Nonmonotonical behavior in correlated site-bond percolation

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Site-bond percolation is addressed in a very general class of correlated site-bond systems. The site-bond model analyzed provides a simple natural picture of disordered media such as porous materials, nonuniform surfaces adsorption potential, conductivity of inhomogeneous systems, and landscapes. The bond (site) percolation threshold exhibits a nonmonotonical behavior, showing maxima and minima as statistical correlation of two-point lattice properties varies from zero to one. Phase diagrams for the general case of correlated site-bond percolation are obtained by Monte Carlo simulation. The continuum percolation limit is recovered for percolation at maximum correlation. [S1063-651X(97)08612-1]

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I. INTRODUCTION

Percolation models have been of central significance in modern statistical physics to gain understanding about phenomena in very different areas of science. Processes such as polymerization, fluid-phase transport in porous media, disease spreading, disordered systems, surface aggregation, forest fires, electron localization [1], hopping conduction in amorphous solids [2], etc. can be readily modeled in terms of percolation concepts [3–5]. Moreover, lattice percolation provides the simplest geometrical model of a phase transition. Dominant concepts such as critical exponents, scaling laws, and universality classes are continuously being checked by investigating percolation models.

There exist two main categories concerning percolation processes: (a) simple percolation [also called random percolation (RP)] in which the elements of a lattice (sites and bonds in general) are occupied at random with a preset probability p; (b) correlated percolation (CP) where occupation probabilities of neighboring sites are not independent of each other (although the term correlated percolation is sometimes strictly reserved for systems with algebraic-law decaying correlations, we use it here with a broader significance). Most studies have dealt with RP. Recent comprehensive review articles on percolation formalism and applications are those in Refs. [3,6–8]. However, correlated percolation turns out to be a more interesting model since it can take into account natural correlations present in realistic systems such as magnetic systems below the critical temperature, insulator-metal alloys, fluid phase in porous media, etc. Two essential mechanisms by which correlations enter percolation can be distinguished: (1) interactions between the units representing occupied sites or bonds (for instance, the Ising model [9–11], dilute magnets [12]; (2) quenched statistical correlations between the metric of lattice elements (pore and throat size in a void porous space, local minima and saddle-point energy in an adsorption potential surface, mountain height and valley depth in landscapes, grain and border conductivity in inhomogeneous solid materials, etc.). Several authors have addressed mechanism (1); however, quenched disorder in percolation has been given significantly less attention [7,13].

It is known that short range correlations do not bring the results out of the RP universality class (although it has been recently argued that beta exponents may be affected [14,15]) while long range correlations may give critical exponents diverse from those of RP [7] (strictly, the correlation function has to decay as $C(r) \propto r^{2H}$, with $H < -1/\nu$ being the RP critical exponent). Concerning the percolation threshold, it has always been accepted that either site or bond correlations decreases the critical fraction at which a spanning cluster appears [6,12]. On one hand, scaling properties and critical exponents different from those of RP are significant from a fundamental viewpoint. On the other hand, cluster morphology and percolation threshold are important concepts in applied science since they determine work regimes and efficiency of technologically relevant processes such as oil-recovery, drying, etc.

Site-bond percolation was studied first in Refs. [16–18]. Furthermore, correlated site-bond percolation has also been investigated since the seminal papers in Ref. [19]. Here we present results for site-bond percolation and phase diagrams for a general class of self-consistent lattice models of inhomogeneous media, so-called dual site-bond model [20–22], that exhibit a nonmonotonic behavior of the percolation threshold not displayed by any other correlated site-bond model in the best knowledge of the authors. Moreover, the characteristics of correlated percolation can be traced to elementary statistical properties of the disordered media in which it develops.

