Percolation threshold parameters of fluids

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Extensive Monte Carlo simulations on three qualitatively different model supercritical fluids (square-well fluid, Lennard-Jonesium, and primitive water) have been performed to examine percolation threshold parameters for continuum (correlated) models and their relation to general results valid for random lattice models; random-site percolation simple-cubic lattice has therefore been considered as well. Two different bond criteria, the configurational and self-bound ones, defining a cluster have been used. In addition to the percolation threshold occupation probability p_c and the percolation threshold fluid density ρ_c, the correlation length exponent ν and the wrapping probability at the percolation threshold $R_{w,c}$ have also been evaluated. It is found that parameters ν and $R_{w,c}$ exhibit not only strong temperature dependence but also, unlike the case of lattice systems, dependence on the nature of the system considered and the employed definition of the cluster.

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I. INTRODUCTION

The concept of percolation [1,2] is used to characterize the effect of connectivity of microscopic elements (such as molecules, pores, conducting elements, people, etc.) in disordered systems (such as fluid, porous media, composite film, and society) on their macroscopic properties (such as condensation, flow of oil, flow of electric current, and information flow). From its origin until today, it deals primarily with lattice systems, i.e., with a grid of sites (edges) occupied with a probability p for which a number of theoretical results have been derived.

An important aspect of percolation theory developed for random lattice systems is universality, which makes it possible to study and understand a variety of systems without bothering about their details. However, there is no need to use a grid for the objects can be placed in a continuum space as well, as, e.g., molecules of fluids. But in this case, unlike uncorrelated systems, no theoretical results are available. Consequently, various conjectures on the behavior of continuous (correlated) systems represent a mere extension of the results obtained originally for lattice systems, particularly for two-dimensional (2D) lattices and special types of three-dimensional (3D) lattices [1]. A question then arises as to what extent such an assumption on similarity in the behavior of uncorrelated and correlated systems may hold true over a wide range of systems and conditions.

From the macroscopic point of view, the fluid phase is homogeneous because its density is, in average, uniform throughout the system. However, on the microscopic level its particles (molecules) may form localized morphological structures called clusters which may be then the object of percolation theory. A number of various characteristics of clusters may be defined and investigated as, e.g., the size, shape, lifetime, etc. [1,3,4]. The primary object of the percolation theory is the probability of occurrence of a percolating cluster (called also an infinite cluster or wrapping, spanning or crossing cluster), i.e., the cluster that spans the entire system. Dealing with fluids, the probability of occurrence of a percolating cluster R depends on density ρ and the size L of the system at hand. It is a monotonously increasing function of density which in the limit of an infinite system becomes the step function. The density at which this occurs is called the percolation threshold [1,5] ρ_c, and the goal is to find or determine this threshold and the corresponding probability R_c. Using molecular simulations as the only currently available tool, the goal is then to estimate the percolation threshold from simulations on finite systems.

The determination of the percolation threshold from results obtained for finite-size systems is not unique and a number of various criteria have been proposed and used (see, e.g., [6]). Although some authors claim that scaling corrections due to finite-size effects are not essential [7], more accurate methods determining the percolation threshold density and other parameters are based on a finite-size scaling analysis; for further details, we refer the reader to [5,8–10]. Typically, it is assumed that for large system size L, the function R exhibits near the percolation threshold the universal behavior as a function of the scaling variable $(\rho-\rho_c)L^{1/\nu}$ with ν called the correlation length exponent; for uncorrelated systems, this is a universal constant for the given dimensionality regardless of the thermodynamic conditions and the nature of systems considered [1,11,12].

In addition to such physical assumptions, there is also a number of rather technical quantities that enter the process of the percolation threshold determination and that may significantly affect the final results. It is, first of all, the identification of a percolating cluster in a finite system and the definition of cluster itself. Concerning the latter issue, there are

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two main concepts: (i) global one, in which the cluster is defined with respect to other clusters and an optimal cluster decomposition is then found using some criterion [13,14] and (ii) local one, in which the cluster is defined by means of bonds between pairs of constituent particles. A cluster is then a collection of particles such that there is a pass (via bonds) between any pair of particles and the entire problem is then reduced to the problem of an appropriate definition of bonds. Different cluster definitions with respect to their relevance are summarized, e.g., in Ref. [15].

