Very large thermal rectification in bulk composites consisting partly of icosahedral quasicrystals

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Abstract
The bulk thermal rectifiers usable at a high temperature above 300 K were developed by making full use of the unusual electron thermal conductivity of icosahedral quasicrystals. The unusual electron thermal conductivity was caused by a synergy effect of quasiperiodicity and by a narrow pseudogap at the Fermi level. The rectification ratio, defined by \( \text{TRR} = |J_{\text{large}}|/|J_{\text{small}}| \), reached very large values exceeding 2.0. This significant thermal rectification would lead to new practical applications for the heat management.

Keywords: quasicrystal, thermal diode, electron thermal conductivity

1. Introduction
Thermal rectifiers \cite{1} have attracted considerable interest because of their ability to manage heat, a large fraction of which is typically lost into the environment. After the first report of thermal rectification \cite{2}, several different mechanisms leading to the thermal rectification were reported: (a) a metal-insulator junction \cite{2}, (b) thermal wrapping \cite{3–7}, (c) thermal strain at the interface \cite{8}, (c) the thermal potential barrier \cite{9}, (d) inhomogeneous mass loading \cite{10} and (e) composite of bulk materials possessing different temperature dependences of thermal conductivity \cite{11–15}. Among these thermal rectifiers, (e) the composite of two bulk materials of different temperature dependences has attracted maximum attention because of its superior characteristics, which make it suitable for practical applications. One of these characteristics is tunable heat flux, controlled by the thickness of the composites. In addition, the geometry of the bulk thermal rectifier is not subject to any significant physical constraints, so it can be easily incorporated into a wide range of mechanical components.

The principle of the bulk thermal rectifier, which is a composite consisting of two solid materials joined together, each possessing thermal conductivities (\( \kappa \)) with different temperature dependences, was theoretically proposed by two different groups in 2006 \cite{11, 12}. These theoretical predictions were subsequently validated by experiments \cite{13–15}. Despite the experimental confirmation of bulk thermal rectification, several problems prevent their use in practical applications. One of the most serious problems is the very low working temperature. Thermal rectification in a bulk material was first observed at low temperatures below 150 K \cite{13–15} because these composites made use of the significant temperature dependence of lattice thermal conductivity, typically observed in crystalline materials below 100 K. Unfortunately, increasing the temperature range over which this large variation in lattice thermal conductivity occurs is difficult, especially up to room temperature (300 K). Another problem is the small magnitude of the thermal rectification ratio (\( \text{TRR} = |J_{\text{large}}|/|J_{\text{small}}| \)) observed for the bulk thermal rectifiers; the largest value observed is less than 1.45 \cite{13–15}, which would not be suitable for applications. To make a practical bulk thermal rectifier, we need to greatly increase both the working temperature and the magnitude of the TRR.
We considered that the serious problems could be removed if we could employ an Al-based icosahedral quasicrystal (IQC) as a main component of the bulk thermal rectifier because it is characterized by drastically increasing thermal conductivity with an increasing temperature above 300 K [16, 17]. Indeed, by making composites consisting mainly of IQC, we succeeded in developing new thermal rectifiers that work at high temperatures above 300 K. In this paper, we shall report the performance of our newly developed thermal rectifier, together with the detailed information about the unusual thermal conductivity of Al-based IQC.

2. Unusual electron thermal conductivity of icosahedral quasicrystals

The thermal conductivity of Al-based IQC is characterized by its small magnitude at low temperatures below 300 K and the drastic increase with increasing temperatures (at high temperatures above 300 K). The former is caused by the small magnitude of both the lattice thermal conductivity and electron thermal conductivity. The small lattice thermal conductivity in IQCs is realized due to the quasiperiodicity and its corresponding phonon dispersions in which optical phonon branches exist in the low energy range [18], causing a significant reduction of group velocity and the enhancement of the Umklapp process of phonon scattering. The quasiperiodicity also contributes to the small electron thermal conductivity by enhancing the scattering probability of electrons into the strongest scattering limit known as the Mott–Ioffe–Regel limit [19]. The very small electronic density of states at the Fermi level [20–22] limits the number of conducting electrons and further reduces the magnitude of electron thermal conductivity [23, 24].