II. MODEL OF CORRELATED SITE-BOND LATTICE

Let us assume a regular lattice of sites with connectivity c, connected by bonds. Disorder is simply introduced by characterizing sites and bonds by their size r_s, r_b (or any other property of sites and bonds applicable to a given physical process), having density frequency functions $f_s(r_s)$, $f_b(r_b)$ and cumulative distributions $S(r_s) = \int_0^{r_s} f_s(r)dr$, $B(r_b) = \int_0^{r_b} f_b(r)dr$, respectively. The most transparent counterpart of our model is a disordered porous medium

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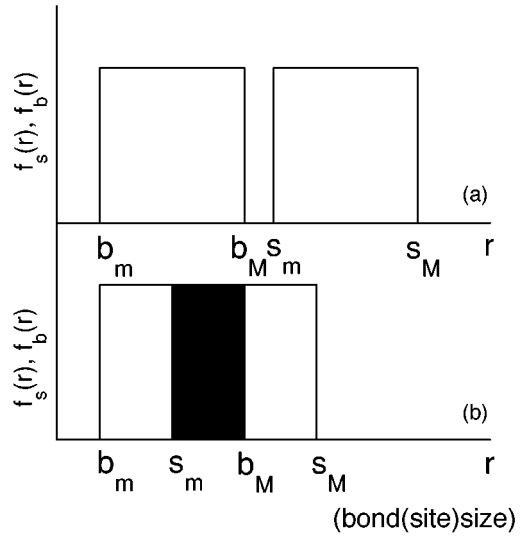

FIG. 1. Site and bond size density frequency distribution functions $f_s(r)$, $f_b(r)$; (a) null degree of overlapping, $\Omega=0$ (random site-bond lattice); (b) finite Ω $(0<\Omega\leqslant 1)$ (correlated site-bond lattice).

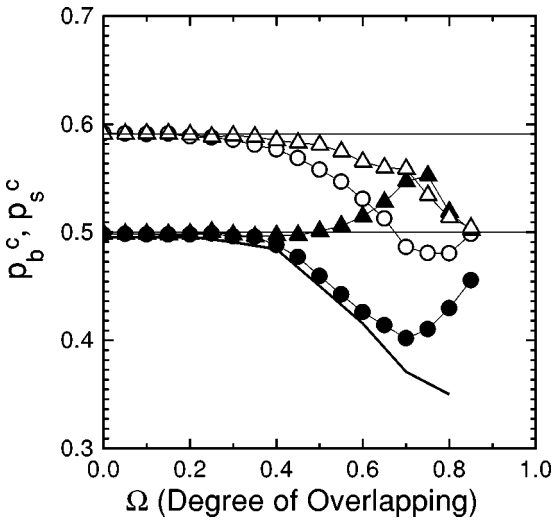

FIG. 2. Percolation threshold p_c vs degree of overlapping Ω between the distributions f_s, f_b. Symbols correspond to MC simulation on a square lattice of $L^2=700\times 700$ sites ($2\times 700\times 700$ bonds). Filled symbols correspond to pure bonds percolation ($p_s=1$) for procedure (P1) (filled triangles) and (P2) (filled circles), respectively. Pure site percolation ($p_b=1$) is shown with open symbols for (P1) (open triangles) and (P2) (open circles). A typical finite-size effect is shown in the line below, for data (filled circles) and $L^2=200\times 200$.

where pore bodies and throats are represented by sites and bonds of our lattice model, respectively [20]. There exists a natural constraint for sites and bonds, named the construction principle (CP), that requires the whole site-bond lattice to be self-consistent, which can be stated as follows: $r_{s_i}\geqslant r_{b_{ij}}$, $j=1,\ldots,c$: *for all* i, where the index i runs over the sites and j runs over the c bonds connected to the i site (sites are always equal in size or larger than bonds connected to them). This inequality can always be fulfilled if f_s and f_b do not overlap at all. However, if f_s and f_b do overlap (see Fig. 1), some sites and bonds cannot be nearest neighbors in the lattice, hence size correlation naturally arises. The elementary correlation can be explicitly described in its simplest way through a joint site-bond density function $f_{sb}(r_s,r_b)=f_s(r_s)f_b(r_b)\phi(r_s,r_b)$, where ϕ denotes a site-bond correlation function. If nothing but maximum configurational entropy (other than CP) is required in order to self-consistently distribute size for sites and bonds over the lattice, ϕ is given by the general expression

$$\phi(r_s,r_b)=\frac{\exp[\int_{B(r_b)}^{B(r_s)}dB/(B-S)]}{B(r_s)-S(r_s)} \quad \text{for } r_s\geqslant r_b$$

and $\phi(r_s,r_b)=0$ otherwise [20,23]. Let us denote Ω, the degree of overlapping between f_s and f_b, as the common area under both curves (shaded area in Fig. 1), then ϕ bears the following general properties: $\lim_{\Omega\to 0}\phi=1$ (random site-bond lattice); $\lim_{\Omega\to 1}\phi\propto\delta(r_s-r_b)$ (heterogeneous set of infinitely large homotatic domains). Ω is an elementary measure of site-bond size correlation.