As regard the identification of a percolating cluster, various spanning rules (the local-bond concept) are discussed at length in Refs. [9,11,16,17]; the primary classification is based on the boundary conditions considered. In our recent paper [18], we examined consequences of different spanning rules considering the supercritical square-well (SW) fluid and concluded that the so-called wrapping probability R_w provided a more accurate estimate of the percolation threshold when compared to the crossing probability. Furthermore, it turns out that finite-size corrections to R_w at the percolation threshold and for sufficiently large system sizes are negligible and that the parameters ν and $R_{w,c}$ (the wrapping probability at the percolation threshold) exhibited, within the simulation error bars, universal behavior. In a subsequent paper [19], the same analysis was applied to results obtained for a qualitatively different model, the primitive model of water, and a similar conclusion was drawn.

All the above findings are in agreement with the statement made long time ago by Geiger and Stanley [20] who claimed that there was no obvious basis for the criticisms against the application of lattice percolation exponents to describe continuum correlated systems as well. This view seems to go back to the Geiger *et al.*'s finding that "… liquid water appears as a uniform space-filling random network" [21] and has been later taken for granted (see, e.g., [11]). Nonetheless, a more careful numerical analysis of our previous results indicates that the validity of the considered universality has been at the edge of accuracy of the computations and that relatively large error bars may hide some significant deviations. In fact, the same observation was made already by Heyes and Melrose [8] long time ago, but they attributed the found deviations from universality to finite-size effects and did not thus question it.

The mentioned uncertainties concerning the assumption of universality have prompted us to examine this assumption in more details. We consider three qualitatively different model fluids, the square-well fluid, the Lennard-Jonesium, and the extended primitive model EPM5–4: all of them for a range of supercritical temperatures and two different definitions of cluster. Great care is also paid to the error analysis. To further support the obtained results and check correctness of the used numerical method, we also consider a simple lattice model for which the literature data are available [10,17,22]. All the obtained results clearly show that the percolation parameters for correlated systems may hardly be considered as universal; they exhibit a strong dependence not only on temperature but also on the nature of the system and particular definition of clusters. The paper is organized as follows. After providing all necessary definitions and explanation of the used methodology in Sec. II, we then present and discuss the results. The algorithm used for the cluster identification is explained in the Appendix.

II. MODELS AND CLUSTER DEFINITIONS

A. Models

We consider two common simple fluid models (i.e., the models without any angular dependence), the SW model and the Lennard-Jones (LJ) model and one model fluid with a strong orientational dependence, the EPM5–4 primitive model [23,24]. The SW potential is defined as

$$u_{\mathrm{SW}}(r;\lambda)=\begin{cases}\infty & \text{for} \quad r<\sigma \\ -\varepsilon & \text{for} \quad r\in\langle\sigma;\lambda\rangle \\ 0 & \text{for} \quad r\geq\lambda,\end{cases} \tag{1}$$

where σ is the diameter of the hard core, ε is the depth of the potential well, and for the range of the potential λ, we choose the value 1.5σ. The considered cutoff LJ potential has the form

$$u_{\mathrm{LJ}}(r)=\begin{cases}4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12}-\left(\frac{\sigma}{r}\right)^{6}\right] & \text{for} \quad r\leq 5\sigma \\ 0 & \text{for} \quad r>5\sigma,\end{cases} \tag{2}$$

where σ and ε are the common potential parameters. The extended five-site primitive waterlike model (EPM5–4) has four tetrahedrally arranged interaction sites of two kinds with the hard-sphere (HS) interaction between the like sites and the SW attraction between the unlike sites (for further details and parameters, see [23]),

