行政院國家科學委員會補助專題研究計畫成果報告
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※ 絕緣態的 Al70Pd22.5Re7.5 準晶物質，Mott 跳躍傳導 ※
※ 變成 Efros-Shklovskii(ES)跳躍傳導的研究 ※
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中文摘要

利用電導數據及磁阻理論，我們發現電阻比 81-118 的準晶的低溫傳導會由 Efros-Shklovskii 跳躍傳導變成 Mott 跳躍。轉變溫度 (crossover temperature) 隨電阻比值下降而下降，高電阻比的準晶其負磁阻的大小在低溫時急劇下降，可能如同在 CdTe 半導體一樣，其傳導是經由庫侖能隙的電子態傳導所致。
A crossover from Mott to Efros-Shklovskii hopping conduction
In insulating Al$_{70}$Pd$_{22.5}$Re$_{7.5}$ quasicrystals – a magnetoresistance study

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ABSTRACT

Combining the conductivity and the magnetoresistance (MR) data with the MR theories, we found that there is a crossover from the Mott to Efros-Shklovskii variable-range hopping conduction in insulating Al_{70}Pd_{22.5}Re_{7.5} quasicrystals (QC's) with the resistivity ratio $\mathcal{R} = \rho(4.2\,\text{K}) / \rho(300\,\text{K})$ equal to 118 and 81. The crossover temperature is seen to decrease with decreasing the sample's $\mathcal{R}$ value. At low temperature the rapid decrease in the magnitude of the negative MR in highly resistive QC's might be correlated with the conduction via the states in the Coulomb gap as observed in CdTe semiconductors.

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I. INTRODUCTION

Recently we found that the temperature dependence of the magnetoresistance (MR) of insulating Al-Pd-Re quasicrystals (QC’s) could be described well by the forward interference and the wave-function shrinkage theories. A crossover from Mott to Efros-Shklovskii (ES) variable–range hopping (VRH) conduction could appear at liquid helium temperatures for Al$_{70}$Pd$_{22.5}$Re$_{7.5}$ QC’s having a resistivity ratio $\rho_\parallel = 77^1$. The transition from the Mott VRH to the ES VRH in disordered systems is usually studied by fitting the dc conductivity measured in a wide range of temperatures with the Mott VRH law or the ES VRH law$^2$. However, the residual zero–temperature conductivity $\sigma (0)$ in insulating Al-Pd-Re QC’s$^3$ makes it difficult to distinguish the Mott VRH from the ES VRH at low temperatures. In this work, we will demonstrate how to identify the transition from the Mott VRH to the ES VRH conduction by combining the conductivity and MR data with the MR theories in VRH regime.

II. THEORIES

A. The forward interference (FI) model

The FI model considers the effect of interference among various hopping trajectories between two hopping sites and predicts a negative MR$^5$. An empirical equation, which predicts a linear field dependence of the MR ratio $r = R(B,T)/R(0,T)$ at low magnetic fields$^5$ and a saturation MR at high magnetic fields$^6$ can be approximately written as$^7$

$$r_{\text{forward}} \approx 1 / \{ 1 + C_{\text{sat}} [ B / B_{\text{sat}} ] / [ 1 + B / B_{\text{sat}} ] \}. \quad (1)$$

For $B >> B_{\text{sat}}$ and $C_{\text{sat}}$ is small, $r_{\text{forward}} \approx 1 - C_{\text{sat}}$, where $C_{\text{sat}}$ is called the saturation constant and $B_{\text{sat}}$ is the effective saturation field.
For the Mott VRH case, $B_{sat}$ is given by

$$B_{sat} \approx 0.7 \left( \frac{T}{T_{Mott}} \right)^{-\frac{3}{2}} \left( \frac{h}{a_0^2} \right)^{\frac{3}{2}} \left( \frac{1}{T_{Mott}} \right)^{\frac{3}{8}}.$$  \hspace{1cm} (2)

B. The wave-function shrinkage (WFS) model

This model takes into account the contraction of the electronic wave function at impurity centers in a magnetic field $B$, which leads to a reduction in the hopping probability between two sites and therefore to a positive $MR^8$. An expression for the MR ratio $r_{wave}$, which is valid for an entire field regime, was given by Schoepf$^9$, i.e.,

$$r_{wave} = \exp \left\{ \xi_C(0) \left[ \frac{\xi_C(B)}{\xi_C(0)} - 1 \right] \right\}, \hspace{1cm} (3)$$

where $\xi_C(0) = (T_{Mott}/T)^{1/4}$ for the Mott VRH case and $\xi_C(B)/\xi_C(0)$ is called the normalized hopping probability parameter. Tabulated values of $\xi_C(B)/\xi_C(0)$ as a function of $B/B_c$ are listed in Ref. 10. $B_c$ is the only fitting parameter in Eq. (3), given for the Mott VRH case by

$$B_c = 6h/\left[ ea_0^2 (T_{Mott}/T)^{1/4} \right], \hspace{1cm} (4)$$

where $a_0$ is the localization length.