Despite that, the lattice thermal conductivity is kept small at high temperatures, and the magnitude of electron thermal conductivity of IQCs drastically increases with the increasing temperature. This unusual electron thermal conductivity is caused by the narrow, deep pseudogap at the Fermi energy.

The electron thermal conductivity is formulated in the context of the linear response theory [25].

\[
\kappa_{\text{el}}(T) = \frac{1}{e^2T} \int_{-\infty}^{\infty} \sigma(\epsilon, T)(\epsilon - \mu)^2 \left( -\frac{\partial f_{\text{FD}}(\epsilon, T)}{\partial \epsilon} \right) d\epsilon - \frac{1}{e^2T} \int_{-\infty}^{\infty} \sigma(\epsilon, T)(\epsilon - \mu)^2 \left( -\frac{\partial f_{\text{FD}}(\epsilon, T)}{\partial \epsilon} \right) d\epsilon \times \left[ \int_{-\infty}^{\infty} \sigma(\epsilon, T)(\epsilon - \mu)^2 \left( -\frac{\partial f_{\text{FD}}(\epsilon, T)}{\partial \epsilon} \right) d\epsilon \right]^2
\]

\[
(1)
\]

Here, \( f_{\text{FD}}(\epsilon, T) \), \( \mu \) and \( e \) represent the Fermi–Dirac distribution function, the chemical potential and the unit charge of electron, respectively. The function \( \sigma(\epsilon, T) \) is known as the ‘spectral conductivity’ that represents the contribution of electronic states existing at \( \epsilon \) to the electrical conductivity. If we use the relaxation time approximation with the isotropic electronic structure, the spectral conductivity is expressed by the following equation

\[
\sigma(\epsilon, T) = \frac{e^2}{3} \int \frac{d\epsilon}{k_B T} \langle \epsilon \rangle^3 \epsilon^2 \left( \right) \tau(\epsilon, T).
\]

(2)

Here, \( N(\epsilon) \), \( \nu(\epsilon) \), and \( \tau(\epsilon) \) are the electronic density of states, the group velocity and the relaxation time, respectively. These three values generally vary with energy and temperature, and the resulting \( \sigma(\epsilon, T) \) also shows significant energy dependence and strong temperature dependence.

When the Seebeck coefficient is smaller than 100 \( \mu V K^{-1} \) [23, 24], the second term of equation (1) is small and can be safely ignored. In such a case, the behavior of electron thermal conductivity is accounted for solely with the first term of equation (1). The integrant in the first term of equation (1) contains \( W(\epsilon) = (\epsilon - \mu)^2 \left( -\frac{\partial f_{\text{FD}}(\epsilon, T)}{\partial \epsilon} \right) \), which behaves as a window function and determines the energy range of electrons that contribute to the thermal conductivity, as shown in figure 1. Obviously, it has two peaks below and above the chemical potential \( \mu \) at around \( \epsilon \approx \mu \pm 2.4k_B T \).

In the case of IQCs, \( \sigma(\epsilon, T) \) has almost temperature independence and becomes directly proportional to \( N(\epsilon) \) because of the quasiperiodicity and consequently introduces a strong scattering limit. Let us now assume that \( N(\epsilon) \) has a

