Breaking the trade-off between thermal and electrical conductivities in the thermoelectric material of an artificially tilted multilayer

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Breaking the trade-off between thermoelectric (TE) parameters has long been demanded in order to highly enhance its performance. Here, we report the ‘trade-off-free’ interdependence between thermal conductivity ($k$) and resistivity ($\rho$) in a TE/metal tilted multilayer and significant enhancement of TE power generation based on the off-diagonal thermoelectric (ODTE) effect, which generates transverse electrical current in response to vertical thermal current. $\rho$ and $k$ can be simultaneously decreased by setting charge flow along more-electrically conductive layer and thermal flow across less-thermally conductive perpendicular direction by decreasing the tilting angle. Moreover, introducing porosity in the metal layer enables to decrease in $k$ without changing $\rho$, because the macroscopic $\rho$ and $k$ of the tilted multilayer is respectively governed by the properties of the TE material and the metal with large dissimilarity. The obtained results reveal new strategies for developing trade-off-free TE materials, which will stimulate practical use of TE conversion for waste-heat recovery.

To reduce worldwide primary energy consumption, much attention is now focused on power generation from enormous amounts of low-temperature waste heat. Its main media is hot water and low pressure steam at temperatures below 200°C commonly found in plants, automobiles, geothermal areas and other locations. Thermoelectric (TE) technology is a suitable solution for waste heat recovery since it can directly convert such low-grade thermal energy to electricity and also construct a power generator into a compact system.

The performance of TE materials is quantified by the figure of merit $ZT = S^2 T / \rho k$ where the individual TE parameters depends on the carrier density as shown schematically in Fig. 1a (Seebeck coefficient: $S$, electric resistivity: $\rho$, thermal conductivity: $k$ and absolute temperature: $T$). Increase in carrier density gives advantages to decreasing $\rho$ and disadvantage to increasing $k$ and decreasing of $S$ (not shown). During the past half century, the ‘trade-off’ between the TE parameters has hampered serious improvement of $ZT$. It has long been demanded to find a way to optimize the TE parameters independently. A recent common approach to overcome the trade-off is to form a so-called ‘PGEC’ (Phonon glass Electron crystal) by nanoscale structuring. The interrelation between $\rho$ and $k$ appears to be almost independent and enhances $\rho^{-1} k^{-1}$ in $ZT$ as schematically shown in Fig. 1b. By tuning the density of the scattering centres, we can selectively scatter phonons with longer mean free paths, and hence, can decrease $k$ without largely degrading $\rho$. However, this PGEC approach can generally only decrease $k$, and the effective range of selective scattering is limited by differences in the phonon and charge mean free path of the material. As an alternative approach beyond the PGECs, here we propose a way to decouple the thermal and charge conducting paths by using the off-diagonal TE (ODTE) effect. This effect typically appears in multilayers with alternate stacks of dissimilar materials such as a TE material and a pure metal. In these multilayer materials, we can change the TE parameters individually and enhance the performance of TE generation. The macroscopic TE properties of these TE/metal multilayers are highly anisotropic between layer parallel and perpendicular directions: $\kappa_\perp \ll \kappa_\parallel$, $\rho_\perp \gg \rho_\parallel$, and $S_\perp \gg S_\parallel$. The thermal and electrical anisotropies originate from the different ways of stack dissimilar components in series and in parallel toward the directions of thermal and charge current. In the layer parallel direction, we observe high thermal and electrical conductivity nearly as high as those of the pure metal components. In contrast, we observe low conductivity similar to the TE material in
the layer perpendicular direction. The TE properties of the multilayer in the layer parallel ($S_{//}$, $\kappa_{//}$ and $\rho_{//}$) and perpendicular ($S_{\perp}$, $\kappa_{\perp}$ and $\rho_{\perp}$) are expressed in

$$\rho_{//} = \frac{1}{\rho_{M}} + \frac{1-a}{\rho_{TE}} \quad \kappa_{//} = a \kappa_{M} + (1-a) \kappa_{TE} \quad S_{//} = \frac{(1-a)\rho_{xx} + a\rho_{xx}}{\frac{1}{\rho_{xx}} + \frac{a}{\rho_{xx}}}$$

$$(1-a)\kappa_{zz} + a\kappa_{zz} \quad S_{\perp} = \frac{\frac{1}{\rho_{xx}} + \frac{a}{\rho_{xx}}}{\frac{1}{\kappa_{zz}} + \frac{a}{\kappa_{zz}}}$$