This description provides a general framework for modeling inhomogeneous porous media, stochastic adsorption fields, etc., with spatial size correlations from the existence of two distinctive elements of the network that are identified, in general, as sites and bonds. It is known that real porous materials, such as the Indiana limestone, show correlation between pore bodies and throats sizes [25]. Futhermore, the simulation of site-bond lattice systems from general $f_s(r)$ and $f_b(r)$ functions turns out to be extraordinarily simple since the recent development of proper MC techniques [23]. It simply reduces to statistical relaxation driven by transitions fulfilling the CP, from an arbitrary initial configuration of site and bonds sizes sampled from f_s and f_b, respectively.

III. CORRELATED SITE-BOND PERCOLATION. RESULTS AND DISCUSSION

Percolation on a square lattice was studied by simulation for uniform site and bond density functions, $f_x(r_x)=1/(x_M-x_m)\forall x_m\leqslant r_x\leqslant x_M$ and $f_x(r_x)=0$ otherwise, where $x=s,b$ holds for sites and bonds, respectively (this is the simplest representation of a highly disordered system). Two main procedures were introduced to define the set of occupied elements: (P1) sites and bonds of increasing size are filled up starting from the smallest size (it corresponds to capillary condensation in porous networks); (P2) elements of decreasing size are filled starting from the biggest size (it resembles mercury intrusion and oil imbibition of a porous media). Accordingly, given p_s,p_b, the fraction of occupied sites and bonds, respectively, the filled phase embodies the elements that fulfill $r_s\leqslant S^{-1}(p_s)$, $r_b\leqslant B^{-1}(p_b)$ for (P1) and $r_s\geqslant S^{-1}(1-p_s)$, $r_b\geqslant B^{-1}(1-p_b)$ for (P2).

For pure bond ($p_s=1$) as well as pure site ($p_b=1$) percolation, the threshold p_b^c,p_s^c exhibits a minimum for (P2) as the overlap Ω increases (so does site-bond correlation) from 0 to 1 (Fig. 2). This turns out to be a remarkable behavior inasmuch as site-bond correlation only promotes cooperation between neighboring elements lowering the threshold for $\Omega<0.7$, while for $\Omega>0.7$ the greater the spatial correlation the more randomlike percolation behaves (typical lattice size was $L^2=700^2$ sites (2×700^2 bonds). Finite-size effects are shown in Fig. 2. The limit $p_b^c(0)=0.5$ is trivial. In addition,

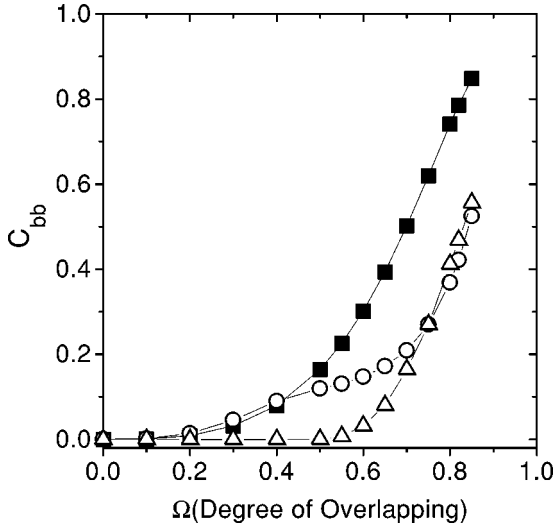

FIG. 3. Nearest neighbor (NN) bond correlation functions, $C_{bb,1}$ (open triangles), $C_{bb,2}$ (open circles), vs Ω corresponding to the domains Γ_1, Γ_2 defined by procedures (P1) and (P2) at percolation, respectively. Filled squares show C_{bb} for all NN bonds of the lattice.

