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EFFECTIVE PROPERTIES OF MACROSCOPICALLY INHOMOGENEOUS MEDIA WITH RESTRUCTURING

The problems of describing kinetic phenomena in macroscopically randomly disordered media have been considered. These are the tasks that P.M. Tomchuk dealt with. Some of the previously obtained results were described in a review by P.M. Tomchuk in the Ukrainian Journal of Physics. In this issue, the current state of the theory that describes a new type of material with a variable structure is presented. One group of such materials includes magnetoelastomers, which have various applications in technology and medicine. The application of the concept of a moving percolation threshold, which was introduced to describe the magnetic and elastic properties of magnetoelastomers, is described.

Keywords: elastic properties, effective medium approximation, self-consistent random heterogeneous medium, two-phase composite material, percolation threshold.

1. Introduction. Description Methods

By macroscopically inhomogeneous media are understood those media in which the characteristic size of inhomogeneities is much larger than any microscopic characteristic length. For example, if the matter concerns the current flow, the inhomogeneity size must be much larger than the characteristic free path lengths of current carriers. In particular, this means that such a macroscopically inhomogeneous medium can be characterized by a local Ohm's law, which relates the electric current density to the electric field strength at every point in the medium.

A macroscopic inhomogeneity, for example, conductivity, can be continuous; then the specific conductivity $\sigma(\mathbf{r})$ is a continuous function of coordinates and a discrete. In the latter case, we speak of two-, three-, and so forth phase media, where, by the phase is understood a set of regions of the same type with this specific conductivity value.

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The main parameters of a transport processes in macroscopically inhomogeneous media are effective kinetic coefficients, which characterize the medium as a whole, i.e., at lengths much larger than the characteristic size of macroscopic inhomogeneities. When calculating the effective kinetic coefficients, the local properties of the medium can be given in two different ways: either by assuming that the dependences of the local kinetic coefficients on coordinates are known (the deterministic approach) or by determining the local kinetic coefficients as random fields (the statistical approach). Each of those approaches has its advantages and disadvantages. The deterministic approach makes it possible to study, as a rule, media with the simplest structure; the difficulties of the statistical approach are associated with the complexity of comparing the examined media and the corresponding random fields of local kinetic coefficients. If following a strict theoretical approach to calculating the effective kinetic coefficients of randomly inhomogeneous media, the task should be divided into two stages: (i) the calculation of the effective kinetic coefficients with fixed dependences of local kinetic coefficients on the coordinates and (ii) a subsequent averaging over various implementations [1–5].

A rather unexpected fact consists in that there are problems aimed at determining effective properties

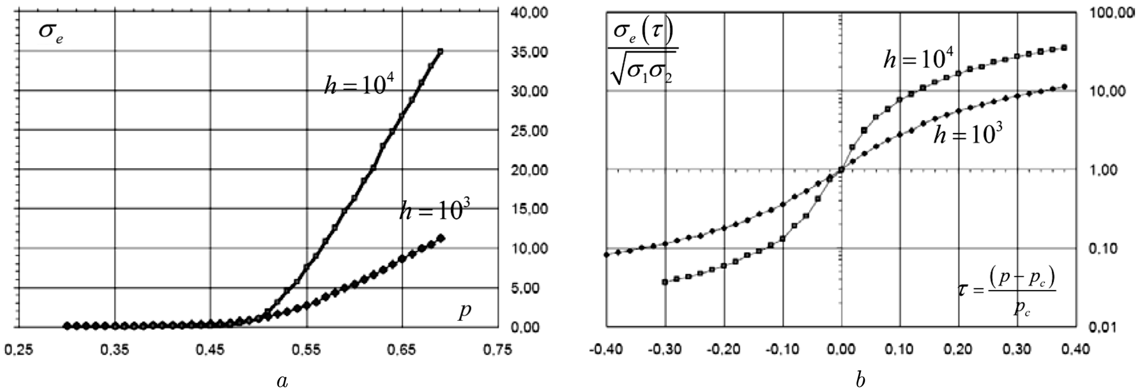

Fig. 1. Curves characterizing the critical behavior of the effective conductivity σ_e of a two-dimensional resistance lattice at various phase conductivity ratios $h = \sigma_2/\sigma_1$: (*a*) in the $\sigma_e - p$ axes and (*b*) in the $\sigma_e - \tau$ axes (adapted from work [5])