$$u_{\mathrm{EPM5-4}}(1,2)=u_{\mathrm{HS}}(r_{\mathrm{OO}};\sigma_{\mathrm{OO}})+\sum_{i\in\{1\},j\in\{2\}}u_{\mathrm{HS}}(r_{ij};0.8\sigma_{\mathrm{OO}}) + \sum_{i\in\{1\},j\in\{2\}}u_{\mathrm{SW}}(r_{ij};0.4\sigma_{\mathrm{OO}}), \tag{3}$$

where the summation in the second term runs over the pairs of the like sites and in the third term over the unlike sites, and the HS interaction is given by

$$u_{\mathrm{HS}}(r;\sigma)=\begin{cases}\infty & \text{for} \quad r<\sigma \\ 0 & \text{for} \quad r\geq\sigma.\end{cases} \tag{4}$$

The potential parameters are used henceforth to scale the appropriate quantities and we may thus set $\sigma=\sigma_{\mathrm{OO}}=1$, and $\varepsilon/k_B=1$ where k_B is the Boltzmann constant.

Referring again to the assumption of universality and as a means to verify the used numerical algorithm (see the Appendix), we consider also the standard site percolation on a simple-cubic lattice. There are two ways to generate different configurations with a linear dimension L: either the number N of randomly occupied sites is fixed or the probability for a given site to be occupied p is fixed. If one imagines that the occupied sites represent the particles then the former choice corresponds to the sampling from a canonical ensemble and the latter to the sampling from a grand canonical ensemble [25]. Another and likely more common interpretation is that the number of occupied sites represents the energy and the respective samplings are then microcanonical and canonical

[16]. Nevertheless, we prefer the first point of view because p and the fluid density ρ have a similar meaning and both are equal to N/L^3.

B. Definitions of cluster

There are two basic concepts to define an existence of a bond between a pair of particles, one based on their proximity in the configurational space (configurational [15] or Stillinger [26] clusters) and the other based on their proximity in the phase space (self-bound [15] or Hill [27] clusters). In the former case, the criterion is specific for each fluid model considered and is defined as follows. SW: $r<\lambda$, LJ: $r<(\frac{26}{7})^{1/6}$ (the inflection point of the potential function [28]), and EPM5–4: $u<0$. All these criteria are based on the relative interparticle distance and in the third case also on the relative orientation.

The Hill clusters are defined by means of the relative energy [15,27]. If $\boldsymbol{v}_i$ and $\boldsymbol{v}_j$ are the velocity vectors of particles i and j, respectively, and m is their mass, then they are considered to be bonded if

$$\frac{m}{4}(\boldsymbol{v}_j-\boldsymbol{v}_i)^2 \leq -u_{ij}. \tag{5}$$

In the case of the site percolation on a simple-cubic lattice, the cluster is defined as a group of occupied sites connected by bonds that are between nearest-neighbor sites.

III. PERCOLATION THRESHOLD AND FINITE-SIZE SCALING

From various spanning rules, we use the wrapping rule for its general features. In a system with periodic boundary conditions, the cluster wraps the system if it is possible to get, starting from any particle of the cluster and moving along interparticle bonds, to an image of that particle in another replica [11]. For the given system size and density, a large number of configurations n is analyzed to obtain an estimate of $R_w(N,\rho)$. As we showed in our previous papers [18,19], the functions $R_w(N=\text{const},\rho)$ for different N intersect in one point, the percolation threshold. Two curves $R_w(N,\rho)$ are therefore sufficient to determine $R_{w,c}$ and hence also the threshold density ρ_c. This estimate as well all other percolation threshold parameters are subject to errors. If the analyzed Monte Carlo (MC) configurations were uncorrelated then the standard error of the mean of the probability R_w should be given by [16]

$$\sigma_{R_w}=\sqrt{\frac{R_w(1-R_w)}{n}}. \tag{6}$$

This is the case of random configurations of the lattice model because they are generated independently of each other. For continuous systems, this would hold only if the analyzed configurations were separated at least by the (unknown) correlation length. Nonetheless, comparison of the evaluated results for fluids with Eq. (6) may provide an indirect estimate of their accuracy.

