Critical Casimir interactions and percolation: The quantitative description of critical fluctuations

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Casimir forces in a critical medium are produced by the spatial suppression of order parameter fluctuations. In this paper we address the question of how fluctuations of a critical medium are related to the magnitude of critical Casimir interactions. Namely, for the Ising model we express the potential of critical Casimir interactions in terms of Fortuin-Kasteleyn site-bond correlated percolation clusters. These clusters are a quantitative representation of fluctuations in a medium. We propose a Monte Carlo method for the computation of the Casimir force potential which is based on this relation. We verify this method by computation of Casimir interactions between two disks for the two-dimensional Ising model. The method is also applied to the investigation of a nonadditivity of the critical Casimir potential. The nonadditive contribution to three-particle interaction is computed as a function of the temperature and the separation between disks. The benefit of the proposed method is that it lets us compute the multiparticle interaction explicitly.

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I. INTRODUCTION

Critical behavior in the vicinity of the second-order phase transition point is characterized by large-scale fluctuations of the order parameter. The appearance of forces due to the spatial confinement of fluctuations in the vicinity of the critical point was predicted by Fisher and de Gennes [1]. This phenomenon in critical media is now known as the critical Casimir (CC) effect [2–4]. One can observe the appearance of CC forces in a critical binary mixture near its consolute point [5]. Two colloidal particles in a fluctuating medium exhibit attraction. The observation of colloidal particle aggregation in a critical binary mixture was described in [6]. The interaction between a colloidal particle and a planar substrate has been measured directly using total internal reflection microscopy [7,8]. Critical depletion in colloidal suspensions has been studied in many experiments [9–13] (see Ref. [14] for a review). Recently, the nonadditivity of CC interactions between three colloidal particles was studied experimentally [15]. Experimentally, it has been shown that, in a lipid membrane consisting of two different types of lipids, the second-order phase transition may occur [16,17]. Such a transition has been observed in membrane-forming plasma vesicles [18].

The analytical computation of CC forces is a difficult problem. A critical binary mixture belongs to the universality class of the Ising model. Therefore, one can use numerical simulations of the Ising model to extract information about CC interactions for particular geometry and boundary conditions. The CC force and its scaling functions for the three-dimensional (3D) Ising universality class with and without the bulk field for the film geometry and various boundary conditions have been studied numerically [19–24]. The CC force between a spherical particle and a plane for the 3D Ising universality class has been studied in Ref. [25]. A numerical algorithm for the computation of CC interactions between two particles in the presence of the negative bulk field for the 3D Ising model has been proposed in [26] and the interaction of a particle with two walls has been studied in [27]. Numerical methods are also used for the computation of critical interactions within the mean-field (MF) universality class. The CC force between two colloidal particles in three dimensions was studied using the conformal transformation in Refs. [28,29]. Mean-field interactions between an elliptic particle and a wall [30] and between two colloidal particles in the presence of the bulk ordering field [31] have been studied. Nonadditive interactions for the MF universality class have been investigated for plane-particle-particle [32] and three-particle [33] geometries.

Experimental observations of the phase separations in 2D membranes demonstrate the need for the study of the critical behavior of the 2D Ising model with appropriate geometries that have the same universality class. For the stripe geometry the CC force may be computed analytically [34–37]. Results for the CC force between two disks for the 2D Ising model with the bulk field have been obtained via the Derjaguin approximation [38]. An alternative method for the computation of the CC interaction between a mobile disk and a wall has been proposed [39]. The interaction between inclusions in a critical 2D membrane has been studied in [40] by using the Bennett method. The torque, acting on a needle near a wall, has been studied in Ref. [41]. The phase diagram of a ternary solvent-solvent-colloid mixture represented by the 2D Ising model with disklike particles has been investigated by using the grand-canonical insertion technique [42]. Recently, fluctuation-induced Casimir interactions in colloidal suspensions at the critical point in the 2D system have been studied by the geometric cluster algorithm [43]. This algorithm is capable of moving the particles and mixing the medium. Then two-particle and multiparticle interactions are extracted from particle distribution functions. For the 2D Ising model at

criticality one can use the power of conformal transformations for analytical evaluation of CC interactions between inclusions [18,43]. Recently, conformal invariance has been used for the investigation of interactions between rodlike particles [44].

