Intelligence thermotics: Correlated self-fixing behavior of thermal metamaterials

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Abstract – It is a challenge to design intelligent thermal metamaterials due to the lack of suitable theories. Here we propose a kind of intelligent thermal metamaterials by investigating a core-shell structure, where both the core and shell have an anisotropic thermal conductivity. We solve Laplace’s equation for deriving the equivalent thermal conductivity of the core-shell structure. Amazingly, the solution gives two coupling relations of conductivity tensors between the core and the shell, which cause the whole core-shell structure to counterintuitively self-fix a constant isotropic conductivity even when the area or volume fraction of the core changes within the full range in two or three dimensions. The theoretical findings on fraction-independent properties are in sharp contrast with those predicted by the well-known effective medium theories, and they are further confirmed by our laboratory experiments and computer simulations. This work offers two coupling relations for designing intelligent thermal metamaterials, and they are not only helpful for thermal stabilization or camouflage/illusion, but they also offer hints on how to achieve similar metamaterials in other fields.

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Introduction. – Thermal metamaterials [1,2] are actually artificial materials or devices that exhibit novel thermal properties based on their geometrical structures or patterns. The earliest example of thermal metamaterials are thermal cloaks designed by coordinate transformation, which guide the heat flow around an object as if the object does not exit [3–8]. In principle, thermal metamaterials are useful to efficiently control the heat flow. However, almost all the existing thermal metamaterials are non-intelligent, which means that the corresponding thermal metamaterials cannot feel and respond to the change of external or internal stimuli in a controlled fashion. On the contrary, if such metamaterials can do so, they can be called intelligent thermal metamaterials.

Owing to the lack of suitable theories, intelligent thermal metamaterials have not been touched in the literature, except for those with dual functions [9–15] or with thermal responsiveness [16,17]. For example, in ref. [16], Li et al. developed a theory of temperature-dependent transformation thermotics, and designed an intelligent thermal cloak. The cloak can feel the change of environmental temperatures, and then it can be automatically switched on or off. Nevertheless, this kind of intelligent thermal cloaks feels and responds to a kind of external stimuli (namely, the change of environmental temperatures), rather than internal stimuli (e.g., the change of internal states). In fact, for external stimuli, a similar intelligence appears in all the other existing intelligent thermal metamaterials [9–17]. Regarding the internal stimuli, no work has been reported to date. Thus, in this work, we start to propose a class of intelligent thermal metamaterials that can feel and respond to a type of internal stimuli, namely, the change of area/volume fractions. In practice, area or volume fractions can be changed due to either expansion caused by heat or contraction caused by cold. For simplicity, this work focuses only on the cases corresponding to the pure change of area or volume fractions in a core-shell structure, where both the core and the shell have an anisotropic thermal conductivity.

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Then we theoretically reveal two coupling relations for counterintuitively self-fixing a constant isotropic thermal conductivity of the core-shell structure as the area or volume fraction varies within the full range. To this end, our theoretical results (namely, fraction-independent properties) are validated by both experiments and simulations in two or three dimensions, which are in sharp contrast to the fraction-dependent properties predicted by the well-known effective medium theories including the Bruggeman formula and the Maxwell-Garnett formula. Such effective medium theories have been extensively adopted in the field of metamaterials from optics/electromagnetics, to acoustics, to mechanics, and to thermics.

**Theory for two dimensions.** – Let us start by considering a core-shell structure embedded in a host, whose center is located in the origin of coordinates. See fig. 1(a). The core (or shell) has radius $r_1$ (or $r_2$) and thermal conductivity $\kappa_1$ (or $\kappa_2$). $\kappa_3$ denotes the host’s thermal conductivity. For the sake of generality, we assume the core and shell to be anisotropic. In cylindrical coordinates $(r, \theta)$, $\kappa_1$ and $\kappa_2$ are both tensorial, which can be represented by diag($\kappa_{rr_1}, \kappa_{\theta\theta_1}$) and diag($\kappa_{rr_2}, \kappa_{\theta\theta_2}$), respectively.

