Effective magnetic permeability of composite materials near the percolation threshold

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A proposed cluster model is used to determine analytically the effective magnetic permeability of a composite system when this permeability is due to generation of Foucault (eddy) currents in an alternating magnetic field. It is shown that an allowance for the inductive interaction of currents removes the divergence of the imaginary part of the effective magnetic permeability near the percolation threshold predicted in earlier investigations [P. G. De Gennes, C. R. Acad. Sci. **292**, 701 (1981); M. J. Stephen, Phys. Lett. A **87**, 67 (1981); D. R. Bowman and D. Stroud, Phys. Lett. **52**, 299 (1984)]. This result is generalized to the behavior of the diamagnetic susceptibility of superconducting composites.

Intensive theoretical and experimental investigations of disordered systems consisting of metal and insulator particles are currently under way. A characteristic feature of such composites is the existence of a critical concentration p_c of metal particles known as the percolation threshold. At this concentration the system first exhibits a nonzero conductivity. Electrophysical properties of disordered systems are characterized by a number of anomalies near the percolation threshold. For example, the permittivity and the inductance of composite materials become infinite as $p \to p_c$ (Refs. 1–5). It is pointed out in Refs. 6–13 that the diamagnetic susceptibility of composite superconductors diverges near the percolation threshold. The generalization of this result to the case of normal composite systems[6,7,11] gives rise to a divergence in the limit $p \to p_c$ ($p < p_c$) for the imaginary part of the magnetic permeability due to generation of eddy currents by an alternating magnetic field.

These investigations of magnetic properties of composite systems have ignored the effects of the inductive interactions of currents, but we shall show that these effects are of fundamental importance close to p_c.

We shall investigate theoretically the effective magnetic permeability

$$\mu_{eff} = \mu'_{eff} + i\mu''_{eff}$$

of composite materials consisting of nonmagnetic conducting inclusions and we shall allow for the inductance effects. The influence of the inductive interaction of currents ensures that μ''_{eff} remains finite in the limit $p \to p_c$. The diamagnetic susceptibility of superconducting composite materials is also finite near p_c. We shall show that the frequency dependence of the effective magnetic permeability $\mu_{\text{eff}}(p, \omega)$ exhibits a resonance.

We shall explain the reason for the possible divergence of the magnetic susceptibility predicted in Refs. 6–13 when the inductive interaction is ignored. Composite systems are characterized by the appearance of large conducting clusters. Eddy (Foucault) currents are induced when an external alternating magnetic field

$$\mathbf{H} = \mathbf{H}_0 \exp(-i\omega t)$$

is applied to systems containing such clusters. The Foucault currents are accompanied by dissipation of the energy of the electromagnetic field governed by the imaginary part of the magnetic polarizability α_{ik} (Ref. 14):

$$Q = {}^1/_2 \omega V \alpha_{ik}'' \operatorname{Re}\{H_i H_k\},$$

where V is the volume of the conducting material in a cluster. A cluster has a strongly branched structure formed by conducting particles. According to Refs. 6–8, the value of α'' for such a cluster is given by

$$\alpha''(\omega) = S_{eff}/(4\pi\delta)^2,$$

where $\delta = c/(2\pi\sigma\omega)^{1/2}$ is the depth of penetration of an electromagnetic field ($\delta \gg a$, where a is the characteristic size of the conducting particles), σ is the conductivity, and S_{eff} is the effective area of a cluster which depends on its topology. Since the magnetic-dipole interaction is ignored, the polarizabilities of the clusters α'' make an additive contribution to the total magnetic susceptibility of the composite system. In this approximation the susceptibility χ is found by summing the relevant series S_{eff} of the individual clusters. The analytically values of S_{eff} can be calculated only for simple two-dimensional structures (such as, for example, a periodic square lattice[8,10] or a Sierpiński carpet[7]). In the case of real two- and three-dimensional systems it is necessary to use numerical methods. The results of a numerical calculation reported in Ref. 11 predict divergence of the value of χ'' near p_c:

$$\chi'' \propto \tau^{-b}, \quad \tau = (p_c - p)/p_c \tag{1}$$

with a critical exponent $b = 1.29$ if the dimensionality is $d = 2$ and $b = 0.35$ if $d = 3$. Since the diamagnetic susceptibility of superconducting composites is governed by the same geometric factor S_{eff} (Refs. 6 and 7), it should diverge in the limit $p \to p_c$ and the critical exponents should be the same.