In the present article we propose a short and elegant expression for CC interactions between objects based on the critical percolation clusters. This method applies to any spatial dimension (including three dimensions). The method introduced is based on the counting of some percolation clusters "touching" immersed objects.

The paper is organized as follows. In the next section we express the interaction potential of the CC force in terms of clusters of Fortuin-Kasteleyn site-bond percolation. Contrary to previously existing methods, the proposed algorithm allows performing the simulation for several objects and for mutual separations between these objects during a single run. We verify the numerical method by computing CC interactions between two disks and comparing them with the results of another numerical approach. Then the proposed method is applied to the computation of the nonadditive interactions between three particles as a function of the inverse temperature. We end with a summary and conclusions.

II. CASIMIR INTERACTION EXPRESSED VIA CRITICAL PERCOLATION CLUSTERS

Let us consider the Ising model on a simple square lattice with periodic boundary conditions. All distances are measured in lattice units. The classical spin $\sigma_i = \pm 1$ is located at a site i of the lattice. The inverse temperature is $\beta = 1/k_{\mathrm{B}}T$. The standard Hamiltonian of the bulk system (no restrictions for spin direction) for a spin configuration $\{\sigma\}$ reads

$$\mathcal{H}_{\mathrm{b}}(\{\sigma\}) = -J \sum_{\langle ij \rangle} \sigma_i \sigma_j, \tag{1}$$

where $J = 1$ is the interaction constant and the sum $\langle ij \rangle$ is taken over all pairs of neighboring spins. The partition function of the model is given by the sum over the total set $\langle \sigma \rangle$ of all spin configurations $Z_b = \sum_{\langle \sigma \rangle} e^{-\beta \mathcal{H}_{\mathrm{b}}(\{\sigma\})}$. The corresponding free energy of the bulk system is $F_b(\beta) = -\frac{1}{\beta} \ln[Z_b(\beta)]$. One can rewrite the partition function in terms of the Fortuin-Kasteleyn correlated site-bond percolation [45] (reviews on percolation theory are given in Refs. [46–48]). For every bond between two spins σ_i and σ_j we introduce the bond variable n_{ij}, which can take values 0 (open) and 1 (closed). We introduce the probability $p = 1 - e^{-2\beta}$ for a bond between two parallel $\sigma_i = \sigma_j$ spins to be closed $n_{ij} = 1$. In our case of homogeneous interactions with the fixed constant $J = 1$, the value of this probability p does not depend on spin indices ij. The probability of a bond between parallel spins being open $n_{ij} = 0$ is $1 - p = e^{-2\beta}$ (the concept of open and closed bonds between parallel spins was proposed by Coniglio and Klein [49]). A bond between two antiparallel $\sigma_i = -\sigma_j$ spins is always open $n_{ij} = 0$. We denote by $\{n\}$ the particular configuration of bond variables. The standard expression for the partition function may be rewritten as the sum over the entire set $\langle n \rangle$ of configurations of open and closed bonds

$$\begin{aligned} Z_b(\beta) &= \sum_{\langle \sigma \rangle} e^{-\beta \mathcal{H}_{\mathrm{b}}(\{\sigma\})} = \sum_{\langle \sigma \rangle} \prod_{\langle ij \rangle} e^{\beta \sigma_i \sigma_j} \\ &= e^{\beta N_b} \sum_{\langle \sigma \rangle} \sum_{\langle n \rangle} \prod_{\langle ij \rangle} \left[(1-p)\delta_{n_{ij},0} + p\delta_{n_{ij},1}\delta_{\sigma_i,\sigma_j} \right] \\ &= e^{\beta N_b} \sum_{\langle n \rangle} \left[p^{n_c(\{n\})} (1-p)^{N_b - n_c(\{n\})} 2^{c(\{n\})} \right], \end{aligned} \tag{2}$$

where N_b is the total number of bonds in the system, $n_c(\{n\})$ is the number of closed bonds, $c(\{n\})$ is the number of clusters of connected spins in the configuration $\{n\}$, and $2 = q$ is the number of spin states for the Ising model. The set $\langle n \rangle$ consists of 2^{N_b} different bond configurations.

