-order transition, the cumulant intersection point can be considered a good estimation of the transition temperature.[52]

Another characteristic of a first-order phase transition is that the minimum of V_L has a nontrivial limit in the infinite lattice at T_c. This value, in the two-Gaussian approximation framework,[60] is given by

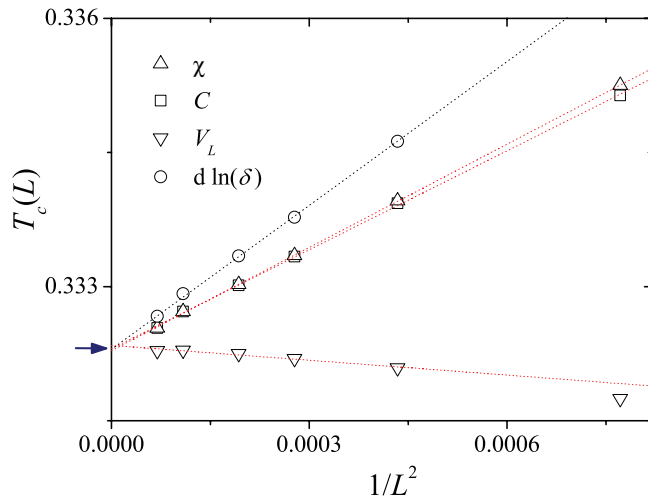$$V_L|_{min} = 1 - \frac{(E_-^2 + E_+^2)^2}{12E_-^2 E_+^2}, \qquad (10)$$

FIG. 4. $T_c(L)$ versus L^{-d} from several quantities as indicated. From extrapolation, one obtains the estimation of the transition temperature. In all cases, dotted lines correspond to linear fits of the data and $L = 36, 48, 60, 72, 96,$ and 120. The arrow corresponds to the critical temperature $T = 0.3323$ obtained in Ref. 45 from the cumulant crossing.

where E_- and E_+ are the energies per site at the low-temperature ordered and at the high-temperature disordered phases, respectively, that are coexisting at the transition. Energy distributions were calculated for three different temperatures for a lattice size of $L = 72$. In Fig. 5, the distributions (a) and (c) correspond to temperatures slightly above and below the transition temperature, while the distribution (b), was performed at the transition temperature $T_c(L = 72) = 0.33305$. All these results show the characteristic two-peak shape that we have fitted by a sum of two Gaussians,

$$D(E) = \frac{a_-}{\sigma_- \sqrt{2\pi}} \exp\left[\frac{(E - E_-)^2}{2\sigma_-^2}\right]$$
$$+ \frac{a_+}{\sigma_+ \sqrt{2\pi}} \exp\left[\frac{(E - E_+)^2}{2\sigma_+^2}\right], \qquad (11)$$

obtaining $E_- = 0.0259(2)$, $\sigma_- = 0.0064(2)$, $a_- = 0.00066(2)$, $E_+ = 0.1084(2)$, $\sigma_+ = 0.0057(2)$, and $a_+ = 0.00057(1)$, at the transition temperature. Evaluating Eq. (10) with these values, it results that $.V_L|_{min} = -0.63(3)$. On the other hand, the value obtained from extrapolation of the MC data (inset of Fig. 3) is $.V_L|_{min} = -0.41(1)$. Disregarding the distance between these two values, it is important to note that both are far from the 2/3 limit expected in the case of a second-order phase transition. The difference can be attributed to finite-size effects, which do not allow a precise fit of the energy distribution by the sum of two Gaussian functions.

