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Tensor of effective susceptibility in random magnetic composites: Application to two-dimensional and three-dimensional cases

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ABSTRACT

The measuring of dynamic magnetic susceptibility by nuclear magnetic resonance is used for revealing information about the internal structure of various magnetoactive composites. The response of such material on the applied external static and time-varying magnetic fields encodes intrinsic dynamic correlations and depends on links between macroscopic effective susceptibility and structure on the microscopic scale. In the current work we carried out computational analysis of the frequency dependent dynamic magnetic susceptibility and demonstrated its dependence on the microscopic architectural elements while also considering Euclidean dimensionality. The proposed numerical method is efficient in the simulation of nuclear magnetic resonance experiments in two- and three-dimensional random magnetic media by choosing and modeling the influence of the concentration of components and internal hierarchical characteristics of physical parameters.

1. Introduction

Nuclear magnetic resonance (NMR) examination of complex, porous and heterogeneous materials yields many opportunities for assessing the ways in which the underlying structure influences observed change in magnetization through the physical processes of relaxation and dispersion [1,2]. NMR provides the basis for magnetic resonance imaging (MRI) and, as a consequence, is a powerful tool for medical applications, where it allows the resolution of abnormal growth, tissue configurations and reactions in the whole body [3,4].

NMR is fundamentally described by the Bloch equation: a set of linear, first-order ordinary differential equations [5]. This model is adequate for describing the evolution of magnetization in simple liquids and gels with a relatively homogeneous composition. However, characterizing highly random multicomponent structure requires a structural model with quantified parameters that predicts the result of the experiment by comparing the measurement to the prediction [6–8]. Interpreting experimental results involves identifying which model structural parameters most strongly affect magnetization while neglecting inessential remaining parameters.

Here we demonstrate that such a parameter as structural irregularity of the components of medium plays a key role in the description of random composites, which appears incredibly complex at the mesoscopic scales typically intermediating between the microscopic molecular dimensions, where material properties such as local magnetization originate, and the macroscopic sample dimensions [9–11].

In a previous study, we used a coarse-graining multi-scale model of random composites, with an expanded theoretical framework on different local constitutive laws to analyze dispersion of fully relaxing composites observed in diffusion, biorheology and elastography [12–14]. Our current work provides further enhancement of earlier proposed Euclidean two-dimensional (*2D*) and three-dimensional (*3D*) models of relaxivity for the problems of resonating magnetoactive composites enabling a link between the macroscopical effective properties of susceptibility to constants of susceptibility and random geometry of components of medium on the mesoscale.

The concept of multi-scale coarse-graining is adapted from statistical physics of critical phenomena [15–17] and, in our context, it implies the global hierarchical similarity between different structurally complex specimens that appears at intermediate distances whenever specimens are sufficiently coarse-grained, such that local differences between statistical realizations of disorder become inessential.

Our paper is arranged as follows. The *Theory* section gives a brief

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description of the function and subtleties of a multi-scale algorithm seen from the design of the hierarchical Bloch equation viewpoint. The section *Results of numerical simulations* explains the parameters we used, details of the calculation, and describes the outcome of our algorithm. We round off the paper with a *Discussion and conclusion* of our work.

2. Theory

2.1. A hierarchical model of binary random magnetic composite: 2D and 3D cases

Binary composite can be modelled as a two-phased random structure, which can be mapped onto *2D* square and *3D* simple cubic lattices and investigated by percolation theory methods [15,18]. Assume that each bond of the lattice is colored "black" with probability p, and "white" with probability q, representing components (or phases) of composite and satisfying the condition:

$$\begin{cases} p+q=1 \\ \tilde{D}(p,q): 0\le p\le 1; 0\le q\le 1 \end{cases} \tag{1}$$

where $\tilde{D}(p,q)$ is a probability domain.

The physical properties of such material can be described by the generalized binary local probability density function:

$$\rho\left(\chi_{(km)}\right)=p\delta\left(\chi_{(km)}-\chi^{(1)}\right)+q\delta\left(\chi_{(km)}-\chi^{(2)}\right), \tag{2}$$

where $\delta(x)$ is a Dirac delta function. In Eq. (2) every bond is bounded with the closest neighbour nodes (km) in the lattices of N inner and N_Γ contact boundary Γ nodes. The bond can either belong to phase (*1*) or (*2*) characterized by parameters of susceptibility $\chi^{(1)}$ and $\chi^{(2)}$ correspondingly.

We model global (or effective) properties of a magnetic material with randomly distributed local properties according to a real space renormalization group method [19,20]. The main idea of this method consists of replacing a lattice of size l of iterative level n and density function $\rho_n(\chi_{(km)})$ into a lattice of size bl on $n+1$ level with density function $\rho_{n+1}(\chi_{(k'm')})=T(\rho_n(\chi_{(km)}))$ and rescaling factor b. To this end, each realization of Eq. (2) should be renormalized to a rudimentary unit of the transformed lattices. Recursive repetition results in a self-similar structure that hierarchically repeats the connectedness of the original bonds. If we consider only the horizontal or vertical spanning of the renormalization element of the lattice, the connectivity in component (*1*) can be determined only by five bonds in *2D* (Fig. 1a) and twelve bonds in *3D* (Fig. 1b). The procedure of renormalization is illustrated in Fig. 1a,b. On the left-hand part of Fig. 1a,b we show a section of the original lattice while on the right the same section is shown after renormalization. The iterative transformation, T, of generalized probability density function of Eq. (2) is defined as [21–23].

