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Effective electrical conductivity of microstructural patterns of binary mixtures on a square lattice in the presence of nearest-neighbour interactions

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HIGHLIGHTS

- The conductivity of binary mixtures using the continuum approach was studied.
- The influence on percolation threshold of interactions and temperature was examined.
- The thermodynamics properties of the binary mixtures was determined.

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ABSTRACT

The effective conductivity and percolative behaviour of microstructural patterns of binary mixtures are studied. Microstructure patterns are not entirely random, but result from the presence of attractive or repulsive interactions and thermal fluctuations. The interactions of the particles with one another lead to the formation of correlations between particle positions, while thermal fluctuations weaken these correlations. A simple lattice model is used, where each site is occupied by a single particle, and interactions can occur only between the nearest neighbours. The Kawasaki algorithm is adopted to create 2D microstructure samples. The microstructure is treated as a continuous medium, which means that the contribution from the flow through 'choke points' is taken into account in the calculation of the effective conductivity. We studied the thermodynamics of the system and its effective conductivity in a wide range of parameters. A change in the percolation threshold when the temperature changed was observed. The direction of the threshold shift depends on the sign of the interaction between the particles. In the high temperature range, we obtained a formula describing the dependence of the percolation threshold on temperature, as well as on the critical exponent.

1. Introduction

Studies of the macroscopic properties of random heterogeneous materials, despite having a long history, are still intensively developed because of their obvious technological importance and many uses in different fields [1,2]. Heterogeneous material is composed of domains of different materials (phases), such as a mixture or a composite, wherein the average domain size (the 'microscopic' length scale) is much larger than the molecular dimensions, but much smaller than the characteristic length of the macroscopic sample. Naturally, the macroscopic properties of heterogeneous materials, such

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as electrical conductivity, depend not only on the conductivity of the individual phases and their volume fractions, but also on the shapes and distribution of the domains (clusters), i.e. a microstructure pattern of random heterogeneous material.

There are very popular models in the study of disordered media, which start with a uniform host material and randomly introduced objects built of material different from the host material and having some given shapes. These objects can overlap freely, their centres being uniformly distributed, whereas their orientations are isotropically distributed only in some cases. Pike and Seager have done pioneer work on percolation and conductivity via computer study [3,4]. The general percolating problem of cutting randomly centred insulating holes in a two-dimensional conducting sheet have been examined by Garboczi et al. [5]. Recently, Matsutani et al. have investigated isotropic electric conductivity of a three-dimensional continuum percolation model consisting of overlapped spheroids of revolution in continuum [6], and Koza et al. have considered a model of overlapping squares or cubes randomly distributed on a regular lattice [7].

In the case of the approaches mentioned above, in which objects are introduced into a regular lattice, the pixels can be treated as the smallest portion of the object's material, as a particle occupying one site of the lattice. In such models, each object is represented by a cluster of pixels. The particles belonging to the cluster are spatially correlated. The length of the correlations depends on the size and the shape of embedded objects. However, spatial correlations also appear in microstructures, whose models are different from those mentioned above. For example, they can be observed in a system consisting of particles interacting with each other. Interactions tend to change the microstructure and consequently lead to the appearance of spatial correlations. Such correlations are not connected with specific objects larger than a particle.

The spatial correlations caused by interactions are important in the adsorption processes. Garcia et al. have studied the binary mixtures adsorbed on square lattices in Ref. [8] and, in Ref. [9], Quintana et al. have dealt with the percolation process caused by the random sequential adsorption on the heterogeneous triangular lattices. In both papers, the Monte Carlo method was used.

A few years ago, Hattori et al. presented a lattice model in conjugation with the Effective Medium Approximation (EMA) that describes electrical percolation in the microemulsion solution with interacting droplets [10]. This is an important point because in a system of interacting particles, the percolation behaviour is influenced not only by the volume concentration, but also by the interactions between particles, which involve correlation of positions. As it was mentioned in Ref. [10], some microemulsion systems show both an electrical percolation phenomenon, whose concentration threshold depends strongly on temperature, and a phase separation. Thus, an analytical prediction of the relationship between the percolation threshold and interactions is of some importance.

In Ref. [11], we extend the model proposed in Ref. [10] to any cluster of size, focusing only on the influence of geometrical features of local configurations at different length scales of clusters, whereas in Ref. [12], within the same framework, but with included interactions, we examined, based on small segments of microstructures (cluster of sizes $L = 2, 3, 4, 5$), in what way the interactions shift the percolation threshold.

In this article, we consider a two-phase, two-dimensional lattice model, in which some particles interact with the nearest-neighbours. The model delivers the patterns imitating the microstructure of mixtures (e.g. microemulsions) in the thermodynamic equilibrium. Unlike previous papers [10–12], we used neither the approximation of the mean field to determine the thermodynamic potentials of the system nor very rough assumptions about the effective conductivity of the microstructure. We apply the Monte Carlo method based on the Kawasaki dynamics [13] to probe the canonical ensemble, in order to determine the thermodynamic properties of the system. The ensemble is also called constant-NVT ensemble (especially when using computer simulation). Acronym NVT means that we focus on simulations of the system of a fixed number of particles (of each type) in a given volume and temperature. We obtained the global solution of the conductivity problem by means of numerical methods. For this purpose we used analytical local solutions, derived in Ref. [14], to build an appropriate linear system of equations. In this manner we minimize the influence of the discretization procedure on the effective conductivity of a sample; consequently, we include the contribution from the flow through 'choke points'. If an effective conductivity is concerned, the approach presented here belongs to the group of the continuum methods, i.e. it relies on the continuum model of the conductivities of the disordered materials [1,2].

We consider a square lattice in $d = 2$ dimensions with N sites. The occupation number c_i is equal to 1 if site i is occupied by a microemulsion droplet and 0 if it is occupied by another droplet. Alternatively and more generally, one may say that the system consists of sites occupied by two types of particles: interacting particles of A type ($c_i = 1$) and non-interacting particles of B type ($c_i = 0$). The Hamiltonian of the system with the constant number n_A of A type particles has the following simple form

$$H = -4J \sum_{\langle i,j \rangle} c_i c_j, \tag{1}$$

where: J is the interaction energy between two adjacent droplets/particles and the summation on $\langle i, j \rangle$ is taken over the nearest neighbour sites. (Factor 4 is needed for agreeing the temperature scale of the model with that of the Ising model.)

The assumptions of the mixture model presented here are analogous to those that underlie the well-known models, such as the Ising model (with conserved magnetization) or the Flory–Higgins model (used to describe binary polymer solutions, molecules in pure liquids and their solutions). Usually, the simplest medium field approximation (MFA) is used to study the thermodynamics of these models. In this paper, we avoided MFA, instead we used the Kawasaki algorithm, which belongs to the Monte Carlo methods. An additional advantage of this approach is that we have obtained microstructure samples using the Kawasaki algorithm, which we then examined for electrical conductivity.