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AMORPHOUS, VITREOUS, POROUS, ORGANIC, AND MICROCRYSTALLINE SEMICONDUCTORS; SEMICONDUCTOR COMPOSITES

Limiting Values of the Quality Factor of Thermoelectric Composites

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Abstract—It is shown that consideration of the effect of thermoelectric phenomena on effective conductivity and thermal conductivity can lead to "rigid" limitations on the effective thermoelectric quality factor. In some cases, a limiting value of the quality factor exists irrespective of how large the quality factor of the phases is.

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1. INTRODUCTION

Three kinetic coefficients (electrical conductivity σ, thermal conductivity κ, and thermopower α) characterize the properties of a homogeneous thermoelectric material, for example, a semiconductor. These coefficients allow one to naturally introduce a dimensionless temperature, or the characteristic temperature for this material

$$\tilde{T} = \frac{\sigma\alpha^2}{\kappa}T. \quad (1)$$

This temperature and, naturally, the combination

$$Z = \frac{\sigma\alpha^2}{\kappa}, \quad (2)$$

which was for the first time introduced by Ioffe [1] and is referred to as the quality factor, determine the characteristics of energy processes of thermoelectric devices, such as efficiency, cooling performance etc.

Currently, researchers are actively searching for new thermoelectric materials with large values of ZT (for example, [2]). Recently, materials with relatively high quality factors have been developed, for example, for $Bi_2Ti_{2.85}Se_{0.15}$ + Cu, $ZT \approx 1$ at $T \approx 300$ K [3]; for $Mg_2Si_{0.6}Sn_{0.4}$, $ZT \approx 1$ at $T \approx 800$ K [4]; for Zn_4Sb_3, $ZT \approx 1.35$ at $T \approx 673$ K [5], and for PbSe–PbTe, $ZT \approx 1.6$ [6]. Some hopes are pinned on the nanostructured materials since theoretical predictions give ZT values of around 10 and larger for Bi nanowires [7].

Taking into account the prospects, assumed thermoelectric devices with considerable quality factors ($ZT \geq 20$) and, correspondingly, high efficiencies and cooling performances are considered in the literature (see, for example, [8]). However, it should be clearly recognized that each actual thermoelectric device is a macroscopic device and, in addition, a macroscopically nonuniform device. Theoretical predictions for possible maximum values of efficiency and cooling performances obtained previously, as a rule, were carried out without taking into account the effect of thermoelectric phenomena on electrical conductivity and thermal conductivity, which is true for low quality factors and ceases to be true at larger values of ZT.

A natural and, as we believe, sufficiently modern question arises: what is the effect of macroscopic inhomogeneity of the material, whose separate parts (phases) possess a considerable quality factor, on the quality factor of the material (device) in general. In this study, we consider a narrower problem on the effective quality factor of a series of two-phase macroscopically nonuniform structures in the case where one of the phases can possess a larger, in theory arbitrarily high, quality factor.

Note that the question on limitations of effective conductivity σ_e or thermal conductivity κ_e, in the absence of thermoelectric phenomena, was resolved long ago. For σ_e and κ_e of randomly inhomogeneous media, the so-called two-side limitations (branches) of Foigt–Rois–Viner, Khashin–Shtrikman, etc. are obtained (see, for example, [9]). A general principle of construction of such branches [10] was generalized in [11] for thermoelectric phenomena. According to results obtained in [11], such forks turn out rather wide, and the effective quality factor is restricted by the highest quality factor of the phases, i.e., increases as the latter one increase. However, attainment of limiting values is not proved.

In this study, we obtained an unexpected answer to the above question: for some cases, the effective quality factor could not be higher than a certain definite value even for an arbitrarily high quality factor of one of the phases.

In sections 2 and 3 of the article, we consider plane-layered and self-dual media; for them, an exact solution of a problem on efficient kinetic coefficients is known. In Section 4, we consider three-dimensional randomly inhomogeneous media. In the conclusion, we discuss physical causes of existence of limiting values of an effective quality factor of an inhomogeneous material.