$p_b^c(\Omega)=0.5$ for $\Omega\rightarrow 1$ would be expected since sites and bonds segregate into infinitely large domains containing elements of equal size with neither concave nor convex boundary, thus mapping the lattice problem onto continuum random percolation [24].

An even more unexpected finding is that $p_b^c(\Omega)$ shows a maximum for (P1) at approximately the same value of Ω as (P2). In other words, the critical fraction is larger than the one for random percolation, which, to the author's best knowledge, has never been observed before in two-dimensional (2D) randomlike percolation. We should bear in mind that these are straightforward consequences of the elementary construction principle (CP) and maximum entropy principle.

The facts can be rationalized by considering the influence of elementary correlations on the morphology of the sets Γ_1, Γ_2 including the elements that belong to the occupied phase defined by (P1) and (P2), respectively. We calculate the elementary correlation function

$$C_{bb,i}=\frac{\langle (r_b-\langle r_b\rangle)(r_b'-\langle r_b\rangle)\rangle}{\langle (r_b-\langle r_b\rangle)^2\rangle},$$

where r_b, r_b' are nearest neighbor bonds $\in\Gamma_i$ and $\langle r_b\rangle$, the statistical mean over the set, for $i=1,2$. All long-ranged correlations can be thought of as merely consequences of this basic correlation, so the topological features of percolation can already be inferred from $C_{bb,i}$. As Fig. 3 shows, $C_{bb,2}$ increases monotonically with Ω, thereby lowering p_b^c for $\Omega\leqslant 0.7-0.75$ as shown in Fig. 2. Within this range $C_{bb,1}$ is almost vanishing, thus in agreement with $p_b^c\approx 0.5$ for $\Omega\leqslant 0.6$; then p_b^c increases, reaching a maximum at $\Omega\approx 0.75$. This extraordinary increment appears because part of phase Γ_1 is embodied and isolated within phase Γ_2 under the form of small clusters (provided that $C_{bb,1}<C_{bb,2}$ and both domains topologically coexist), hence, it does not contribute to percolation as it would do for RP. When $C_{bb,1}>C_{bb,2}$ this

