Metal-insulator transition and localization in quasicrystalline $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ alloys

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Metal-insulator transition was observed in icosahedral quasicrystals $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ as x changed from 5 to 0. It was found that conductivity data of the insulating samples ($x \leqslant 3.5$) could be analyzed in terms of variable range hopping, i.e., $\sigma=\sigma_0 \exp[-(T_0/T)^p]$, with $p=\frac{1}{2}$ for $x=2$, 2.5, 3, and 3.5 and $p=\frac{1}{4}$ for $x=0$, over the temperature range from 0.45 to 10 K. The localization length was found to be of the order of intercluster distance in the quasicrystalline structure. At decreasing x, magnetoresistance (MR) was found to change smoothly from weak-localization behavior in the metallic regime to anomalous features in the insulating regime which are distinctly different from those seen in other well-studied insulators. The persistence of positive MR in insulating $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ alloys suggests that the backscattering effect plays an important role for localized states in quasicrystalline materials. Specific heat measurement on the insulating $x=2$ sample yielded $\gamma=0.38$ mJ/g atom K^2, and carrier density was found to be $\sim 10^{20}$ cm^{-3} at 4.2 K from Hall effect measurement, which are at least as large as those of metallic Al-Cu-Fe quasicrystals. The finding of spatially localized states in quasiperiodic alloys cannot be explained satisfactorily by existing theories. [S0163-1829(96)08542-6]

I. INTRODUCTION

Thermodynamically stable icosahedral quasicrystals such as Al-Cu-(Fe,Ru,Os) and Al-Pd-(Mn,Re) (Ref. 1) with high structural quality are suitable for investigation of the intrinsic physical properties of quasicrystals. Several reports have shown their interesting electronic properties such as very low conductivity for metallic alloys, positive temperature dependence of the conductivity, and highly sensitive, alloy-compositional dependence of the conductivity.[2] The lowest conductivity at 4.2 K reported for each alloy is ~ 100 $\Omega^{-1}\,cm^{-1}$ for Al-Cu-Fe,[3] ~ 30 $\Omega^{-1}\,cm^{-1}$ for Al-Cu-Ru,[4] ~ 110 $\Omega^{-1}\,cm^{-1}$ for Al-Pd-Mn,[5] and ~ 0.5 $\Omega^{-1}\,cm^{-1}$ for Al-Pd-Re.[6–8] Al-Cu-(Fe,Ru) and Al-Pd-Mn alloys are marginally metallic or in proximity to a metal-insulator transition (MIT) with their low-temperature conductivities near Mott's minimum metallic conductivity. Their magnetoresistance are adequately described by quantum corrections in the weakly localized regime with inelastic scattering times comparable to those of metallic glasses,[3,4,9] thus they are on the metallic side of the MIT. The conductivity of *i*-phase Al-Pd-Re is comparable to that of doped semi-conductors inside the insulating region of the MIT with $\sigma(T\rightarrow 0)\rightarrow 0$, which indicates that it is an insulator. Meanwhile, it has much higher carrier concentration and density of states than those of the corresponding semiconductor systems, which makes the transport properties of the insulating Al-Pd-Re quasicrystal quite unique. Results from optical conductivity measurement of *i*-Al-Pd-Re and other quasicrystalline alloys give indications that quasiperiodicity favors carrier localization;[10] however, it is still uncertain whether the electron wave functions which lead to the insulating state in *i*-Al-Pd-Re are spatially localized or in critical states (i.e., decay in a power law). Further studies of this insulating quasicrystal system and the transition from insulating to marginal metallic behavior are necessary to help shed light on the intrinsic electronic properties of quasicrystals.