Clusters of parallel spins connected by closed bonds are called physical clusters (contrary to geometrical clusters of parallel spins). A detailed derivation of the partition function and expressions of physical quantities such as energy, specific heat, magnetization, and magnetic susceptibility in terms of physical clusters have been provided by De Meo *et al.* [50]. The Fortuin-Kasteleyn correlated site-bond percolation is the basis of the Swendsen-Wang numerical cluster algorithm for spin models [51]. We write Eq. (2) in the form of the Ising model (e.g., $\sigma_i = \pm 1$ and $q = 2$ is the number of spin states). It is also possible to rewrite it for the q-state Potts model ($\sigma_i = 0, 1, \ldots, q$) with an arbitrary value of q. This means that this approach generalizes CC interactions for the Potts model.

Let us clarify the definition of a cluster of connected spins. Two spins σ_i and σ_j are connected if the bond between these spins is closed $n_{ij} = 1$; otherwise, these spins are not connected. The set of all spins connected to each other by closed bonds is called a cluster. By the definition of closed bonds, all spins in a given cluster are parallel. However, not every pair of parallel neighboring spins belongs to the same cluster. Let us note that, in the last line of Eq. (2), the sum is taken only over the set $\langle n \rangle$ of all possible configurations of open and closed bonds (a total of 2^{N_b} configurations). At the same time, for every particular configuration $\{n\}$, the contribution from all possible -1 and $+1$ cluster configurations is given by the term $q^{c(\{n\})} = 2^{c(\{n\})}$, which counts the number of clusters $c(\{n\})$ for this configuration. The example of such a bond configuration on the square lattice is given in Fig. 1(a), where closed bonds are shown by solid lines and open bonds are not indicated. The directions of all spins in Fig. 1(a) are not specified because each cluster can take both possible directions -1 and $+1$. Let us now consider the system with an immersed particle (or particles). In terms of the Ising model this means that all spins of such a particle are fixed [25,26,39,43]. Figure 1(b) shows the example of a disk of radius $R = 2.5$ with the center located at the point (100,100). We denote by {col} the set of all fixed spins in our colloidal particle(s); in the current example $\sigma_k = +1$ for spins with coordinates $(x_k - 100)^2 + (y_k - 100)^2 \leqslant R^2$. The partition function Z_c of the system with an immersed colloidal particle (or particles) may be expressed via the Hamiltonian of a bulk system $\mathcal{H}_{\mathrm{b}}$ with the application of the constraint $\delta_{\sigma_k,1}$ to all fixed spins. On the other hand, this partition function may be expressed via the third line of Eq. (2) if we take into account that all clusters c_c that contain spins of the colloidal