2. EFFECTIVE CHARACTERISTICS OF MACROSCOPICALLY INHOMOGENEOUS MEDIA

The main characteristics of macroscopically inhomogeneous media (which are homogeneous on average) are the effective kinetic coefficients, which relate by definition the average thermodynamic flows and forces. For thermoelectric phenomena, the flows are the electric current density j and heat-flow density q, and forces are the electric field strength E and temperature gradient ∇T, which are related to each other as

$$\mathbf{j} = \sigma\mathbf{E} - \sigma\alpha\nabla T,$$
$$\mathbf{q} = -\kappa\left(1 + \frac{\sigma\alpha^2}{\kappa}T\right)\nabla T + \sigma\alpha T\mathbf{E}. \quad (3)$$

Correspondingly, for effective electrical conductivity, thermal conductivity, and thermovoltage, we have

$$\langle\mathbf{j}\rangle = \sigma_e\langle\mathbf{E}\rangle - \sigma_e\alpha_e\langle\nabla T\rangle,$$
$$\mathbf{q} = -\kappa_e\left(1 + \frac{\sigma_e\alpha_e^2}{\kappa_e}T\right)\langle\nabla T\rangle + \sigma_e\alpha_e T\langle\mathbf{E}\rangle, \quad (4)$$

where angle brackets mean averaging over the bulk. It should be noted that the second equation in relations (4) is true only at small temperature gradients; i.e., here we assume that $|\nabla T|L \ll Z^{-1}$, where L is the sample size.

Depending on the configuration of arrangement of the phases, the effective kinetic coefficients σ_e, κ_e, and σ_e can be both scalars and tensors even in the case of phase isotropy.

The effective quality factor is determined as

$$Z_e = \frac{\sigma_e\alpha_e^2}{\kappa_e}; \quad (5)$$

the possible limitations of Z_eT at any values of local kinetic coefficients σ, κ, and α will be considered in this article.

3. PLANE-LAYERED MEDIA

Effective kinetic coefficients for the plane-layered media were obtained in [12, 13]. Here, we will consider a two-phase medium consisting of alternating layers with the values of local kinetic coefficients σ_1, κ_1, and α_1 for the first phase and σ_2, κ_2, and α_2 for the second phase. For further consideration, it is convenient to write the components of the tensors of effective electrical conductivity, thermal conductivity, and thermopower as

$$\sigma_{xx}^e(p) = \frac{\sigma_1\sigma_2}{p\sigma_2 + (1-p)\sigma_1}\frac{1}{1+\tilde{Z}(p)T},$$
$$\sigma_{zz}^e(p) = p\sigma_1 + (1-p)\sigma_2,$$
$$\kappa_{xx}^e(p) = \frac{\kappa_1\kappa_2}{p\kappa_2 + (1-p)\kappa_1},$$
$$\kappa_{zz}^e(p) = [p\kappa_1 + (1-p)\kappa_2][1+\tilde{Z}(1-p)T], \quad (6)$$
$$\alpha_{xx}^e(p) = \frac{\kappa_2\alpha_1 p + \kappa_1\alpha_2(1-p)}{p\kappa_2 + (1-p)\kappa_1},$$
$$\alpha_{zz}^e(p) = \frac{\alpha_1\sigma_1 p + \alpha_2\sigma_2(1-p)}{p\sigma_1 + (1-p)\sigma_2},$$

where the x axis is directed normally to the layers, the z axis is arranged in parallel to the layers, and p is the concentration of the first phase,

$$\tilde{Z}(p)T = p(1-p) \times \frac{\sigma_1\sigma_2(\alpha_1-\alpha_2)^2}{[p\sigma_2 + (1-p)\sigma_1][p\kappa_2 + (1-p)\kappa_1]}T. \quad (7)$$

Introduced in [14], the so-called internal quality factor $\tilde{Z}(p)$ characterizes the effect of thermoelectric phenomena on the effective thermal conductivity and electrical conductivity. Note that, in $\sigma_{xx}^e(p)$ and in $\kappa_{zz}^e(p)$, this effect is determined by the same function (7).

