Magnetoresistance of $Al_{70}Pd_{22.5}Re_{7.5}$ quasicrystals in the variable-range hopping regime

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The temperature dependence of the magnetoresistance (MR) of insulating $Al_{70}Pd_{22.5}Re_{7.5}$ quasicrystals taken at liquid-helium temperatures can be explained by the theories of MR—the forward interference and the wave-function shrinkage—in the variable-range hopping (VRH) regime. By analyzing the MR data with the theories mentioned above, a crossover from Mott VRH conduction to Efros-Shklovskii VRH conduction at liquid helium temperatures was identified in a highly resistive $Al_{70}Pd_{22.5}Re_{7.5}$ sample. The rapid decrease in the negative MR of highly resistive samples at low temperatures might be attributed to the conduction via the states in the Coulomb gap.

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I. INTRODUCTION

Bulk $Al_{70}Pd_{22.5}Re_{7.5}$ quasicrystals (QC's) with a resistivity ratio $\mathfrak{R}=\rho(4.2\,\mathrm{K})/\rho(300\,\mathrm{K})<13$ are found to be metallic.[1] Their low-temperature magnetoresistance (MR) is positive because of a strong spin-orbit scattering and can be described well by quantum interference theories (QIT).[2,3] This suggests that electrons in metallic QC's transport via a diffusive process. In contrast, for a $Al_{70}Pd_{22.5}Re_{7.5}$ QC with $\mathfrak{R}\geqslant 18$, its MR appears small and negative at low magnetic fields and changes to large and positive values at higher magnetic fields; the QIT theories fail to explain its MR,[3–6] and its conductivity exhibits Mott variable range hopping (VRH) behavior.[1,7] This signals that the metal-insulator transition has been crossed over.[1]

The common features of the MR of insulating $Al_{70}Pd_{22.5}Re_{7.5}$ QC's ($\mathfrak{R}\geqslant 18$) including negative MR at low fields and positive MR at high fields are also widely observed in disordered systems like amorphous Ni_xSi_{1-x},[8] Y_xSi_{1-x},[9] and Mo_xGe_{1-x}.[10] Since the origins of the negative and positive MR are relatively complicated, it is not an easy task to account for those MR data. Recently, a numerical model taking into account the forward interference process and the wave-function shrinkage process was successful in interpreting the low-field MR of amorphous Ni_xSi_{1-x} (Ref. 8) films. These two theoretical models are known to be formulated for describing the MR of highly disorderedly localized systems in the VRH regime. The fact that insulating Al-Pd-Re QC's at low temperatures exhibit Mott VRH behavior implies that disorder (structural and/or chemical) plays an important role in the electronic transport properties of QC's. Therefore, we believe that this numerical model can be applied to insulating QC's too. In this work we will report how to use this numerical model to fit the MR data of $Al_{70}Pd_{22.5}Re_{7.5}$ QC's in the VRH regime.

II. THEORIES

A. Forward interference (FI) model

The FI model (also called the forward-directed path approach) was first proposed by Nguyen, Spivak, and Shklovskii (NSS).[11] They consider the effect of interference among various hopping paths between hopping sites. These paths include sequence of scatterings of tunneling electrons by the impurities located within a cigar-shaped domain of length R_h (hopping distance) and width $(R_h a_o)^{1/2}$, where a_o is the localization length. Averaging numerically the logarithm of the conductivity over many different possible paths in the presence of the magnetic field, NSS obtained a negative MR which is linear in magnetic field B in the low field limit. Later, using a critical percolating method instead of logarithmic average, Sivan and co-workers[12] obtained a negative MR which is quadratic in B at small fields and which saturates to a negative value at high fields. Realistically, the linear field dependence of MR is often observed at low fields; the quadratic field dependence of the MR is only ocassionally seen in some samples at very weak fields. Therefore, an empirical equation, which describes the MR ratio $r=R(B,T)/R(0,T)$ caused by the interference effects and neglects the quadratic term in B, can be expressed approximately by the relation[8]