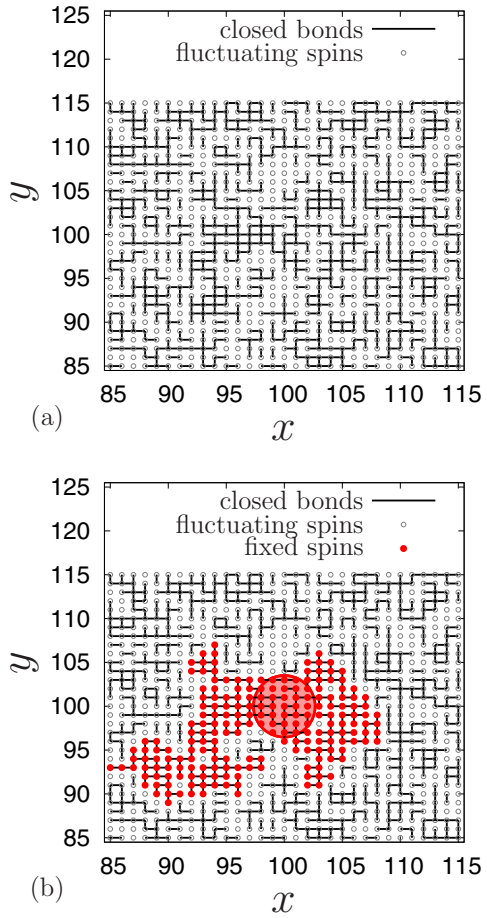

FIG. 1. (a) Clusters of spins, connected by closed bonds. Each cluster can take either -1 or $+1$ orientation; these fluctuating spins are denoted by open circles. (b) Same configuration of clusters with the inserted disk of radius $R = 2.5$ with the center at the point (100,100). All clusters which contain spins within the circle should be fixed at $+1$. Fixed spins of these clusters are denoted by closed circles.

particle cannot fluctuate. The partition function of the system with an immersed particle (or particles) is expressed as

$$Z_c(\beta) = \sum_{\langle\sigma\rangle} \prod_{k\in\{\mathrm{col}\}} \delta_{\sigma_k,1} e^{-\beta\mathcal{H}_b(\{\sigma\})} = e^{\beta N_b} \sum_{\langle n\rangle} \left[p^{n_c(\{n\})}(1-p)^{N_b - n_c(\{n\})} 2^{c(\{n\})-c_c(\{n\})}\right]. \quad (3)$$

The only difference from the partition function of the bulk system without a particle (or particles) Z_b in Eq. (2) is the term $2^{-c_c(\{n\})}$. This term reflects the fact that all c_c clusters of the current configuration $\{n\}$, which contain spins of colloidal particles, are fixed and do not contribute to the partition function. The free energy of a system with a particle (or particles) is $F_c(\beta) = -\frac{1}{\beta}\ln[Z_c(\beta)]$. Therefore, we can express the free-energy difference (in $k_B T$ units) of the insertion of a particle (or particles) as

$$\beta[F_c(\beta) - F_b(\beta)] = -\ln\left[\frac{Z_c(\beta)}{Z_b(\beta)}\right] = -\ln\langle 2^{-c_c}\rangle_\beta, \quad (4)$$

where $\langle 2^{-c_c}\rangle_\beta$ is 2 to the power $-c_c$, where c_c is the number of clusters touching the immersed object computed as a thermal average $\langle\cdots\rangle_\beta$ at the inverse temperature β with respect to the bulk (empty) system. The proposed method can also be applied to systems with various types of boundary conditions and the presence of the bulk field. Let us note that we do not specify how many particles are immersed in a bulk system. This means that the simple expression (4) may be applied to the computation of CC interactions for various geometries: particle-particle, multiparticle, wall-particle, etc. Moreover, this numerical method can also be applied to an arbitrary spatial dimension and for the $q \neq 2$ Potts model.

III. MODEL DESCRIPTION AND NUMERICAL VERIFICATION

Let us demonstrate the application of the proposed method to the computation of CC interactions between two disks in the 2D Ising system. This example has a practical interest, because it describes the interaction between protein inclusions in a lipid membrane at a critical concentration of membrane components [16–18,38,42,43].

We consider the 2D Ising model on a square lattice with the periodic boundary conditions (BCs). All distances are measured in lattice units; the system size is 200×200. For the reference bulk system with the free energy $F_b(\beta)$, the fluctuating spin $\sigma_i = \pm 1$ is located at each site of the lattice. For the system with immersed particles with the free energy $F_c(\beta, D)$, two disks (disk 1 and disk 2) of half-integer radius $R = 3.5$ are immersed in the system at the distance D [see Fig. 2(a)]. Spins in these disks are fixed at $+1$. This choice corresponds to the symmetry-breaking BCs with a completely ordered surface and it is usually referred to as a $(++)$ BC (see [22] for details). Let us denote by $c_{12}(D)$ the number of all clusters which contain spins of two disks separated by the distance D. We define the insertion free-energy difference for two disks as

$$U_{\mathrm{ins}}^{12}(\beta, D) = \beta[F_c(\beta, D) - F_b(\beta)] = -\ln\left\langle 2^{-c_{12}(D)}\right\rangle_\beta, \quad (5)$$