In contrast to the extensive studies of metal-insulator transition in crystalline and amorphous semiconductors, so far there has been no report on MIT in the quasicrystalline environment. This is because the structural quality of quasicrystalline samples is very sensitive to even a tiny amount of impurities. However, by utilizing the structural similarities between the *i*-phase Al-Pd-Re and Al-Pd-Mn alloys and by realizing the fact of the former being an insulator and the latter a metal, we are able to achieve metal-insulator transition by varying the composition of $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ alloys from $x=0$ to 5 while maintaining their well-ordered quasicrystalline structure. In this paper, we will report on the low temperature conductivity and magnetoresistivity of $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ alloys, as well as Hall effect and specific-heat measurement of insulating $Al_{70.5}Pd_{21}Re_{6.5}Mn_{2.0}$. Our data will be analyzed in light of theories applicable to insulators and quasicrystals. The implications of our results on the electronic states in quasicrystals will be discussed.

II. EXPERIMENTAL PROCEDURE

Polycrystalline alloy ingots of $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ ($0 \leqslant x \leqslant 5$) were made by melting appropriate quantities of high-purity elements in an arc furnace under argon atmosphere.[7] Bar-shaped samples ($1\times 1.5\times 5$ mm^3) were cut from ingots. First, they were annealed between 860 and 950 °C for 12 h, then between 600 and 650 °C for 4–12 h to obtain high-quality *i* phase with enhanced insulating behavior as described in detail previously.[11] The phase purity of the samples was confirmed by powder x-ray diffraction on a SCINTAG diffractometer with a resolution $\leqslant 0.13°$, using Cu $K\alpha$ radiation (Fig. 1). Resistivity was measured from 0.45 to 295 K using the standard four-probe technique with silver paint contacts. dc Hall effect measurement was performed using the six-lead method, in a field up to 4 T at selected temperatures between 4.2 and 300 K. Specific heat measure-

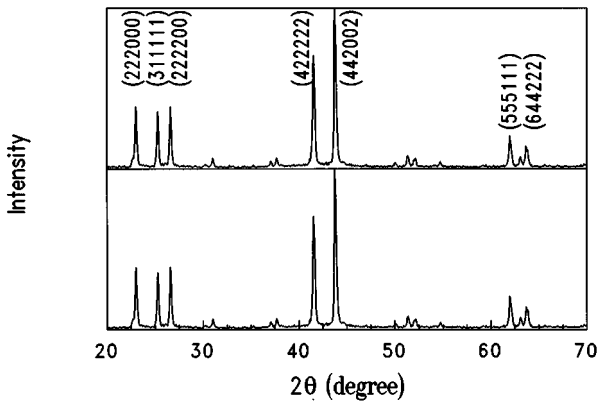

FIG. 1. Cu $K\alpha$ powder x-ray diffraction pattern of i-$Al_{70.5}Pd_{21}Re_{6.5}Mn_{2.0}$ (upper pattern) and i-$Al_{70.5}Pd_{21}Re_{4.5}Mn_{4.0}$ (lower pattern). Only major peaks are labeled for clarity; the 6D index together with more details were explained in Refs. 6 and 7.

ment was carried out on ~100 mg samples using the thermal relaxation method at temperatures from 1 to 8 K.[7]

III. RESULTS AND DISCUSSION

A. Low-temperature conductivity

Figure 2 presents temperature dependence of conductivity for $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ in the form of σ vs T. It is clear from the trend of the low temperature conductivity as well as from the values of σ (0.45 K) vs Mn concentration (Fig. 3) that metal-insulator transition occurred in the region $4 \geqslant x \geqslant 3$ in these alloys, as the concentration of Mn was reduced. MIT has been extensively studied in disordered systems such as doped semiconductors, where the conductivity of the material changes from that of a metal (finite σ as $T \to 0$) to that of an insulator ($\sigma \to 0$ as $T \to 0$) as a function of impurity concentration or other external parameters such as magnetic field, pressure, and strain.[12] It is well known that in an insulating semiconductor, as the density of one-electron impurities N increases towards N_c (the critical concentration), the spatial overlaps of localized electron states become larger,