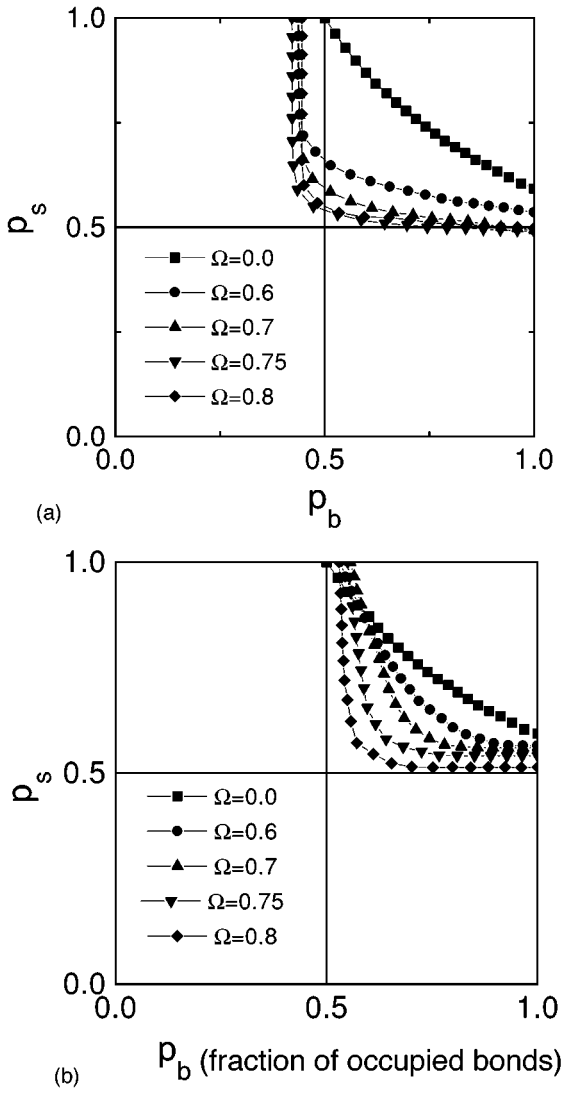

FIG. 4. Phase diagram for the percolating-nonpercolating phases as a function of the fraction of occupied sites, p_s (vertical axis) and bonds p_b (horizontal axis) (a) for (P2); (b) for (P1). As shown in the insets, the various symbols correspond to an increasing value of the degree of overlapping Ω between $f_s(r)$ and $f_b(r)$.

effect reverses and both percolation thresholds tend to 0.5 for $\Omega\rightarrow 1$ where the domains Γ_1 and Γ_2 turn out to be topologically equivalent. For pure-site percolation ($p_b=1$) also a local minimum appears for $R2$. The effect is smoother than the one for bonds because of the lower connectivity of sites.

All the observed extrema showed up at approximately the same value of Ω regardless of the particular mechanism to filling up elements of the lattice. This clearly displays a distinctive characteristic of the disordered media underlying percolation. The phase diagram for the percolation-nonpercolation transition (Fig. 4) shows the general case of site-bond percolation for both (P1) and (P2) rules. It can be observed that a mimimum of p_b^c is reached for (P2) even though the site occupation probability is other than one (for $p_s\leqslant 0.5$ percolation is not attainable in any way). Figure 4(b) displays a rich phase diagram where maxima in p_c^b are still possible for $p_s\geqslant 0.9$.

A simplistic physical effect of the nonmonotonic behavior

of $p_b^c(p_s^c)$ discussed above can be visualized by considering imbibition and drainage in 2D structures [26]. Although percolation does not rigorously match the characteristics of a fluid phase in a porous medium it is accepted that the error caused by modeling invasion percolation by means of RP is often negligible [4]. If we think of a 2D structure initially filled by a nonwetting (NW) phase that is displaced by a wetting (W) phase sequentially invading the smallest elements connected to the interface, the latter will break through the structure, forming a spanning cluster when the fraction of invaded elements reaches p_b^c (the limiting case of pure bond percolation is depicted by full triangles in Fig. 2). Since at this stage the fraction of the NW phase expelled from the network is also p_b^c (the remaining gets trapped in the form of clusters surrounded by the W fluid), the result in Fig. 2 means that there exists a correlated structure ($\Omega \approx 0.75$) for which the volume recovered (relative to the total volume) reaches a maximum before the invading fluid flows throughout it. Conversely, for drainage the NW phase displaces the W fluid from the largest pore connected to the interface as the external pressure is increased. The effect of topological correlation on percolation properties for drainage would rather match the results shown by circles in Fig. 2 (full circles and empty circles denote pure bond and pure site percolation, respectively). Henceforth, there exists a measure of correlations, $\Omega \approx 0.7$, for which the percolation threshold is minimum for drainage and maximum for imbibition in the same porous structure.

IV. CONCLUSIONS

Percolation features of a very general class of inhomogeneous site-bond lattices have been presented. All topological properties of the lattice elements are not *ad hoc* assumptions but straightforward consequences of two essential principles, that is, the construction principle and the maximum entropy principle. There is a dominant result in the present work that follows from the basic assumptions, namely, the correlation and topology of the set of small elements in the inhomogeneous network are, in general, distinct from those of the large elements. This is a general outcome of the dual site-bond model, which is expected to be valid in lattices that are not two dimensional. A precise resolution of small and large pore-size correlations in a void-space network of porous materials would be desirable to check the pertinence of the present description to real systems.

The study addresses an unexpected behavior of simple percolation properties of a disordered system conceived upon general grounds. The distinct behavior of the percolating phase of large sites and bonds compared with the corresponding to small ones, caused by the spatial size correlation between the lattice entities, appears as a unique characteristic of the inhomogeneous media that should considerably influence (and hence could be confirmed) physical processes taking place within the porous space like capillary condensation, mercury intrusion, imbibition, and drainage fluid phase morphologies.

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