\[
\text{Figure 1. Three-window-functions limiting the energy range of electrons that contribute electron transport properties. The electron thermal conductivity is mainly determined by the function at the top panel.}
\]
deep, narrow pseudogap of a few hundred meV in width, and
the Fermi energy is located at the energy where $N(\varepsilon)$ pos-
sesses the smallest magnitude in the pseudogap. At a low
temperature, $\kappa_{el}$ should be kept very small because of the very
small magnitude of $N(\varepsilon)$ at $\varepsilon \sim \mu \pm 2.4 k_B T$, while it dras-
tically increases with the increasing temperature because the
electronic density of states drastically increases with being
apart from the Fermi energy; consequently, $N(\varepsilon)$ at
$\varepsilon \sim \mu \pm 2.4 k_B T$ drastically increases with the increasing
temperature. This is the reason why we observe a large
evolution of electron thermal conductivity, and this mechan-
ism was quantitatively investigated using the Al-Re-Si and
Al-Mn-Si 1/1-cubic approximants [23, 24].

Dolinšek et al [26] reported that $L(T) = \kappa_{el}(T\sigma(T))$ of
Al-Cu-Fe IQC becomes almost constant above 300 K in their
calculation using equation (1). Their result is certainly
inconsistent with our interpretation of the unusual evolution
of the thermal conductivity of IQCs. We should comment on
this inconsistency and mention that the model of spectral
conductivity used by Dolinšek et al was too simplified for
estimating thermal conductivity at high temperatures above
300 K. Their spectral conductivity consisted of two Lor-
entzian functions in the same manner as that reported by
Landauer and Sollbrig [27], and the detailed shape was
determined so as to reproduce the measured transport prop-
erties below 300 K using the equations deduced from the
linear response theory. Their spectral conductivity should be
highly reliable in the narrow energy range near the Fermi
energy because of the function fitting on experimental data.
However, it should not be reliable at the energy range apart
from the Fermi energy where high temperature properties are
determined because the transport properties below 300 K do
not contain the information about the spectral conductivity at
that energy range. This should be the reason why Dolinšek
et al [26] reported behavior of electron thermal conductivity
that is certainly inconsistent with our interpretation.

The reliability of our argument on the unusual increase of
electron thermal conductivity can be confirmed in the
experimental facts: (1) the magnitude of the Seebeck coeffi-
cient is closely related with the evolution of thermal con-
ductivity at high temperatures [22], (2) the electron thermal
conductivity of 1/1-cubic approximants shows almost the
same behavior of that calculated from the electronic density of
states determined theoretically and experimentally [23, 24]
and (3) the behavior of thermal conductivity sensitively varies
with carrier concentration [23, 24, 28]. These facts lend great
support to our scenario of an unusual increase of electron
thermal conductivity at high temperatures.

3. Thermal conductivity of Al-Cu-Fe icosahedral
quasicrystals

As a result of the analyses on the electronic structure and its
relation to the unusual behavior of electron thermal con-
ductivity [23, 24], we realized that the Al-based IQC and their
Corresponding approximants show a significant increase of
thermal conductivity with the temperature, provided that
those IQCs and their approximants contain 3d transition metal
elements as one of the main constituent elements, rather than
4d or 5d transition metal elements. This tendency is caused by
the narrower width of the pseudogap in IQC, which contains
3d transition metal elements. The narrower width of the
pseudogap for the IQC and their approximants containing 3d
transition metal elements is also understood from the behavior
of the Seebeck coefficient, which increases with the increasing
temperature and starts to decrease after becoming maxi-
mal at the $T_{\text{peak}}$. The peak temperature $T_{\text{peak}}$ roughly
represents the width of the pseudogap, and the IQCs and their
approximants containing 3d elements possess lower $T_{\text{peak}}$
that those containing 4d and/or 5d elements [29, 30].

These considerations, together with the very small electronic
density of states at the Fermi energy reported for Al-Cu-Fe
IQC [20], prompted us to employ the Al-Cu-Fe IQC for the
most appropriate material possessing a drastic increase of
electron thermal conductivity with increasing temperatures.