To complete the picture, the behavior of the order parameter as a function of the temperature is shown in Fig. 6, and the distributions of the order parameter, for the same parameters of Fig. 5, are shown in Fig. 7. In the first case, the curves of δ present a steep variation around T_c (indicated with an arrow). As expected, the larger the lattice size, the more steep the order parameter becomes. In the case of Fig. 7, the distributions show the characteristic two-peak shape predicted for a first-order phase transition.[52]
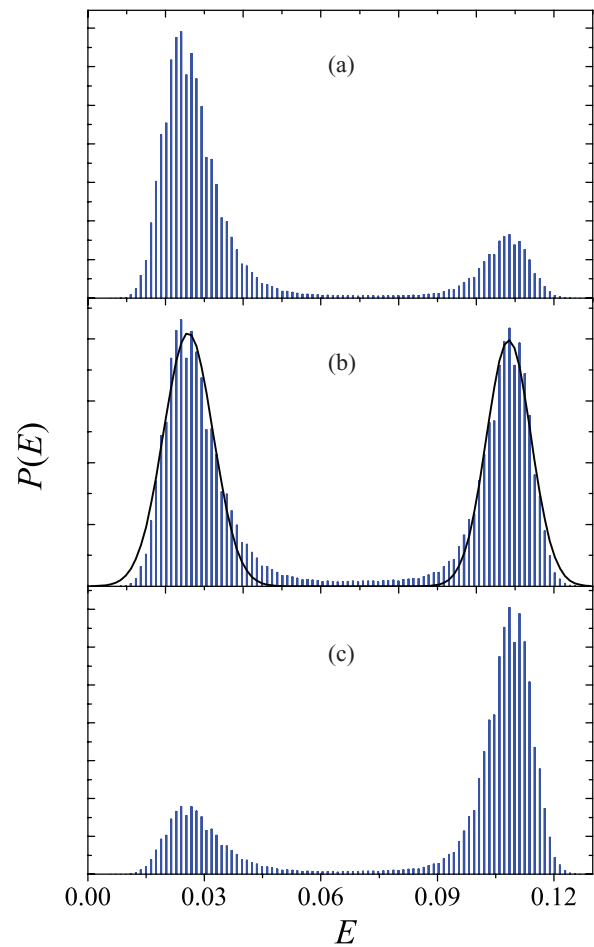

FIG. 5. Energy distribution for a system size of $L = 72$ for three different temperatures: (a) $T = 0.3327$, (b) $T = 0.3331$, and (c) $T = 0.3334$. In the second one, the continuous curve represents the best double Gaussian fit.
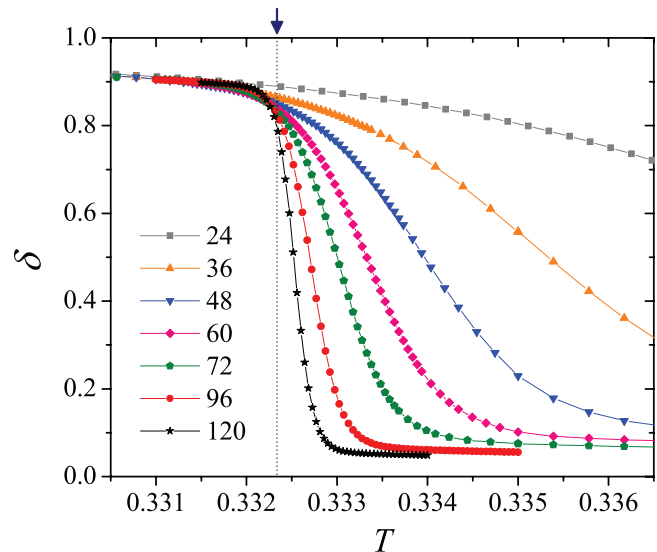