which depends on the distance between disks D and is expressed via the logarithm of the average value of 2 to the power of $-c_{12}(D)$ in accordance with Eq. (4). The insertion energy itself does not provide information about the disk interaction because it is defined up to a certain constant. Let us take as a normalization constant the insertion energy $U_{\mathrm{ins}}^{12}(\beta, D_{\max})$, which corresponds to the maximal possible separation $D_{\max}$ between particles for the finite system with periodic BCs. In our case $D_{\max} = L - 2R$, where $L = 200$ is the system size. We introduce the CC interaction potential

$$U_{12}(\beta, D) = U_{\mathrm{ins}}^{12}(\beta, D) - U_{\mathrm{ins}}^{12}(\beta, D_{\max}) = \ln\left\langle 2^{-c_{12}(D_{\max})}\right\rangle_\beta - \ln\left\langle 2^{-c_{12}(D)}\right\rangle_\beta, \quad (6)$$

whose derivative with respect to the distance provides the Casimir force $f_{\mathrm{Cas}} = -\partial U_{12}/\partial D$ between two disks.

As a first step we numerically verify the algorithm by computing the interaction potential $U_{12}(\beta, D)$ between disks 1 and 2 in accordance with Eq. (6). We use the hybrid Monte Carlo (MC) algorithm [52]. Each step consists of one update in accordance with the Swendsen-Wang algorithm [51]

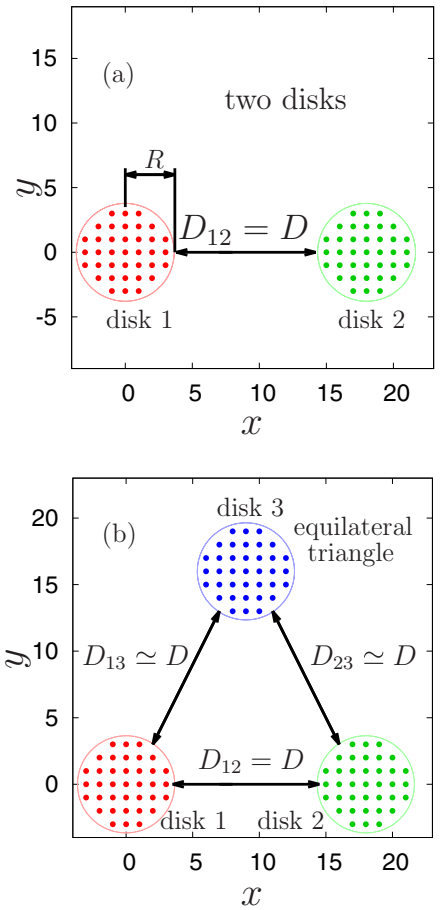

FIG. 2. System geometry for (a) two disks of radius $R = 3.5$ at the separation $D_{12} = D$ for the numerical verification of the algorithm and (b) equilateral triangle of disks at separations $D_{12} = D$ and $D_{13} = D_{23} \simeq D$ for the study of nonadditive interactions.

(which is efficient in the vicinity of the critical point) followed by $L^2/5$ attempts of Metropolis spin updates [53] at random positions (which is efficient out of criticality). The averaging is performed over 8×10^8 MC steps split over ten series for the evaluation of the numerical inaccuracy. Let us note that, in accordance with our algorithm, we can simultaneously perform computations for a set of disk separations D. In Fig. 3 we plot with symbols the interaction potential U_{12} between two disks as a function of the inverse temperature β for various separations $D = 1, 3, 5, 7, 11, 19$. In the same figure we also plot with lines the potential computed by the alternative reference method. The reference method (ref.) is based on the numerical integration of local magnetization over the locally applied field [26,54]. We observe the perfect agreement between the results computed in the two different ways. This confirms Eq. (6) and verifies the numerical code used for simulations. The position of the critical value of the inverse temperature $\beta_c = \ln(\sqrt{2}+1)/2$ is shown by the vertical dashed line. The maximum of the attractive interaction is slightly shifted to the high-temperature region, as is typical for (++) BCs [22]. The proposed method can be straightforwardly expanded for systems with bulk and surface fields and systems with several values of the interaction constants.