Let us determine the effective quality factor along and across the layers correspondingly as

$$Z_\parallel^e = \sigma_{xx}^e(\alpha_{xx}^e)^2/\kappa_{xx}^e, \quad Z_\parallel^e = \sigma_{zz}^e(\alpha_{zz}^e)^2/\kappa_{zz}^e. \quad (8)$$

Substituting the relation for effective kinetic coefficients from relations (6) into relations (8), it is easy to see that both $Z_\parallel^e$ and $Z_\perp^e$, even at an arbitrary high quality factor, for example, of the second phase Z_2T, are limited, i.e., do not exceed certain limiting values. These limiting relations look especially simple in the case where thermoelectric properties of the first phase can be disregarded, i.e., when $\alpha_1 = 0$,

$$\lim_{\alpha_2\to\infty} Z_\parallel^e(p)T = \frac{\kappa_1}{\kappa_2}\frac{1-p}{p}, \quad Z_2T \longrightarrow \infty,$$
$$\lim_{\alpha_2\to\infty} Z_\perp^e(p)T = \frac{\sigma_2}{\sigma_1}\frac{1-p}{p}, \quad Z_2T \longrightarrow \infty. \quad (9)$$

More exactly, for $Z^e_{\parallel}$, the fulfillment of conditions $\alpha_2 \gg \alpha_1$ and $\kappa_1\alpha_2(1-p) \gg \kappa_2\alpha_1 p$ is sufficient, and for $Z^e_{\perp}$, $\alpha_2 \gg \alpha_1$ and $\sigma_2\alpha_2(1-p) \gg \alpha_1\sigma_1 p$ is sufficient.

Therefore, the effective quality factor of the plane-layered media is restricted by certain limiting values, and no arbitrarily large value of the quality factor of one of the phases allows it to exceed them.

4. SELF-DUAL MEDIA

As self-dual media, we assume such two-dimensional isotropic media, for which a mutual replacement of the phases does not vary the values of their efficient kinetic coefficients [15]. Specifically, such media are media of the chessboard type (black squares denote one phase, and white squares denote the other one), and randomly inhomogeneous media at the leakage threshold, when concentration of both phases $p = 1/2$. Numerous examples of such media are given in [16, 17].

For the self-dual media, the exact relations for effective kinetic coefficients suitable for arbitrary large inhomogeneity are known. The effective conductivity with no allowance made for thermoelectric phenomena was obtained in [15]: $\sigma^e_D = \sqrt{\sigma_1\sigma_2}$. Relations for σ_e, κ_e, and α_e, taking into account the thermoelectric phenomena, are given in [18], and for σ_e and κ_e, the explicit relations are given in [19]. In a form convenient for the further consideration, they can be written as

$$\sigma_e = \frac{\sigma^e_D}{\sqrt{1+\tilde{Z}T}}, \quad \kappa_e = \kappa^e_D\sqrt{1+\tilde{Z}T},$$
$$\alpha_e = \frac{\alpha_1\sqrt{\sigma_1\kappa_2}+\alpha_2\sqrt{\sigma_2\kappa_1}}{\sqrt{\sigma_1\kappa_2}+\sqrt{\sigma_2\kappa_1}}, \tag{10}$$

where the effect of thermoelectric phenomena on σ_e and κ_e, similarly to the case of the plane-layered media, is determined by the same combination of local kinetic coefficients, or the internal quality factor, which can be written for the self-dual media as

$$\tilde{Z}T = \frac{\sigma^e_D}{\kappa^e_D} \times \frac{(\alpha_1-\alpha_2)^2}{(\sqrt{\sigma_2/\sigma_1}/\sqrt{\kappa_2/\kappa_1}+\sqrt{\kappa_2/\kappa_1}/\sqrt{\sigma_2/\sigma_1})}T, \tag{11}$$
$$\kappa^e_D = \sqrt{\kappa_1\kappa_2}.$$