FIG. 6. Order parameter, δ, versus T for different sizes. The arrow indicates the transition temperature obtained by extrapolation in Fig. 4.
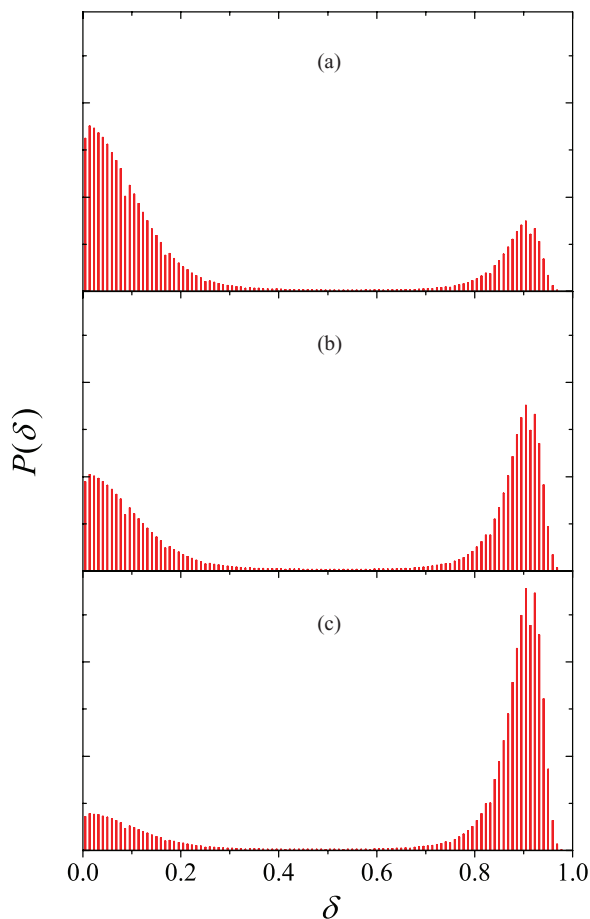
FIG. 7. Order parameter distribution for a system size of $L = 72$ and the same three temperatures around T_c as Fig. 5.
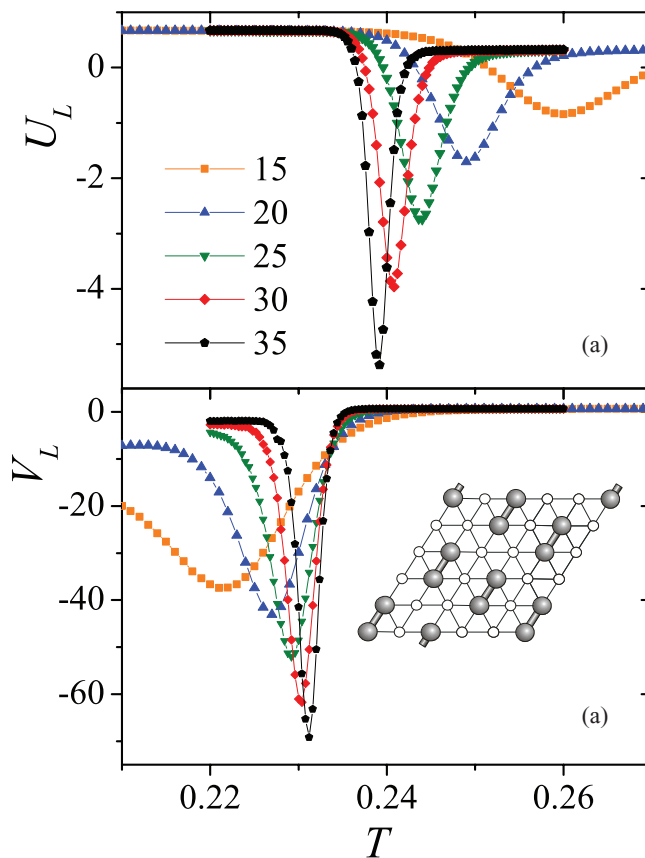

FIG. 8. (a) Order parameter cumulant, U_L, versus T for different sizes, for the case of dimers ($k = 2$) adsorbed on the triangular lattice. (b) The same for the energy cumulant, V_L. The inset shows a snapshot of the ordered phase observed in this case (the symbols are as in Fig. 1).

The finite-size scaling study was also carried out for $k = 3$ and $k = 4$, with an effort reaching almost the limits of our computational capabilities. In both cases, the parameters used in the simulations were as follows: a number of replicas of $m = 80$, $n_1 = 10^5$ MCS*s and $n_2 = n_{MCS} = 5 \times 10^6$ MCSs for lattice sizes ranging from 24 to 120. The curves corresponding to the order parameter cumulant, U_L, the energy cumulant, V_L, and the minimum of V_L versus L are analogous to those for dimers (Figs. 2 and 3) and will not be shown here for brevity. The behavior of $.V_L|_{\min}$ for increasing sizes goes to the values 0.20(1) and 0.35(1) for trimers and tetramers, respectively. The corresponding values obtained from Eq. (10), are 0.08(1) and 0.32(1). As can be seen, in both cases they are far from 2/3, as would be expected in the case of a second-order transition.

