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The Metal-Insulator Transition in Al–Pd–Re Quasicrystals

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The metal-insulator (M-I) transition in Al–Pd–Re quasicrystals (QCs) was studied by annealing method. We found that increasing the resistivity ratio r=ρ(4.2 K)/ρ(300 K) of Al–Pd–Re QCs can greatly reduce the value of the electron correlation gap Δ and therefore drives the system towards and through the M-I transition. In the insulating state, the conductivity $\sigma(T)$ between 0.5 $\sim$ 7 K can be analyzed with $\sigma(T) = \sigma_0 \exp[-(T_0/T)]^{1/4}$ and the negative magnetoconductance observed at low temperature is mainly attributed to the decrease of the localization length ξ with increasing field.

KEYWORDS: quasicrystals, metal-insulator transition

§1. Introduction

Recently we found[1)] that McMillan's scaling theory[2)] of the metal-insulator (M-T) transition in amorphous materials, taking into account of localization, correlation and screening, can be applied to quasicrystals (QCs) equally well, that is, the low-temperature conductivity on the metallic side of the M-I transition varies as

$$\sigma(T) = \sigma(0) \cdot \left(1 + \sqrt{\frac{T}{\Delta}}\right) = \sigma(0) + m\sqrt{T}, \qquad (1)$$

where the electron correlation gap Δ is proportional to the square of the zero-temperature conductivity $\sigma(0)$ and Δ goes to zero at the M-I transition.

We also found that among the studied QCs, series of Al–Cu–(Fe, Ru) and Al–Pd–Mn QCs with the Δ value in the range of 0.1 $\sim$ 10 eV will be always in the metallic state, while series of Al–Pd–(Re, Mn) QCs with much smaller values of Δ (10^{-3} $\sim$ 10^{-5} eV) are near the M-I transition and can make a transition from metals to insulators on choosing annealing conditions appropriately.

Our findings clearly indicate that like amorphous alloys,[3-6)] granular metal films,[7)] and doped semiconductors,[8)] both localization and electron-electron interactions play important roles in the low-T electron transport in QCs. For the above systems, localization of electrons is known to arise from disorder. On the contrary, perfect Al–Cu–(Fe, Ru) and Al–Pd–(Mn, Re) QCs are strictly ordered materials with very sharp X-ray diffraction peaks; and improving (not deteriorating) the perfection of the quasilattice order is found to lead to the increase of resistivity. Thus the origin of localization in QCs is not clear yet, but it is unimportant here. The important point for our studies here is that eq. (1) must be valid in QCs.

Equation (1) is derived from the relation of the density of states versus energy, given by

$$N(E) = N(0)\left(1 + \sqrt{\frac{E}{\Delta}}\right), \qquad (2)$$

where $N(0)$ is the density of states at the Fermi level. Tunneling experimental measurements on Al–Pd–Re and Al–Cu–Fe QCs have shown that eq. (2) also holds in these QCs.[9)] Thus the validity of eq. (1) in QCs is justified from both conductivity and tunneling measurements.

In this work, we will study the M-I transition in Al–Pd–Re QCs by annealing method. Annealing the sample at low temperature has been found to be able to increase the resistivity ratio r=ρ(4.2 K)/ρ(300 K) and to push the sample closer to the M-I transition.[1)] This method has an advantage in preserving the nature of the system on both sides of the M-I transition.

§2. Experiments

Sample fabrication was performed as described in ref. 10. The resistivity was measured between (0.3 K or 1.5 K) to 15 K under a magnetic field 0 $\sim$ 7 T using a Linear Research LR-400 AC bridge ($\sim$15.9 Hz) with a PC/AT-base automatic data-acquisition program; and a measuring current $I_{\rm rms}$ of 3 μA$\sim$0.3 μA was used. The bar-shaped samples for this measurement have dimensions about 0.9×1.5×6 mm^2. Both the cooling and warming rates in the cycle measurement were controlled to be slow enough to ensure that the resistivity $\rho(T)$ curves measured during both the cooling and the warming processes can overlap each other.