We can propose a fairly simple model which makes it possible to determine analytically the magnetic susceptibility of a composite system. Each cluster represents a set of doubly connected regions. Then doubly connected clusters have a scale-invariant drop structure[15] (see Fig. 1). If we ignore the inductance effects, these doubly connected clusters interact with an alternating magnetic field **H** independently of one another. We shall assume that, relative to the field **H**, a doubly connected cluster is equivalent to a contour

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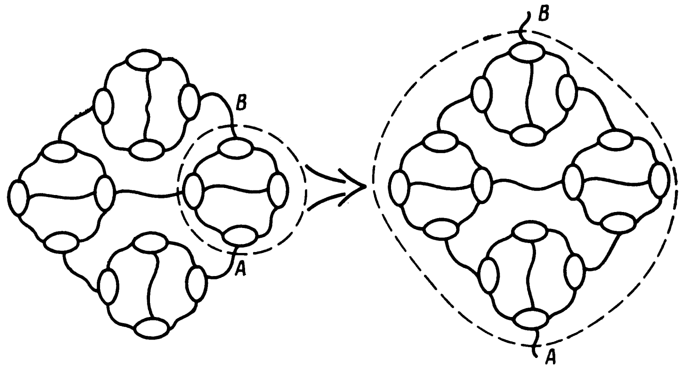

FIG. 1. Fragment of a cluster with a scaling invariant drop structure.

of size identical with the characteristic size l of this cluster. This assumption is justified by the calculations of Ref. 7, where it is shown that the largest contour dominates the magnetic polarizability of a cluster.

The imaginary part of the magnetic susceptibility of a system of noninteracting clusters is described by the expression

$$\chi''=\int_a^\infty \bar{\alpha}''(l)F(l)\,dl, \tag{2}$$

where $F(l)$ is the size distribution of doubly connected clusters and $\bar{\alpha}(l)$ is the total magnetic polarizability of a doubly connected cluster, which under the above approximations is proportional to l^3.

The form of the function $F(l)$ can be deduced from scaling relationships. If $p=p_c$, then the composite system has no specific scale and, consequently, we find that $F(l)\propto l^{-\varkappa}$. We shall now divide the system into cells of size $l \gg a$. We shall assume that a cell is conducting if a cluster of size greater than l passes through it. Then, the fraction of conducting cells is

$$p'\propto l^d \int_l^\infty F(l')\,dl' \propto l^{d+1-\varkappa}.$$

In view of the scaling invariance, the value of p' should be independent of l. This condition is satisfied if $\varkappa = d+1$. This value of $\varkappa$ can be obtained also from the relationships between the critical exponents.[16] If $p \to p_c$ the distribution function is truncated at scaling lengths equal to the correlation length ξ ($\xi \to \infty$ when $p \to p_c$). In the subsequent analysis it will be convenient to consider the function $F(l)$ represented in the form

$$F(l)=Bl^{-(d+1)} \Big/ \int_a^\xi l^{-(d+1)}\,dl, \tag{3}$$

where B is a certain normalization constant independent of ξ.

Integrating Eq. (2), we find the imaginary part of the magnetic susceptibility near p_c:

$$\chi''\propto \begin{cases} \xi, & d=2 \\ \ln\xi, & d=3 \end{cases}. \tag{4}$$

Since $\xi \propto \tau^{-\nu}$ (Ref. 16), the value of χ'' diverges for $p \to p_c$ in accordance with the power law which has the exponent $b=\nu=1.33$ when $d=2$, and in accordance with the logarithmic law when $d=3$. The critical exponent b for two-dimensional systems agrees with the numerical calculations reported in Ref. 11 [see Eq. (1)]. In the $d=3$ case the authors of Ref. 11 state that the value of χ'' is still a power-law function of τ. In our opinion, this is not in agreement with the data reported there. Figure 2 is based on the graphs and analytic expressions of Ref. 11 and it gives the dependences $\chi''(\tau)$ on logarithmic and semilogarithmic scales. We can see that the divergence of the magnetic susceptibility is described quite well by a logarithmic law in a wide range of concentrations. Therefore, Eq. (4) describes also the numerical results obtained for the $d=3$ case.

