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Thermal and electrical transport properties of Ag-In-Yb quasicrystals: An experimental study

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The thermal and electrical transport properties, including electrical resistivity (ρ), thermopower (S), and thermal conductivity (κ) of $Ag_{42}In_{42}Yb_{16}$, $Ag_{42.25}In_{42.25}Yb_{15.5}$, and $Ag_{42.5}In_{42.5}Yb_{15}$ icosahedral quasicrystals (QCs) are reported. Great similarities were found in measured transport properties between these ternary and their binary analogies Cd-Yb QC's. This finding indicates that the complete replacement of Cd by equal amount of Ag-In does not alter the electronic band structure significantly. Comparison between the previously reported binary Cd-Yb QC, Cd_6Yb cubic approximant, and present ternary Ag-In-Yb QC suggests that the peculiar transport properties observed in the quasicrystalline Cd-Yb alloys may not be due to chemical disorder or quasiperiodicity. These findings provide valuable information for better understanding of the transport properties in quasicrystalline alloys.

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I. INTRODUCTION

The stable Cd-based family of binary quasicrystals (Cd-Yb and Cd-Ca) has attracted considerable attention due to their unusual physical and structural properties.[1–4] Cd-based alloys are the only known thermodynamically stable quasicrystals with only two metallic elements, which have the advantage of requiring less parameter for theoretical investigations than the ternary ones. Further, from the experimental point of view, these binary systems are more appropriate for studying their physical and structural properties associated with the quasicrystalline structure due to their high thermodynamic stability, less parasitic phases, and less chemical disorder. Such advantages of these binary systems lead to the possibility of observing real intrinsic effect on their physical properties, rather than the influence of extrinsic effect in the earlier known ternary systems. Depending on the composition of the constituent elements, these binary systems are usually found to possess an approximant crystalline (AC) phase and in some cases a slight deviation from the particular composition results in a quasicrystalline (QC) phase to AC or vice versa. For example, Cd_6Yb and Cd_6Ca crystallize in bcc cubic phase, whereas $Cd_{5.7}Yb$ and $Cd_{5.7}Ca$ form in *i*-phase QC's.[1] Some of the unique features associated with this class of Cd-Yb binary QC are, for example, less chemical disorder, lowest Debye temperature, large electronic specific heat coefficient, higher thermal conductivity, low thermopower, and relatively high electrical conductivity, compared to those of the ternary Al-based and other *i*-phase quasicrystals.[3,4] In order to understand the observed unusual physical properties, the crystalline approximants were also subjected to intense investigations due to the fact that they possess similar atomic compositions and local structures as the quasicrystalline phase.[5,6]

Quasicrystals are generally found to exhibit moderate electrical resistivity (between conductors and semiconductors), reasonably high thermopower, and low thermal conductivity. In contrast, the Cd-based binary QC's have orders of magnitude smaller electrical resistivity, low thermopower, and high thermal conductivity in comparison with Al based ternary system. This clearly indicates that the underlying mechanism behind the electronic transport in binary systems may be different from that of the ternary systems.[7,8] One notable difference between the binary and ternary alloys is the origin of the pseudogap. It has been suggested that the formation of a pseudogap in ternary system is mainly due to the Hume-Rothery mechanism (quasi-Brillouin-zone-Fermi-sphere interaction) and *sp*-*d* hybridization. Recent electronic structure calculations have revealed that in binary Cd (Yb/Ca) system, hybridization between Cd 5p and unoccupied Yb/Ca d states leads to a dip/pseudogap with Fermi level pinned to the shoulder of the occupied states in DOS.[9–14] From the absence of pseudogap in the hypothetical Cd_6Mg alloy (Mg has no low lying unoccupied d states), it has been concluded that the hybridization of d states near the Fermi level is essential for the pseudogap formation in Cd-based compounds.[9–11] However, experimental investigations pointed out that the pseudogap originates most likely due to the Hume-Rothery mechanism, whereas the *sp*-*d* hybridization enhances the formation of pseudogap.[5,15]