The probability R_w is a function of two variables, density ρ (or p in the case of lattice systems), and the system size characterized either by N or $L=(N/\rho)^{(1/3)}$. However, as it has been already mentioned, it may be expressed as a function of a single (scaling) variable $x=(\rho-\rho_c)L^{1/\nu}$. Thus, as a result of the finite-size scaling analysis, a single scaling function $R_w(x)$ is obtained near the percolation threshold density ρ_c and for sufficiently large system sizes. In other words, all the curves $R_w(\rho)$ plotted for different system sizes should fall on a single curve $R_w(x)$ independent of different system sizes. To obtain the threshold parameters, the scaling function $R_w(x)$ is parametrized and the least-square analysis is then applied (to the entire collection of data for different N). For the parametrization, we use the following function:

$$R_w(x)=\frac{1}{1+\exp\left(\sum_{i=0}^{5}a_i x^i\right)}, \tag{7}$$

where a_i, ρ_c, and ν [entering Eq. (7) via the variable x] are adjustable parameters. As a first estimate of ρ_c, the point of the intersection of the curves $R_w(\rho)$ vs ρ for different system sizes may serve.

The mean and its standard error of each adjustable parameter are calculated using a Monte Carlo method as follows [29]. 100 different data sets are generated by randomly adding Gaussian noise [an algorithm analogous to Eq. (8) may be used] to the original data set with a distribution of the width proportional to σ_{R_w} different for every data point. On each of these data sets, the same fit procedure is performed and resulting adjustable parameters are evaluated. Using their means and Eq. (7), the wrapping probability at the percolation threshold $R_{w,c}$ equals $R_w(0)$.

IV. COMPUTATIONAL DETAILS

In the case of fluid models, we carried out common metropolis MC simulations in a canonical ensemble (NVT) [30,31]. The parameters of the simulations were set so as to maintain the acceptance ratio, approximately, around 1/3. The systems were equilibrated by performing at least $6000N$ MC steps (for the SW and LJ fluids) and $30\,000N$ MC steps (for the EPM5–4 fluid), respectively. Control quantities were monitored to keep the simulations under control [32]. Equilibrium configurations used for the detection of the presence of the wrapping cluster were separated by $3N$ MC steps.

In MC simulations, the mass and velocity vectors are not involved. Nonetheless, the Hill clusters may yet be identified by assigning velocity components (v_x, v_y, and v_z) to every particle selected at random from the Gaussian distribution characterized by the temperature of the system [15]. We employ the following algorithm (a modification of the Box-Muller algorithm [33]) using the transformation of uniformly distributed pseudorandom numbers $\gamma_i \in \langle 0;1 \rangle$:

$$v_{xy}=\sqrt{\frac{k_BT}{m}}\sqrt{-2\ln(1-\gamma_1)},$$

$$v_x=v_{xy}\cos(2\pi\gamma_2),$$

TABLE I. The number of analyzed configurations for different fluid models (in multiples of 10^5). The numbers of generated distributions of the Hill clusters at every configuration are enclosed in parentheses.

Fluid model	$N=1000$	$N=2000$	$N=4000$
	Configurational clusters		
SW	128	64	32
LJ	64	32	16
EPM5–4	512	256	128
	Hill clusters		
SW	128(1)	64(1)	32(1)
LJ	2(32)	2(16)	2(8)
EPM5–4	256(100)	128(100)	64(100)

$$v_y = v_{xy} \sin(2\pi\gamma_2),$$

$$v_z = \sqrt{\frac{k_B T}{m}} \sqrt{-2 \ln(1-\gamma_3)} \cos(2\pi\gamma_4), \tag{8}$$

with an arbitrary value of m assigned to the molecules. This procedure may be independently repeated so that a large number of distributions of the Hill clusters may be examined at the same configuration.