$$\rho_{n+1}\left(\chi_{(k'm')}\right)=\int_E \prod_{\substack{k\in S\backslash S_\Gamma \\ m\in S}}^{k\neq m}\left(d\chi_{(km)}\rho_n(\chi_{(km)})\right)\delta\Big(\chi_{(k'm')}-\Big\langle F\Big(\Big\{\chi_{(km),n}\Big\}\Big)\Big\rangle\Big), \tag{3}$$

where $E|_{2D}=\{E^{(1)}\cup E^{(2)}\}$ and $E|_{3D}=\{E^{(1)}\cup E^{(2)}\}\otimes\{E^{(1)}\cup E^{(2)}\}\otimes\{E^{(3)}\cup E^{(4)}\}\otimes\{E^{(3)}\cup E^{(4)}\}$.

S is a surface covering N and N_Γ nodes, and $S\backslash S_\Gamma$ is a surface where nodes N_Γ are excluded. $\langle F(\{\chi_{(km),n}\})\rangle$ is the effective susceptibility function of a lattice, delineated via a rudimentary element bounded by nodes (km), which was averaged over all configurations E in a way to preserve the invariant form of Eq. (2) for every nth scale of recursive building of the hierarchical lattice. In the *2D* case E, consisting of 2^5 elements, can be easily represented as unification, $\cup$, of connected, $E^{(1)}$, and disconnected, $E^{(2)}$, clusters (Fig. 1a). The set E in the *3D* case can be calculated as a direct product, $\otimes$, of elements of sets $E^{(1)}$, $E^{(2)}$, and possible configurations $E^{(3)}$ and $E^{(4)}$ for a single bond (Fig. 1b). Thus renormalizing lattice in the *3D* case is considered as a combination of two *2D* lattices connected through two bonds. In *3D* the number of elements in a set E is equal 2^{12}.

The transformation of p in Eq. (3) corresponds to connectedness function, $R(p)$, and can be expressed by the polynomial equations

$$R(p)|_{2D}=p^5+5p^4(1-p)+8p^3(1-p)^2+2p^2(1-p)^3, \tag{4}$$

and

$$\begin{aligned} R(p)|_{3D}=\;& p^{12}+12p^{11}(1-p)+66p^{10}(1-p)^2+220p^9(1-p)^3 \\ &+493p^8(1-p)^4+776p^7(1-p)^5+856p^6(1-p)^6 \\ &+616p^5(1-p)^7+238p^4(1-p)^8+48p^3(1-p)^9 \\ &+4p^2(1-p)^{10}. \end{aligned} \tag{5}$$

Thus the iterative recursion of p_n in Eq. (3) is defined as

$$p_{n+1}=R(p_n), \tag{6}$$

and then Eq. (3) can be written as

$$\begin{aligned} \rho_{n+1}\left(\chi_{(k'm')}\right)=\;& R(p_n)\delta\Big(\chi_{(k'm')}-\Big\langle F\Big(\Big\{\chi_{(km),n}\Big\}\Big)\Big\rangle^{(1),n+1}\Big)+(1 \\ &-R(p_n))\delta\Big(\chi_{(k'm')}-\Big\langle F\Big(\Big\{\chi_{(km),n}\Big\}\Big)\Big\rangle^{(2),n+1}\Big) \end{aligned} \tag{7}$$

where the function of equivalent susceptibility is separated into components (*1*) and (*2*) depending on the connectedness of bonds in set E.

The roots p_* of Eqs.(4) and (5) in the physical meaningful range of $p_*\in[0,1]$ are $p_*|_{2D}=\{0,0.5,1\}$ and $p_*|_{3D}=\{0,0.2085,1\}$. The first and third roots represent a lattice fully occupied by phases (*2*) or (*1*), respectively. The second root is unstable, i.e. a small perturbation in the probability p leads to stable homogeneous components [21]. The number of iterations for building a hierarchical lattice is defined by the constraint $|p_n-p_{n+1}|\le\varepsilon$, where $\varepsilon>0$ is an infinitely small number. Due to this condition, the two components of a composite are indistinguishable after multi-scale hierarchical averaging:

$$\begin{bmatrix} \lim\limits_{n\to\infty} R_b(p_n)\Big|_{p_0\in D_{(0,1)}(p,q)}=0, & \lim\limits_{n\to\infty}\chi^{(2),n}\Big|_{p_0\in D_{(0,1)}(p,q)}=\chi^{(eff)} \\ \lim\limits_{n\to\infty} R_b(p_n)\Big|_{p_0\in D_{(1,0)}(p,q)}=1, & \lim\limits_{n\to\infty}\chi^{(1),n}\Big|_{p_0\in D_{(1,0)}(p,q)}=\chi^{(eff)} \end{bmatrix} \tag{8}$$

leading to a convergence of binary properties to solitary

$$\lim_{n\to\infty}\rho_n\left(\chi_{(km)}\right)\Big|_{\forall p_0\in D(p,q)}=\delta\left(\chi_{(km)}-\chi^{(eff)}\right), \tag{9}$$

with effective properties corresponding to component (*1*) or (*2*) depending on selection of initial probability p_0 at the first iteration