It therefore follows that the adopted approximation, i.e., the representation of doubly connected clusters by their contours, makes it possible to determine the behavior of the magnetic susceptibility near the percolation threshold and the results are not only in qualitative but also in quantitative agreement with those obtained in earlier investigations. The simplicity of the proposed model makes it possible to allow for the inductive interaction of the eddy currents, which has been ignored completely in earlier treatments.

Clusters in a composite system are distributed randomly in space. The inductive interaction between them will be allowed for using the effective-medium method.[17,18] In this approximation it is assumed that the influence of all the other clusters on the selected one reduces to the replacement of the environment by some effective medium whose magnetic properties can be described by an effective magnetic permeability μ_{eff}. The example of a periodic model is used in the Appendix to show that the frequency dependence $\chi''_{\text{eff}}(\omega)$ deduced by the effective medium method agrees

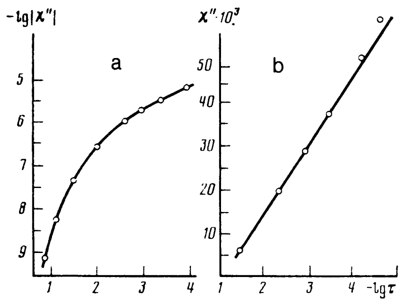

FIG. 2. Functions $\chi''(\tau)$ plotted in logarithmic (a) and semilogarithmic (b) scales using the numerical results of Ref. 11 for the $d=3$ case.

quite well with the exact results. A self-consistent equation for the parameter $\mu_{\rm eff}$ is

$$\mu_{eff}=1+4\pi\int_a^\xi F(l)\bar{\alpha}(l,\mu_{eff})\,dl. \quad (5)$$

The magnetic polarizability of a doubly connected cluster $\bar{\alpha}(l,\mu_{\rm eff})$ in a medium characterized by $\mu_{\rm eff}$ is described by

$$\bar{\alpha}=-Al^3\mu_{eff}/(\tilde{Z}+\mu_{eff}\tilde{L}), \quad (6)$$

where

$$\tilde{Z}=ic^2Z/\omega l,\quad \tilde{L}=L_e/l\approx\ln(l/a),$$

Z is the impedance of the cluster and L_e is the external part of the self-induction of the cluster. The value of Z and the product of the constants (AB) can be found from the condition that in the range $p\ll p_c(\xi\to a)$ Eq. (5) describes the magnetic permeability of noninteracting conducting inclusions:

$$\mu_{eff}=1+4\pi p\langle\alpha\rangle,$$

where $\langle\alpha\rangle$ is the polarizability of a conducting inclusion averaged over its various shapes. It is assumed that Eq. (6) is valid for clusters of any size. Therefore, we are ignoring a weak dependence of Z on ξ.

Assuming, for the sake of simplicity, that the conducting inclusion is spherical and that $\xi\gg a$, we obtain the following equation for $\mu_{\rm eff}$:

$$\mu_{eff}=1-12\pi p\ln\left[1-\alpha_s\ln((\xi/a)\mu_{eff})\right], \quad (7)$$

where α_s is given by the expression[14]

$$\alpha_s=-\frac{3}{8\pi}\left[1-\frac{3}{k^2}+\frac{3\operatorname{ctg}k}{k}\right],\quad k=\frac{(1+i)a}{\delta}.$$

Equation (7) makes it possible to find the asymptote of $\mu_{\rm eff}$ for $\alpha_s\xi\to\infty$:

$$\mu_{eff}(\alpha_s\xi\to\infty)=\frac{1-\exp(1/12\pi p)}{\alpha_s\ln(\xi/a)}. \quad (8)$$

It follows from Eq. (8) that at any fixed frequency the magnetic susceptibility tends to zero on approach to the percolation threshold. The frequency dependence $\mu_{\rm eff}(\omega)$ exhibits a resonance (Fig. 3). The frequency at the maximum of the imaginary part of the magnetic permeability ω_r decreases on approach to the percolation threshold: the dependences $\mu'_{\rm eff}(\omega)$ and $\mu''_{\rm eff}(\omega)$ shift toward lower frequencies. A numerical solution of Eq. (7) shows that if $p\to p_c$ ($\xi\to\infty$), then the resonance value $\mu''^{r}_{\rm eff}(p)=\mu''_{\rm eff}(\omega_r,p)$ tends to a certain constant limit, the value of which depends weakly on the threshold concentration p_c and for spherical inclusions it does not exceed 0.5. Therefore, an allowance for the inductive interaction of doubly connected clusters removes the divergence of the imaginary part of the effective magnetic permeability. The behavior of this permeability differs fundamentally from that of the permittivity, because the value of the latter diverges on approach to the percolation threshold. This difference is due to the fact that the physical analog of the permittivity is the quantity $1/\mu$. Therefore, an allowance for the interaction between clusters (in contrast to the magnetic permeability) increases the total permittivity of the system ε and is responsible for its divergence in the limit $p\to p_c$.