Figure 2(a) shows the thermal conductivity of Al-Cu-Fe
IQC. The mother ingots of Al-Cu-Fe IQC were prepared by
induction melting under a pressurized argon atmosphere.
The ingots were crushed into powders and sintered using a pulse-
current sintering technique for making dense samples free of
voids and cracks [28, 31]. Since electron thermal conductivity
of IQC sensitively varies with the carrier concentration, we
prepared several different samples possessing different carrier
concentrations. We also measured the Seebeck coefficient of
the samples (see figure 2(b)) because the magnitude of the
Seebeck coefficient is supposed to be small when the electron
thermal conductivity shows a drastic increase at high tem-
peratures. The magnitude of the Seebeck coefficient becomes
small at $\text{Ag}_{61.5}\text{Cu}_{26.5}\text{Fe}_{12}$ where the ratio of thermal con-
ductivity at 1000 K to that at 300 K, $\kappa_{\text{1000K}}/\kappa_{\text{300K}}$ possesses
the largest value of 8.9 [28]. Therefore, we decided to employ
this $\text{Ag}_{61.5}\text{Cu}_{26.5}\text{Fe}_{12}$ IQC as one of the main components of the
thermal rectifier.

4. Materials possessing thermal conductivity that
decreases with increasing temperatures

We selected Si, $\text{Al}_2\text{O}_3$, $\text{CuGeTe}_2$ and $\text{Ag}_2\text{Te}$ as the materials
possessing thermal conductivity that decrease with increasing
Figures. Figure 3 shows the thermal conductivity of these
materials plotted as a function of temperature.

The downward trend in thermal conductivity with increasing
Figures observed for $\text{Al}_2\text{O}_3$ and Si is easily
understood as a consequence of the lattice thermal con-
ductivity of an insulator possessing a high Debye temperature
($\Theta_D$). These values have been reported as $\Theta_D = 1000–1100$ K
for $\text{Al}_2\text{O}_3$ [32] and $\Theta_D = 600–650$ K for Si [33]. In such
materials, lattice thermal conductivity moderately decreases with
the temperature as a result of the intensified Umklapp process of phonon-phonon scattering.

The temperature-dependent behavior of $\kappa$ in CuGaTe$_2$, on
the other hand, is difficult to interpret. Its Debye temperature
was reported to be very low: $\Theta_D = 226$ K [34], and this low
$\Theta_D$ indicates that lattice vibrations at high temperatures
should not be considered as ‘conducting wave packets’ but rather as ‘intensely exited, localized oscillators.’ In materials such as these, the lattice thermal conductivity should have no temperature dependence at high temperatures ($T > \Theta_D$), but this consideration is not the case with the experimentally observed variation of $\kappa$ of CuGaTe$_2$. Although the mechanism that produces large reductions in $\kappa$ with the increasing temperature is not yet well understood, we are justified in employing CuGaTe$_2$ as one of the components of our rectifier partly because its $\kappa$ is very small to be comparable with that of the IQC.

The thermal conductivity of Ag$_2$Te shows a big reduction at 420 K with increasing temperatures [35]. At temperatures above 420 K, silver ions start to wander in the samples [36]. The mobile silver ions presumably prevent the propagation of the wave packet or directly prohibit the existence of well-defined wave packets; therefore, the lattice thermal conductivity shows very small values.

5. Calculation of thermal rectification ratio TRR

$$\text{TRR} = \left| \frac{J_{\text{large}}}{J_{\text{small}}} \right|$$

is determined not only by the temperature dependence of the thermal conductivity of two constituent materials but also by their length ratio $x = L_{\text{IQC}}/(L_{\text{IQC}} + L_X)$ ($X = \text{Si, Al}_2\text{O}_3$, CuGeTe$_2$, or Ag$_2$Te). Before preparing the composite samples, we estimated the optimal length ratio $x_{\text{opt}}$ for the maximum TRR obtainable for the given set of materials. The calculation method was reported previously [31].