§3. Results and Discussion

$Al_{70}Pd_{22.5}Re_{7.5}$ samples with a resistivity ratio ranging from 1.3 to 33.8 were prepared by varying annealing conditions as described in Table I. X-ray diffraction patterns reveal that all the prepared samples are single-phased. Figure 1 shows $\sigma(T)$ versus $T^{1/2}$ for typical samples QC6a–d. By fitting the straight portion of α versus $T^{1/2}$ curves to eq. (1), we obtain the values of conductivity $\sigma(0)$ extrapolated to T=0, m and Δ, which

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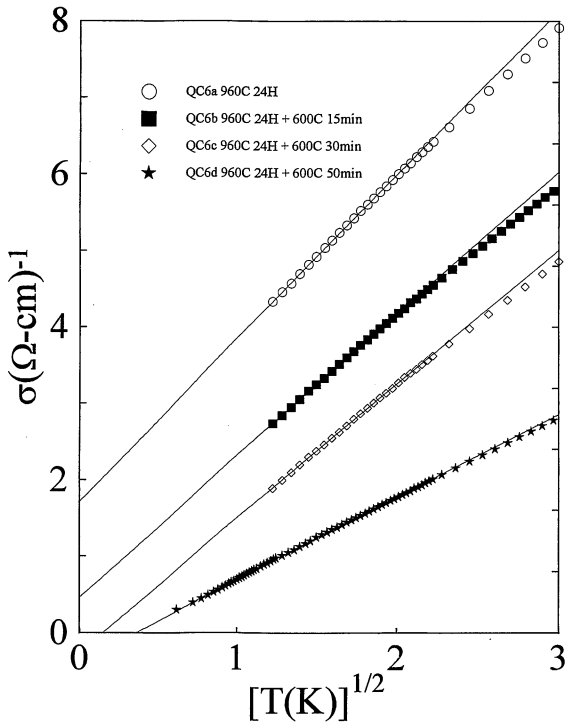

Fig. 1. Conductivity as a function of $T^{1/2}$ for samples QC6a-d.

are listed in Table I. We can see that increasing the resistivity ratio r=$\rho(4.2\,\mathrm{K})/\rho(300\,\mathrm{K})$ can decrease the values of $\sigma(0)$ and Δ quite rapidly. Double-logarithmic plot of the Δ spanning about six orders of magnitude against the zero-temperature resistivity $\rho(0) = 1/\sigma(0)$ is shown in Fig. 2(b). Δ follows the relation: $\Delta \propto \rho(0)^{-\nu}$ with $\nu \cong 2$. The solid line (the theoretical fit) is nearly parallel to the dashed line obtained from amorphous alloys, but shifts to higher resistivity. This may suggest that the electron-electron interactions in QCs are in general weaker than those in amorphous alloys. The obtained $\sigma(0)$ is seen to decrease almost linearly with increasing r and to go continuously to zero at r between 16.4 and 22.4. More experimental data are needed to determine accurately the critical resistivity ratio r_c at which the M-I transition occurs.

The increase of r arises from the improvement of structural perfection, as revealed in the reduction in the widths of diffraction peaks, has been widely observed in Al–Cu–(Fe, Ru) QCs[11] and it is also observed by us in Al–Pd–Re samples (samples QC1–5) with lower values of r. But no significant difference in the widths of the diffraction peaks was detected among samples QC6a–d ($13.8 \leq r \leq 33.6$) prepared with an additional low-T annealing (see Table I). The increased r in these samples might be attributed to the increased chemical order rather than quasilattice order due to structural relaxation during annealing.[12] If we assume that the increase of r in Al–Pd–Re indeed arises from the improved quasilattice and/or chemical order, then the M-I transition which occurs at $r = r_c$ may be regarded as occurring at a certain degree of structural imperfection. Structural disorder may persist into the insulating regime, although it might not be easily detected macroscopically. Consequently, to know how disorder affects the wave function of the electronic states near the Fermi level is important to fully understand the origin of the M-I transition in QCs.

Samples QC6a–d are taken as an example to study the M-T transition in Al–Pd–Re QCs in detail. They were cut from the same ingot but annealed at 600°C with a annealing time equal to 0, 15, 30, 50 min, respectively. Increasing the annealing time is seen to increase the resistivity and resistivity ratio significantly and moves the specimen close to and across the M-I transition. As seen in Fig. 1, samples QC6a and 6b are in a metallic state with a finite value of $\sigma(0)$ equal to 1.8 and 0.5 $(\Omega\text{-cm})^{-1}$, respectively. Samples QC6c and 6d subjected to a longer annealing time 30 and 50 min., respectively, are already pushed into a insulating state. This is judged by the following characteristics: (1) extrapolation of $\sigma(T)$ versus $T^{1/2}$ curves down to T=0 yields a negative $\sigma(0)$; (2) these two samples have a positive magnetoconductance (MC), which is a signature of hopping conduction in the insulating regime (see Fig. 3 and discussions below). We plotted $\ln[\sigma(T)]$ versus $T^{-1/4}$ and $T^{-1/2}$ between (15 $\sim$ 1.5 K) and (15 $\sim$ 0.5 K), respectively, for samples QC6c and 6d (see Fig. 4) and found that the data are fitted better with Mott's variable-range hopping (VRH) law:

$$\sigma(T) = \sigma_0 \exp[-(T_0/T)^{1/4}]. \qquad (3)$$

It should be mentioned here that the conductivity is fitted to eq. (3) without the need to include an extra term $\sigma(0)$ as Guo *et al.* did for analyzing their conductivity data.[13] The discrepancy in both results may result from the large difference in the values of r associated with respective samples. For example, the value of r ($\sim$100) for their most resistive sample[14] is much larger than that