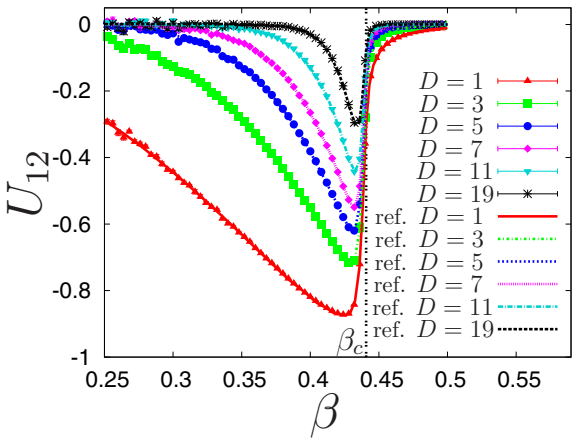

FIG. 3. Numerical verification of the CC interaction potential U_{12} as a function of the inverse temperature β between two disks at distances $D = 1, 3, 5, 7, 11, 19$ computed via percolation clusters (symbols) and by using the reference method [26] (lines).

The method also provides insight into the type of interaction, that is, whether it should be attractive or repulsive. The number of touching clusters for a short distance $n_{12}(D)$ is less than this number for a large distance $n_{12}(D_{\max})$. Therefore, $\ln\langle 2^{-c_{12}(D_{\max})}\rangle_\beta < \ln\langle 2^{-c_{12}(D)}\rangle_\beta$ and the interaction is attractive $U_{12}(\beta; D) < 0$.

IV. NONADDITIVITY OF THREE-PARTICLE INTERACTIONS

Let us now study the nonadditivity of the interaction in the three-particle system with the geometry of an equilateral triangle. This geometry is shown in Fig. 2(b) and it has been used in an experiment [15] and in the investigation of nonadditivity for the MF universality class [33]. Now we add the third disk at the separation D_{13} from the first disk and at the separation D_{23} from the second disk. The separations are selected to be almost equal on the lattice $D_{13} = D_{23} \simeq D_{12} \equiv D$. Later on for the equilateral triangle we select the odd distance D so that the projection of the center of the third disk on the x axis is located exactly between the centers of disks 1 and 2.

We denote by c_{12}, c_{13}, c_{23}, and c_{123} the number of percolation clusters which contain spins of disks 1 and 2, disks 1 and 3, disks 2 and 3, and all disks 1–3, respectively. The potential of three-particle interactions is expressed as

$$U_{123}(\beta, D) = \ln\left\langle 2^{-c_{123}(D_{\max})}\right\rangle_\beta - \ln\left\langle 2^{-c_{123}(D)}\right\rangle_\beta. \qquad (7)$$

We also introduce the sum of pair interactions $U^s_{123} = U_{12} + U_{13} + U_{23}$. For the additive potential we expect $U_{123} = U^s_{123}$. In Fig. 4(a) we plot (with symbols) U_{123} as a function of the inverse temperature β for different separations between disks $D = 1, 3, 5, 7, 11, 19$. In the same figure we plot with lines the sum of pair potentials U^s_{123}. The difference between U_{123} and U^s_{123} demonstrates the nonadditive nature of multiparticle CC interactions. Let us underline that for the fixed value of β, all quantities $\langle 2^{-c_{123}(D)}\rangle_\beta$, $\langle 2^{-c_{12}(D)}\rangle_\beta$, $\langle 2^{-c_{13}(D)}\rangle_\beta$, and $\langle 2^{-c_{23}(D)}\rangle_\beta$ for all sets of separations D are computed during the single simulation of the bulk system. This means that we