The $x$ dependence of TRR was calculated for the composite thermal rectifiers consisting of (a) Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC/Si, (b) Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC/Al$_2$O$_3$, (c) Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC/CuGeTe$_2$, and (d) Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC/Ag$_2$Te by assuming that the composites are placed between two heart reservoirs kept at ($T_H$, $T_L$) = (900 K, 300 K) for (a)–(c), and ($T_H$, $T_L$) = (543 K, 300 K) for (d). The resulting TRR is plotted as a function of $x$ in figures 4(a1)–(a4).

The calculations predict that very large values of TRR exceeding 2.0 can be obtained in three composites: IQC/Si, IQC/Al$_2$O$_3$, and IQC/CuGaTe$_2$, placed between two heat sources.
reservoirs kept at 900 K and 300 K. Each composite has its own optimum value of $x$ ($x_{\text{opt}}$) in which the largest value of $\text{TRR}_{\text{calc}}$ is attained. The $x_{\text{opt}}$ values were 0.08, 0.21 and 0.55 for IQC/Si, IQC/Al$_2$O$_3$ and IQC/CuGaTe$_2$, respectively.

It is also worthwhile to mention that the large value of $\text{TRR}$ exceeding 1.75 is obtainable even at the small temperature difference from 543 K to 300 K in the composite of ICQ/Ag$_2$Te when the length ratio is $x=0.65$. Although the predicted value of $\text{TRR}_{\text{calc}}=1.75$ is slightly smaller than those of the other three composites, the much smaller temperature difference of $\Delta T=243$ K than $\Delta T=600$ K of the other composites could have advantages in practical applications.

The conditions that determine $x_{\text{opt}}$ are clearly understood from the $x$-dependence of the interface temperature, as shown in figures 4(b1)-(b4).

At $x_{\text{opt}}$, the interface temperature ($T_{\text{boundary}}$) possesses the same value for two different sample configurations: one configuration with the IQC at the high-temperature side and the other with the IQC at the low-temperature side. This result indicates that the ratio of the thermal resistance of the IQC at the high temperature side to that of the one of the other composite.

Figure 4. (a) Thermal rectification ratio, (b) boundary temperature and (c) measured heat flux of the (1) IQC/Si, (2) IQC/Al$_2$O$_3$, (3) IQC/CuGaTe$_2$ and (4) IQC/Ag$_2$Te composites.
materials at the low temperature side becomes equal to the ratio of thermal resistance for the one of the other materials at the high temperature side to that of the IQC at the low temperature side.

In addition, $x_{\text{opt}}$ requires a longer length of the material with the larger thermal conductivity, and vice versa. We obtained the smallest value of $x_{\text{opt}}$ in the IQC/Si composites, because Si possesses a much larger $\kappa$ than the Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC. $x_{\text{opt}}$ is in the vicinity of 0.5 for the IQC/CuGaTe$_2$ and IQC/Ag$_2$Te composites because of the comparable $\kappa$-values of the two components. The calculated $x_{\text{opt}}$ the boundary temperature at $x_{\text{opt}}$ and the estimated maximum value of $\text{TRR}_{\text{calc}}$ are listed in Table 1.

### 6. Heat flux measurement for determining the TRR

To experimentally confirm the very large values of the TRR predicted from our calculations, we directly measured the heat flux in the IQC/Si, IQC/Al$_2$O$_3$, and IQC/CuGaTe$_2$ composites placed between two heat reservoirs kept at $T_L \approx 300$ K and $T_H = 900$ K in vacuum and that in the IQC/Ag$_2$Te placed at $T_L \approx 300$ K and $T_H = 550$ K.

The samples have cylindrical shapes 20 mm in height and 10 mm in diameter, and the length ratios $x$ were fixed at $x_{\text{opt}}$ for all of the composites. The heat flux $J$ in the composite was estimated using a simple apparatus comprised of a heater, one of the composites, a copper block and a water-cooled block placed in a hand-press. The entire measurement system was sealed in a chamber, and the measurements were conducted under vacuum better than 10 Pa [28, 31]. To obtain good sealed in a chamber, and the measurements were conducted under vacuum better than 10 Pa [28, 31].

