Thermoelectric Properties of Al–Ga–Pd–Re Icosahedral Quasicrystals

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Various compositions of Al–Ga–Pd–Re icosahedral quasicrystals (QCs) were synthesized using arc-melting and annealing methods, and their thermoelectric properties were investigated. With the same trend in the Seebeck coefficient as Al–Pd–Mn and Al–Pd–Re QCs, the Al–Ga–Pd–Re QC has a similar pseudogap electronic structure near the Fermi level. More particularly, we found that a sample with a nominal composition of $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ exhibited a higher Seebeck coefficient of $90 \mu\text{V K}^{-1}$ at 373 K, and is thus a highly efficient thermoelectric material. This dense sample, having neither cracks nor pores, shows a 1.5 times higher dimensionless figure of merit ZT of 0.18 compared with sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$. ZT enhancement through Ga substitution for Al is enabled through an increase in both electrical conductivity and Seebeck coefficient, and a decrease in phonon thermal conductivity. This behavior is discussed in terms of the precipitation of metallic secondary phases and the “weakly bonded rigid heavy clusters (WBRHCs)” scheme applicable to cluster-based solids including quasicrystals.
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1. Introduction

Thermoelectric materials can directly convert waste heat from automobiles, facilities, and power plants into electrical energy. Recently, thermoelectric power generation from solar heat was also tested.¹⁾ It has been demonstrated that solar heat conversion technique is a potential environmentally friendly and sustainable power generation for our future energy saving society. However, the conversion efficiency of the thermoelectric power generators is not high enough for practical application. The potentiality of the thermoelectric materials is indicated in terms of the dimensionless figure of merit, $ZT = S^2\sigma T/\kappa$, where S , σ , κ , and T are the Seebeck coefficient, the electrical conductivity, the total thermal conductivity, and the temperature.²⁾ One of the criteria for the practically applicable thermoelectric materials is over unity ZT ones. To obtain a high ZT material, one must search for a large power factor, $S^2\sigma$, materials. The power factor is mainly governed by the local electronic structure in the vicinity of the Fermi level, E_F . Most of high $S^2\sigma$ and high ZT materials are pseudogap or narrow-gap compounds such as doped-PbTe,^{3,4)} Zn_4Sb_3 ,⁵⁾ skutterudites,⁶⁾ clathrates,⁷⁾ silicides,⁸⁾ and gallides compounds.⁹⁾

Al–Pd–Mn and Al–Pd–Re icosahedral quasicrystals (QCs) are potential thermoelectric materials¹⁰⁾ because their aperiodic complex crystal structure brings glass-like low thermal conductivities of about $1 \text{ W m}^{-1} \text{ K}^{-1}$. These quasicrystals also have large Seebeck coefficients of over $90 \mu\text{V K}^{-1}$ because of the pseudogap formed near E_F . In pioneering studies of the quasicrystals as thermoelectric materials, Pope *et al.*¹¹⁾ and Kirihaara *et al.*¹²⁾ reported the thermoelectric properties of ternary Al–Pd–Mn and Al–Pd–Re QCs, respectively. Our group has proposed a guiding principle, i.e., “weakly bonded rigid heavy clusters (WBRHCs)”, for improving the thermoelectric properties in icosahedral cluster solids.¹³⁾ Ru and Fe substitution for Re in an Al–Pd–Re QC yield $ZT_{\text{max}} = 0.15$ ¹⁴⁾ and $ZT_{\text{max}} = 0.21$ ¹⁵⁾ respectively, and Ga substitution for Al in an Al–Pd–Mn QC yields $ZT_{\text{max}} = 0.26$.^{16,17)}