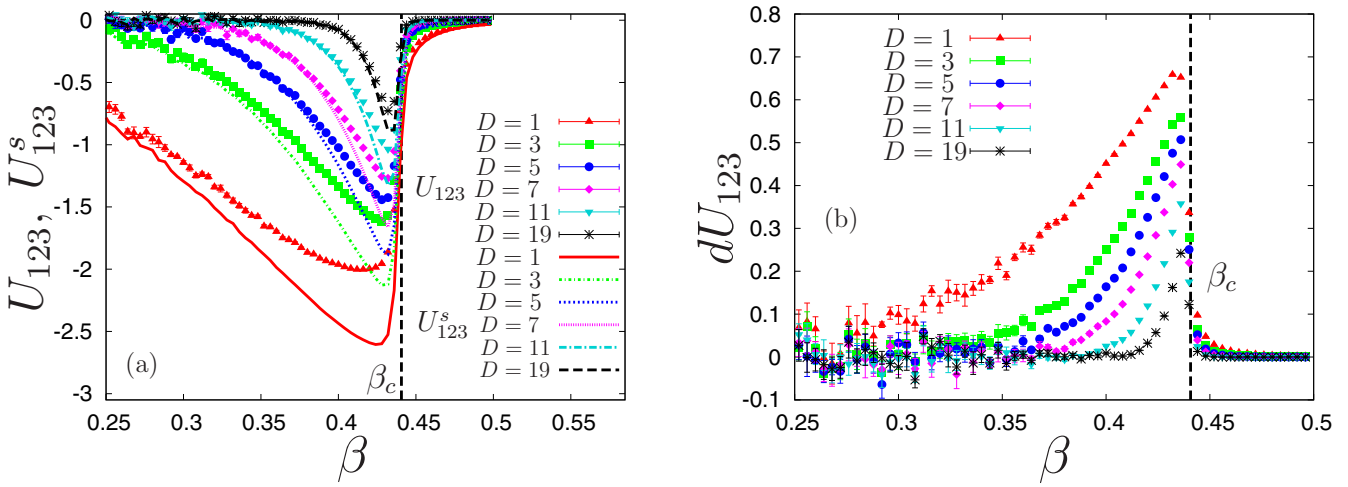

FIG. 4. (a) Multiparticle CC interaction potential U_{123} (symbols) between three disks located in vertices of the equilateral triangle with a side of length $D = 1, 3, 5, 7, 11, 19$ as a function of the inverse temperature β in comparison with the sum of three pair potentials $U^s_{123} = U_{12} + U_{13} + U_{23}$ (lines). (b) Excess nonadditive contribution to the CC interaction potential $dU_{123} = U_{123} - U^s_{123}$ as a function of the inverse temperature β for three disks at the same separations.

should not perform a separate simulation for each value of D. In Fig. 4(b) we plot the nonadditive difference $dU_{123} = U_{123} - U^s_{123}$ as a function of β for the same systems. The nonadditive part of the interactions is positive. This means that the presence of the third particle decreases the interaction between a pair of particles. In Fig. 4(b) we see that the nonadditive part of three-particle interactions has a maximum approximately at the same point, as the minimum of the two-particle interaction potential. All these potentials have wide tails to the high-temperature region, while interactions in the low-temperature region decrease very fast as we separate from the critical point. This behavior qualitatively coincides with results for the MF system [32,33] and with experimental results [15] for similar geometries.

In Figs. 5(a) and 5(b) we plot the multiparticle interaction potential and its excess nonadditive contribution as functions of the separation D for various values of the inverse temperature β, respectively. Such a presentation is more convenient for experimenters who typically measure the potential as a function of separation for the given temperature [7]. The results in Fig. 5(a) qualitatively coincide with the results shown in Fig. 4 of Ref. [15]. The nonadditivity of three-particle interactions for the 2D system at the critical point was previously studied in Ref. [43]. In that article the information about interaction potentials was extracted from particle distribution functions. Contrary to that approach, our method provides the possibility to compute the interaction potential directly, simultaneously for several values of separation D.