$$r_{\mathrm{forward}}\approx 1/\{1+C_{\mathrm{sat}}[B/B_{\mathrm{sat}}]/[1+B/B_{\mathrm{sat}}]\}, \tag{1}$$

where the two fitting parameters are the saturation constant C_{sat} and the effective saturation magnetic field B_{sat} given for the Mott VRH case by[12]

$$B_{\mathrm{sat}}\approx 0.7\left(\frac{8}{3}\right)^{3/2}\left(\frac{1}{a_0^2}\right)\left(\frac{h}{e}\right)\left(\frac{T}{T_{\mathrm{Mott}}}\right)^{3/8} \tag{2}$$

In the low-field limit,

$$r_{\mathrm{forward}}\approx 1-C_{\mathrm{sat}}\frac{B}{B_{\mathrm{sat}}}. \tag{3a}$$

This gives

$$\frac{\Delta R(B,T)}{R(0,T)}=\frac{R(B,T)-R(0,T)}{R(0,T)}\approx -C_{\mathrm{sat}}\frac{B}{B_{\mathrm{sat}}}. \tag{3b}$$

B. Wave-function shrinkage model

This theory considers the contraction of the electronic wave function at impurity centers in a magnetic field, which leads to a reduction in the hopping probability between two sites and therefore to a positive MR. The authors of Refs. 13 and 14 proposed this theory but only gave the expressions for the low- field and high- field limits. Later, Schoepe[15] developed a method of calculations for the entire field regime, obtaining an expression for r_{wave} under the assumption that the density of states at the Fermi level $N(E_F)$, and the localization length a_o are independent of the magnetic field. r_{wave} is given as

$$r_{\mathrm{wave}}=\exp\{\xi_C(0)[\xi_C(B)/\xi_C(0)-1]\}, \tag{4}$$

where $\xi_C(0)=(T_{\mathrm{Mott}}/T)^{1/4}$ for the Mott VRH case and $\xi_C(B)/\xi_C(0)$ is the normalized hopping probability parameter. Detailed descriptions of the calculations of $\xi_C(B)/\xi_C(0)$ as a function of B/B_c are given in Ref. 15. Tabulated values of $\xi_C(B)/\xi_C(0)$ as a function of B/B_c for Mott and Efros-Shklovskii (ES) VRH cases are listed in Ref. 16. The only fitting parameter in Eq. (4) is B_c, given for the Mott VRH case by

$$B_c=6\hbar/[ea_o^2(T_{\mathrm{Mott}}/T)^{1/4}]. \tag{5}$$

In the low-field limit, Eq. (4) can be simplified to[17]

$$r_{\mathrm{wave}}\approx 1+t_2\frac{B^2}{B_c^2}\left(\frac{T_{\mathrm{Mott}}}{T}\right)^{1/4} \tag{6}$$

Then

$$\frac{\Delta R(B,T)}{R(0,T)}\approx t_2\frac{B^2}{B_c^2}\left(\frac{T_{\mathrm{Mott}}}{T}\right)^{1/4}, \tag{7}$$

where $t_2=(5/2016)\times 36\approx 0.0893$ is a numerical constant.

Assuming that the MR contributions from Eqs. (1) and (4) are additive, the total MR ratio r_{total} can be written as

$$r_{\mathrm{total}}=\exp\{\xi_C(0)[\xi_C(B)/\xi_C(0)-1]\} + 1/\{1+C_{\mathrm{sat}}[B/B_{\mathrm{sat}}]/[1+B/B_{\mathrm{sat}}]\}-1. \tag{8}$$

The last term, -1, is needed to assure that r_{total} is equal to 1 when $B=0$. We will use Eq. (8) to fit the MR data of $Al_{70}Pd_{22.5}Re_{7.5}$ QC's.