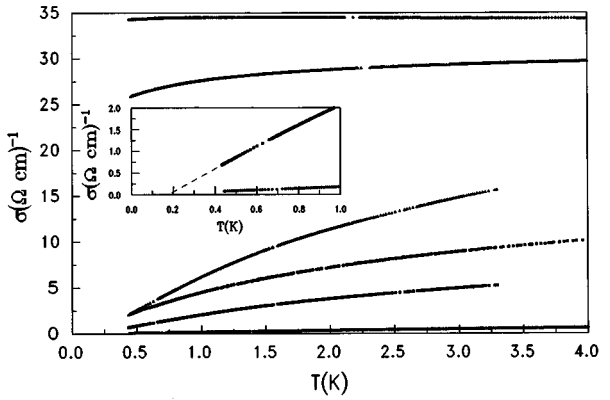

FIG. 2. Conductivity versus temperature of $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$, $x=0$, 2, 2.5, 3, 4, 5 from bottom to top. Inset shows $\sigma(T)$ of $x=0$ (lower one) and $x=2$ (upper one) samples at low temperature. Dotted line indicates extrapolation of $\sigma(T)$ to zero value at a finite temperature.

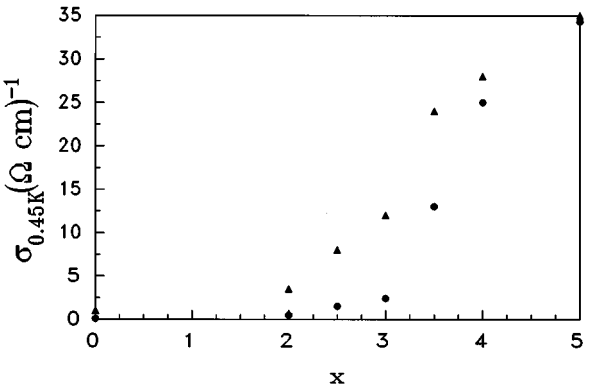

FIG. 3. Values of σ (0.45 K) versus Mn concentration for samples with (●) and without (△) low-T annealing.

and extended states eventually form, which turns the system into a metal. In $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ quasicrystals, the changing of Mn concenration may not be viewed as straightforward as the changing of the impurity concentration since the carrier density was found to be $\sim 10^{20}$ cm^{-3} for $x=0$–5 (Ref. 7 and to be discussed later), which is substantially higher than those in doped semiconductors. Thus the mechanism responsible for the transition may be more involved. To reveal the wave-function properties of electrons near MIT in this quasicrystalline environment, we further analyze our conductivity data of the alloys.

First, the $\sigma(T)$ of $Al_{70.5}Pd_{21}Re_{8.5}$ ($x=0$) was analyzed since its σ had the lowest value among the alloys, suggesting it being most insulating. Previously, we reported $\sigma(T) \sim T$ for the most insulating samples over the temperature range 0.45–3 K with nearly zero extrapolation of $\sigma(0) (\leqslant 0.01\ \Omega^{-1}\ \text{cm}^{-1})$.[7] During our study of insulating Al-Pd-Re quasicrystals, about two dozen samples were measured, and their low-T conductivity consistently showed an apparent linear T dependence and small residual $\sigma(0) (0.005–0.03\ \Omega^{-1}\ \text{cm}^{-1})$. As the Mn concentration of $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ increased, the $\sigma(T)$ showed a clearly different trend with higher temperature coefficient. Below 1 K, $\sigma(T)$ could be extrapolated to zero value at a finite temperature (inset of Fig. 2), which could be analyzed in terms of the variable-range hopping (VRH) model, i.e., $\ln \sigma \propto T^{-1/2}$ for $x=2$–3.5, to be shown later. The VRH behavior observed in less insulating systems ($x=2$–3.5) prompted us to reanalyze $\sigma(T)$ of the most insulating $Al_{70.5}Pd_{21}Re_{8.5}$ ($x=0$) system. Due to inherent difficulties in obtaining perfect samples in a complex ternary alloy system, the apparent $\sigma(0)$ in the $x=0$ samples could be attributed to tiny imperfections in the structural quality of samples. It is also possible that the residual $\sigma(0)$ is due to the existence of a small amount of structural defects in the realistic three-dimensional (3D) quasicrystal, since it is impossible to achieve perfect quasicrystalline structure in real alloys. To reveal the intrinsic behavior, we presume the small $\sigma(0)$ is a constant at the low temperature range, thus $\sigma' = \sigma - \sigma(0)$ vs temperature was analyzed. $\sigma'(T)$ was found to follow Mott's variable-range-hopping behavior for all samples over the temperature range 0.45–10 K, i.e., $\sigma' = \sigma_0 \exp[-(T_0/T)^{1/4}]$. The values of T_0 extracted from the plot [Fig. 4(a)] were found to be $\sim 10^3$ K. With the density of states estimated from specific heat measurements ($\gamma=0.11$