In the series of Cd-R (R is a rare-earth element), where R=Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu, the QC phase could be realized only with Yb, whereas in the remaining substitutions only AC's could be obtained. A further search for simi-

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lar classes of Cd-Yb binary quasicrystals by varying the constituent elements and composition has led to the discovery of single phase and thermodynamically stable icosahedral quasicrystal $Ag_{42}In_{42}Yb_{16}$ by substituting Ag and In in equal proportion in place of Cd in the $Cd_{84}Yb_{16}$ alloy.[16] Similar to that of the Cd-Yb system, *i*-phase Ag-In-Yb QC's can only be realized in a very narrow range of composition, with Ag and In having an atomic number one less and one more than Cd, respectively.[17,18] The Ag-In-Yb *i*-phase QC is found to be isostructural with Cd-Yb, with Ag-In occupying the Cd atomic sites. In this study we have investigated thermal and electrical transport properties including electrical resistivity (ρ), thermopower (S), and thermal conductivity (κ) on $(AgIn)_{100-x}Yb_x$ QC's of three different compositions ($x=42$, 42.25, and 42.5), and compared these results with previously reported Cd-Yb *i*-phase QC's. From our present measurements, we have noticed substantial similarities in transport behaviors between these ternary Ag-In-Yb, binary Cd-Yb QC's, and crystalline Cd_6Yb approximant. Such a result indicates that the peculiar transport properties observed in binary Cd-Yb QC's may not be due to chemical disorder or quasiperiodicity.

II. EXPERIMENT

Alloys of $(AgIn)_{100-x}Yb_x$ ($x=42$, 42.25, and 42.5) were prepared by arc melting a mixture of high purity Ag (99.99%), In (99.99%), and Yb (99.99%) under argon atmosphere, followed by the annealing at 400–500 °C for 50–100 h. X-ray-diffraction spectra were recorded using a rotating-anode x-ray generator (Cu $K\alpha$, 50 KV, 120 mA) with a graphite (002) monochromator. Typical x-ray diffraction patterns for these samples are shown in Fig. 1. The diffraction peaks can be indexed by Elser's method, and the results revealed that the prepared alloys were of *i*-phase QC's. Electrical resistivity measurements were carried out by a standard four probe technique. The measurements of thermopower were performed with a dc pulse technique. Seebeck voltage was detected using a pair of thin Cu wires electrically connected to the sample with silver paint at the same positions as the junctions of differential thermocouple. The stray thermal e.m.f.'s are eliminated by applying long current pulses (~100 s) to a chip resistor that serves a heater, where the pulses appear in an off-on-off sequence. The thermal conductivity was carried out in a closed-cycle refrigerator, using a direct heat-pulse technique. Samples were cut to a rectangular parallelepiped shape with typical sizes of $1.5 \times 1.5 \times 5.0\ mm^3$. One end was thermally glued to a copper block (heat sink) and a calibrated chip resistor (100Ω), which served as a heat source, was glued to the other end. The temperature difference was measured by an *E*-type differential thermocouple with junctions thermally attached to two well-separated positions along the sample. The temperature difference was controlled to be less than 1 K to minimize the heat radiation. During measurements the sample space is maintained in a good vacuum (better than 10^{-4} Torr). All experiments were performed on warming at a rate slower than 20 K/h. The reproducibility of S and κ measurements is better than 2%, while the absolute accuracy of κ is approximately 20%, mainly due to the error in the determination of sample dimensions.