For investigating the site percolation on the simple-cubic lattice, we use the sampling from a grand canonical ensemble. We do not use any sophisticated algorithm for generating different configurations and for cluster identification such as Newman's and Ziff's algorithm [34]. We simply generate a new configuration by randomly occupying each site of an initially empty lattice with an independent probability p. Then the clusters are identified using the algorithm presented in the Appendix and which may be generally applied both to the lattice and continuum systems.

V. RESULTS AND DISCUSSION

The models and methodology described in Secs. II, III, IV, and VI and in the Appendix have been used to examine the behavior of the percolation threshold parameters and their potential universality. We have performed MC simulations using both the Stillinger and Hill definitions of the cluster. Three different system sizes have been considered: for the fluid models, N has been set to 1000, 2000, or 4000, and the respective number of analyzed configurations different for particular definitions of cluster is listed in Table I. For the simple-cubic lattice size, L has been set to 16, 32, and 64, respectively, and the respective n have been $\{64,8,1\}\times 10^5$. Further, for each fluid model we have investigated configurations for a number of supercritical temperatures T (see Table II) for a set of densities ρ near the percolation threshold density ρ_c; for the lattice system then a set of occupation probabilities p near the percolation threshold occupation probability p_c.

In Fig. 1 we show the dependence of the wrapping probability in dependence on the size of systems, and in Fig. 2

TABLE II. Results for the percolation threshold fluid density ρ_c, the percolation exponent ν, and the wrapping probability at the percolation threshold $R_{w,c}$ for the considered models. Numbers in parenthesis denote the standard error of the mean of the last digits.

Square-well fluid			
T	ρ_c	ν	$R_{w,c}$
	Configurational clusters		
1.25	0.1238453(49)	1.00755(85)	0.41207(22)
1.30	0.1265005(41)	0.99667(59)	0.41590(19)
1.40	0.1310388(36)	0.98565(53)	0.42209(16)
1.60	0.1379264(23)	0.97122(34)	0.43027(11)
2.00	0.1469017(16)	0.95124(29)	0.436193(75)
2.50	0.1537498(15)	0.94277(22)	0.439863(70)
4.00	0.1636471(14)	0.93263(21)	0.443757(68)
6.00	0.16900440(94)	0.93012(17)	0.445549(49)
	Hill clusters		
1.25	0.240056(53)	1.0670(26)	0.27295(77)
1.30	0.266927(28)	1.0508(11)	0.33700(43)
1.40	0.308647(14)	0.97207(67)	0.40590(22)
1.60	0.3706048(60)	0.91732(32)	0.437442(85)
2.00	0.4718585(82)	0.88900(30)	0.450848(98)
2.50	0.589187(16)	0.87218(42)	0.45233(16)
3.20	0.771646(87)	0.8408(15)	0.44760(49)
Lennard-Jones fluid			
T	ρ_c	ν	$R_{w,c}$
	Configurational clusters		
1.35	0.243568(33)	1.0943(23)	0.27327(62)
1.40	0.253411(21)	1.0759(15)	0.31902(44)
1.60	0.2737133(81)	1.01078(66)	0.39003(19)
2.00	0.2901569(47)	0.96331(40)	0.42299(12)
2.50	0.2990854(36)	0.94563(33)	0.432922(99)
	Hill clusters		
1.35	0.387867(64)	1.1046(26)	0.35266(66)
1.40	0.413878(32)	0.9799(12)	0.40706(36)
1.60	0.487745(12)	0.90414(43)	0.44393(13)
2.00	0.619912(18)	0.87972(51)	0.45109(15)
2.50	0.801749(65)	0.84206(98)	0.44799(34)
EPM5–4 model of water			
$1/T$	ρ_c	ν	$R_{w,c}$
	Configurational clusters		
5.00	0.143679(15)	0.9718(18)	0.42240(57)
4.00	0.2195969(56)	0.91950(70)	0.45297(22)
3.00	0.3087252(28)	0.90051(33)	0.45812(10)
2.00	0.4130068(24)	0.88838(27)	0.462602(79)
1.00	0.5352741(32)	0.88446(27)	0.462888(94)
	Hill clusters		
5.00	0.147970(16)	0.9131(21)	0.44866(61)
4.00	0.2302540(50)	0.90699(64)	0.45218(19)
3.00	0.3369448(42)	0.89384(43)	0.45910(14)
2.00	0.4902213(52)	0.87904(35)	0.46185(13)
1.20	0.711210(13)	0.86692(67)	0.45831(23)