III. EXPERIMENTS

Ingots of $Al_{70}Pd_{22.5}Re_{7.5}$ alloys were obtained by arc melting of a mixture of high purity Al (99.99%), Pd (99.99%), and Re (99.99%) in a purified argon atmosphere. Icosahedral $Al_{70}Pd_{22.5}Re_{7.5}$ QC's with a wide range of $\Re$ 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. Low-temperature annealing (~600 °C) is an important step for preparing good-quality samples with $\Re > 40$.[1,18] The quality of the quasicrystalline samples was assessed by x-ray diffraction patterns. The resistance and MR were measured, using a Linear Research LR-700 AC resistance bridge (~15.9 Hz). Sample fabrications and experimental measuring techniques are described in detail in Ref. 19.

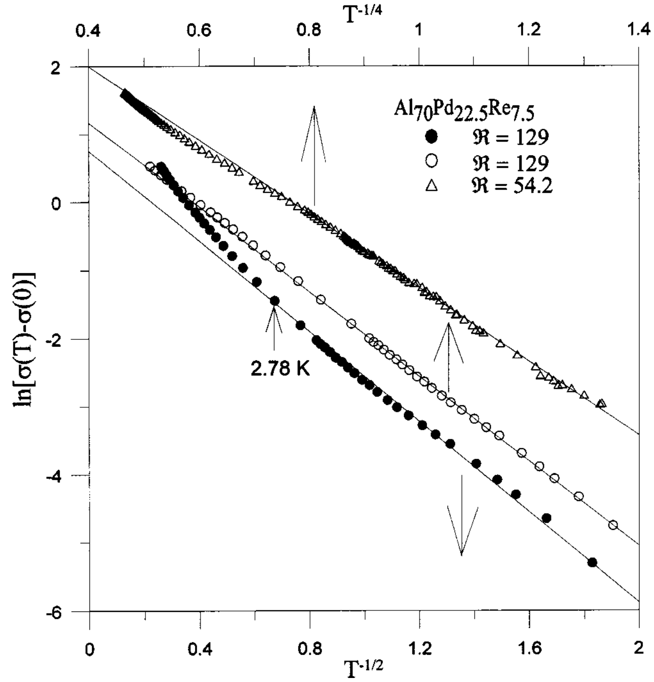

FIG. 1. $\ln[\sigma(T)-\sigma(0)]$ is plotted against $T^{-1/4}$ and $T^{-1/2}$ for sample $\Re=129$, against $T^{-1/4}$ for sample $\Re=54.2$.

IV. RESULTS AND DISCUSSION

Guo and Poon first found that the low-T conductivity $\sigma(T)$ for highly resistive $Al_{75}Pd_{21}Re_{8.5}$ QC's can be fitted with a modified Mott law, i.e.,

$$\sigma(T)=\sigma(0)+\sigma_0\exp[-(T_{\mathrm{Mott}}/T)]^{1/4}. \tag{9}$$

This was confirmed later by Wang *et al.*[6] Recently, we found the conductivity $\sigma(T)$ between 0.05 and 1.6 K for the $Al_{70}Pd_{22.5}Re_{7.5}$ QC with a value of $\Re$ as low as 13.2 obeys Mott's VRH law, and obtained its value of T_{Mott} to be about 3.1 K. This confirms that the previously determined critical resistivity ratio $\Re_c\approx 12.8\pm 0.5$ for the metal-insulation transition in $Al_{70}Pd_{22.5}Re_{7.5}$ QC's is reliable.[1] The significance of these results will be presented in a future paper shortly. According to our studies, the value of $\sigma(0)$ decreases as the $\Re$ value of the sample is increased; and the effects of $\sigma(0)$ on the low-T $\sigma(T)$ can only be seen clearly at the temperature about two orders of magnitude lower than the value of T_{Mott}. In this article we will focus on studying the MR of the $Al_{70}Pd_{22.5}Re_{7.5}$ QC's with $\Re=54.2$ and 129. Both samples are supposed to be well inside the insulating regime.