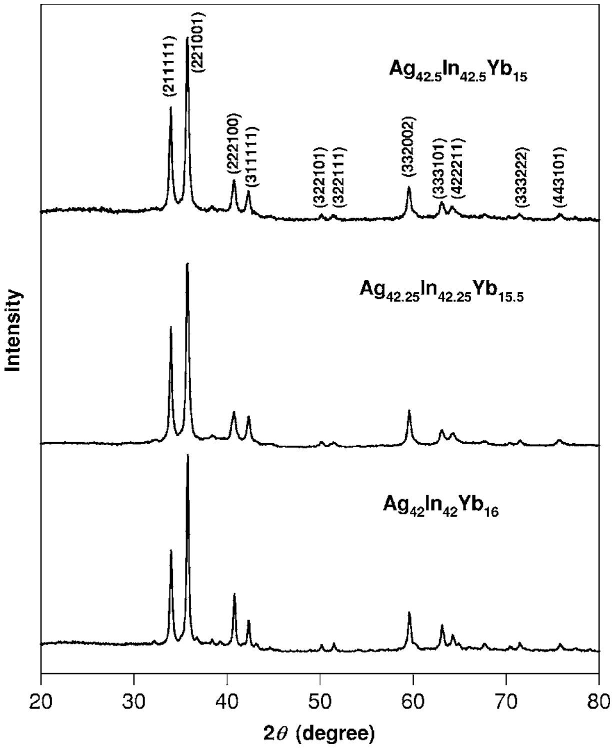

FIG. 1. The x-ray diffraction patterns recorded with Cu $K\alpha$ radiation for $Ag_{42}In_{42}Yb_{16}$, $Ag_{42.25}In_{42.25}Yb_{15.5}$, and $Ag_{42.5}In_{42.5}Yb_{15}$ quasicrystals.

III. RESULTS AND DISCUSSIONS

A. Resistivity

The temperature-dependent electrical resistivity $\rho(T)$ for $Ag_{42}In_{42}Yb_{16}$, $Ag_{42.25}In_{42.25}Yb_{15.5}$, $Ag_{42.5}In_{42.5}Yb_{15}$, and $Cd_{84}Yb_{16}$ QC's are shown in Fig. 2. For $Cd_{84}Yb_{16}$ alloy, the data has been taken from Ref. 4. The $\rho(T)$ characteristic of all three Ag-In-Yb alloys is found to be nearly identical, except the variations in the magnitude. The room-temperature resistivity $\rho(RT)$ for different alloys shows nearly equal magnitude, between 200 to 250 $\mu\Omega$ cm. It is seen that the electrical resistivity decreases with decreasing Yb concentration, presumably due to an increase in the carrier concentration. All the studied Ag-In-Yb samples show a rather weak $\rho(T)$ with a negative temperature coefficient, while $Cd_{84}Yb_{16}$ exhibits a small shallow maximum at around 50 K. This low-temperature resistivity maximum was commonly observed in Cd-Yb quasicrystals,[19] which was attributed to weak localization and spin-orbit interaction.[3] However, in the Ag-In-Yb alloys no such shallow maximum was noticed.

It is worthwhile mentioning that the magnitude of electrical resistivity of the previously known ternary *i*-phase QC alloys could be an order of magnitude larger than their AC phase.[20–22] Such a high value of electrical resistivity in the quasicrystalline phase has been attributed to the quasi-

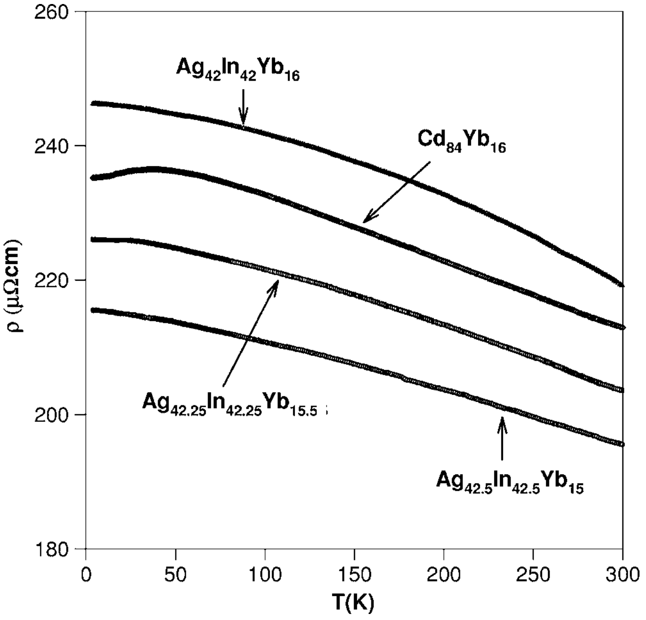

FIG. 2. Electrical resistivity as a function of temperature for $Ag_{42}In_{42}Yb_{16}$, $Ag_{42.25}In_{42.25}Yb_{15.5}$, $Ag_{42.5}In_{42.5}Yb_{15}$, and $Cd_{84}Yb_{16}$ quasicrystals.