The effective quality factor of the self-dual media, as follows form relations (10) and (11), can be written as

$$Z_eT = \left(\frac{\sigma^e_D}{\kappa^e_D}\right)\left(\frac{\alpha_1\sqrt{\sigma_1/\kappa_1}+\alpha_2\sqrt{\sigma_2/\kappa_2}}{\sqrt{\sigma_1/\kappa_1}+\sqrt{\sigma_2/\kappa_2}}\right)^2 \times \frac{T}{1+(\sigma^e_D/\kappa^e_D)^3(\alpha_1-\alpha_2)^2/(\sigma_1/\kappa_1+\sigma_2/\kappa_2)^2}. \tag{12}$$

It is seen from relation (12) that, at an arbitrarily high quality factor of the second phase ($Z_2T \longrightarrow \infty$), the value of Z_eT is restricted. This is especially clearly seen for the case where the thermovoltage of the first phase can be disregarded assuming $\alpha_1 = 0$ (metal), and $\sigma_1/\kappa_1 = \sigma_2/\kappa_2$:

$$Z_eT = \frac{(1/4)Z_2T}{1+(1/4)Z_2T}. \tag{13}$$

It follows from relation (13) that in this case

$$Z_eT \le 1. \tag{14}$$

If $\sigma_1/\kappa_1 \ne \sigma_2/\kappa_2$, but α_2, as above, is large ($\alpha_2 \gg \alpha_1$, $\alpha_2 \gg \alpha_1\sqrt{(\sigma_1/\sigma_2)(\kappa_2/\kappa_1)}$, and $\alpha_2^2 \gg (\sigma_1/\kappa_1+\sigma_2/\kappa_2)^2/T(\sigma^e_D/\kappa^e_D)^3$), the limiting value is

$$Z_eT = \frac{\sigma^e_D}{\kappa^e_D}\left[\left(1+\frac{\kappa_1\sigma_2}{\kappa_2\sigma_1}\right)/\left(1+\sqrt{\frac{\kappa_1\sigma_2}{\kappa_2\sigma_1}}\right)\right]^2,$$
$$Z_2T \longrightarrow \infty. \tag{15}$$

Therefore, at arbitrarily large α_2, i.e., at an arbitrarily high quality factor of the second phase, the effective quality factor Z_eT is saturated.

5. THREE-DIMENSIONAL RANDOMLY INHOMOGENEOUS MEDIA

For randomly inhomogeneous media, the solution of a problem of effective kinetic coefficients over the entire range of variations in the concentrations can be obtained only in an approximate form. In [20], the so-called isomorphism method was suggested, which allows one, in some cases, to reduce the problem of effective kinetic coefficients of the thermoelectric composite to a simpler problem on effective conductivity in the absence of thermoelectric phenomena. In [21], this method was developed and applied, specifically, to thermoelectric composites taking into account the effect of thermoelectric phenomena to electrical con-