Eqs. (1) and (2)\textsuperscript{9,10,11}, where $S_{TE}$, $\kappa_{TE}$ and $\rho_{TE}$ are the TE properties of TE material and $S_{M}$, $\kappa_{M}$ and $\rho_{M}$ are the TE properties of the metal and $a$ is the volume occupancy by the metal layer. The ODTE effect is developed in such an anisotropic material by tilting the laminated planes by $\theta$ against the material surface normal. This then generates a transverse TE current ($I_{Q}$) in response to a vertical thermal current ($I_{J}$) as shown in Fig. 1c. Finite element method analysis revealed that a microscopic $I_{Q}$ flows diagonally in each layers, and thus, generating diagonal $I_{Q}$ in each layers as well, which leads to generation of a macroscopically transverse $I_{Q}$ as a whole\textsuperscript{10,15}. TE parameters in tilted multilayer are theoretically formulated by a tensor as Eq. (3)\textsuperscript{13}, where $A$ corresponds to each one of the TE parameters ($S$, $\rho$ and $\kappa$) and $\theta$ is the tilting angle. In the ODTE effect, the figure of merit is described as $Z_{zz}T = S_{zz}T_{zz}^{-1} \rho_{xx}^{-1}$ using $\rho_{xx}$, $\kappa_{zz}$ and $S_{zz}$\textsuperscript{10,12,14}. The TE properties in the multilayer can be manipulated by controlling $\theta$ and combination of dissimilar components, which is a different optimizing way from the conventional TE materials. In this paper, we theoretically

$$\begin{pmatrix} A_{xx} & A_{zz} \\ A_{xx} & A_{zz} \end{pmatrix} = \begin{pmatrix} A, \cos^2 \theta + A_{\perp} \sin^2 \theta & \frac{1}{2} (A_{\perp} - A_{\perp}) \sin 2\theta \\ \frac{1}{2} (A_{\perp} - A_{\perp}) \sin 2\theta & A, \sin^2 \theta + A_{\perp} \cos^2 \theta \end{pmatrix}$$

and experimentally demonstrate trade-off-free interdependences of TE parameters in a Bi$_{0.5}$Sb$_{1.5}$Te$_3$/Ni artificially tilted multilayer and significant enhancements in $Z_{zz}T$ and output power, accomplished by controlling $\theta$ and degree of material porosity.

Equation (3) shows there are two mechanisms which possibly yield trade-off-free interdependences between $\rho_{xx}$ and $\kappa_{zz}$: (1) the coefficient of $A_{xx}$ and $A_{zz}$ is opposite for $\rho_{xx}$ and $\kappa_{zz}$. We can simultaneously decrease $\rho_{xx}$ to $\rho_{//}$ and $\kappa_{zz}$ to $\kappa_{\perp}$ by decreasing $\theta$ ($\sin^2 \theta \ll \cos^2 \theta$); and (2) the extremely large difference between $A_{xx}$ and $A_{zz}$ in $\rho_{xx}$ and $\kappa_{zz}$. $\kappa_{zz}$ is governed by $\kappa_{//}$, which can be approximated by the thermal property of the metal. In contrast, $\rho_{xx}$ is governed by $\rho_{\perp}$, which can be approximated by the electrical properties of TE material. In Fig. 1d and 1e, we schematically show how to introduce the trade-off-free interdependences according to mechanisms (1) and (2), respectively. In Fig. 1d, by decreasing $\theta$, we see that the laminated planes become more parallel to the direction of the transverse $J_{e}$ and thereby, $\rho_{xx}$ approaches to $\rho_{//}$. Simultaneously, the direction of $I_{J}$ becomes closer to orthogonal to the laminated planes, which decreases $\kappa_{zz}$ to $\kappa_{\perp}$. In such a way, we can realize a material with $\kappa_{zz}$, similar to TE materials, and higher electrically conductivity, similar to pure metals. Figure 1e shows that increasing the porosity and changing $\kappa$ of the metal layer yields reduction in $\kappa_{zz}$ without changing $\rho_{xx}$ of the multilayer. This independent change occurs because $\rho_{xx}$ of the multilayer is mainly governed by $\rho$ of the TE material and $\kappa_{zz}$ is mainly governed by $\kappa$ in the metal. The interrelation from mechanisms (1) and (2) simultaneously decrease $\kappa_{zz}$ and $\rho_{xx}$ as well as $\kappa_{zz}$ without degrading $\rho_{xx}$, respectively, which can largely increase $\kappa_{zz}^{-1} \rho_{xx}^{-1}$ in $Z_{zz}T$.