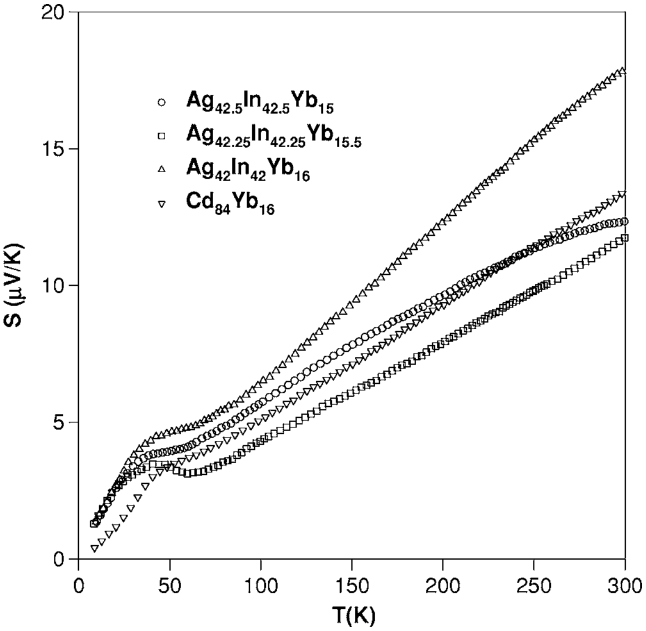

FIG. 3. Thermopower as a function of temperature for ternary Ag-In-Yb and binary $Cd_{84}Yb_{16}$ quasicrystals. A great resemblance is observed between $Ag_{42}In_{42}Yb_{16}$ and $Cd_{84}Yb_{16}$.

periodicity and chemical disorder.[23] In contrast, in the present study, the resistivity values between Cd-Yb quasicrystals and Cd_6Yb approximant are actually comparable. Further, the crystalline Cd_6Yb approximant and the present Ag-In-Yb *i*-phase QC alloys differ only by the temperature coefficient, where the former is positive and the latter is negative.[3,19,24] Therefore, the negative temperature coefficient in resistivity commonly observed in QC alloys is most likely due to the quasiperiodicity rather than the chemical disorder.

B. Thermopower

The temperature-dependent thermopower $S(T)$ of Ag-In-Yb *i*-phase QC alloys is shown in Fig. 3. The $Cd_{84}Yb_{16}$ sample data is also included for comparison. The thermopower is positive for all samples in the temperature range we investigated; seemingly, the hole-type carriers dominate the thermoelectric transport in these alloys. However, in a strict sense, in addition to the majority carriers, the sign of S depends on the position of E_F in DOS, which will be discussed in later section. As seen in Fig. 3, the $S(T)$ characteristics for all studied samples are nearly identical, except for a small variation in the magnitude. Especially, a striking resemblance exists between the ternary *i*-phase QC alloy $Ag_{42}In_{42}Yb_{16}$ and binary $Cd_{84}Yb_{16}$. From 50 to 300 K, S increases monotonically as the temperature rises, revealing the diffusion thermoelectric transport.

Around 40 K a broad maximum or plateau develops for each of the samples. Nonlinearities in thermopower in this temperature range are often associated with electron-phonon effects, which typically reach their maximum value at a temperature that is some fraction of the effective Debye temperature T_D^*. The phonon drag thermopower[25] S_g at low temperatures varies as the specific heat (i.e., as T^3) in the simplest model, but the behavior is often complex depending on the Fermi surface details. Phonon drag has been suggested[3,26] as the explanation for the thermopower nonlinearities in quasicrystal samples below 50 K. In our data, however, the thermopower extrapolates approximately linearly to zero at temperature decreases to zero, a feature more usually associated with metallic diffusion thermopower rather than phonon drag. We therefore compare our data to the behavior expected for electron-phonon enhancement of diffusion thermopower.