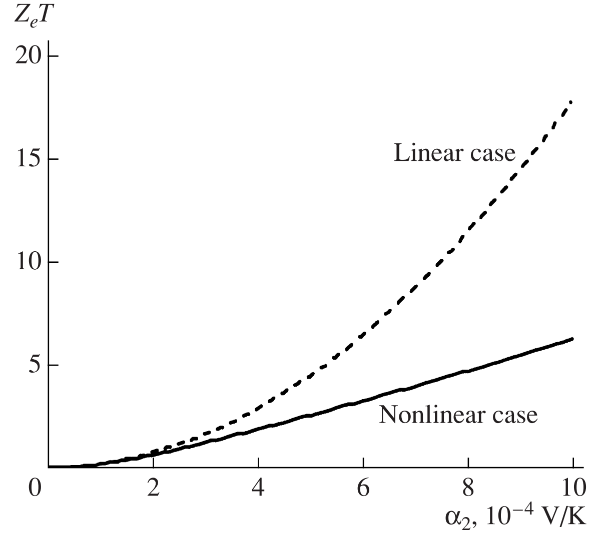

Fig. 1. Dependence of the effective quality factor on the thermopower of the second phase. A drastic lag of the dependence of the effective quality factor on the thermopower of the second phase compared with the linear case is shown. The parameters of calculation: $\sigma_1 = 5 \times 10^6\ \Omega^{-1}\ \mathrm{m}^{-1}$, $\sigma_2 = 3.2 \times 10^4\ \Omega^{-1}\ \mathrm{m}^{-1}$, $\alpha_1 = 10^{-9}$ V/K, $\kappa_1 = 119\ \mathrm{V}^2/(\Omega\ \mathrm{m\ K})$, $\kappa_2 = 0.637\ \mathrm{V}^2/(\Omega\ \mathrm{m\ K})$, $p = 0.3$, and $T = 673$ K.

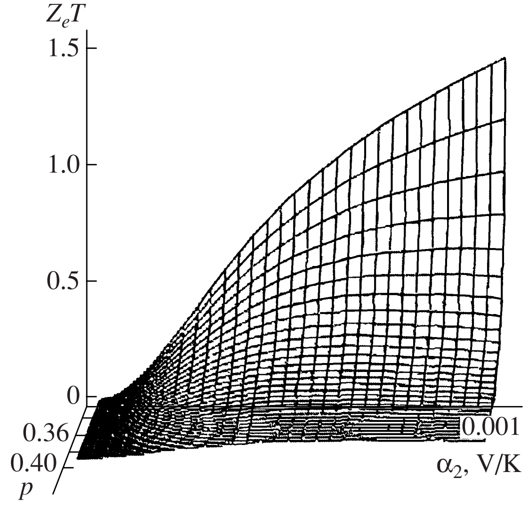

Fig. 2. Dependence of the effective quality factor on the thermopower of the second phase α_2 and concentration of the first phase p. Attainment of saturation of the effective quality factor with increasing α_2 at $p > p_c$ is clearly observed. As an example, the following parameters of calculation are chosen: $\sigma_1 = 5 \times 10^6\ \Omega^{-1}\ \mathrm{m}^{-1}$, $\sigma_2 = 3.2 \times 10^4\ \Omega^{-1}\ \mathrm{m}^{-1}$, $\alpha_1 = 10^{-9}$ V/K, $\kappa_1 = 119\ \mathrm{V}^2/(\Omega\ \mathrm{m\ K})$, $\kappa_2 = 0.637\ \mathrm{V}^2/(\Omega\ \mathrm{m\ K})$, $p = 0.3$, and $T = 673$ K.

ductivity and thermal conductivity. According to this method [21],

$$\left.\begin{aligned} \sigma_e &= \frac{(\mu\sigma_1 - \sigma_2) f_\lambda - (\lambda\sigma_1 - \sigma_2) f_\mu}{\mu - \lambda} \\ \alpha_e &= \frac{(\mu\sigma_1\alpha_1 - \sigma_2\alpha_2) f_\lambda - (\lambda\sigma_1\alpha_1 - \sigma_2\alpha_2) f_\mu}{(\mu\sigma_1 - \sigma_2) f_\lambda - (\lambda\sigma_1 - \sigma_2) f_\mu} \\ \kappa_e &= \frac{\sigma_1\kappa_1(\mu - \lambda) f_\lambda f_\mu}{(\mu\sigma_1 - \sigma_2) f_\lambda - (\lambda\sigma_1 - \sigma_2) f_\mu} \end{aligned}\right\}, \quad (16)$$

where

$$\begin{Bmatrix}\mu \\ \lambda\end{Bmatrix} = \frac{1}{4\sigma_1\kappa_1} \times \{[(\sqrt{\sigma_1\kappa_2} + \sqrt{\sigma_2\kappa_1})^2 + \sigma_1\sigma_2 T(\alpha_1 - \alpha_2)^2]^{1/2} \pm [(\sqrt{\sigma_1\kappa_2} - \sqrt{\sigma_2\kappa_1})^2 + \sigma_1\sigma_2 T(\alpha_1 - \alpha_2)^2]^{1/2}\}^2. \quad (17)$$