In the limit of the nearly-free-electron model, the electrical conductivity, σ , and Seebeck coefficient, S , are expressed by the following equations.^{15,18)}

$$\sigma = \frac{ne^2\tau}{m^*}, \quad (1)$$

$$S = \frac{\pi^{2/3}}{3^{2/3}} \frac{k_B^2}{e\hbar^2} T \left(\frac{m^*}{n^{2/3}} \right), \quad (2)$$

where n , e , m^* , τ , k_B , and $\hbar$ represent the carrier concentration, the unit charge, the effective mass of electron, the relaxation time, the Boltzmann constant, and the reduced Planck constant, respectively. Therefore, the power factor, $S^2\sigma$, is simply proportional to m^* and τ , and inversely proportional to $n^{1/3}$:

$$S^2\sigma \propto \tau \cdot m^* \cdot n^{-1/3}. \quad (3)$$

It is expected that an increase in m^* will enhance $S^2\sigma$ by weakening the intercluster bonds and strengthening the intracluster bonds.^{13–15)} In general, the thermal conductivity, κ , can be divided into two contributions from electron and phonon:

$$\kappa = \kappa_{\text{el}} + \kappa_{\text{ph}}. \quad (4)$$

From the simple harmonic oscillator approximation, the phonon thermal conductivity, κ_{ph} , can be written as^{15,19)}

$$\kappa_{\text{ph}} = \frac{1}{3} C v_s^2 \tau_{\text{ph}} = \frac{1}{3} C \frac{a^2 K}{M} \tau_{\text{ph}}, \quad (5)$$

where C , v_s , a , K , M , and τ_{ph} are the specific heat, the speed of sound, the distance between clusters, the force constant of bond between clusters, the mass of cluster, and the relaxation time of phonon, respectively.

As for Al–Ga–Pd–Mn QCs,^{16,17)} whereas the strength of intercluster bonds was seen to weaken with Ga substitution, confirmed by measurements of the speed of sound, there were no significant changes in σ and S for up to 4% Ga substitution of Al sites. The main reason for the ZT enhancement from 0.18 to 0.26 was the lowering of phonon thermal conductivity by both weakening the inter-cluster bonds and alloying.

We expected a similar ZT enhancement in Al–Pd–Re QCs to that in Al–Pd–Mn QCs, may occur from substituting Ga

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Fig. 1 Nominal composition diagram of (Al+Ga)–Pd–Re system. The samples are identified by nos. 1–13.

for Al atoms to weaken the intercluster bonds. In this study, the thermoelectric properties of various Al–Ga–Pd–Re QCs compositions were investigated. Since Ga substitution for Al was limited to less than 4 at%, as found in Al–(Ga)–Pd–Mn QCs,^{16,17)} therefore, we prepared uniform Ga concentrations with 4 at% for all samples.

2. Experimental Procedure

The investigated nominal composition diagram of Al–Ga–Pd–Re QCs is displayed in Fig. 1. Mother ingots were synthesized using an arc-melting technique (NEV-ACD-05, NISSIN GIKEN Co., Japan) under an argon atmosphere, and then sealed in a quartz tube under an argon atmosphere. The ampoules were placed in an electric furnace and kept at 1198 K for 48 h. The bulk was crushed to an average particle size of less than 45 μm with an agate mortar and a stainless sieve, and the powder was placed in a carbon die of diameter 10 mm for spark plasma sintering (SPS) (SPS-515S, Fuji Electronic Industrial Co., Japan). The temperature of the specimen was increased from room temperature to a consolidation temperature of 1198 K for 10 min and held for 10–15 min. Pressure of 57 MPa was applied during sintering which was conducted in an argon atmosphere to prevent oxidation.

The characterization of the samples was performed by X-ray diffraction (XRD) (Rint2000, Rigaku Co., Japan) measurements with Cu $K\alpha$ radiation. The local compositions were examined by conventional scanning electron microprobe-energy dispersive X-ray (SEM-EDX) (JSM-5600, JEOL Ltd., Japan) measurements. The electrical conductivity and Seebeck coefficient were measured in a helium atmosphere at temperatures between 373 and 1000 K using the four-probe and steady-state temperature gradient methods, (ZEM-1, ULVAC-RIKO Co., Japan). The thermal conductivity was obtained by measuring geometrical and powder densities using the He pycnometer (AccuPyc 1330, SHIMADZU Co., Japan), specific heat, and thermal diffusivity from 373 to 973 K using the laser flash method (TC-7000, ULVAC-RIKO Co., Japan).