The metallic diffusion thermopower S_d is enhanced at low temperatures (similar to the specific heat) by the electron-phonon coupling,[27,28] and the resulting temperature dependence of S_d was calculated as[29]

$$S_d = [1 + a\bar{\lambda}(T/T_D^*)]S_u, \tag{1}$$

where S_u is the unenhanced diffusion thermopower, usually taken as linear in temperature at low temperature), a is the thermopower enhancement in the low-temperature limit, and $\bar{\lambda}(T/T_D^*)$ is the temperature-dependent electron-phonon enhancement of thermopower normalized by its value at $T=0$, i.e., $\bar{\lambda}(0)=1$ (details were given earlier[30]). The decrease of this enhancement at higher temperatures gives rise to a characteristic "knee" or change of slope in S_d centered around a temperature of approximately 0.25 T_D^*. The total thermopower is[25]

$$S = S_d + S_g. \tag{2}$$

In quasicrystals the lack of long-range order gives rise to strong disorder scattering of the charge carriers and of phonons, which strongly reduces the phonon-drag ther-

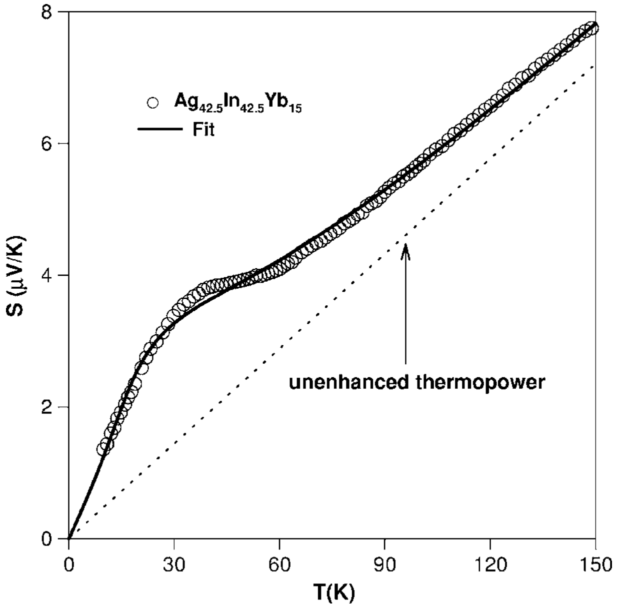

FIG. 4. An example of the fit to expression (1) for the enhancement of the linear metallic diffusion thermopower at low temperatures by the electron-phonon interaction, with parameter values as discussed in the text. The dashed line is the unenhanced thermopower.

mopower as electron-phonon scattering is only a small fraction of the total scattering for both electrons and phonons. It is in such cases that the electron-phonon enhancement is clearly seen.[28] Disordered metallic alloys (glassy metals) with resistivities greater than 100 $\mu\Omega$ cm typically show the electron-phonon enhancement of thermopower with phonon drag strongly suppressed. The high measured resistivities of more than 200 $\mu\Omega$ cm for Ag-In-Yb and Cd-Yb quasicrystals (Fig. 2) suggest that phonon drag is likely to be suppressed in these materials.