**Results and Discussion**

We have fabricated tilted multilayers of Bi$_{0.5}$Sb$_{1.5}$Te$_3$ (BST) and Nickel (Ni) in a tubular structure using spark plasma sintering (SPS) and then measured the thermal and electric properties\textsuperscript{16}. A tilted layer structure is observed in the cross-section of the tubular shaped measured sample with a thickness ratio $t_{BST}/t_{Ni} \sim 1$ as depicted in Fig. 2. The optical micrograph in the inset of Fig. 2 reveals an existence of joining layer which assures a fine mechanical and electrical junction between the layers. A typical size of the tubular sample is 11 cm-long with an outer and an inner diameter of 14 and 10 mm, respectively. We performed thermal and electrical measurements in custom equipment that mimicked a shell/tube heat exchanger introducing a temperature difference ($\Delta T$) between tube inside.
contrasts with the interrelations in the ordinal TE materials shown. The tilted multilayer of Bi$_{0.5}$Sb$_{1.5}$Te$_3$ and Ni is fabricated by the SPS method. The inset shows optical image around the junction between the two layers.

and outside. The $\Delta T$ was maintained by flowing hot water (95°C and 20 L min$^{-1}$) in the inner side of the BST/Ni tube, and chilled water (10°C and 20 L min$^{-1}$) in the outer side of the BST/Ni tube.

**Dependence on layer tilt angle.** The simultaneous decrease in $\rho_{xx}$ and $\kappa_{zz}$ is theoretically demonstrated by their $\theta$ dependences in the multilayer. We can favourably decrease $\rho_{xx}$ and $\kappa_{zz}$ to $\rho_{yy}$ and $\kappa_{\perp}$ with lowering $\theta$ as already shown in Fig. 1d. To quantitatively elucidate such an interrelation and its effect on $Z_{xz}T$, we calculated the $\theta$ dependence of TE parameters and $Z_{xz}T$ based on Eq. (3). The theoretical result reveals more than 90% decrease in both $\kappa_{zz}$ (25.4 $\rightarrow$ 2.1 W m$^{-1}$ K$^{-1}$) and $\rho_{xx}$ (0.73 $\rightarrow$ 0.04 m$\Omega$cm) with lowering $\theta$ from 90° to 0° in Fig. 3a. This simultaneous decrease in $\kappa$ and $\rho$ is in contrast with the interrelations in the ordinal TE materials shown in Fig. 1a and 1b. Consequently, $Z_{xz}T$ enhances with decreasing $\theta$; $Z_{xz}T$ of 0.4 at $\theta$ $\sim$ 10°, as shown in Fig. 3b. In this calculation, we used the TE parameters ($S$, $\rho$ and $\kappa$) of pelletized BST and Ni fabricated by the same SPS condition as before: 500°C and 50 MPa. (Bi$_{0.5}$Sb$_{1.5}$Te$_3$; $S$ $\sim$ 210 $\mu$V/K, $\rho$ $\sim$ 1.2 m$\Omega$cm and $\kappa$ $\sim$ 1.1 W m$^{-1}$ K$^{-1}$). Ni; $S$ $\sim$ $\sim$ -20 $\mu$V/K, $\rho$ $\sim$ 17 m$\Omega$cm and $\kappa$ $\sim$ 51 W m$^{-1}$ K$^{-1}$ at room temperature).