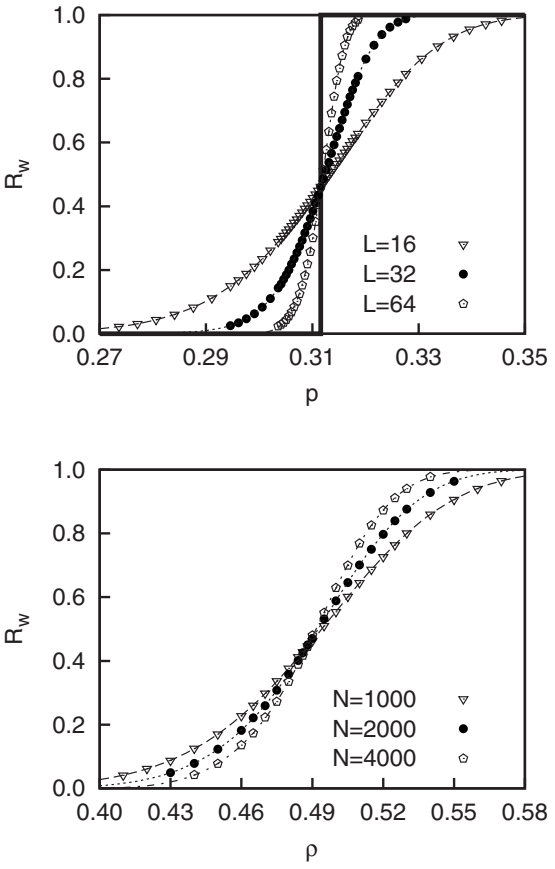

FIG. 1. The wrapping probability R_w as a function of probability p for a given site to be occupied in a simple-cubic lattice of linear dimension L (the upper graph) and as a function of density for the Lennard-Jones fluids and Hill clusters at temperature $T=1.6$ and different number of particles N (lower graph). The thick solid line represents the probability R_w in the limit $L\to\infty$.

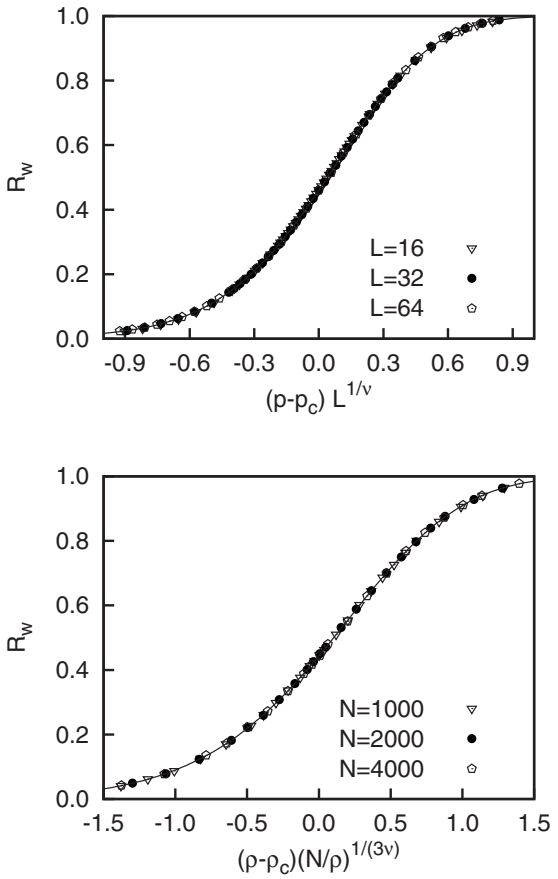

FIG. 2. The same as Fig. 1 for R_w as a function of the scaling variable. The solid line corresponds to the fit by Eq. (7)