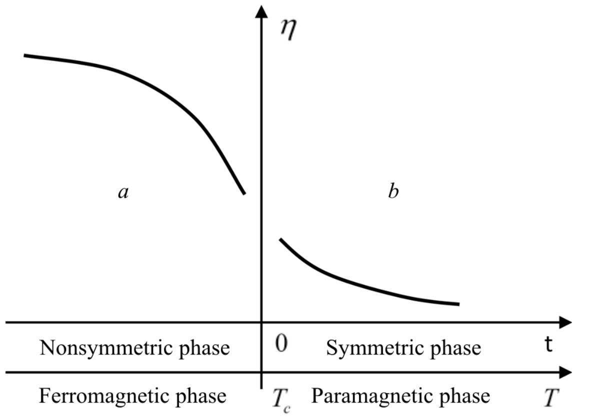

Fig. 2. Dependence of the order parameter on the temperature $t = T - T_c$, where T_c is the critical temperature (adapted from work [5])

in the media with not macroscopic, but microscopic inhomogeneities, which are reduced to the above-mentioned macroscopic problems. Such problems include, for example, the problem of high-temperature jump conductivity in doped semiconductors.

The problem of calculating effective kinetic coefficients was posed for the first time as long ago as by Maxwell and developed by Clausius, Mosoti, Lorenz, and Garnett [6]. The obtained approximations work well, only if the concentration of inclusions is low. However, they have been and are used in a huge number of applications describing the properties of a wide variety of composites.

In the 1930–1940s, in order to calculate the effective kinetic coefficients, the method of self-consistency belonging to the mean field theory was applied: the Bruggeman approximation [7] and – independently, but later – the Landauer one [8]. This approximation is often called the effective medium approximation (EMA). The EMA was developed for various physical properties, in particular, for conductivity (by Bruggeman and Landauer) and for elastic moduli (Budiansky) [9,10].

In the case of two-phase strongly inhomogeneous media, if the concentration of the well-conducting component (phase) grows and reaches a certain value, the effective conductivity drastically increases (see Fig. 1). A qualitative explanation of this phenomenon consists in that there emerges a continuous path (an infinite cluster) in the well-conducting phase [2–5]. Such a concentration value was called the flow threshold, and the phenomenon itself was called percolation. After Broadbent and Hammersley introduced the flow threshold notion and found that (i) various geometric and physical characteristics of percolation systems depend on their proximity to the flow threshold, (ii) this dependence has a stepwise form, and (iii) the critical indices that describe this dependence are universal, the idea of that this is percolation of the 2nd kind could not help but arise. This means that, near the percolation threshold, the effective conductivity can be understood as (and described in terms of) an order parameter of second-order phase transitions (see Fig. 2).

The percolation approach (and the development of a hierarchical model of the percolation structure [5]) made it possible to give a visual description for a wide class of problems: thermo- and galvanomagnetic phenomena, flicker noise, Abrikosov vortex pinning, Anderson localization in percolation structures, high-temperature jump

Recently, there appeared a new class of macroscopically inhomogeneous media. For their description, the above-mentioned approaches had to be modified.

2. Magnetically Active Elastomer

At the beginning of the 2010s, a magnetically active elastomer (MAE) was created [11, 12]. It consisted of two phases: phase 1 is carbonyl iron (balls a few microns in size and with no own magnetic moment), and phase 2 is polydimethylsiloxane (a soft, highly elastic matrix). Separately, the phases reacted to the magnetic field in a simple, obvious way. Namely, there emerged magnetic moments in the metal balls, and the matrix practically did not react to the magnetic field. Quite unexpectedly, the combination of those phases demonstrated a number of unusual (extraordinary!) properties in the magnetic field. For example, the effective value of the shear modulus of the composite increased by more than 1,000 times, when a relatively low magnetic field of about 0.6 T was switched on. In other words, a piece of soft rubber became a hard plate. Another unexpected property was the dependence of the effective dielectric permittivity of MAE on the magnetic field (despite the fact that the materials of both phases do not change their dielectric properties in the magnetic field).

During the following years, there appeared a large number of theoretical works attempting to construct a theoretical model describing the set of detected effects. In the models of the first type [13], the behavior of MAE in a magnetic field was related to the interaction between the magnetic moments induced on inclusions (carbonyl iron balls). The elastic stress that emerged between the interacting balls due to their magnetic moments changed the elastic state of the sample as a whole and thereby changed the effective elastic properties of the material (including the shear modulus). Despite the fact that such an interaction exists and affects the effective properties, such a model is clearly insufficient to describe the MAE. First, the calculations showed that the resulting increase of elastic moduli is not large. Second, which is more important, the proposed mechanism does not affect the dielectric constant in a magnetic field.