We experimentally demonstrate the reduction in both $\rho_{xx}$ and $\kappa_{zz}$ and enhancement of output power in the $\theta$ dependence. We found that the resistance monotonically decreased from 6.7 to 2.4 m$\Omega$ as $\theta$ decreased from 55° to 23°, as shown in Fig. 3c. These results are consistent with the behavior of $\rho_{xx}$ shown in Fig. 3a. We also show in Fig. 3c the $\theta$ dependence of the exchanged thermal power ($Q$). The values of $Q$ were experimentally obtained by the relation of $Q = C_w \rho_{ww} F_{cw} T_{diff}$, where $C_w$ and $\rho_{ww}$ is the heat capacity and density of the water, $F_{cw}$ is the cold water flowing rate and $T_{diff}$ is the $\Delta T$ between cold water inlet and outlet. Here we see that $Q$ monotonically decreases from 1.8 to 1.0 kW with decreasing $\theta$ as shown in Fig. 3c. This behavior implies that $\kappa_{zz}$ actually decreases with decreasing $\theta$. We note that $Q$ is expressed by the relation of $Q = UA \Delta T$, where $U$ and $A$ is the overall thermal transfer coefficient and heat transfer surface area, respectively$^{35,37}$. In this condition, the thermal resistance ($R$) along the tube radial direction is described as ($UA$)$^{-1}$ = $R_{H}$ + $R_{zz}$ + $R_{xx}$, where $R_{H}$ and $R_{zz}$ are the hot and the cold side interfacial thermal resistance, respectively; $R_{zz}$ is the bulk thermal resistance of the BST/Ni tube. Assuming that $R_{H}$ and $R_{zz}$ are similar between the fabricated samples, the change in the overall thermal transfer coefficient should largely owe to of the change in $R_{xx}$.

The $\rho_{xx} - \kappa_{zz}$ trade-off interrelation. The decrease in $\kappa_{zz}$ by decreasing $\theta$ is also demonstrated by the increase in voltage as shown in Fig. 3d. In the $\theta$ dependence of generated voltage, we observe values ranging from 0.2–0.28 V with a peak at $\theta$ $\sim$ 30°, which is shifted away from the -$S_{xz}$ maximum around 45° as shown.
in Fig. 3b. This again is a signature of decrease in $\kappa_{zz}$ by decreasing $h$. The simultaneous decrease in $\rho$ and $k$ then steeply and significantly enhanced the output power up to 7.8 W at $\theta \sim 23^\circ$ as shown in Fig. 3d.

**Porosity dependence.** The independent interrelation between $\kappa_{zz}$ and $\rho_{xx}$ is theoretically examined by varying the TE properties of each layer component, as already shown in Fig. 1e. $\kappa_{zz}$ and $\rho_{xx}$ of the multilayer are respectively governed by the properties of Ni and BST, which allowed us to independently control the two quantities. We have investigated how porosity in either layer of BST and Ni affects the multilayer’s TE behaviour based on Eq. (3). In this calculation, we assumed that the TE parameters in the Eqs. (1) and (2) varies with porosity and obey the following relations: $\kappa \sim (1-p) \kappa_0$ and $\rho \sim \rho_0/(1-p)$. Here $p$ is the degree of porosity expressed as $p = (d_0 - d)/d_0 $ where $d$ is the density of the respective porous Ni or BST layer and $d_0$, $\kappa_0$, $\rho_0$ and $S_0$ are the properties of the fully densified components. ($\text{Bi}_0.5\text{Sb}_1.5\text{Te}_3$; $d_0 \sim 0.8 \text{ g/cm}^3$, $\rho_0 \sim 1.2 \text{ m}\Omega\text{cm}$, $\kappa_0 \sim 1.1 \text{ Wm}^{-1}\text{K}^{-1}$ and $S_0 \sim 210 \mu\text{V/K}$; Ni; $d_0 \sim 8.9 \text{ g/cm}^3$, $\rho_0 \sim 7 \mu\text{Ωcm}$, $\kappa_0 \sim 90 \text{ Wm}^{-1}\text{K}^{-1}$ and $S_0 \sim -20 \mu\text{V/K}$ at room temperature). The calculated results show almost independent behaviours between $\kappa_{zz}$ and $\rho_{xx}$ on $p$ by introducing porosity in either the Ni layer or the BST layer as shown in Fig. 4a. Increasing $p$ of the Ni layer (red solid lines) decreases $\kappa_{zz}$ and barely changes $\rho_{xx}$. In contrast, increasing $p$ of the BST layer (blue solid lines) barely changes $\kappa_{zz}$ and increases $\rho_{xx}$. These results predict that $\kappa_{zz}$ and $\rho_{xx}$ in the multilayer are indeed governed by the Ni and BST properties, respectively. We found that the TE parameter variations by introducing porosity in the Ni layer can increase $Z_{xx}T$. Figure 4b shows that $Z_{xx}T$ increases with increasing $p$ over the whole investigated range because $\rho_{xx}$ remains unchanged and $\kappa_{zz}$ favourably decreases with increasing $p$ in the BST layer because $\kappa_{zz}$ remains unchanged and $\rho_{xx}$ unfavourably increases. We therefore see that introducing porosity does not always enhance $Z_{xx}T$. In the case of BST/Ni multilayers, manufacturing porous structure only in the Ni layer is necessary for higher power generation.