3. Characterization

We obtained dense samples with relative density close to 100% after SPS; e.g., the powder and geometric densities of $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QC are both 6.40 g cm^{-3} . Figure 2 shows XRD patterns of arc-melted and annealed Al–Ga–Pd–Re QCs (samples are identified by nos. 1–13). Except for sample no. 7 ($\text{Al}_{68}\text{Ga}_4\text{Pd}_{19}\text{Re}_9$) and no. 12 ($\text{Al}_{67}\text{Ga}_4\text{Pd}_{20.5}\text{Re}_{8.5}$), small amounts of precipitations of metallic $\text{Al}_{11}\text{Re}_4$ and/or Al_3Pd_2 were observed. Although the local compositions were examined by SEM-EDX, the observed sample compositions deviated from nominal ones due to the weight loss during arc-melting and widely scattered depending on the probe position. When discussing the composition dependence of Seebeck coefficient, we adopted the nominal composition instead of the SEM-EDX analyzed one to compare with other quasicrystals,²⁰⁾ because there were no systematic composition dependence in the latter case.

4. Seebeck Coefficients of Arc-melted and Annealed Al–Ga–Pd–Re Quasicrystals

Figure 3 shows the temperature dependence of the Seebeck coefficient for arc-melted and annealed Al–Ga–Pd–Re QCs. In general, arc-melted and annealed Al–Pd–Re QCs contain many cracks and pores that dramatically decrease the electrical conductivity but these defects do not affect the value of the Seebeck coefficient.²⁰⁾ Therefore, we first measured S for various compositions of Al–Ga–Pd–Re QCs to determine a base material for further evaluations of the electrical and thermal conductivities.

It is well-known that for Al–Pd–Re QCs the magnitude of S and its temperature dependence strongly depend on the composition.¹²⁾ For Al–Ga–Pd–Re QCs, S at 373 K varies widely $-5 < S_{373\text{K}} < 90\text{ }\mu\text{V K}^{-1}$, as shown in Fig. 3. This can be qualitatively understood by the difference in the electronic density of states at E_F , $N(E_F)$;

$$S \propto -\frac{1}{N(E_F)} \cdot \left[\frac{dN(E)}{dE} \right]_{E=E_F}. \quad (6)$$

For quasicrystals, $N(E)$ is difficult to calculate because of the absence of periodicity. Instead, we adopt the average valence electron number per atom (e/a) which is frequently used in discussions of the physical properties^{10,12)} and phase stabilization^{21,22)} of quasicrystals. Recently suggested valences of transition metals, such as $(e/a)_{\text{Mn}} = 1.05$, $(e/a)_{\text{Re}} = 1.31$, and $(e/a)_{\text{Pd}} = 0$, will be adopted in discussing phase stabilization.²²⁾ However, when discussing physical properties, such small differences in valences between Pd (group-10) and Mn or Re (group-7), which are three groups separated in the periodic table, are difficult to apply when considering composition changes within the pairing. Therefore, we have adopted the Raynor's valences of the transition metals, $(e/a)_{\text{Mn}} = -3.66$, $(e/a)_{\text{Re}} = -3.66$, and $(e/a)_{\text{Pd}} = 0$,²³⁾ for all e/a calculations. Alternatively, the valence electron concentration (VEC) has often been used to explain the E_F shift with changes in physical properties.²⁴⁾ When using VEC, a substitution of Al (VEC = 3) for Mn or Re (VEC = 7) is responsible for the shift in E_F to higher energy because of the increase in VEC, though our

Fig. 2 X-ray diffraction patterns of arc-melted and annealed Al-Ga-Pd-Re QCs: (left) nos. 1–6, (right) nos. 7–13. Arrows show the secondary phases of $\text{Al}_{11}\text{Re}_4$ and Al_3Pd_2 .