The measured heat fluxes are plotted in figures 4(c1)–(c4) as a function of heating time. Large differences in heat flux were observed between the measurements made in the two opposite directions, and the heat flux is always larger when the IQC is located at the high-temperature side. The experimentally observed TRR ($\text{TRR}_{\text{exp}}$) values for the IQC/Si, IQC/Al$_2$O$_3$, IQC/CuGaTe$_2$, and IQC/Ag$_2$Te composites were $1.81 \pm 0.16$, $2.03 \pm 0.16$, $2.20 \pm 0.13$ and $1.65 \pm 0.20$, respectively. These values are much larger than those of any other bulk thermal rectifiers previously reported [13–15, 28, 31, 37].

### 7. Discussions

The $\text{TRR}_{\text{exp}}$ values were nearly consistent with the $\text{TRR}_{\text{calc}}$ values for the IQC/CuGaTe$_2$ and IQC/Ag$_2$Te devices, whereas the other two rectifiers possessed $\text{TRR}_{\text{exp}}$ values that were slightly smaller than $\text{TRR}_{\text{calc}}$. This fact is clearly confirmed by superimposing the $\text{TRR}_{\text{exp}}$ data on the $\text{TRR}_{\text{calc}}$ data in figure 4(a). Additionally, by calculating the ratio of $\text{TRR}_{\text{exp}}$ to $\text{TRR}_{\text{calc}}$, we discovered that this ratio is closely related to the averaged thermal conductivities of component materials, estimated from the temperature distribution in the composites and the temperature dependence of $\kappa$ for each material. The values of $\text{TRR}_{\text{exp}}/\text{TRR}_{\text{calc}}$ were 0.97, 0.96, 0.92 and 0.86 for IQC/CuGaTe$_2$, IQC/Ag$_2$Te, IQC/Al$_2$O$_3$ and IQC/Si, respectively. Their averaged $\kappa$ for the two different directions of heat flow were calculated to be (2.78 W m$^{-1}$ K$^{-1}$, 6.08 W m$^{-1}$ K$^{-1}$), (1.55 W m$^{-1}$ K$^{-1}$, 2.63 W m$^{-1}$ K$^{-1}$), (7.48 W m$^{-1}$ K$^{-1}$, 15.97 W m$^{-1}$ K$^{-1}$), and (19.53 W m$^{-1}$ K$^{-1}$, 40.93 W m$^{-1}$ K$^{-1}$), respectively. Very small values of $\text{TRR}_{\text{exp}}/\text{TRR}_{\text{calc}}$ were obtained for the IQC/Si composite, which possesses the largest values of averaged $\kappa$, whereas $\text{TRR}_{\text{exp}}/\text{TRR}_{\text{calc}}$ reached nearly equal to unity in the IQC/CuGaTe$_2$ and IQC/Ag$_2$Te composites, which possess the very small values of averaged $\kappa$.

We firstly considered the effect of radiation emitted from the sidewall of the samples. The amount of radiation loss is not negligibly small but occasionally reaches a seriously large value. It is strongly affected by the dimension and thermal conductivity of the samples and becomes large for long, narrow samples of lower thermal conductivity. In the present samples with cylindrical shapes, we confirmed that the radiation loss was less than 10%, and the variation of TRR due to the radiation was less than a few %. Therefore, we safely ignored the effect of radiation loss in our samples.