this probability function as a function of the scaling parameter x. As it is discernible, the larger the system size, the more abrupt change of R_w from zero to unity. Figure 1 thus confirms the previous finding, namely, that when the spanning cluster is defined by wrapping then all the curves for different system sizes intersect in one point which corresponds to the percolation threshold. This feature also expresses the fact that the finite-size corrections to R_w at the percolation threshold and for sufficiently large system sizes are negligible. Figure 2 then demonstrate the validity of the scaling feature of the function R_w. A nearly perfect collapse of all three curves into one may be considered as a convincing proof that the used regression model [Eq. (7)] and consequently also the used numerical method for the determination of the percolation threshold properties, parameter ν, and $R_{w,c}$, is sufficiently accurate. We may conclude that systematic errors arising from the choice of the regression model are not larger than the statistical ones listed also in Table II. It is also worth mentioning that such a universal scaling behavior of R_w is observed only if the wrapping definition is used for the occurrence probability R.

The most important results, values of the percolation exponent ν, and the critical wrapping probability $R_{w,c}$ are listed in Table II. They have been determined from the fitting procedure specified in Sec. III and, as it is seen, they are specific for each percolation process. These results are evidently subject to errors. First, we note that at least 17 (up to 55) R_w data points for each system size, such that the minimum of the mean of R_w is less then 0.05 and its maximum is greater then 0.95, have been used. Second, the accuracy of the wrapping probability R_w determination itself depends primarily on the number of analyzed configurations n. In Fig. 3 we show that there exists a very good correspondence between the conventionally calculated standard error of the mean of the probability R_w and its theoretical estimation from Eq. (6). Such a correspondence cannot be observed for the correlated configurations that are frequently obtained from routine (not very careful) simulations on the fluid models.

To further support the obtained results, we compare in Table III our results for the site percolation on a simple-cubic lattice with data available from literature. As it is seen, the results are in mutual agreement although we have investigated only a quite small sample of simple-cubic lattice configurations with relative small system sizes. From this fact, we conclude that the used methodology is sufficiently efficient and accurate.

As we have already mentioned, it is not unusual to find in literature claims that the percolation exponent ν is a universal parameter for a given dimensionality, i.e., it has the same value both for random percolation models and fluids. However, this does not seem to be the case because the obtained

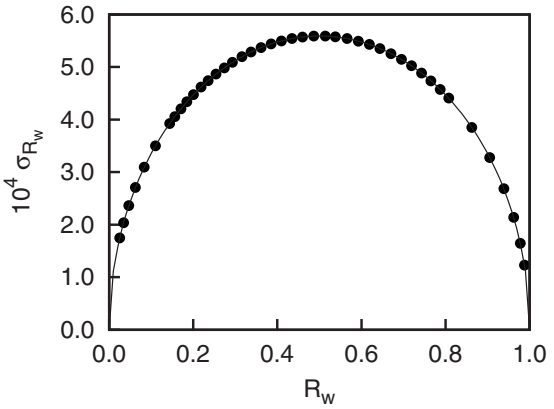

FIG. 3. The standard error of the mean of the probability R_w for the simple-cubic lattice with $L=32$ for which $n=8\times10^5$ configurations was generated and investigated for the presence of the wrapping cluster. The full circles are simulation results and the solid line represents results obtained using Eq. (6).

results show its rather strong dependence on (i) the interparticle interaction, (ii) the temperature, and also (iii) the particular definition of cluster. For example, if we compare the values for a specific fluid model at a given temperature, e.g., ν for the LJ fluid at $T=2.5$, we have 0.94563(33) for the configurational clusters whereas for the Hill cluster we have 0.84206(98). The same conclusion is valid also for the wrapping probability at the percolation threshold $R_{w,c}$ that has also been supposed to be a universal parameter for a given dimensionality. Contrary to these assumptions, we may conclude that real fluids (i.e., correlated systems) belong to the different universality class than usual random percolation systems. As regard the percolation threshold density ρ_c, it also exhibits specific values for the individual systems but this finding cannot be surprising.