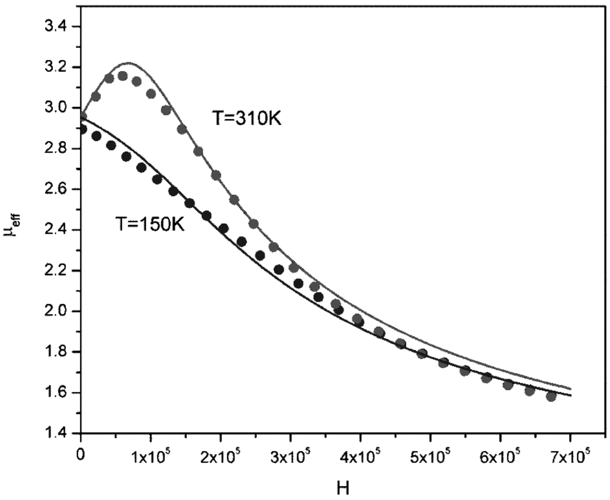

Fig. 3. Dependences of the effective magnetic permeability on the magnetic field A/m (adapted from work [19])

The models of the second type [14] are based on the rotation of inclusions so that the induced magnetic moment directed along the easy magnetization axis of the particle becomes oriented as close as possible along the external magnetic field direction. Such a rotation "twists" the matrix, creates stresses and strains in it, and, as was shown, increases the effective elastic moduli. Using the Padé approximant method, it was shown that a large growth of the shear modulus value is possible [15]. However, as in the first-type models, the indicated shortcomings are also present here.

In works [16–18], when measuring the effective magnetic permeability at various temperatures, it was experimentally shown that the properties – for example, the effective magnetic permeability – behave according to the standard mean field theory at liquid nitrogen temperature (when the motion of inclusions is impossible), but there appear peculiarities at room temperature (see Fig. 3 [19]). In works [20, 21], the measurements of this type were reproduced, which confirmed the results of works [16–18].

When introducing a MAE sample into a magnetic field, the particles should shift and change their positions. Later [22, 23], using microwave examination of samples, the displacement of inclusions in a magnetic field was directly shown. In particular, this displacement means that the theoretical description of the effective properties of the MAE in the framework of the known theories (the mean field theory, the percolation theory, and so forth), where the composite

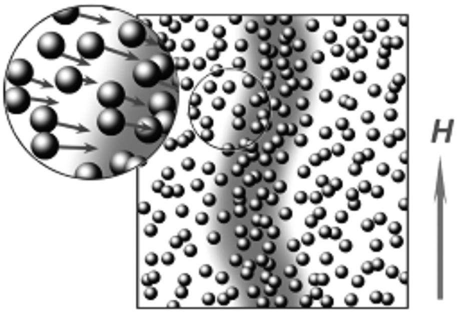

Fig. 4. Concentration growth in a pre-percolation cluster in a magnetic field

structure does not change when an external field is applied, is impossible. So, a fundamental change in the theoretical approach is required.

In work [24], when analyzing experimental data, it was proposed to use the percolation theory and, within its framework, specify the dependence of the percolation threshold on the magnetic field. This approach contains a fundamentally new idea; however, it is quite approximate (fitting) for the description of MAE because the percolation dependences of the effective coefficients are applicable (valid) only in a very narrow interval and provided a large ratio between the magnetic permeabilities (or conductivities, dielectric constants, and so forth).

To build a model that involves the restructuring (displacement of particles of the first phase inclusions when an external magnetic field is turned on), it is necessary to determine, under what forces, this occurs.

In experiments with the MAE, the concentration of inclusions is high and close to but less than the percolation threshold, and it is necessary to consider that a "pre-percolation" structure has already been formed in the medium. There are the so-called "lattice beasts": clusters of finite size composed of inclusion particles; some of them, as the concentration grows further, form a structure (a grid) of an infinite cluster [4]. Accordingly, the internal magnetic field in the sample can no longer be considered uniform; its magnitude will increase in the cluster regions and decrease in the gaps. At the same time, the Kelvin force proportional to the square gradient of the magnetic field will act on the magnetic moments of the inclusion particles. The particles will be attracted to the finite clusters, thus increasing their density (the local concentration); see Fig. 4.

The main idea of the theoretical description of the effective properties in the case where the inclusion particles move is that the growth of their concentration in the pre-percolation clusters can be interpreted as a reduction of the percolation threshold. That is, the difference between the percolation threshold and the concentration, $p_c - p$, decreases, but not at the expense of increasing p, but of decreasing p_c. With such a description, the percolation threshold is no longer a constant, but it is a function of the magnetic field, and this function decreases with the increasing magnetic field.