Finally, motivated by the results obtained for the square lattice, we revisit previous studies of the critical behavior in a submonolayer 2D gas of repulsive linear k-mers on a triangular lattice at coverage $k/(2k+1)$.[51] In this case, at low temperature, the k-mers are aligned along one of the three lattice directions (the corresponding ordered structure is shown in the inset of Fig. 8). A generalization of Eq. (2) for this case is[53]

$$\delta = \frac{1}{2}\left(\left|\frac{N_1 - N_2}{N}\right| + \left|\frac{N_1 - N_3}{N}\right| + \left|\frac{N_2 - N_3}{N}\right|\right), \quad (12)$$

where N_x ($x = 1, 2, 3$) represents the number of k-mers aligned along one of the three axes of the lattice and $N = N_1 + N_2 + N_3$. The simulations were carried out with the following parameters: for $k = 2$, lattice sizes of $L = 15, 20, 25, 30$ and 35, $m = 100$, $n_1 = 10^5$ MCS*s and $n_2 = n_{MCS} = 2 \times 10^6$ MCSs; for $k = 3$, lattice sizes of $L = 14, 21, 28, 35$ and 42, $m = 120$, $n_1 = 10^5$ MCS*s and $n_2 = n_{MCS} = 5 \times 10^6$ MCSs. As can be seen in Figs. 8 and 9, both U_L and V_L show a deep minimum similar to that previously observed in the square lattice, showing that a first-order phase transition occurs for $k \geq 2$. As the system shows *large finite size effects,* it has not been possible to calculate an appropriate value of the $V_L|_{\min}$ in the thermodynamic limit.

In summary, we have reinvestigated the critical properties of repulsive linear k-mers on two-dimensional square and triangular lattices. The new results, obtained by using an efficient exchange algorithm and high-performance *computational capabilities*, revealed that (1) the nature of the phase transition occurring at half coverage in a system of repulsive rigid k-mers on a square lattice changes from second order (Ising universality class) for $k = 1$ to first order for $k \geq 2$, (2) in the case of triangular lattices at coverage $k/(2k+1)$, the phase transition changes from second order (three-state Potts universality class) for $k = 1$ to first order for $k \geq 2$, and (3) the critical exponents calculated in Refs. 45 and 53 for $k \geq 2$ should be reinterpreted in terms of effective exponents.[52]
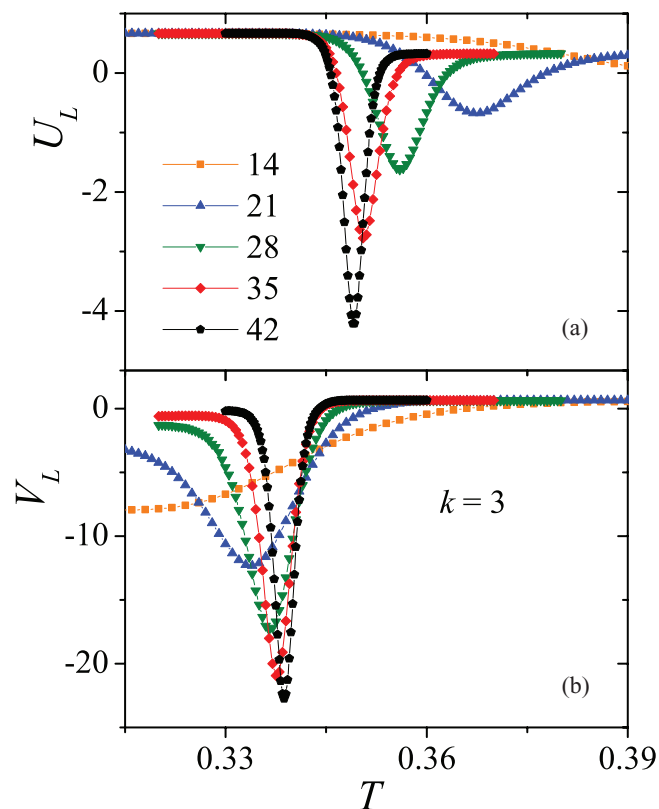
FIG. 9. The same as Fig. 8 for the case of trimers ($k = 3$) adsorbed on the triangular lattice.

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