We have fitted our low-temperature thermopower data to the enhancement expression (1). An example is shown in Fig. 4. The electron-phonon enhanced thermopower is predicted by Eq. (1) to extrapolate approximately linearly to zero at $T=0$, in agreement with the experimental data. The energy of the phonons with which the electrons interact most strongly is fitted as $k_BT_D^* \sim 8$ meV ($T_D^* \sim 100$ K); the shape of the thermopower is not very sensitive to the form of phonon spectrum assumed.[28] This value of T_D^* is of the right order of magnitude since the Debye temperature T_D for the Ag-In-Yb samples would be expected to be similar to the value $T_D = 140$ K measured for icosahedral Cd-Yb with the same crystal structure. The size of the enhancement coefficients (e.g., $a \sim 1.4$ for the fit shown) is rather large. An even larger electron-phonon coupling was inferred[31] from the thermopower in the quasicrystalline plane for $Al_{73}Ni_{17}Co_{10}$ quasicrystals and ascribed to an increase in anharmonic components of the phonon system in the absence of translational invariance. (This study also used the calculation of the temperature dependence of electron-phonon enhancement.[30]) An additional enhancement of low-temperature thermopower could also arise from additional renormalization effects besides energy renormalization that have a similar temperature dependence.[30]

We note that while the electron-phonon enhancement fit does give the overall behavior, the reduction in thermopower above the knee is sharper than expected theoretically. It seems possible, therefore, that phonon drag may still play a role in causing the thermopower peak or plateau. In that case, the less pronounced plateau near 40 K in $Cd_{84}Yb_{16}$ and $Ag_{42}In_{42}Yb_{16}$ (compared to the other Ag-In-Yb samples) may be associated with the greater resistivity in these two samples, indicating stronger disorder scattering and so greater reduction in phonon drag. A similar nonlinear behavior of $S(T)$ around a temperature of 40 K was also noticed in our earlier measurements in $AlPdRe_{1-x}Mn_x$ quasicrystals.[32]

It is well known that the thermopower of quasicrystals changes with respect to a small variation in chemical composition and different heat treatments applied to the samples, as it is very sensitive to changes in the electronic structure at the Fermi surface. Particularly, the deepening of pseudogap and the location of Fermi level within the pseudogap as a function of transition metal concentration have been accounted for the sign reversal and significant variations in magnitude of thermopower for Al-based ternary QC alloys.[33–36] In Al-based ternary alloys, the Fermi level is located at the minimum of the pseudogap and shifts according to the concentration of valence electrons.[21,37] Nevertheless, the situation in Cd-based binary and Ag-In-based ternary QC's is different, as the temperature-dependent thermopowers for these alloys are quite similar. In the following, we attempt to understand the overall thermoelectric transport of the studied samples with the pseudogap scenario. For a metallic alloy, the linear variation of thermopower is often discussed using the well-known Mott formula (unenhanced electronic diffusion contribution)

$$S_e = \frac{\pi^2 k_B{}^2}{3e} T \left(\frac{1}{\sigma(E)} \frac{\partial\sigma(E)}{\partial E} \right)_{E=E_F}$$

which is valid for a one-band model with an energy-independent relaxation time, where e is the elementary charge with negative sign and $\sigma(E)$ is the energy-dependent electrical conductivity. S can be positive or negative, depending on the sign of $\partial\sigma(E)/\partial E$, which in turn depends on the position of E_F across the pseudogap.[34] Electrical conductivity in the metallic phase is known to be directly proportional to the electronic density of states (DOS) at E_F and the thermopower is then proportional to $\partial N(E)/\partial E$ at E_F. In the Al-based transition metal containing ternary quasicrystals, the flexible valence electron ratio (1.75–1.65) leads to a significant deepening of the pseudogap and variations in DOS at Fermi level.[35,38] As a result, significant composition-dependent variations in S and ρ have been observed.[33,36] On the other hand, our present thermopower measurements on Ag-In-Yb QC's strongly indicate that the composition change in the Cd-Yb alloys has a minor effect on their electronic structure. Indeed, electronic band structure calculations have suggested the insensitive nature of DOS towards the precise position of Cd atoms within the dodecahedral cavity.[6] This is further evident from the reported x-ray-diffraction studies, which indicate that the effect of the replacement of Cd by Mg (more than 10%) on the band struc-