In the low field limit, one can easily obtain from Eq. (3)

$$r_{wave}^{-1} = \frac{\Delta R(B, T)}{R(0, T)} = \frac{[R(B, T) - R(0, T)]/R(0, T)}{\approx 0.0893 \frac{B^2}{B_c^2} \left( \frac{T_{Mott}}{T} \right)^{1/4}}, \hspace{1cm} (5)$$

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as predicted\textsuperscript{11}.

If the effects of the FI and the WFS are independent, Eq. (1) and Eq. (3) are additive, and then the total MR ratio \( r_{\text{total}} \) can be given as

\[
 r_{\text{total}} = \exp \left\{ \xi_c (0) \left[ \frac{\xi_c (B)}{\xi_c (0)} - 1 \right] \right\} 
+ \left\{ 1 + \frac{1}{1 + C_{\text{sat}} [B / B_{\text{sat}}]} \right\} - 1. 
\]

The last term, \(- 1\), insures that \( r_{\text{total}} \) equals 1 when \( B = 0 \).

III. EXPERIMENTAL PROCEDURES

Ingots of Al\textsubscript{70}Pd\textsubscript{22.5}Re\textsubscript{7.5} alloys were obtained by arc melting of a mixture of Al (99.99\%), Pd (99.99\%), and Re (99.99\%) in a purified argon atmosphere.

High-quality Al\textsubscript{70}Pd\textsubscript{22.5}Re\textsubscript{7.5} QC’s with \( n_f = 118 \) and 81 were prepared by annealing the ingots in vacuum at 950°C for 24-28 hs and subjected a further annealing at 600°C for 3 hs. The conductivity was measured between 0.3 K and 20 K, while the MR was measured in the temperature range \( 1.5 \text{ K} < T \leq 4.2 \text{ K} \) and in a magnetic field of \( 0 \text{ T} \leq B \leq 6.5 \text{T} \). Detailed description of the sample fabrications and experimental measuring techniques can be found in Ref. 12.

IV. RESULTS

The low-T conductivity for insulating Al\textsubscript{70}Pd\textsubscript{22.5}Re\textsubscript{7.5} QC’s can be fitted with a modified VRH Mott law\textsuperscript{3}, i.e.,

\[
\sigma (T) = \sigma (0) + \sigma_e \exp \left[ - \left( \frac{T_{\text{Mott}}}{T} \right) \right]^{1/4}. 
\]
Figure 1 plots \( \ln[\sigma(T) - \sigma(0)] \) against \( T^{-1/4} \) for Al\(_{70}\)Pd\(_{22.5}\)Re\(_{7.5}\) QC's with \( R = 118 \) and 81. We extracted the value of \( T_{Mott} = 1411 \) K and 1243 K, and the localization length \( a_0 = 41.3 \) Å and 44 Å, respectively, for samples with \( R = 118 \) and 81.

Figures 2a and 2b show the MR ratio \( r = R(B, T)/R(0, T) \) versus magnetic field B for samples \( R = 118 \) and 81. It is seen that the MR is negative at low fields and appears positive at higher fields. At each temperature, there is a MR ratio minimum \( r_{min} \); as the temperature is lowered, the position of the minimum shifts to the low field side and the magnitude of \( r_{min} \) decreases.