Near the percolation threshold the value of $\mu''^{r}_{\rm eff}$ of a percolation system with conducting inclusions of spherical shape ($p_c=1/3$) is several times greater than $\mu^{r''}$ of a system of particles which are not in contact (curve 3 in Fig. 3). In the case of percolation systems containing conducting particles of ellipsoidal shape the value of the threshold concentration p_c decreases on increase in the eccentricity of the particles[19] and can generally be as small as we please. However, at the percolation threshold the values of the effective magnetic permeability depend weakly on p_c. Clearly, when the concentration is appropriate in a system of nonconducting particles, the effective magnetic properties may vanish practically completely. Therefore, the effective magnetic properties of composite systems with low values of p_c can only be due to the formation of anomalously large percolation clusters.

We shall conclude by noting that the analogy between the behavior of normal and superconducting composites in alternating and static magnetic fields mentioned above allows us to conclude that inclusion of the mutual induction effects ensures that the diamagnetic susceptibility of superconducting composite systems remains finite near the percolation threshold.

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APPENDIX

We shall consider a simple periodic model consisting of conducting rings of radius r with a transverse cross-section area πa^2 on the assumption that their centers are located

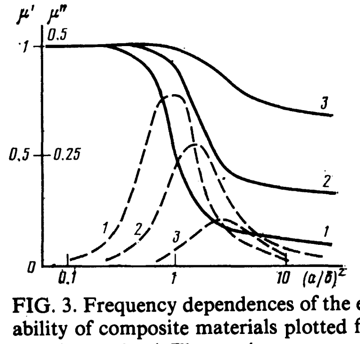

FIG. 3. Frequency dependences of the effective magnetic permeability of composite materials plotted for different values of the correlation size ξ. The continuous curves are the dependences $\mu'_{\rm eff}(\omega)$ and the dashed curves are the dependences $\mu''_{\rm eff}(\omega)$: 1) $\xi/a=10^3$; 2) $\xi/a=10$; 3) $\xi\sim a$ (inclusions which are not in contact).

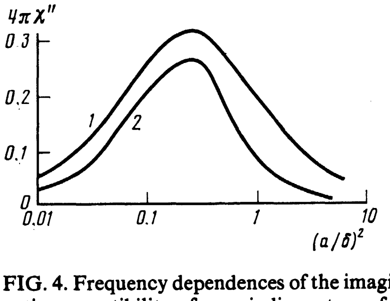

FIG. 4. Frequency dependences of the imaginary part of the magnetic susceptibility of a periodic system of conducting rings deduced from the exact solution (1) and by the effective medium method (2) on the assumption that $r/a=100$ and $nr^3=1$.

along one line ($\mathbf{z}$ axis) and they are separated by a distance h from one another with their planes perpendicular to this line. The magnetic susceptibility of such a system subjected to an alternating magnetic field $\mathbf{H}\|\mathbf{z}$ can be written in the form

$$\chi=Nm/HV, \quad V=\pi Nhr^2, \quad m=J\pi r^2/c,$$

where N is the number of rings and J is the current induced by the field $\mathbf{H}$ in one ring. In the approximation that $\delta \gtrsim a$ the value of the current J is given by

$$J=\frac{iHr}{4c[(\delta/a)^2-i\mathscr{L}]}, \tag{A1}$$

where

$$\mathscr{L}=\frac{1}{4\pi r}\left[L_0+2\sum_{k=1}^{N}L_{0k}(kh)\right].$$

Here L_0 and L_{0k} are the self-inductance and mutual inductance coefficients. Using Eq. (A1), we can represent the magnetic susceptibility in the form

$$\chi=\chi_0/(1+\rho),$$
$$\chi_0=\frac{i}{4}\frac{r}{h}\left(\frac{a}{\delta}\right)^2, \quad \rho=-i\mathscr{L}\left(\frac{a}{\delta}\right)^2. \tag{A2}$$

If we ignore the inductive interaction, we find that $\chi=\chi_0$. The value of χ_0 is purely imaginary and it diverges on reduction in the distance h between the rings. The limit $h\to 0$ corresponds qualitatively to the behavior of a composite system near the percolation threshold.