Table I. The name of samples, annealing conditions, resistivity ratio and the values of $\sigma(0)$, m and Δ for $Al_{70}Pd_{22.5}Re_{7.5}$ quasicrystals.

Sample	Annealing conditions	$\rho(4.2\,\mathrm{K})/\rho(300\,\mathrm{K})$	$\sigma(0)$ $(\Omega\text{-cm})^{-1}$	m $(\Omega\text{-cm}\,\mathrm{K}^{1/2})^{-1}$	Δ (μeV)
QC1	960°C (50 h)	1.3	252.2	2.5	846142
QC2	950°C (24 h)	6.2	14.4	2.2	3533
QC3	940°C (24 h)	7.2	12.3	3.1	1314
QC4	950°C (40 h)	8.9	9.2	3.0	797.6
QC5	980°C (24 h)	11.6	3.8	3.4	110
QC6a	960°C (24 h)	13.8	1.8	2.1	62.4
QC6b	960°C (24 h)+600°C (15 min)	16.4	0.5	1.8	5.9
QC6c	960°C (24 h)+600°C (30 min)	22.4			
QC6d	960°C (24 h)+600°C (50 min)	33.8			

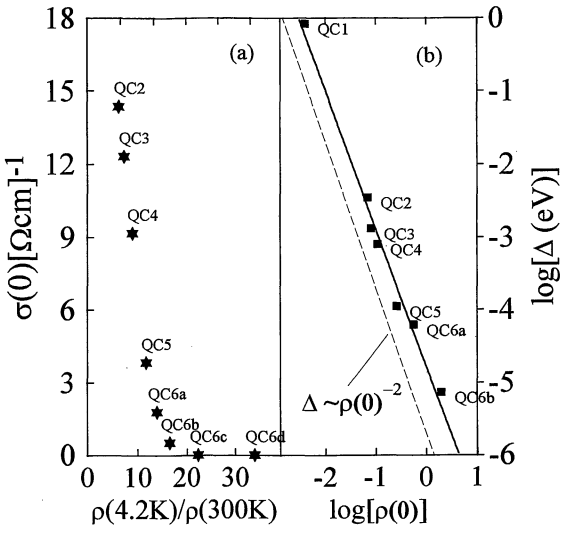

Fig. 2. (a) Conductivity $\sigma(0)$ extrapolated to T=0 plotted against the resistivity ratio $\rho(4.2\,\mathrm{K})/\rho(300\,\mathrm{K})$ for the samples marked. The value for QC1 is off the scale. (b) Double-logarithmic plot of Δ values $\rho(0)$ for the sample marked.

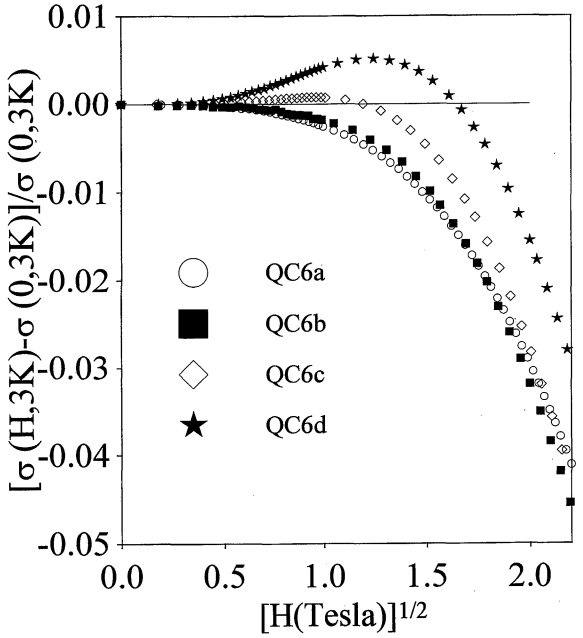

Fig. 3. Magnetoconductance $[\sigma(H)-\sigma(0)]/\sigma(0)$ as a function of square root of magnetic field $H^{1/2}$ for samples QC6a-d at T=3 K.