VI. CONCLUSIONS

We have performed extensive MC simulations on three qualitatively different models of fluids over a range of supercritical temperatures and on one lattice model with the goal to examine the behavior of some percolation threshold parameters and their potential dependence on thermodynamic conditions. To avoid getting results valid only for one specific concept of the cluster definition, we have considered also two different concepts of bonds: the configurational and self-bound clusters. After performing a careful numerical analysis, we have obtained results which clearly show the difference in the behavior of uncorrelated and correlated systems. We have thus to conclude that the percolation threshold in fluids (i.e., continuum correlated systems) cannot be characterized by universal exponents of random percolation, contrary to the claims found, e.g., in Ref. [35].

TABLE III. Results for the percolation threshold occupation probability p_c, the percolation exponent ν, and the wrapping probability at the percolation threshold $R_{w,c}$ for the site percolation on simple-cubic lattice. Numbers in parenthesis denote the standard error of the mean of the last digits.

Reference	p_c	ν	$R_{w,c}$
[22]	0.3116080(4)		
[10]	0.3116081(13)	0.8765(18)	
[17]	0.3115(3)	0.877(12)	
This work	0.3116004(35)	0.87555(49)	0.46063(11)

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APPENDIX: ALGORITHM FOR THE WRAPPING CLUSTER IDENTIFICATION

Decomposition of the configuration in clusters begins with creating a neighbor list, i.e., a list of directly connected (bonded) particles. The list (made separately for each reference particle) must contain not only the particle's label but also its relative location, i.e., the vector connecting the reference particle and the nearest image of the bonded particle. Then we can construct clusters, i.e., make lists containing the particle's label and its absolute location in an infinite system. The goal is to find in such a list (cluster) the same particle but at a different location. Then the cluster is wrapping because it contains both a particle and also its image.

To construct a new cluster, proceed as follows:

(1) Find particle k not associated with any cluster during the cluster construction process (for the first cluster construction, set simply $k\leftarrow1$). Start with making the list of particles directly bonded to k, $\{k_1\equiv k,k_2,\ldots,k_\kappa\}$.

(2) Move to the next member of the list (if it does not

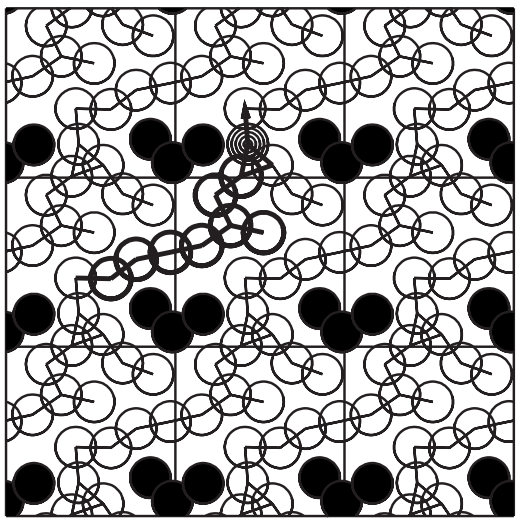

FIG. 4. An example of two configurational clusters (overlap of disks defines the bond) in a continuous 2D system. Filled disks represent a three particle, not percolating cluster. Disks connected by lines represent a wrapping cluster.

exist, the construction of a new cluster is completed) and find particles directly bonded to it and keep adding them to the list, $\{k_1, k_2, \ldots, k_\kappa, k_{\kappa+1}, \ldots\}$. Before adding a new member $k_{\kappa+\lambda}$ to the list, check if this particle already exists in the list: (a) if it does not exist, add it to the list and (b) if it exists then compare their location. If it is in the same location do not add it to the list. If it is located in another position then it is its image and a wrapping cluster has been identified.

Repeat this step until the construction of a new cluster is completed.

Repeat these steps until all particles have been incorporated into any cluster or a wrapping cluster has been found. In Fig. 4 we present a simple example of the wrapping cluster identification.

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