By introducing Ni porosity, we experimentally show the independent interrelation between $\kappa$ and $\rho$, and enhanced power generation in the BST/Ni multilayer. Figure 4c summarizes the $p$ dependence of electrical resistance, voltage, and the output power of the porous BST/Ni multilayer tube. We see that the measured resistance was almost constant at ~3.5 mΩ over the range of $p$. On the other hand, the voltage independently increased from 260 to 305 mV with increasing $p$ of the Ni layer. It is reasonable to consider that increase in $p$ of the Ni layer decreased $\kappa_{zz}$ of the entire multilayer while barely impacting $\rho_{xx}$ which is consistent with the prediction in Fig. 4a. As a result, the output power increased to 6.6 W in the 11-cm-long tube. Provided that $p$ could be further increased, it appears the output power could be further enhanced according to the theoretical result in Fig. 4b. In this measurement, we used the multilayer with a porous Ni layer controlled by the pressure ($P_{\text{spS}}$) in the SPS sintering. Figure 4d and 4e show transmission electron microscopy (TEM) images of the dense BST and porous Ni layers, respectively, in the multilayer fabricated by SPS at 500°C and 50 MPa. These micrographs revealed porosity only in the Ni layer. This contrastive texture has been caused by differences in the optimal sintering temperature of BST and Ni. The volume fraction of porosity in the Ni increased from 8 to 50% with decreasing $P_{\text{spS}}$ and we simultaneously observe the decreasing $\kappa$ and increasing $\rho$ in doubly. Table 1 lists the details of $P_{\text{spS}}$ dependence of densification ($d$ and $p$) and consequent variations in $\kappa$ and $\rho$ in the Ni.

**Enhancement of the power generation.** We summarize the enhancement of output power ($P$) and efficiency ($\eta$) of the fabricated

![Figure 4](image-url)
multilayers. We experimentally suppressed $k_{zz}$ largely by 73% without degrading $\rho_{xx}$ in the multilayer through the optimizations by the trade-off-free interdependences. In an 11-cm long BST/Ni multilayer tube, $P$ significantly increased to a maximum of 7.9 W as shown in Fig. 5a. The power density per heat transfer area reached as high as 2.5 kW/m² with a relatively small $\Delta T = 85$ K at temperatures below 100°C. In Fig. 5b, $\eta$ ($\eta = P/Q$) of the multilayer also steeply increased up to 0.83% due to increase in $P$ and decrease in $Q$ as shown in Fig. 5c. The reduction in $Q$ originated from the decrease in $k_{zz}$ of the multilayer. The remarkable feature in our results is the high capability of the multilayer to capture heat effectively ($Q \sim 0.9–1.8$ kW) and to generate high electric power. The highly heat conductive Ni layer (40 ~ 80 Wm⁻¹K⁻¹) yields an excellent heat exchange property with a maximum $Q$ of 1.8 kW. These high $Q$ values go far beyond the conventional TE devices and make it possible to generate large $P$ in spite of the relatively low $\eta$, that is, low $Z_{xx}T$ as shown in the Fig. 3b. From a practical perspective, we can use this material as a bifunctional TE device with efficient heat exchange and high power generation in a standard heat exchanger. Note that, to achieve high $P$, it is important to control $k_{xx}$ of the multilayer. According to Fig. 5a, $k_{xx} \sim 5$ Wm⁻¹K⁻¹ is desirable in the BST/Ni multilayer. We found the differences in peak position between the variances of $P$ and $\eta$ because $Q$ is also $k_{zz}$-dependent. This result shows that to achieve higher power generation, it is more important to control $k_{xx}$ appropriately rather than to enhance $Z_{xx}T$ simply by decreasing $k_{zz}$ of the multilayer.