In relation (17), sign (+) is related to μ, and sign (–) is related to λ. Functions f_λ and f_μ are defined as follows. Let us write the effective electrical conductivity σ_e (with thermoelectric phenomena disregarded) of the system under consideration in the form $\sigma_e = \sigma_1 f(p, h)$, where $h = \sigma_2/\sigma_1$, σ_1 and σ_2 are the electrical conductivities of the first and second components, and p is the concentration of the first component. In this case, functions f_λ and f_μ are obtained from $f(p, h)$ by the substitution $h \longrightarrow \lambda$ and $h \longrightarrow \mu$. Therefore,

$$f_\lambda = f(p, \lambda), \quad f_\mu = (p, \mu). \quad (18)$$

Relations (16) are true for both two-dimensional and three-dimensional isotropic binary systems of an arbitrary structure. All information on the form of their inclusions and arrangement is contained in the function f. If the function $f(p, h)$ is known for any binary conducting system without thermoelectric phenomena at all values of p and h, then formula (16) gives a complete solution of the problem on thermoelectric phenomena of such a system.

To find the function $f(p, h)$, let us use the approximation of the mean field [22, 23], which is a good approximation over the entire range of variations in the concentrations. For the three-dimensional randomly inhomogeneous medium, in this approximation,

$$f(p, h) = \frac{\sigma_e}{\sigma_1} = \frac{1}{4}\{[3p(1-h) + 2h - 1] + \sqrt{[3p(1-p) + 2h - 1]^2 + 8h}\}. \quad (19)$$

Figure 1 shows an increase in Z_eT with an increase in the thermopower of the second phase α_2. For comparison, the same dependence is represented, but for the linear case, if in relations (16) and (17), the local quality factor is omitted, $(\sigma_i\alpha_i^2/\kappa_i)T \ll 1$. A drastic lag of Z_eT from the linear case is clearly seen. Figure 2 shows

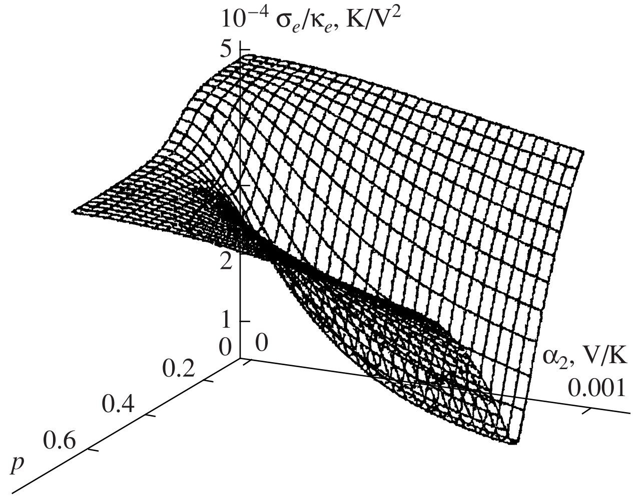

Fig. 3. Ratio σ_e/κ_e as a function of the concentration of the first phase p and thermopower of the second phase α_2 for a three-dimensional randomly inhomogeneous medium. As an example, the following parameters of calculation are chosen: $\sigma_1 = 5\times10^6\ \Omega^{-1}\ \mathrm{m}^{-1}$, $\sigma_2 = 3.2\times10^4\ \Omega^{-1}\ \mathrm{m}^{-1}$, $\alpha_1 = 10^{-9}$ V/K, $\kappa_1 = 119$ V^2/(Ω m K), $\kappa_2 = 0.637$ V^2/(Ω m K), $p = 0.3$, and $T = 673$ K.

the attainment of saturation of the effective quality factor with an increase in α_2 in the range $p > p_c = 1/3$.