In this case, for a passive and stable heat transport process, Laplace’s equation has the following form:

$$\frac{1}{r}\frac{\partial}{\partial r} \left( r \kappa_{rr} \frac{\partial T}{\partial r} \right) + \frac{1}{r} \frac{\partial}{\partial \theta} \left( \kappa_{\theta\theta} \frac{\partial T}{\partial \theta} \right) = 0,$$

where $T$ denotes the temperature. The general solution of eq. (1) is given by

$$T = A_0 + B_0 \ln r + \sum_{m=1}^{\infty} \left[ A_m \cos(m\theta) + B_m \sin(m\theta) \right] r^m \sqrt{\frac{m^2}{m^2 - 1}} + \sum_{n=1}^{\infty} \left[ C_n \cos(n\theta) + D_n \sin(n\theta) \right] r^{-n} \sqrt{\frac{n^2}{n^2 - 1}}.$$

It is worth noting that eq. (2) is valid under the condition that both $\kappa_{rr}$ and $\kappa_{\theta\theta}$ are either positive or negative. Here it is necessary for us to remark that apparently negative thermal conductivities can only be achieved by adding external work as required by the second law of thermodynamics, which means that the heat (e.g., in electric refrigerators) can be driven to transport from a region with a low temperature to another region with a high temperature; more relevant comments can be found in the appendix.

For clarity, we use $T_1$, $T_2$ and $T_3$ to denote the temperature distribution in core, shell and host. To focus on the core-shell structure, we simplify the host to be homogeneous and isotropic. As a result, we obtain $T_3$ as

$$T_3 = A_0 + A_1 r \cos \theta + C_1 r^{-1} \cos \theta.$$  

Meanwhile, this system has the corresponding boundary conditions [18],

$$\begin{cases}
T_1 \text{ is finite,} \\
T_1(r_1) = T_2(r_1), \\
T_2(r_2) = T_3(r_2), \\
-\kappa_{rr_1} \left. \frac{\partial T_1}{\partial r} \right|_{r=r_1} = -\kappa_{rr_2} \left. \frac{\partial T_2}{\partial r} \right|_{r=r_1}, \\
-\kappa_{rr_2} \left. \frac{\partial T_2}{\partial r} \right|_{r=r_2} = -\kappa_{rr_3} \left. \frac{\partial T_3}{\partial r} \right|_{r=r_2}, \\
T_3(r \to \infty) = -\nabla T_0 | r \cos \theta,
\end{cases}$$

where $\nabla T_0$ represents the external thermal field.

For convenience of comparison, throughout this work we set $\kappa_3$ to have the same value as the equivalent thermal conductivity $\kappa_e$ of the core-shell structure. The temperature distribution in the host is given by eq. (3). By applying the boundary conditions, we can derive the undetermined constants $A_1$ and $C_1$. Then we set $C_1$ to be zero to ensure the temperature distribution in the host is undistorted. Namely, $\nabla T_0$ is maintained uniform without being affected by the core-shell structure. As a result, we obtain the expression for $\kappa_e$ as

$$\kappa_e = \kappa_3 = c_1 \kappa_{rr_1} + c_2 \kappa_{rr_2} + \left( c_1 \kappa_{rr_1} - c_2 \kappa_{rr_2} \right) p^2,$$

where $p = r_1^2/r_2^2$ is the area fraction of the core in the core-shell structure, and $c_i = \sqrt{\kappa_{\theta\theta_i}/\kappa_{rr_i}}$ with $i = 1$ or 2. In view of eq. (5), if we set

$$c_1 \kappa_{rr_1} - c_2 \kappa_{rr_2} = 0,$$

they yield, respectively,

$$\kappa_e = c_1 \kappa_{rr_1} = c_2 \kappa_{rr_2}.$$

Clearly, eqs. (8) and (9) show that the resulting equivalent thermal conductivity $\kappa_e$ is independent of the core’s area fraction, thus being a constant. In other words, the area fraction can take any value within the full range.

![Fig. 1: Schematic graph showing the core-shell structure in (a) two dimensions and (b) three dimensions.](Image)
from $0^+$ to 1, which, however, does not affect the value of $\kappa_e$. More remarks can be added herein: in the case of isotropic core and shell, eq. (9) predicts an equivalent thermal conductivity of the core-shell structure that equals the thermal conductivity of the core, which echoes with the condition required for the phenomenon of partial resonance in electromagnetic fields [19]. Here it is worth mentioning that the authors of ref. [19] discuss only isotropic cases, which differ from the anisotropic cases considered in this work.