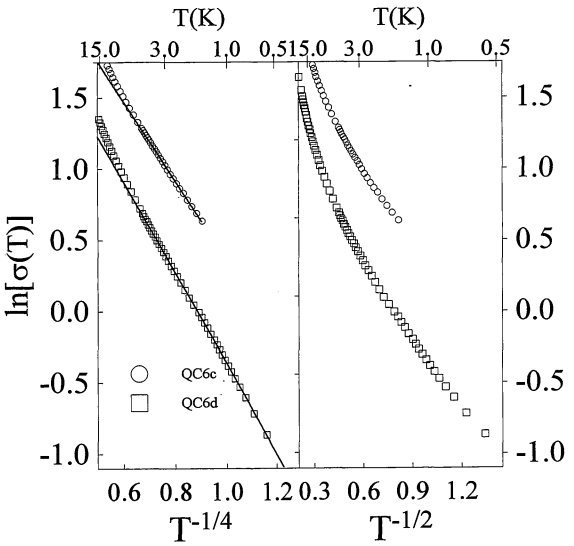

Fig. 4. $\ln[\sigma(0)]$ plotted against $T^{-1/4}$ and $T^{-1/2}$ for samples QC6c and 6d. ○: QC6c; □: QC6d. The straight lines are theoretical fits.

(~34) for QC6d. Their samples with much larger value of r, as we discussed earlier, are more free of structural disorder such that their hopping conduction may deviate from the Mott's V.R.H. behavior because Mott's law is, in principle, only valid for disordered systems. T_0 in eq. (3) is determined to be 61 K and 107 K, respectively, for samples QC6c and 6d. The localization length ξ calculated from $\xi = (18/k_B N(E_F)T_0)^{1/3}$, taking the density of state at the Fermi level $N(E_F) \sim 0.1$ (mJ/g-atom-K^2),[14] is 118 A and 99 A, respectively, for QC6c and 6d. It must be noted that the obtained ξ's are much larger than the separation (~20 A) of the ideal clusters which are assumed to be the structure units between which electrons hop in QCs, a conduction process proposed by Janot[15] based on the self-similar structure of QCs.

MC measurements at 3 K for samples QC6a–d are shown in Fig. 3. A negative MC observed in samples QC6a and 6b is a common feature of quasicrystalline metals with strong spin-orbit (SO) scattering. For samples QC6c and 6d, MC is positive at low fields, but changes to be negative at high fields and the latter sample with smaller ξ has larger positive value of $\Delta\sigma(H)/\sigma(0)$ and smaller negative value of $\Delta\sigma(H)/\sigma(0)$ at H=7 T. Positive MC can be attributed to the quantum interference among the various forward paths associated with hopping as explained by Nguyen *et al.*[16] However, there is a debate concerning whether a negative MC should appear in the system with strong SO scattering. For example, theories,[17] which are based on forward-directed-path formalism where returning-loops are neglected, predicted a positive MC in the strongly localized system, regardless of the strength of SO scattering. But some authors[18,19] pointed out that it is only true for the system in which the electron hopping distance $R_h \gg \xi$; for the system in the barely insulating regime, returning-loop effects are not negligible such that a large enough field (depending on temperature and ξ) in the presence of SO scattering can change the sign of MC due to the decrease of ξ. This has been well demonstrated in amorphous Y_xSi_{1-x}[20] and widely observed in amorphous alloys like WRe,[21] Mo_xGe_{1-x}[22] and Au-doped In_2O_3,[18] where SO scattering is strong.

For the studied Al–Pd–Re QCs, we get $R_h/\xi \approx 0.4(T_0/T)^{1/4}$=1.6–0.8 (between 0.5–7 K) for sample QC6d. This means that at certain temperature, $R_h < \xi$, indicating that this sample is in the critical regime approaching the M-I transition.[23] The SO scattering length $L_{so} = (D\tau_{so})^{1/2}$ for sample QC6c is about 3.2 A. Here the value of the diffusion constant $D \cong 10^{-6}$ (m^2/s) is determined using Einstein relation and the SO relaxation time $\tau_{so} \sim 10^{-13}$ s is estimated from the Al–Pd–Re QCs with the resistivity ratio ~6, based on the theory of weak localization. Because L_{so} is much smaller than