We eventually realized that the departure of $\text{TRR}_{\text{exp}}/\text{TRR}_{\text{calc}}$ from unity would be related to the contact resistance for heat flow at the interfaces between the heat reservoirs and the devices because it is capable of significantly reducing the highest temperature or significantly increasing the lowest temperature of the composite. The thermal conductivity of the Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC and the CuGaTe$_2$ was small enough so that the effect of the contact resistance could be safely ignored; therefore, $\text{TRR}_{\text{exp}}/\text{TRR}_{\text{calc}}$ was nearly equal to unity for the IQC/CuGaTe$_2$ composite. In the case of the IQC/Si composite, on the other hand, the thermal conductivity of Si is

### Table 1. Thermal rectification ratio of the present composites.

| Material A          | Material B | $T_L$/K | $T_H$/K | $\text{TRR}_{\text{exp}}$ | $\text{TRR}_{\text{calc}}$ | $\text{TRR}_{\text{exp}}/\text{TRR}_{\text{calc}}$ |
|---------------------|------------|---------|---------|---------------------------|---------------------------|-----------------------------------------------|
| Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC Si | 900        | ~300    | 1.81 ± 0.16 | 2.10                      | 0.86                      |
| Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC Al$_2$O$_3$ | 900        | ~300    | 2.01 ± 0.13 | 2.17                      | 0.92                      |
| Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC CuGaTe$_2$ | 900        | ~300    | 2.20 ± 0.13 | 2.26                      | 0.97                      |
| Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC Ag$_2$Te | 550        | ~300    | 1.65 ± 0.16 | 1.75                      | 0.96                      |
so large that the temperature distribution in the samples is greatly affected under the presence of contact resistance. The highest temperature of the composite would be reduced significantly when Si is located at the hot side, while the lowest temperature would be increased at the opposite configuration. The temperature difference between the two edges of the composite is certainly reduced, and the value of the TRR is also reduced.

The contact resistance between two component materials, on the other hand, does not seriously affect the value of the TRR because the value is dominated by the thermal conductivity near the highest and lowest temperatures, whereas the interface of two component materials always stays in the middle temperature range.

If our consideration is correct, we still have a chance to observe a large TRR exceeding 2.0 even for the IQC/Si composite by tuning the heater power using the temperature exactly at the interface rather than that in the heater block. This requires the modified experimental setup; we are now in progress on it, and the results will be reported elsewhere in the near future.

Before the closing discussion, it would be worthwhile to mention the further increase of the TRR. Let us assume a composite consisting of two materials, one of which possesses thermal conductivity that linearly increases with temperature, and the other linearly decreases. For the sake of easy calculation, the increasing ratio and decreasing ratio of thermal conductivity in two materials were assumed to be the same as $\frac{\kappa_T}{\kappa_L}$ for Material A and $\frac{\kappa_L}{\kappa_H}$ for Material B, where $N > 1$. The value of TRR$_{\text{model}}$ was calculated as a function of $N$ under this condition, and the resulting values were plotted in figure 5.

Obviously, the TRR increases with increasing $N$ and gradually approaches the maximal value 3. The Al-Cu-Fe IQC possesses $N = 7.0$ under the condition of $T_H = 900$ K and $T_L = 300$ K, and the TRR $\sim 2.2$ observed for the composite consisting mainly of this IQC is very close to the TRR$_{\text{model}}$ at $N = 7.0$. These facts indicate that our samples have already stayed in very good condition for the thermal rectifier and that it would not be very easy to obtain much larger TRR values exceeding 3.0.

Nevertheless, if we employed materials possessing thermal conductivity that increases with temperature much more drastically than those with linear dependence, the maximum value should be increased to a value larger than 3.0. The thermal conductivity of IQCs possesses such a behavior; therefore, we keep studying toward the goal of developing a thermal rectifier possessing a TRR exceeding 3.0 using IQCs. The result will be reported elsewhere in the near future.

8. Conclusion

In this study, we developed a new thermal rectifier working at high temperatures above 300 K and possessing a large TRR exceeding 1.65 using Al$_{61.5}$Cu$_{26.5}$Fe$_{12}$ IQC together with one of the following materials: Si, Al$_2$O$_3$, CuGeTe$_2$ or Ag$_2$Te. The values of TRR obtained in this study were definitely the largest among those ever reported; therefore, some of these composites could be used in practical applications, leading to the efficient use of energy.

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