Figure 1 shows $\ln[\sigma(T)-\sigma(0)]$ plotted against $T^{-1/4}$ for samples $\Re=54.2$ and 129. The value of T_{Mott} is extracted by fitting the $\sigma(T)$ data to Eq. (9). Using the value of T_{Mott}, we can calculate the value of the localization length a_0 from the relation. $a_0=[18/k_BN(E_F)T_{\mathrm{Mott}}]^{1/3}$, where $N(E_F)$ was evaluated with the value of $\gamma\approx 0.1(\mathrm{mJ/g\,atm\,K^2})$ determined from specific-heat measurements.[18] The ratio of the hopping distance R_h to the localization length a_0 can be estimated from the relation $R_h/a_0\approx 0.4(T_{\mathrm{Mott}}/T)^{1/4}$. Table I lists

TABLE I. The values of T_{Mott}, T_{ES}, the localization length a_0, and the ratio of the hopping distance to the localization length, R_h/a_0, between 1.5 and 4.2 K.

$Al_{70}Pd_{22.5}Re_{7.5}$	T_{Mott} (K)	T_{ES} (K)	a_0 (Å)	R_h/a_0(1.5∼4.2 K)
$\Re$=54.2	517	-	56	1.8–1.4
$\Re$=129	1509	11	41	2.3–1.7

the values of T_{Mott}, a_0, and R_h/a_0 between 1.5 and 4.2 K. From the small values of a_0, samples $\Re$=54.2 and 129 seem to be strongly localized systems; however, their values of R_h/a_0 between 4.2 and 1.5 K are only 1.8–1.4 and 2.3–1.7, respectively. Therefore, these two QC's, in fact, cannot be classified as strong insulators.

Figures 2(a) and 2(b) show $\Delta R(B,T)/R(0,T)$ versus B^2 for samples $\Re$=54.2 and 129. At low fields, positive MR is seen to depend on B quadratically, as predicted by Eq. (7).