An allowance for the inductive interaction of the currents has the effect, as demonstrated by Eq. (A2), that for any finite value of ω we have $\chi''\to 0$ when $h\to 0$. If $h=2a$ and $\delta\approx a$, then Eq. (A2) is numerically the same as the familiar formula for the polarizability of a conducting cylinder with its axis parallel to an alternating magnetic field.[14]

We shall now calculate the magnetic susceptibility of this system by the effect medium method. The equation describing the relative value of $\mu_{\rm eff}$ is simple:

$$\mu_{eff}=1+4\pi n\bar{\alpha}(\mu_{eff}), \tag{A3}$$

where $n=1/\pi hr^2$ is the number of rings per unit volume and $\bar{\alpha}$ is the polarizability of a ring:

$$\bar{\alpha}=-\pi\mu_{eff}r^3/4[\mu_{eff}\tilde{L}_e+\tilde{Z}],$$
$$\tilde{L}_e=L_e/4\pi r=\ln(8r/a)-2, \quad \tilde{Z}=i(\delta/a)^2+{}^1/_4.$$

Equation (A3) readily yields the magnetic susceptibility:

$$\chi_{eff}=\frac{1}{8\pi\tilde{L}_e}[W+(W^2+4\tilde{Z}\tilde{L}_e)^{1/2}], \quad W=\tilde{L}_e-\tilde{Z}-\pi nr^3.$$

Figure 4 shows the dependences $\chi''(\omega)$ and $\chi''_{\rm eff}(\omega)$. We can see that the results obtained by the effective medium method are in satisfactory agreement with the exact results described by Eq. (A2).

[1]V. E. Dubrov, M. E. Levinshteĭn, and M. S. Shur, Zh. Eksp. Teor. Fiz. **70**, 2014 (1976) [Sov. Phys. JETP 43, 1050 (1976)].
[2]D. M. Grannan, J. C. Garland, D. B. Tanner, Phys. Rev. Lett. **46**, 375 (1981).
[3]A. L. Efros and B. I. Shklovskii, Phys. Status Solidi B **76**, 475 (1976).
[4]A. P. Vinogradov, A. M. Karimov, A. T. Kunavin, A. N. Lagar'kov, A. K. Sarychev, and N. A. Stember, Dokl. Akad. Nauk SSSR **275**, 590 (1984) [Sov. Phys. Dokl. **29**, 214 (1984)].
[5]A. P. Vinogradov, A. N. Lagar'kov, and A. K. Sarychev, Pis'ma Zh. Eksp. Teor. Fiz. **40**, 296 (1984) [JETP Lett. **40**, 1083 (1984)].
[6]P. G. De Gennes, C. R. Acad. Sci. **292**, 701 (1981).
[7]M. J. Stephen, Phys. Lett. A **87**, 67 (1981).
[8]R. Rammal, T. C. Lubensky, and G. Toulouse, Phys. Rev. B **27**, 2820 (1983).
[9]R. Rammal, T. C. Lubensky, and G. Toulouse, J. Phys. Lett. **44**, L65 (1983).
[10]R. Rammal and J. C. Angles d'Auriac, J. Phys. C **16**, 3933 (1983).
[11]D. R. Bowman and D. Stroud, Phys. Rev. Lett. **52**, 299 (1984).
[12]W. Y. Shih, C. Ebner, and D. Stroud, Phys. Rev. B **30**, 134 (1984).
[13]C. Ebner and D. Stroud, Phys. Rev. B **31**, 165 (1985).
[14]L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media*, Pergamon Press, Oxford (1960).
[15]A. P. Vinogradov and A. K. Sarychev, Zh. Eksp. Teor. Fiz. **85**, 1144 (1983) [Sov. Phys. JETP **58**, 665 (1983)].
[16]D. Stauffer, Phys. Rep. **54**, 1 (1979).
[17]S. Kirkpatrick, Rev. Mod. Phys. **45**, 574 (1973).
[18]A. K. Sarychev and A. P. Vinogradov, Phys. Status Solidi B **117**, K113 (1983).
[19]S. A. Zuev and V. F. Sidorenko, Teor. Mat. Fiz. **62**, 76 (1985).

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