Fig. 3 Seebeck coefficient, S , as a function of temperature for the arc-melted and annealed Al-Ga-Pd-Re QCs.

experimental results show the E_F shifts to lower energy. The reason of this discrepancy is that transition metals such as Mn or Re supply not only more valence electrons but also more electronic states, including d -states, than Al. Because d -states electrons barely contribute to the electrical conduction, the effective number of valence electrons apparently will decrease. The negative e/a values of transition metals reflect these mentioned effects, though the reason for the absolute value is not clear.

Fig. 4 Seebeck coefficient at 523 K, $S_{523\text{K}}$, as a function of valence electron number per atom (e/a) for Al-Ga-Pd-Re, Al-Pd-Re,^{12,20)} and Al-Pd-Mn²⁰⁾ QCs. The solid line is drawn to guide the eyes.

Figure 4 shows the e/a dependence of S at 523 K for various compositions of the Al-Ga-Pd-Re QCs, together with those for Al-Pd-Re^{12,20)} and Al-Pd-Mn²⁰⁾ QCs. Both Al-Pd-Re and Al-Pd-Mn QCs give a similar trend with the maximum $S_{523\text{K}}$ near $e/a = 1.80$ and minimum $S_{523\text{K}}$ above $e/a = 1.86$. From eq. (6), this trend can be understood by assuming a pseudogap near E_F . Both quasicrystals exhibit p -type conduction, i.e., holes are the dominant charge carrier, indicating that E_F is located below the bottom of the

pseudogap. With increasing e/a , the magnitude of S decreases because of a shift in E_F to the bottom of the pseudogap. If E_F is found just at the bottom, the magnitude of S will be minimized. Indeed, $\text{Al}_{72.5}\text{Pd}_{19}\text{Re}_{8.5}$ ($e/a = 1.864$) shows a value that is nearly zero.¹²⁾

Al–Ga–Pd–Re QCs also exhibit a strong e/a dependence in $S_{523\text{K}}$. Although the maximum $S_{523\text{K}}$ value is almost identical to that of Al–Pd–Re and Al–Pd–Mn QCs, its e/a dependence is slightly shifted. It is possible that precipitation of the metallic secondary phases of $\text{Al}_{11}\text{Re}_4$ and Al_3Pd_2 from Ga substitution for Al, decreases S ; this is also observed for 4 at% Ga substituted Al–Ga–Pd–Mn QCs.¹⁶⁾ Nevertheless, the trend can be understood by the rigid-band-like change with pseudogap near E_F .

For further evaluation of the thermoelectric properties, $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ (no. 1) QC with the maximum $S_{523\text{K}}$ of $95 \mu\text{V K}^{-1}$ was selected.

5. Thermoelectric Properties of Sintered $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ Quasicrystal

XRD patterns of (a) an arc-melted and annealed sample and (b) a sintered sample were compared (Fig. 5). Although small amounts of the metallic secondary phases of $\text{Al}_{11}\text{Re}_4$ and Al_3Pd_2 are still present, the primary strong peaks are identified as belonging to the icosahedral phase. Peak broadening after SPS was also observed in $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC.²⁰⁾

The electrical conductivities, σ , of (a) the arc-melted and annealed, and (b) the sintered $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QCs, together with the sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC,²⁰⁾ are plotted in Fig. 6. The σ for the sample before SPS yields a low value of $134 \Omega^{-1} \text{cm}^{-1}$ that arises because of inner cracks and pores. After SPS, σ improved by about six fold without changing the temperature dependence. Compared with $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC,²⁰⁾ σ increased about 1.4 times in the measured temperature range. This increase can again be attributed to the precipitation of metallic secondary phases.