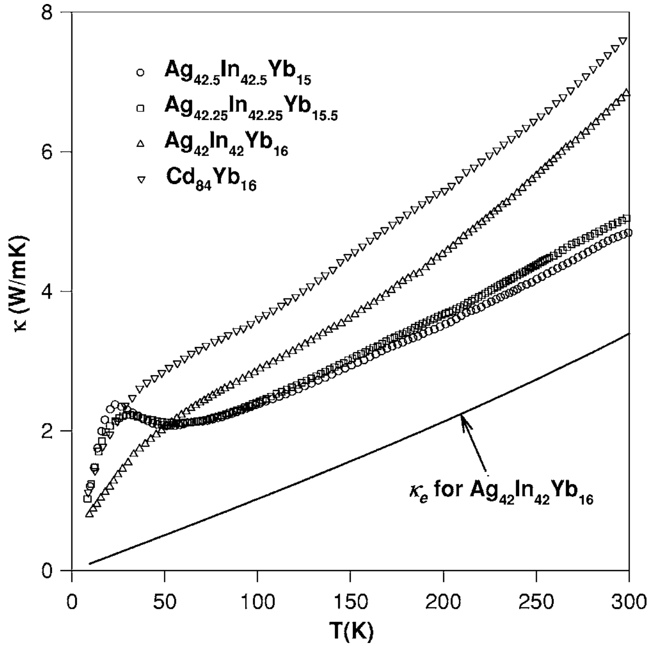

FIG. 5. Thermal conductivity as a function of temperature for Ag-In-Yb and $Cd_{84}Yb_{16}$ quasicrystals. The solid lines represent the electronic contribution to the total thermal conductivity calculated by Wiedemann-Franz's law for $Ag_{42}In_{42}Yb_{16}$ quasicrystal.

ture at the Fermi level is very small.[39] Therefore, slight variations in the precise position of Ag-In are unable to deepen the pseudogap or to shift E_F to influence the DOS significantly to induce corresponding variations in the electronic transport properties. It is worth mentioning that the average atomic radius of Ag-In is the same as that of Cd (0.151 nm).[40] A recent structural study has proven that $Ag_{42}In_{42}Yb_{16}$ is isostructural with Cd_6R with Ag-In occupying the Cd atomic sites.[17] In addition, when Cd is completely replaced by Ag-In, the valence electron ratio also remains the same (2) and hence the position of E_F is not altered. Therefore, both $Ag_{42}In_{42}Yb_{16}$ and $Cd_{84}Yb_{16}$ should show similar $S(T)$ behavior, as we observe experimentally. Another notable point is that temperature-dependent thermopowers for *i*-CdYb and its approximant are quite similar even thought they have very different long-range orders. This clearly indicates that $S(T)$ may be less sensitive to quasiperiodicity and chemical disorder in this class of quasicrystal.