Since thermal conductivities of the core and shell are coupled in eqs. (6) and (7), we may call the two equations as two coupling relations. To distinguish the two relations, we name eq. (6) (or eq. (7)) as positive (or negative) relation. The physical meanings of both relations are to make the equivalent thermal conductivity of the core-shell structure independent of the area/volume fraction $p$.

**Theory for three dimensions.** – The above theory for two dimensions (fig. 1(a)) can be extended to three dimensions (fig. 1(b)). Now $\kappa_1$ and $\kappa_2$ can be reformulated as $\kappa_1 = \text{diag}(\kappa_{rr_1}, \kappa_{\theta\theta_1}, \kappa_{\varphi\varphi_1})$ and $\kappa_2 = \text{diag}(\kappa_{rr_2}, \kappa_{\theta\theta_2}, \kappa_{\varphi\varphi_2})$ in spherical coordinates ($r, \theta, \varphi$). For simplicity, we assume that the system has an axial symmetry, namely, $\kappa_{\theta\theta} = \kappa_{\varphi\varphi}$. So, $T$ is independent of $\varphi$. Then, the governing Laplace equation can be written as

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \kappa_{rr} \frac{\partial T}{\partial r} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left( \sin \theta \kappa_{\theta\theta} \frac{\partial T}{\partial \theta} \right) = 0. \quad (10)$$

Similarly to the procedure for two dimensions, the equivalent thermal conductivity of the core-shell structure in three dimensions can be obtained as

$$\kappa_e = \kappa_3 = \kappa_{rr_2} \times \frac{l_{21}(l_{11} \kappa_{rr_1} - l_{22} \kappa_{rr_2}) - l_{22}(l_{11} \kappa_{rr_1} - l_{21} \kappa_{rr_2}) \rho(l_{21} - l_{22})/3}{(l_{11} \kappa_{rr_1} - l_{22} \kappa_{rr_2}) - (l_{11} \kappa_{rr_1} - l_{21} \kappa_{rr_2}) \rho(l_{21} - l_{22})/3},$$

\[ (11) \]

where $\rho = r_1^3/r_3^3$ is the volume fraction of the core in the core-shell structure, $l_{11} = (-1 + \sqrt{1 + 8\kappa_{\theta\theta_1}/\kappa_{rr_1}})/2$, and $l_{12} = (-1 - \sqrt{1 + 8\kappa_{\theta\theta_1}/\kappa_{rr_1}})/2$. Here $i = 1$ or 2.

Similarly, eq. (11) yields two coupling relations,

$$l_{11} \kappa_{rr_1} - l_{21} \kappa_{rr_2} = 0, \quad (12)$$
$$l_{11} \kappa_{rr_1} - l_{22} \kappa_{rr_2} = 0, \quad (13)$$

which, respectively, lead to

$$\kappa_e = l_{11} \kappa_{rr_1} = l_{21} \kappa_{rr_2}, \quad (14)$$
$$\kappa_e = l_{11} \kappa_{rr_1} = l_{22} \kappa_{rr_2}. \quad (15)$$

Clearly, the equivalent thermal conductivity $\kappa_e$ in eq. (14) (or eq. (15)) is also independent of the core’s volume fraction. Also, for clarity, we call eq. (12) (or eq. (13)) as positive (or negative) coupling relation.

**Laboratory experiments and computer simulations.** – In order to validate our theoretical analysis, now we are in a position to perform corresponding laboratory experiments and computer simulations.

In general, eqs. (5) and (11) can predict the equivalent thermal conductivity of the core-shell structure in two or three dimensions. When the two coupling relations are satisfied (namely, eqs. (6), (7) for two dimensions and eqs. (12), (13) for three dimensions), the equivalent thermal conductivity of the core-shell structure will not change with the area or volume fraction. For comparison, we first utilize the commercial software COMSOL Multiphysics (https://www.comsol.com/) to perform finite-element simulations (fig. 2 and fig. 3(a3)–(c3)), which is followed by laboratory experiments (fig. 3(a1)–(c1) and fig. 3(a2)–(c2)).