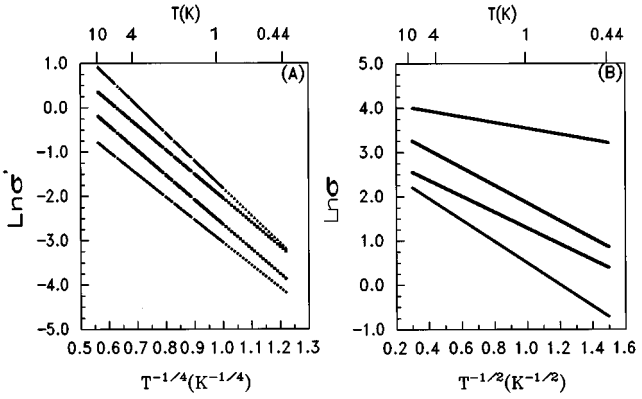

FIG. 4. (a) Mott's variable range hopping, $\ln \sigma' \propto T^{-1/4}$, where $\sigma' = \sigma - \sigma(0)$, observed on $Al_{70.5}Pd_{21}Re_{8.5}$ samples with varying $\sigma(0)$ (0.005–0.03 Ω^{-1} cm^{-1}) and T_0 (700–1500 K). (b) $\ln \sigma \propto T^{-1/2}$ behavior observed for $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ alloys with $x=3.5$, 3, 2.5, 2 (top to bottom), and T_0' varies from 1–4 K.

mJ/g atom K^2), we obtained the localization length $\xi \sim 25$–35 Å from $\xi^3 = 18/k_B TN(E_F)$, and $R_h/\xi \propto 2T^{-1/4}$, where R_h is the hopping length. It is reasonable to expect that in the temperature range where hopping is observed, R_h should be larger than ξ. R_h/ξ is in the range of 2.5–1.2 over the temperatures 0.45–10 K, which satisfies this condition. It should be noted that the localization length $\xi \sim 30$ Å and the hopping length $R_h \sim \xi$ are of the same scale as the size of the basic ''cluster'' and intercluster distance, respectively, in the quasiperiodic structure.[13]

A distinctive feature arose as Mn concentrations were increased ($x=2$, 2.5, 3, 3.5). The conductivity shows $\sigma = \sigma_0 \exp[-(T_0/T)^{1/2}]$ behavior between 0.45 and 10 K [Fig. 4(b)]. For variable-range hopping, $\ln \sigma \propto T^{-1/4}$ if the density of states near the Fermi energy is constant or a slowly varying function of energy.[14] By considering the long-range Coulomb interactions between localized states, Efros and Shlovskii[15] demonstrated that the density of states tended to zero near the Fermi level, giving rise to a parabolic Coulomb gap and a conductivity which depended on temperature as $\ln \sigma \propto T^{-1/2}$. The values of T_0' were found to be $\sim$1–4 K, which is small compared to the fitted temperature range. It should be noted, however, that comparable values were obtained from doped semiconductors such as n-type Ge:As (Ref. 16) and CdSe.[17] Attempts to subtract a small residual $\sigma(0)$ in the range of those used for Al-Pd-Re alloys were found to have no obvious influence on the trend shown in Fig. 4(b), since most of the alloys' conductivities had much higher value compared with those of Al-Pd-Re alloys. The transition from ''$\frac{1}{4}$'' ($x=0$) to ''$\frac{1}{2}$'' ($x=1$–4) behavior can be attributed to the emerging importance of electron-electron interactions as the Mn concentration increases, and the Coulomb gap becomes comparable to the hopping energy. A similar transition has been observed in doped semiconductors as well. For GaAs, the $T^{-1/4}$ law was observed for lightly doped samples which changed to the $T^{-1/2}$ law as the doping level increases.[18]