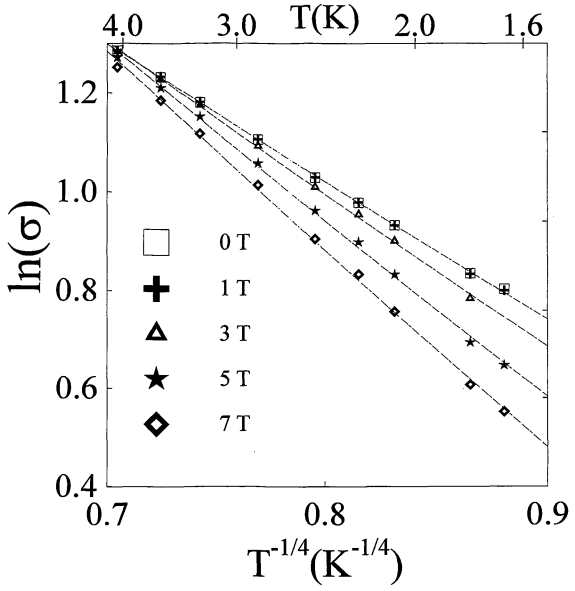

Fig. 5. Logarithm of $\sigma(T)$ as a function of $T^{-1/4}$ at various magnetic fields for sample QC6c.

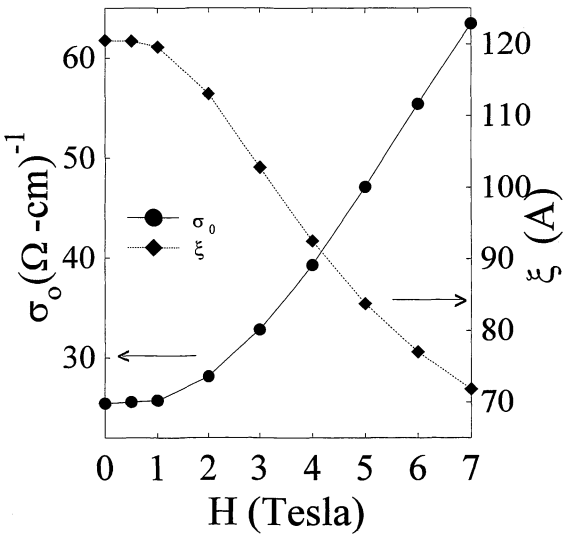

Fig. 6. Prefactor σ_0 and localization length ξ as a function of magnetic field for sample QC6c. The mark • denotes the value of the prefactor σ_0 in eq. (3), while ♦ denotes the value of the localization length ξ.

$\xi \cong 118$ A for QC6c, our samples are in a situation where theories[19] predicted to have a negative MC caused by the decrease of ξ. To see whether this indeed also occurs in QCs, the conductivity for sample QC6c as a function of temperature and magnetic field H was measured. Plotting ln $\sigma(T)$ against $T^{-1/4}$ at various fields, as shown in Fig. 5, reveals that the $\sigma(T)$ under the magnetic field also obeys Mott's law. The values of the prefactor σ_0 in eq. (3) and the localization length ξ are calculated and displayed in Fig. 6. It is seen that as the magnetic field H is increased, σ_0 increases steadily while ξ decreases monotonically. Thus the negative MC at high fields is indeed chiefly dominated by the reduction in ξ because $\sigma(T, H)$ depends on $\xi(H)$ exponentially, while it depends on $\sigma_0(H)$ linearly.

§4. Conclusion

The similarities in the low-T conductivity and the approach to M-I transition among QCs, amorphous alloys, granular metal films and doped semiconductors strongly suggest that both localization and electron-electron interactions must be taken into consideration more seriously in understanding the low-T electronic conduction in QCs. The effect of disorder (structural and even chemical)-induced localization on the VRH conduction in QCs may not be completely neglected.

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Note added in proof—After completing the paper, we became aware of a paper by Ahlgren *et al.*[24] which reports that the conductivity $\sigma(T)$ for AlPdRe samples with a resistivity ratio r ranging from 10.4 ~ 56.5 is finite at T=0. If that is true, there will be

no M-I transition in Al–Pd–Re QCs. However, our results clearly show that the AlPdRe with a value of $r \geq 22.4$ would have a positive MC at low temperature, a positive MC, so far as we know, cannot be allowed by any existing theories to appear in a system like Al–Pd–Re QCs with a high resistivity in a metallic state in the presence of strong SO scattering. On the other hand, the consistency among the perdiction of McMillan's theory (that is, the M-I transition occurs at Δ=0), the appearance of positive MC, and the observed VRH conduction leads us to believe preferably that our samples have been through the M-I transition. Nevertheless the residue conductivity at T=0 still remains unanswered. Therefore, it is suggested that more theoretical and experimental studies on the electronic properties at much lower temperature $T < 0.3$ K are needed to resolve this problem.