In order to illustrate the effect of large local quality factors more clearly, let us consider a formal transition to the infinite quality factor of the second phase by tending its thermovoltage coefficient to infinity, i.e., $\alpha_2 \longrightarrow \infty$. In this case, we obtain

$$\lim_{\alpha_2\to\infty}(Z_eT) = \begin{cases} \dfrac{9}{2}\dfrac{\sigma_2\alpha_2^2}{\kappa_2}T\left(p-\dfrac{2}{3}\right)\left(p-\dfrac{1}{3}\right), & p < p_c, \\ \dfrac{\sigma_2}{\sigma_1}\dfrac{2/3-p}{p-1/3}, & p_c < p < 1-p_c, \\ \dfrac{9}{2}\dfrac{\sigma_1\alpha_1^2}{\kappa_2}T\left(p-\dfrac{2}{3}\right)\left(p-\dfrac{1}{3}\right), & p > 1-p_c. \end{cases} \quad (20)$$

From relation (20), it is immediately seen that, in the region $p > p_c$, at the arbitrary high local quality factor of the second phase, the effective quality factor remains finite. It should be noted that in deriving relation (20), we assumed the fulfillment of the following inequalities: $\alpha_2^2(p-p_c) \gg 1$ and $\alpha_2^2[p-(1-p_c)] \gg 1$.

6. CONCLUSIONS

A possible saturation of Z_eT with an increase in α_2 is directly associated with the fact that the effective quality factor involves the ratio σ_e/κ_e. In the absence of thermoelectric phenomena, this ratio is independent of local thermopower coefficients, and in this case, if $\sigma_1/\kappa_1 = \sigma_2/\kappa_2$, is independent of the phase concentration. The account of thermoelectric phenomena for the above-considered cases decreases the effective conductivity and increases the effective thermal conductivity, which leads to a decrease in the ratio σ_e/κ_e and thereby to a decrease in Z_eT. In randomly inhomogeneous media, the lowest ratio σ_e/κ_e by concentration is attained at the percolation threshold (Fig. 3). It is known from the study of distribution of the current density and its moments that (see, for example, [24]) precisely at the percolation threshold, the regions with the largest values of above parameters exist in the medium.

The variation in σ_e/κ_e due to the thermoelectric phenomena is directly associated with the intrinsic quality factor $\tilde{Z}T$. The last one differs from zero only if the medium is inhomogeneous by thermopower, i.e., $\alpha_2 \neq \alpha_1$ (see relations (7) and (11)). Precisely from here, the qualitative interpretation follows that Z_eT rises more slowly (and has saturation) compared with the linear case, where the effect of thermoelectric phenomena on σ_e/κ_e is disregarded. Inhomogeneity by thermopower means that the macroscopically inhomogeneous medium can be qualitatively represented as the set of thermocouples connected to each other. The higher the quality factor of the phases, the "more active" transformation of heat energy into electrical one within the medium. The emerging eddy currents are not lead out and dissipate the energy inside the medium, thereby decreasing the "external" quality factor Z_eT. At $\alpha_2 = \alpha_1$, the temperature gradient already does not induce thermoelectric currents within a medium inhomogeneous by electrical and thermal conductivity, and the internal quality factor equals zero. For the emergence of closed eddy currents, both phases should be conducting. For example, one phase should be semiconductor, and the second one should be metal or semiconductor with another thermopower coefficient. In the case where one of the phases has low (in the limit zero) conductivity, thermoelectric fields ($\alpha\nabla T$) emerging in the randomly inhomogeneous medium cannot induce the eddy currents, renormalization of σ_e/κ_e is absent, and, ultimately, saturation of Z_eT is absent.

Therefore, at a high local quality factor, macroscopic inhomogeneities can substantially limit the thermoelectric quality factor of the material.

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