We should mention that the annealing effect still exists for Al-Pd-Re-Mn alloys. Low temperature annealing (600–650 °C) turned alloys into more insulating states,[11] except for the metallic samples, for which no annealing effect was found. The samples ($x=2$–3.5) without low temperature annealing still showed $\ln \sigma \propto T^{-1/2}$ law, though with a higher σ. We can view the samples without low-T annealing as in less insulating states, even closer to the MIT. One interesting feature was observed for $x=3.5$ alloy, whose $\sigma(T)$ showed metallic behavior without low-T annealing (only annealed at 940 °C), and annealing (additional annealing at 650 °C) turned it into an insulator with $\ln \sigma \propto T^{-1/2}$ [top curve in Fig. 4(b)]. In other words, we also observed thermal annealing tuned MIT.

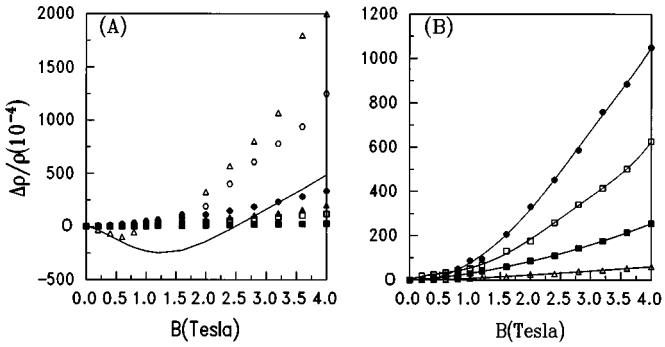

FIG. 5. (a) Magnetoresistance of $x=2$ sample at temperatures 1.5, 2.5, 4.2, 7.5, 10, and 20 K (top to bottom). Solid line is MR data for $x=0$ sample at $T=2$ K. (b) Magnetoresistivity of $x=4$ samples at temperatures 1.5, 2.5, 4.2, and 10 K (top to bottom). Solid lines are fitted to weak-localization theory.

B. Magnetoresistance

To further explore the electron transport process in the localized state, magnetoresistivities were measured for $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$. Magnetoresistance (MR) for the Al-Pd-Re sample had been shown to have distinctive features with prominent presence of both positive and negative contributions at low temperature and in low field. Figure 5(a) shows the MR of Al-Pd-Re at 2 K as a solid line; the detailed description and discussions were given elsewhere.[19] For $x=2$ sample [Fig. 5(a)], it can be noted that the negative contribution to MR was much reduced, though its effect at low temperature was still evident. The MR data became all positive at higher temperatures, and its magnitude and curvature started to resemble those of marginally conducting i-phase quasicrystals (i-QC's) which could be described by quantum corrections due to the weak localization. However, the parameters extracted from force fitting of MR data to weak localization (WL) theory showed spin-orbit scattering time $\tau_{so} \sim 10^{-12}$ sec and inelastic scattering time $\tau_{ie} \sim 10^{-14}$ sec at 4.2 K, which is orders of magnitude lower than those of i-Al-Cu-Fe (Refs. 3 and 9) and Al-Cu-Ru (Ref. 4) systems. This is unreasonably small for Al-Pd-Re-Mn ($x=2$) with comparable structural order as other iQC's. Besides, the $T=1.5$ K data could not be fitted to WL due to the residual negative MR and the robust curvature. Thus the $x=2$ sample still has reminiscent feature of the $x=0$ sample, which cannot be described by WL. As Mn content increased, the MR became positive over the whole temperature range [Fig. 5(b)], and fitting to WL yielded $\tau_{so} \sim 10^{-12}$ sec, $\tau_{ie} \sim 4 \times 10^{-11} T^{-1.4}$ sec for $x=3$; $\tau_{so} \sim 10^{-12}$ sec, $\tau_{ie} \sim 1.2 \times 10^{-10} T^{-1.7}$ sec for $x=4$, 5. Thus MR data over the