The algorithm described also provides insight into the effect of the presence of the third particle on the interaction between the first and second particles. The appearance of the third particle fixes some clusters n_3 on the lattice. Some of

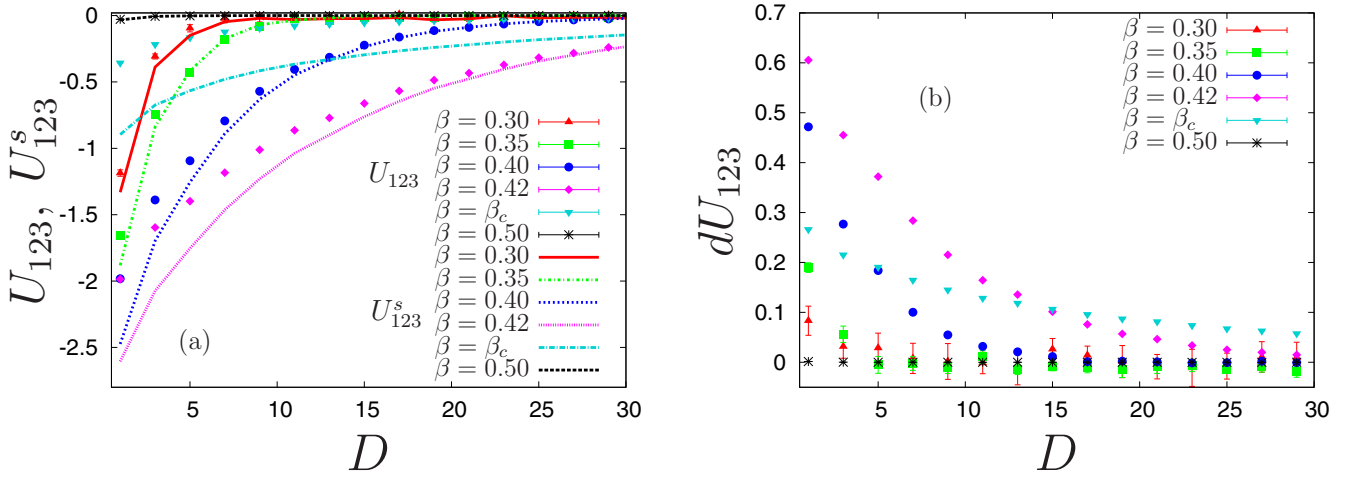

FIG. 5. (a) Multiparticle CC interaction potential U_{123} (symbols) between three disks located in vertices of the equilateral triangle as a function of the side length D for various values of the inverse temperature $\beta = 0.3, 0.35, 0.4, 0.42, \beta_c, 0.5$ in comparison with the sum of three pair potentials $U^s_{123} = U_{12} + U_{13} + U_{23}$ (lines). (b) Excess nonadditive contribution to the CC interaction potential $dU_{123} = U_{123} - U^s_{123}$ for $\beta = 0.3, 0.35, 0.4, 0.42, \beta_c, 0.5$.

these clusters should also be counted among the n_{12} clusters which are fixed by particles 1 and 2. Let us denote by $n_{12}(D)\backslash n_3$ the number of clusters which are fixed by a pair of particles under the condition that some of the clusters are already fixed by the third particle. This number is by definition less than the number n_{12} of clusters which are fixed at the same position by the pair of particles without the third particle. Therefore, the intensity of the attraction between a pair of particles will be reduced by the presence of the third particle of the same type.

V. CONCLUSION

In the present paper we expressed the free-energy change for the insertion of objects into a critical system in terms of percolation clusters intersecting the volume of inserting objects. A numerical algorithm for CC interactions which is based on the counting of the number of percolation clusters was proposed. The algorithm provides an explicit expression for the Casimir potential without numerical integration. This percolation-based algorithm applies to any spatial dimension (including the important case of the 3D Ising model). This algorithm was numerically verified for the computation of the CC force potential between two disks for the 2D Ising model. The proposed method allows one to obtain results for several separations between objects during a single run. The proposed method was also applied to the study of multiparticle interactions in critical media and to express the nonadditive part of the interaction potential in terms of percolation clusters. The expression obtained gives us qualitative information about the sign of the nonadditive contribution to the multiparticle interaction, while the numerical realization of the proposed algorithm enables us to perform a quantitative computation of the interaction potential. These results are in qualitative agreement with Ref. [15], where it was shown that the presence of the additional particle decreases the absolute value of the interaction between two particles with the same surface properties.

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