Figure 2 shows the finite-element simulations for two dimensions. We investigate the effects of both the positive relation (eq. (6)) in fig. 2(a1), (b1) and the negative relation (eq. (7)) in fig. 2(a2), (b2). Note that fig. 2(a2), (b2) has a shell with apparently negative thermal conductivity. For reference, fig. 2(c1), (c2) only has a core (without shell). All the hosts in the six panels fig. 2(a1)–(c2) are set to be the same, but the area fraction of the core is increased from fig. 2(a1) to fig. 2(c1) (or from fig. 2(a2) to fig. 2(c2)). Clearly, fig. 2(a1)–(c2) displays that the host’s temperature distribution is identical to each other in the six panels. This behavior confirms the theoretical prediction that the equivalent thermal conductivity of the
Fig. 3: Experimental demonstrations of fig. 2(a1)–(c1). (a1)–(c1) are three experimental samples, each having a size of 24 × 24 cm and a thickness of 0.3 mm. The core-shell structure in (a1) or (b1) is composed of arrays made of ellipses with two different sizes: the large size is for the core, and the small size is for the shell. The core (or shell) is composed of red copper drilled with air ellipses, each having a major/minor semi-axis of 0.24/0.09 cm (or 0.18/0.045 cm). For reference, the pure core is used in (c1), which has the same radius as the core-shell structure shown in (a1) and (b1). The host in (a1)–(c1) is composed of red copper drilled with 204 air circular holes, each with radius 0.415 cm. In (a1)–(c1), the thermal conductivities of the core and shell are diag(194, 304) W/(m·K) and diag(300, 197) W/(m·K); for (a2) and (b2), \( \kappa_1 = \text{diag}(100, 417, 417) \) W/(m·K) and \( \kappa_2 = \text{diag}(200, 269, 269) \) W/(m·K). (a2)–(c2) (or (a3)–(c3)) are experimental measurements (or simulation results) of the three samples shown in (a1)–(c1), respectively. Other parameters: the thermal conductivity of red copper and air is 397 W/(m·K) and 0.026 W/(m·K), respectively; for (a1), \( r_1 = 1.8 \) cm and \( r_2 = 0.6 \) cm; for (b1), \( r_1 = 4.2 \) cm and \( r_2 = 6.0 \) cm. Panel (d) displays the experimental setup with a sample.

core-shell structure is independent of the core’s area fraction under the two conditions described by eq. (6) (positive relation) and eq. (7) (negative relation).

Meanwhile, we perform the experimental demonstration for the simulation results shown in fig. 2(a1)–(c1); see fig. 3. Figure 3(a1)–(c1) displays our three experimental samples, which are fabricated with copper by laser cutting. The left (or right) edges of the three samples are respectively put into the hot (or cold) sinks with constant temperatures. Then we use the FLIR E60 infrared camera to detect the temperature distribution of the samples, and the measurement results are shown in fig. 3(a2)–(c2), accordingly. By analyzing the temperature distribution in the host (see fig. 3(a2)–(c2)), we confirm that the equivalent thermal conductivities of the core-shell structures shown in fig. 3(a1)–(c1) are (approximately) the same indeed. Furthermore, fig. 3(a3)–(c3) shows the computer simulations corresponding to the three samples shown in fig. 3(a1)–(c1), which just echoes with the experimental results (fig. 3(a2)–(c2)) and the theoretical analysis (fig. 2(a1)–(c1)). Figure 3(d) is an experimental setup.

As far as fig. 2(a2)–(c2) is concerned, we have adopted a shell with apparently negative thermal conductivity. Actually, one may resort to external energy to achieve apparently negative thermal conductivities [20–22]; see also figs. 5 and 6 in the appendix. Apparently negative thermal conductivities and adding extra sources are equivalent only on the level of phenomena. In other words, apparently negative thermal conductivity can automatically generate a local high (or low) temperature, which is impossible. To make it possible, we manually give a local high (or low) temperature to achieve the same phenomena without violating the second law of thermal dynamics. The shells with additional linear heat sources (fig. 5(a1), (b1)) or point heat sources (fig. 5(a2), (b2)) work with the same effect as the shell with apparently negative thermal conductivity (fig. 2(a2), (b2)). The temperatures of the sources are presented in tables 1, 2 and 3 in the appendix.