C. Thermal conductivity

The temperature-dependent thermal conductivity $\kappa(T)$ of the $Ag_{42}In_{42}Yb_{16}$, $Ag_{42.25}In_{42.25}Yb_{15.5}$, $Ag_{42.5}In_{42.5}Yb_{15}$, and $Cd_{84}Yb_{16}$ alloys is shown Fig. 5. The room temperature κ for Ag-In-Yb QC's was also found to be comparable, between 5 to 7 W/m K. The notable difference is the low temperature maximum. Below 50 K, both $Ag_{42.25}In_{42.25}Yb_{15.5}$ and $Ag_{42.5}In_{42.5}Yb_{15}$ develop a well defined maximum, whereas such maximum is absent in $Ag_{42}In_{42}Yb_{16}$ and $Cd_{84}Yb_{16}$. The low-temperature maximum found in the *i*-phase QC alloys has been attributed to the generalized Umklapp process.[41] In general, the impurity and defect scattering could suppress the low-temperature maximum in κ.[8,26] In all our Cd-Yb samples, Cd impurity was clearly observed (see Ref. 4). In $Ag_{42}In_{42}Yb_{16}$ samples, there are a few small peaks in the x-ray-diffraction pattern which can not be indexed by Else's scheme, although they are not as apparent as in Cd-Yb samples. On the other hand, no apparent defects or impurity contents could be detected by x-ray and mass spectrometer analyses on $Ag_{42.25}In_{42.25}Yb_{15.5}$ and $Ag_{42.5}In_{42.5}Yb_{15}$ samples. In addition, the distinct maximum in κ was also observed in our previously studied $Cd_{85}Yb_{15}$ and Cd_6Yb samples, but no maximum in κ was seen by Muro *et al.* in their $Cd_{84.6}Yb_{15.4}$ and Cd_6Yb samples.[8] This suggests that extrinsic effects such as impurity phase may have strong influences on the measured thermal conductivity at low temperatures in these alloys. Another intriguing feature of these alloys is that κ increases monotonically with temperature above 100 K. The increase of thermal conductivity at high temperatures is commonly observed for quasicrystals, attributed to the activation of localized phonon states. Janot proposed a model[42] taking into account the high-energy and localized vibration modes with a hierarchical variable-range hopping mechanism and suggested that the lattice thermal conductivity is expected to follow a power-law-dependent, i.e., $\kappa_L \propto T^{\gamma}$ with $\gamma=1.5$. To examine the validity of this model to these Ag-In-Yb quasicrystals, the data above 150 K were fitted by the expression $\kappa_L=\alpha+\beta T^{\gamma}$. These fitting processes yield $\gamma=1.43\pm0.17$, which is in good agreement with the theoretical prediction.[42]

It has been pointed out that the thermal conductivity of quasicrystals should still follow Wiedemann-Franz's law over a wide temperature range,[43] therefore we have estimated the electronic thermal conductivity κ_e of these Ag-In-Yb alloys by using $\kappa_e\rho/T=L_0$. Here ρ is the dc electrical resistivity and the Lorenz number $L_0=2.45 \times 10^{-8}\ \mathrm{W}\,\Omega\,\mathrm{K}^{-2}$. A representative κ_e for $Ag_{42}In_{42}Yb_{16}$ is illustrated in Fig. 5 as the solid line. From this estimation, a substantial electronic thermal conductivity κ_e $(\sim 40\%)$ to their total thermal conductivity is found. In contrast to other *i*-phase ternary alloys, the electronic contribution in these Ag-In-Yb alloys is significantly higher due to the low magnitude of electrical resistivity.[3] However, such a finding is consistent with the binary Cd-Yb quasicrystals. The average atomic weight and valence of Ag-In is nearly equivalent to Cd and hence it is reasonable that there is no significant variation in $\kappa(T)$ among different compositions. On the other hand, the low value of Debye temperature and high value of heat capacity found in the Cd based *i*-phase alloys has been attributed to the soft lattice.[3] Therefore, thermal conductivity may shed further light on this aspect in the comparative study between the Ag-In–and Cd-based alloys.

IV. CONCLUSIONS

In conclusion, we have presented the measurements of electrical resistivity, thermopower, and thermal conductivity of binary $Cd_{84}Yb_{16}$ and ternary $Ag_{42}In_{42}Yb_{16}$, $Ag_{42.25}In_{42.25}Yb_{15.5}$, $Ag_{42.5}In_{42.5}Yb_{15}$ alloys by replacing Cd by Ag-In. It is found that binary $Cd_{84}Yb_{16}$ and ternary $Ag_{42}In_{42}Yb_{16}$ alloys show nearly identical transport behaviors. Significant similarity in the transport properties could be noticed between different compositions of Ag-In-Yb quasicrystals, except the low-temperature variations in thermopower and thermal conductivity. These findings suggest that Ag-In can be treated as isoelectronic to Cd. Further, similar transport properties between the *i*-phase binary Cd-Yb and ternary Ag-In-Yb alloys clearly indicate that the peculiar transport properties such as a negative temperature coefficient in electrical resistivity may not be due to chemical disorder. In addition, the resemblance in temperature-dependent thermopower between the ternary, binary, and approximant phases (Cd_6Yb) suggests that the electronic structure in this class of quasicrystals is insensitive to composition and quasiperiodicity.

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