In this work, we proposed two following approaches to achieve a trade-off-free interrelation between $k$ and $\rho$ using the ODTE effect in a TE/metal tilted multilayer. (1) Decreasing both $k$ and $\rho$ by directing $J_Q$ and $I_p$ simultaneously towards the favourable thermally less conductive and electrically more conductive direction. (2) Independently varying $k_{zz}$ and $\rho_{xx}$ given that $k_{xx}$ and $\rho_{xx}$ are governed by the properties of Ni and BST, respectively. Theoretical and experimental results validated these proposed approaches and suggested the potential of BST/Ni artificially tilted multilayers for high power generation. Through these optimizations, we demonstrated that $k_{xx}$ and $\rho_{xx}$ can be suppressed largely, and significantly increase the output power and efficiency to 2.5 kW/m² and 0.83%, respectively, with a relatively small $\Delta T \sim 85$ K at temperature below 100°C. Present findings resolve the conflict between $k$ and $\rho$ and establish sure design strategies applicable to all effective TE/metal tilted multilayers. The excellent heat exchange capability and high power generation in this multilayer provides an innovative TE application for power generator in the heat exchange system using enormous low temperature waste heat.

**Methods**

**Fabrication of the tubular tilted multilayers.** We have fabricated the tubular shaped tilted multilayer using BST and Ni by spark plasma sintering (SPS) method. This method allows sintering powders, joining two materials and tubular shaping simultaneously. To obtain the tilted multilayer structure, we alternately stacked compacted powders of BST and Ni with tilting along the axial direction before sintering. We used cold pressing to compact the powders into a conical ring, which allowed us to easily stack the layers and achieve a uniform tilt angle. The tilt angle was controlled by the tapered angle of the conical rings (15° ~ 45°). SPS sintering was performed at a fixed temperature ~ 500°C and pressures ~ 30–150 MPa under vacuum. The fabricated tubular composites were typically 35 mm long. The cross-section of the sample shows a well-defined tilted layered structure. The composite exhibited no cracks or coarsened voids and had high mechanical stability. We produced 110-mm-long TE tubes by joining four of these tubular composites with Sn–Bi solder paste.

**Experimental setup to measure the TE properties of the tilted multilayers.** The thermal (exchange thermal power) and electric (resistivity and voltage) measurements were performed using a custom equipment, which simulated a standard shell/tube heat exchanger. The temperature difference between inside and outside of the tube was maintained by flowing hot and cold water, which was controlled by a liquid heater and a chiller unit. The actual water temperatures were measured by a thermocouple sensor attached to the apparatus. Water flow meters were placed in the cold and hot water lines. Resistance and voltage were measured using Cu electrodes, screwed into the exchanger unit to make the electric contact between the materials of BST/Ni.

**Figure 5 | Summary of performance enhancement in the fabricated multilayer by trade-off free optimizations.** Experimentally obtained (a) output power ($P$), (b) efficiency ($\eta$) and (c) thermal exchange power ($Q$) of the fabricated multilayers as a function of $k_{xx}$.

**Table 1 | $P_{sp}$ dependence of the Ni density, degree of the porosity, and $\rho$ and $k$ at room temperature of pelletized Ni samples fabricated at the same SPS condition used for fabricating the BST/Ni tube**

| Ni (SPS pressure) | Density (g/cm³) | $\rho$ (Ω cm) | $k$ (W/mK) |
|------------------|----------------|--------------|------------|
| 30 MPa           | 6.0            | 0.48         | 21         | 40         |
| 50 MPa           | 6.7            | 0.32         | 17         | 51         |
| 100 MPa          | 7.5            | 0.19         | 11.3       | 67         |
| 150 MPa          | 8.2            | 0.08         | 9.7        | 80         |

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**Author contributions**

A.S., H.T. and H.K. fabricated the BST/Ni multilayers. A.S. and K.T. measured the TE properties of the fabricated multilayers. A.S. analyzed the data based on a model developed by T.K. and drafted the manuscript, which was further revised by all authors. T.K. and Y.Y. directed the work. H.A. jointly discussed the experimental results.

**Additional information**

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