Besides, we perform finite-element simulations for three dimensions; see fig. 4. Figure 4(a1)–(c1) (or fig. 4(a2)–(c2)) shows simulation results based on eq. (12) (or eq. (13)). Evidently, the temperature distribution outside the core-shell structure is also identical from fig. 4(a1) to fig. 4(c2), which agrees with the results shown in fig. 2 for two dimensions.
Discussion and conclusion. – This work proposed a scheme of correlated self-fixing behavior in thermal conduction. It has potential applications in thermal stabilization, for which one needs to overcome thermal fluctuations resulted from the changes of area/volume fractions due to thermal expansion or contraction, thermal stress concentration, etc.

Also, the uniform temperature distribution in the host of figs. 2–4 can help to hide the core-shell structure from being detected by infrared camera, which is useful for thermal camouflage or illusion [23–33].

Moreover, as mentioned before, the coupling mechanisms of eq. (7) for two dimensions and eq. (13) for three dimensions are similar to the partially resonant composites in electrostatics [19]. In ref. [34], Milton et al. studied the cloaking effects associated with such partially resonant composites in electrostatics. Due to the similar dominant equation, related cloaking effects can be expected in thermotics as well.

In summary, we have solved Laplace’s equation associated with appropriate boundary conditions, which helps to propose a class of intelligent thermal metamaterials based on a core-shell structure with anisotropic thermal conductivities. We have revealed two coupling mechanisms (i.e., eqs. (6) and (7) for two dimensions or eqs. (12) and (13) for three dimensions), which counterintuitively cause the equivalent thermal conductivity of the core-shell structure to be always fixed at a constant value when the area or volume fraction of the core is changed within the full range (namely, from 0+ to 1). Our theoretical results have been verified by both experiments and finite-element simulations. Nevertheless, our results are valid for steady states, rather than for unsteady states since we did not take into account the effects of mass density and heat capacity [35]. Besides thermotics, the present work also offers a different method on how to achieve similar intelligent metamaterials in other fields including electrostatics, magnetostatics and particle dynamics, which mathematically share the same dominant equation.

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Appendix. –

Approaches to achieving apparently negative thermal conductivities: computer simulations. Since the negative coupling relations (eqs. (7) and (13)) contain apparently negative thermal conductivities, we have to adopt external energy to obtain them, in order not to violate the second law of thermodynamics. By considering the uniqueness theorem, we can get the exact temperature values on the two boundaries of the shell. As a result, fig. 5 shows that adding appropriate linear heat sources (fig. 5(a1), (b1)) or point heat sources (fig. 5(a2), (b2)) works in the same way the shell with apparently negative thermal conductivity (fig. 2(a2), (b2)).
Table 2: The required temperature value of each point heat source at the inside boundary of the shell in fig. 5(a2): there are 16 point heat sources at the inside boundary and the point heat sources are numbered in a clockwise direction from 1 to 16.

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<th>Source ($r_1 = 1.8$ cm)</th>
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<tr>
<td>2</td>
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Table 3: The required temperature value of each point heat source at the inside boundary of the shell in fig. 5(b2): there are 16 point heat sources at the inside boundary and the point heat sources are numbered in a clockwise direction from 1 to 16.

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Approaches to achieving apparently negative thermal conductivities: laboratory experiments. We further perform experiments to show the feasibility of applying external heat sources to achieve apparently negative thermal conductivities; see fig. 6. Figure 6(a) is an experimental setup. In the experiment, the conductivity of the whole sample is made of copper with thermal conductivity $54$ W/(m · K), but we add external line heat sources to keep the temperature constant on the boundaries of $X = 4.5$ cm and $X = 17$ cm. Figure 6(b) is the temperature distribution of the sample shown in fig. 6(a), which is detected by using the Flir E60 infrared camera. Panel (c) is the simulation result of the temperature distribution in the sample shown in (a). Panel (d) shows the simulation result of the temperature distribution in the same sample (but without linear heat sources on the boundaries at $X = 4.5$ cm and $X = 17$ cm), whose middle region between $X = 4.5$ cm and $X = 17$ cm has been set to have a thermal conductivity with negative value, $-40$ W/(m · K). Clearly, both fig. 6(c) and fig. 6(d) have the same temperature distribution. This behavior means that one can use additional energy to achieve apparently negative thermal conductivities indeed.
REFERENCES