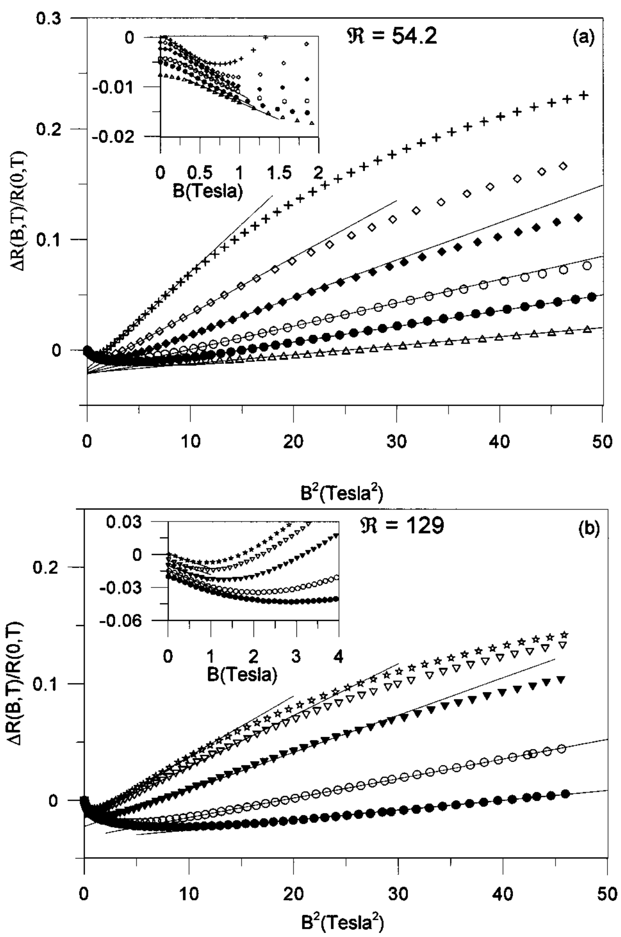

FIG. 2. (a) $\Delta R(B,T)/R(0,T)$ as a function of the square of the magnetic field B^2 for sample $\Re$=54.2. Inset: $\Delta R(B,T)/R(0,T)$ as a function of the magnetic field B. Solid lines are drawn for guiding the eyes. The data from the bottom to top are 4.271 K (△), 3.478 K (●), 3.052 K (○), 2.495 K (◆), 2.047 K (◇), and 1.558 K (+). (b) $\Delta R(B,T)/R(0,T)$ as a function of B^2 for sample $\Re$=129. Inset: $\Delta R(B,T)/R(0,T)$ as a function of B. Solid lines are drawn for guiding the eyes. The data from the bottom to top are 4.290 K (●), 3.294 K (○), 2.328 K (▼), 1.998 K (▽), and 1.875 K (☆).

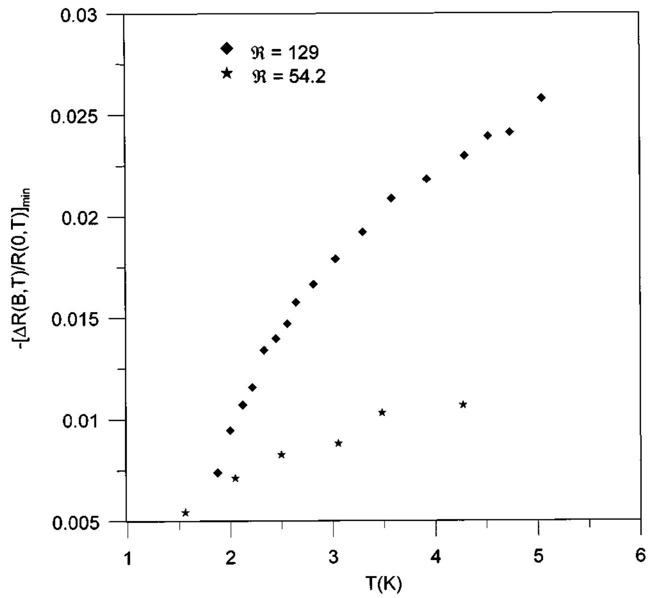

FIG. 3. The experimental values of $-[\Delta R(B,T)/R(0,T)]_{min}$ as a function of temperature T for samples $\Re$=54.2, and 129.

The insets in Figs. 2(a) and 2(b) show $\Delta R(B,T)/R(0,T)$ versus B. It is seen that there is a minimum at each temperature; and as the temperature is lowered, the position of the minimum (B_{min}) shifts to the low-field side with decreasing temperature, also the magnitude of $\Delta R(B,T)/R(0,T)$ at the minimum, $[\Delta R(B,T)/R(0,T)]_{min}$, decreases. Below B_{min}, $\Delta R(B,T)/R(0,T)$ is linearly dependent on B, obeying the prediction of Eq. (3). For sample $\Re$=54.2, it is noted that $\Delta R(B,T)/R(0,T)$ exhibits a quadratic field dependence at very weak fields; and it is pronounced at higher temperatures and tends to disappear at low temperatures, consistent with the prediction of Ref. 20. In fact, the MR of highly resistive Al-Pd-Re QC's has also been studied by Wang *et al.*[6] and Rodmar *et al.*[3]; some of the MR behaviors observed here were also reported. However, no interpretation of the MR based on the MR theories in the VRH regime was considered.

The above analysis amazingly reveals that all the general features of the MR in the low-field limit predicted by the theories are observed in QC's with a single composition. This strongly suggests that applying these theories to insulating QC's is appropriate.

In the low-field limit, one can easily show that from Eqs. (3) and (7) that

$$[\Delta R(B,T)/R(0,T)]_{min} \propto -C_{sat}^2. \qquad (10)$$

The experimental values of $[\Delta R(B,T)/R(0,T)]_{min}$ versus T for samples $\Re$=54.2 and 129 are presented in Fig. 3. It indicates that the C_{sat} for both samples decreases with decreasing temperature and the C_{sat} for sample $\Re$=129 decreases quite rapidly at low temperatures. Equation (8) consists of three fitting parameters C_{sat}, B_{sat}, and B_c. In making the least-squares fitting of Eq. (8) for the Mott VRH case to the MR data, we give the C_{sat} at each temperature an initial value obtained from Eq. (10), and let C_{sat}, B_{sat}, and B_c vary.