From the temperature dependence of the Seebeck coefficient (Fig. 7), there was no significant difference in S for $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QCs before and after SPS, confirming that the sample's microstructure has no influence on S .²⁰⁾ S of $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QC above 500 K is slightly higher than that of $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC,²⁰⁾ which in accordance with the WBRHCs principle is expected after weakening of inter-cluster bonds by Ga substitution for Al. Compared with sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC,²⁰⁾ the power factor, $S^2\sigma$, was dramatically enhanced by Ga substitution owing to the increase in σ and S , as seen in Fig. 8.

The thermal conductivities, κ_{total} , of sintered $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ and $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ ²⁰⁾ QCs are presented in Fig. 9(a). Unfortunately, κ_{total} does not change through Ga substitution for Al up to 573 K, but slightly decreases above 673 K. After subtracting κ_{el} from κ_{total} using the well-known Wiedemann-Frantz law ($\kappa_{\text{el}} = L_0\sigma T$), κ_{ph} is plotted against temperature (Fig. 9(b)). Ga substitution of Al is considered to bring a suppression of κ_{ph} for $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QCs similar to Al–Ga–Pd–Mn QCs^{16,17)} that is almost cancelled by increases in the precipitation of metallic secondary phases. Here, we assumed that the Lorenz number, L_0 , is $2.45 \cdot 10^{-8} \text{ V}^2 \text{K}^{-2}$.

Fig. 5 X-ray diffraction patterns of $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QCs (a) before and (b) after SPS. Arrows show the secondary phases of $\text{Al}_{11}\text{Re}_4$ and Al_3Pd_2 .

Fig. 6 Electrical conductivity, σ , as a function of temperature for $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QCs (a) before and (b) after SPS, and for the sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC.²⁰⁾

Fig. 7 Seebeck coefficient, S , as a function of temperature of (a) before spark plasma sintering (SPS) and (b) after SPS for $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QC and for sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC.²⁰⁾

Finally, we present the estimated dimensionless figures of merit, ZT , as a function of temperature for sintered $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ and $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ ²⁰⁾ QCs (Fig. 10). The ZT value increased and the peak ZT shifts to a higher temperature. The

Fig. 8 Power factor, $S^2\sigma$, as a function of temperature (a) before and (b) after SPS for $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QC as well as for sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC.²⁰⁾

Fig. 10 Dimensionless figure of merit, ZT , as a function of temperature for sintered $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QC, and sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC.²⁰⁾

Fig. 9 (a) Total thermal conductivity, κ_{total} , and (b) phonon thermal conductivity, κ_{ph} , as a function of temperature for sintered $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QC and for sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QC.²⁰⁾

maximum ZT value, ZT_{max} , of $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ QC is 0.18 at 673 K, which is significantly higher than that of sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$ QCs ($ZT_{\text{max}} = 0.12$ at 473 K).^{12,20)} We con-

firmed that Ga substitution of Al is a suitable means to enhance ZT in Al-based QCs.

6. Conclusions

Various compositions of Al–Ga–Pd–Re icosahedral QCs were synthesized using arc-melting and annealing methods, and their thermoelectric properties were investigated. The Seebeck coefficients are well characterized by the e/a factor, showing the same trend as Al–Pd–Mn and Al–Pd–Re QCs. We found that the sample with nominal composition of $\text{Al}_{66}\text{Ga}_4\text{Pd}_{21}\text{Re}_9$ exhibited the highest Seebeck coefficient of $95 \mu\text{V K}^{-1}$. We also confirmed that the sample's microstructure has a critical influence on the electrical conductivity, but has little effect on the Seebeck coefficient. A dense sample without cracks or pores showed a 1.5-times higher ZT of 0.18 compared with that for sintered $\text{Al}_{71}\text{Pd}_{20}\text{Re}_9$. The ZT enhancement through Ga substitution of Al is enabled by an increase in both the electrical conductivity and Seebeck coefficient and a decrease in the phonon thermal conductivity, which are attributed to precipitation of metallic secondary phases and the WBRHCs, a scheme applicable to all cluster-based solids including quasicrystals.

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