For a quantitative description of the effective properties of such media (beyond a narrow critical concentration interval belonging to the percolation theory), an EMA modification can be applied. In the standard EMA, for example, when describing the concentration behavior of the effective dielectric constant, the percolation threshold p is given by the equation

$$\frac{\varepsilon^e - \varepsilon_1}{\varepsilon_1 + 2\varepsilon^e} p + \frac{\varepsilon^e - \varepsilon_2}{\varepsilon_2 + 2\varepsilon^e} (1-p) = 0, \tag{1}$$

and in the three-dimensional case, $p = 1/3$.

In work [26], an EMA modification was proposed to describe the effective galvanomagnetic properties of composites. Her the percolation threshold p_c can be given any value. The corresponding equation looks like

$$\frac{\frac{\varepsilon^e - \varepsilon_1}{\varepsilon_1 + 2\varepsilon^e}}{1 + c(p, p_c) \frac{\varepsilon^e - \varepsilon_1}{\varepsilon_1 + 2\varepsilon^e}} p + \\ + \frac{\frac{\varepsilon^e - \varepsilon_2}{\varepsilon_2 + 2\varepsilon^e}}{1 + c(p, p_c) \frac{\varepsilon^e - \varepsilon_2}{\varepsilon_2 + 2\varepsilon^e}} (1-p) = 0, \tag{2}$$

where

$$c(p, p_c) = (1 - 3p_c) \left(\frac{p}{p_c}\right)^{p_c} \left(\frac{1-p}{1-p_c}\right)^{1-p_c}. \tag{3}$$

Now, when describing the effective properties of the MAE, this percolation threshold becomes dependent on the external magnetic field. This dependence was taken in the form proposed in work [25]

$$p_c(|\langle \mathbf{H} \rangle|) = p_c(0)\, e^{-\frac{(|\langle \mathbf{H} \rangle|)}{H_c}}, \tag{4}$$

where H_c is a characteristic normalizing field, which is usually called critical.

This approach made it possible to describe the dependence of the effective magnetic permeability on the magnetic field (Fig. 5) (the cited authors also considered the nonlinear dependence of the magnetic permeability of inclusions on the magnetic field, which is associated with the nonlinearity of the magnetization curve); see the solid curve and its comparison with experimental data in Fig. 3 [2].

In work [27], the equations of the mean field theory for the dielectric permittivity were written with regard for the anisotropy, and the dependence of magnetic permeability on the magnetic field was found.

An approach similar to the moving threshold approach was used to describe the elastic properties of the MAE. The equations of mean field theory for a two-phase randomly inhomogeneous medium (the Budiansky approximation) [28] look like

$$\begin{cases}\Omega_1 p + \Omega_2\,(1-p) = 0,\\ \Theta_1 p + \Theta_2\,(1-p) = 0,\end{cases} \tag{5}$$

where

$$\begin{aligned}
\Omega_i &= \frac{\frac{G_i}{G_e}\frac{1+\nu_i}{1+\nu_e}\frac{1-2\nu_e}{1-2\nu_i} - 1}{1+\alpha_e\left(\frac{G_i}{G_e}\frac{1+\nu_i}{1+\nu_e}\frac{1-2\nu_e}{1-2\nu_i} - 1\right)},\\
\Theta_i &= \frac{\frac{G_i}{G_e} - 1}{1+\beta_e\left(\frac{G_i}{G_e} - 1\right)},\\
\alpha_e &= \frac{1}{3}\,\frac{1+\nu_e}{1-\nu_e}, \quad \beta_e = \frac{2}{15}\,\frac{4-5\nu_e}{1-\nu_e},
\end{aligned} \tag{6}$$

$i = 1$ and 2, G_i are the phase shear moduli, and ν_i are Poisson's moduli.

Like the Bruggeman–Landauer approximation, the Budiansky approximation must be modified by introducing a moving percolation threshold. For this purpose, term (3) has to be changed. In general, the EMA approximation for the elastic problem with a moving percolation threshold was introduced in works [29, 30] as follows:

$$\left.\begin{aligned}\frac{\Omega_1}{1+s(p,\tilde{p}_c)\Omega_1}p + \frac{\Omega_2}{1+s(p,\tilde{p}_c)\Omega_2}\,(1-p) = 0\\ \frac{\Theta_1}{1+s(p,\tilde{p}_c)\Theta_1}p + \frac{\Theta_2}{1+s(p,\tilde{p}_c)\Theta_2}\,(1-p) = 0\end{aligned}\right\}, \tag{7}$$

where

$$s(p,\tilde{p}_c) = (1-2\tilde{p}_c)\left(\frac{p}{\tilde{p}_c}\right)^{\tilde{p}_c}\left(\frac{1-p}{1-\tilde{p}_c}\right)^{1-\tilde{p}_c}. \tag{8}$$