First, we used Eq. (6) for the Mott case to fit the MR data. Eq. (6) consists of three fitting parameters \( B_{sat} \), \( C_{sat} \) and \( B_c \). For the detailed fitting procedures, one can refer to Ref.1. The fitting results are shown in Fig. 2, where the theoretical fits are compared with the experimental data. It is clearly seen that except the MR data at lower temperature and in the high field regime, the MR data can be described well by the theories. The obtained values of \( B_{sat} \) are plotted against \( T^{3/8} \) for samples \( R = 118 \) and 81, as seen in Fig. 3a. The \( B_{sat} \)'s are seen to follow the Mott VRH behavior between 2.76 K and 4.2 K for sample \( R = 118 \), and between 2.594 K and 4.2 K for sample \( R = 81 \); but they deviate from the Mott law for \( T < 2.76 \) K and 2.594 K, respectively. The obtained \( B_c \)'s, ranging from 12 \( \sim \) 27 T, obey the Mott law in the same temperature ranges as \( B_{sat} \)'s do. We have repeatedly emphasized that the electron–electron interaction (EEI) plays a crucial role in the metal-insulator transition of Al-Pd-Re QC's\(^4\). Therefore, taking into consideration the EEI, the hopping conduction may follow the ES law at low temperature. Thus, we try to fit the MR data below 2.76 K and 2.594 K with Eq. (6) for the ES VRH case. Here \( (T/T_{Mott})^{3/8} \) in \( B_{sat} \) is replaced by \( (T/T_{ES})^{3/4} \) and \( (T/T_{Mott})^{1/4} \) in \( B_c \) by \( (T/T_{Mott})^{1/2} \cdot \xi_e(0) \) for the ES VRH case is \( (T_{ES}/T)^{1/2} \). The value of \( T_{ES} \approx 7.4 \) K was obtained by fitting the conductivity \( \sigma(T) \) to a modified ES VRH law, i.e.,
\[ \sigma (T) = \sigma (0) + \sigma_0 \exp \left[ - \left( \frac{T_{ES}}{T} \right)^{1/2} \right]. \] (8)

One can see in Fig. 1 that at low temperatures the conductivity \( \sigma (T) \) can also be fitted well with Eq. (8).

The obtained values of B_{sat} versus \( T^{3/4} \) are displayed in Fig. 3b. We can clearly see that at lower temperatures the temperature-dependent B_{sat} for both samples does follow the ES law instead of the Mott law. The extracted B_C's also follow the ES law at lower temperature. The crossover temperatures from the Mott VRH to the ES VRH conduction are determined to be around 2.32 K and 1.91 K, respectively, for samples \( \mathcal{R} = 118 \) and 81.

The values of C_{sat}, which are related to the magnitude of the negative MR, are extracted from theoretical fits and are plotted against T, as presented in Fig.4. The value of C_{sat} is larger for the sample with a larger value of \( \mathcal{R} \). C_{sat} is seen to decrease with decreasing temperature and to drop rapidly at lower temperatures. The rapid decrease in C_{sat} at low temperatures was also observed in doped CdTe semiconductors and was attributed to the conduction via the states in the Coulomb gap^{13}.

V. CONCLUSION

We found that there is a crossover from the Mott VRH to the ES VRH conduction in insulating Al_{70}Pd_{22.3}Re_{7.5} QC's with \( \mathcal{R} = 118 \) and 81. This suggests that there is a Coulomb gap in the density of states at the Fermi level in highly resistive Al-Pd-Re QC's. As the sample's \( \mathcal{R} \) value is lowered, the crossover temperature drops and the crossover region expressed by the temperature interval marked by the arrows becomes wider (see Fig 4).
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REFERENCES


Figure Captions

Fig. 1  \( \ln \left[ \sigma (T) - \sigma (0) \right] \) is plotted against \( T^{-1/4} \) and \( T^{-1/2} \) for samples \( \mathfrak{R} = 118 \) and \( 81 \). The solid lines are least-squares fits.

Fig. 2  The MR ratio \( r = R(B, T) / R(0, T) \) at different temperatures versus magnetic field B. Solid lines and dash lines are theoretical fits obtained by using Eq. (6) for the Mott VRH case and for the ES VRH case to fit the data, respectively. (a) sample \( \mathfrak{R} = 118 \), (b) sample \( \mathfrak{R} = 81 \). The curves have been shifted downward from one another to present clarity between the different data and fits.

Fig. 3  (a) \( B_{\text{sat}} \) as a function of \( T^{3/8} \) for samples \( \mathfrak{R} = 118 \) and \( 81 \) in the Mott VRH regime. The solid lines are least-squares fits.

(b) \( B_{\text{sat}} \) as a function of \( T^{3/4} \) for samples \( \mathfrak{R} = 118 \) and \( 81 \) in the ES VRH regime. The solid lines are least-squares fits.

Fig. 4  Extracted values of \( C_{\text{sat}} \) as a function of temperature T for samples \( \mathfrak{R} = 118 \) and \( 81 \). The crossover region is between the two arrows marked in the figure. The solid lines are drawn for guiding the eyes.
Fig 1
Fig 2
Fig 3
Fig 4