whole temperature range could be fitted to WL with τ_{so} and τ_{ie} comparable to other *i*-phase quasicrystals.[3,4,9] It should be mentioned that for the fitting, we need to know the diffusion constant D, which can be calculated from $D=\sigma/e^2N(E_F)$, with $N(E_F)$ estimated from specific heat measurement. Since γ=0.38 mJ/g atom K^2 for $x=2$ and $\gamma\sim$0.5 mJ/g atom K^2 for Al-Pd-Mn alloy,[20] it is reasonable to take $\gamma\sim$0.4 mJ/g atom K^2 for all the alloys ($x=2$–5).

Overall, the MR data of $Al_{70.5}Pd_{21}Re_{8.5-x}Mn_x$ showed a smooth transition from the peculiar feature of Al-Pd-Re ($x=0$) to quantum interference corrections due to weak localization as the increase of Mn concentration drove the system through a metal-insulator transition. For alloys on the insulating side of the MIT with conductivity showing VRH behavior, the dominant presence of positive MR was unexpected. Unlike in disordered metals, where the self-crossing diffusion paths (returning-loops) of electrons give rise to weak localization and the presence of spin-orbit scattering can lead to positive MR if the scattering is mitigated by a B field, the mechanism for MR in an insulating system is different. In VRH regime, MR is believed to be dominated by the quantum interference between different Feynman paths along which spatial overlap between pairs of sites may occur.[21] This is known as ''oriented-path interference,'' and it was found that the interference between all possible paths within a cigar-shaped domain of length R_h and width $(R_h\xi)^{1/2}$ might influence considerably the hopping probability between two sites. By averaging numerically the logarithm of the conductivity over many random impurities in the presence of a magnetic field, negative MR was found. It is expected that the returning-loop effect is negligible when the hopping length $R_h\gg\xi$, hence the accompanying spin-orbit contribution to MR is negligible as well. Based on these criteria, positive MR is not favored in insulators.

Our MR data for Al-Pd-Re ($x=0$) showed the presence of both positive and negative MR at low field. The negative MR was attributed to the ''oriented path interference'' and the positive one was ascribed to the anomalous diffusion of slowly decaying electron function.[19] We noticed that as Mn concentration increased, which drove the system toward MIT, the negative MR disappeared as the contribution from ''direct path interference'' was reduced. However, the observation of positive MR in the insulating regime needs an explanation. The consistency of the presence of positive MR in all the insulating samples suggests it is an intrinsic property of the insulating *i*-QC. We believe that the unique quasicrystalline structure plays an important role for the positive MR. Since unlike other insulating systems, where the randomization of scattering and atomic potential leads to carrier localization at random sites (Anderson localization), the structure of quasicrystal is self-similar.[13] Thus the localized electron may have a higher probability of finding a site with similar energy nearby than in other insulating environments. As a result, the hopping length R_h tends to be shorter. For Al-Pd-Re, we find $R_h/\xi\sim1.2\rightarrow2.5$ over the temperature range 0.45 to 10 K. The direct path interference and the neglect of spin-orbit scattering is justified when $R_h/\xi\gg1$. However, this is not so for *i*-QC with $R_h\sim\xi$. Thus when we consider MR for macroscopic samples by averaging the Feynman paths over scale $\sim R_h$, backscattering (returning loop) should also be taken into consideration. The backscattering together with spin-orbit scattering certainly can yield positive MR in the presence of a magnetic field. It should be remembered that the role of backscattering in systems such as InO with $R_h/\xi\geqslant10$, was still being considered to be important.[22] Meanwhile, it is a fact that almost all quasicrystals' MR can be well described by quantum interference effects in the WL regime even for the marginally conducting systems with $D\sim10^{-1}$–10^{-2} cm^2 s^{-1}. The universal structural similarity in quasicrystal seems to provide the basis for the popularity of returning-loop effects for the MR of quasicrystals.