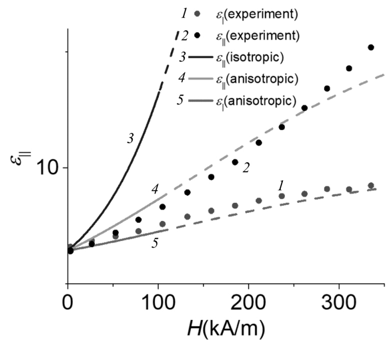

Fig. 5. Dependences of the components of the effective permittivity tensor on the magnetic field

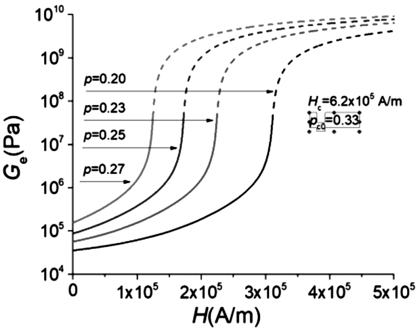

Fig. 6. Dependences of the effective elastic modulus on the external magnetic field

The obtained dependences of the shear modulus on the magnetic field demonstrate the experimentally found gigantic growth; see Fig. 6.

Of course, the proposed model is an approximation where a number of phenomena are not taken into account; for example, the appearance of anisotropy at the locations of inclusions (the so-called geometric anisotropy).

3. Conclusions

It is obvious that the MAE – the composite with interaction via both "magnetic" and elastic forces, and with a structural rearrangement–can hardly be described in the framework of one simple model for various phenomena: elasticity, magnetic permeability, dielectric properties, and so on. At different field, concentration, and temperature values, different mechanisms (particle–particle dipole interaction, rotation of a particle in an external magnetic field, displacement of particles due to the local field nonuniformity, *etc.*) can play different roles. More complicated mechanisms are also possible; for example, the dipole interaction between the conglomerates of several particles, their rotation, and displacement. Currently, we may only state that according to the studies described above, the main mechanism that allows the phenomena to be explained in a unique way (qualitatively, and sometimes quantitatively) is the displacement of inclusion particles under the action of internal nonuniform magnetic fields and to be described as an approach to the percolation threshold (the percolation threshold shift).

In the proposed model of the moving percolation threshold, the dependence of the percolation threshold on the external magnetic field with a characteristic constant, the critical field H_c was introduced. Of course, this is a fitting constant that is chosen to satisfy experimental data. A confirmation of the logic character and consistency of the theory of the moving percolation threshold consists in that the value of this constant, which is chosen, for example, to describe the effective properties of magnetic permeability, remains practically the same for other physical phenomena, for example, elastic properties.

Here is a brief list of the problems and tasks that can be studied and solved using the concept of the moving percolation threshold:

• the account of the structural anisotropy of inclusions in elasticity problems;

• the study of interaction between the inclusions near pre-percolation structures in order to calculate the critical field value and its dependence on the medium parameters;

• the development of a model for the strictions of a finite-size MAE sample;

• the study of temporal processes in MAE, the determination of the relaxation time and the imaginary parts of elastic moduli.

To a great extent, the development of the theory of macroscopically inhomogeneous media by a group of researchers, including the author, is indebted to P.M. Tomchuk, my participation in the seminars held under his supervision, and constant personal communication. The author is grateful to B.I. Lev, M. Shamonin, V.M. Kalita, and I.V. Bezsudnov for numerous fruitful discussions of the raised problems and relevant comments.

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ЕФЕКТИВНІ ВЛАСТИВОСТІ
МАКРОСКОПІЧНО НЕОДНОРІДНИХ
СЕРЕДОВИЩ ІЗ РЕСТРУКТУРИЗАЦІЄЮ

У невеликому огляді розглянуто проблеми опису кінетичних явищ у макроскопічно випадково невпорядкованих середовищах. Завдання, якими займався П.М. Томчук. Частину раніше одержаних результатів було описано в огляді П.М. Томчука в Українському фізичному журналі. Описано сучасний стан теорії, створеної для дослідження нового типу матеріалів із змінною структурою. Одним із таких матеріалів є магнітоеластомери, що мають різноманітні застосування в техніці та медицині. Описано використання введеного поняття рухомого порогу протікання для опису магнітних та пружних властивостей магнітоеластомерів.

Ключові слова: пружні властивості, наближення ефективного середовища, самоузгоджене випадкове гетерогенне середовище, двофазний композитний матеріал, поріг перколяції.