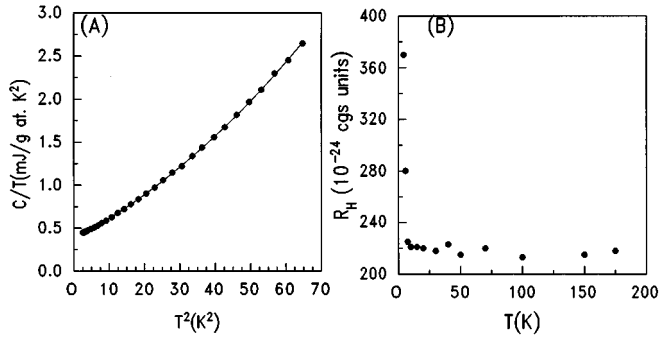

FIG. 6. (a) Specific-heat data of $x=2$ sample, plotted in the form of C/T versus T^2. The solid line is a fit to the expression $C=\gamma T+\beta T^3+\delta T^5$, where γ=0.38 mJ/g atom K^2, β=0.023 mJ/g atom K^4, and $\delta=2.1\times10^{-4}$ mJ/g atom K^6. (b) Hall coefficient versus temperature of the same sample.

C. Electronic states

Specific heat measurement provides information about the electronic density of states at the Fermi level. Figure 6(a) shows the specific heat of $Al_{70.5}Pd_{21}Re_{6.5}Mn_{2.0}$ alloy, plotted in the form of C/T vs T^2, which is fitted to the standard expression $C=\gamma T+\beta T^3+\delta T^5$. It yields γ=0.38 mJ/g atom K^2 and a Debye temperature Θ=440 K. The γ value is comparable to those of metallic and semimetallic *i*-Al-Cu-Fe (Ref. 5) and Al-Pd-Mn alloys.[20] The Hall coefficient R_H is shown in Fig. 6(b). It gives a Hall number $\sim10^{20}$ cm^{-3}, commonly seen in semimetallic quasicrystals.[2] Based on the electron density estimated from Hall number and γ value, which are orders of magnitude larger than those of doped semiconductors, Al-Pd-Re-Mn alloys may just be semimetals. However, the VRH behavior suggests that these *i* phases are quasiperiodic insulators with spatially localized states. Knowing the mean interelectron spacing to be $\sim$20–30 Å, the intrinsic (i.e., intracluster, to be compared with Bohr radii of dopants in a semiconductor) localization length must be of the order of $\sim$10 Å for the MIT to occur. Theoretical calculations support that quasicrystalline structure leads to reduction of conductivity,[23–26] some of the calculations imply the existence of insulating quasicrystals; however, the wave functions are generally believed to be critical, i.e., decay like a power law: $\psi\sim L^{-\alpha}$.[23,24] So far, theories predict the existence of localized states only for some specific energies in 3D system. It is plausible that the structural randomness in realistic 3D system may help turn the critical states into spatially localized ones as long as the randomness does not alter the electronic density of states. For 2D system, Yamamoto and Fujiwara[26] show that intro-

ducing randomness to the 2D Penrose tiling decreases the localization length and the conductance, thus favoring the tendency of localization. However, this conclusion may not be directly applicable to 3D case. Further theoretical and experimental studies on localization in 3D quasicrystals are necessary.

IV. SUMMARY

The metal-insulator transition has been observed for Al-Pd-Re-Mn alloys, conductivity of the insulating samples can be described by variable range hopping, the localization length is found to be of the order of the intercluster distance in the quasicrystalline structure. The positive MR observed in both the insulating and metallic alloys over the whole range of MIT together with the fact that the MR of other metallic quasicrystals can be well described by WL indicate that quasicrystal structure favors the returning-loop (backscattering) effect, and it is suggested that the returning-loop effect is favored in quasicrystal as a result of self similarity in the structure. Specific heat and Hall effect measurements suggest that the electronic densities of states in insulating and metallic *i* alloys are comparable in magnitude. The spatially localized states indicated by the VRH conductivity cannot be satisfactorily explained with current theories. Structural imperfection in quasicrystalline alloys is proposed to be the factor leading to the localized states.

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