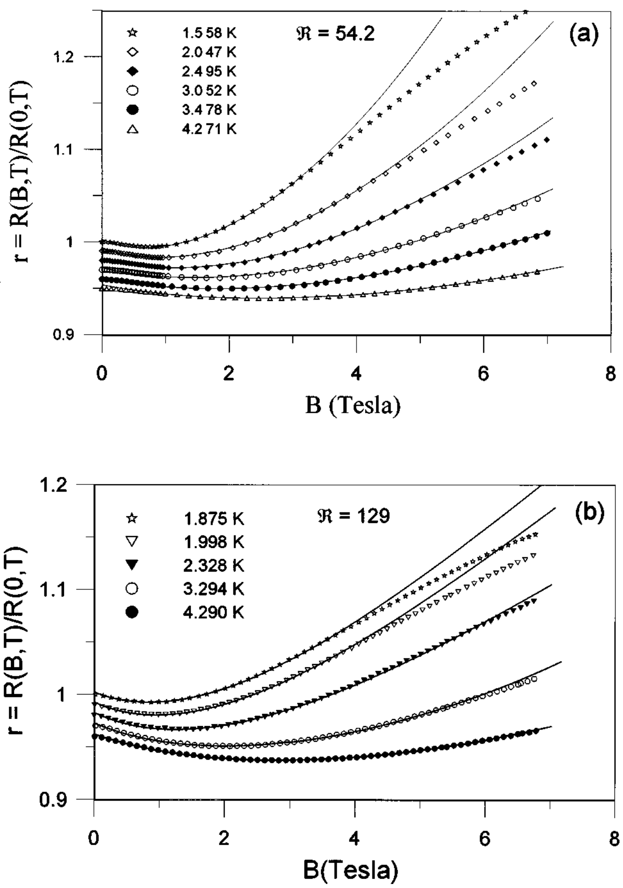

FIG. 4. (a) The MR ratio $r = R(B,T)/R(0,T)$ at different temperatures plotted against the magnetic field B for sample $\Re = 54.2$. Solid lines are theoretical fits, obtained by using Eq. (8) for the Mott VRH case to fit the data. (b) The MR ratio, $r = R(B,T)/R(0,T)$, at different temperatures plotted against B for sample $\Re = 129$. Solid lines at 4.290 and 3.294 K are theoretical fits, obtained by using Eq. (8) for the Mott VRH case to fit the data, while solid lines at 2.328, 1.998, and 1.875 K are obtained by using Eq. (8) for the ES VRH case to fit the data (see the discussions in the text). Note: The curves in (a) and (b) have been shifted downward from one another to present clarity between the different data and fits.

During fitting procedures, we found that the key to get a good fit is to make detailedly accurate measurements of $[\Delta R(B,T)/R(0,T)]_{\min}$, because C_{sat} is closely related to B_{sat}. Figure 4 shows the comparison between the MR ratio data and the theoretical fits for samples $\Re = 54.2$ and 129. It can be seen that the transition from a negative MR at low fields to a positive MR at higher fields can be described well by the theories. But in the high-field region where the theoretical curves keep rising, the experimental data tends to saturate. This suggests that (1) the field dependence of $N(E_F)$ and a_0 neglected in deriving Eq. (4) must be taken into account; but this effect should be small because the values of a_0 for the studied samples are already small; and (2) the FI model should be corrected for returning-loop effects in $Al_{70}Pd_{22.5}Re_{7.5}$ samples where the value of R_h is not much larger than that of a_0 (see Table I).[21] It has been shown that in the system like Al-Pd-Re QC's where the spin-orbit scattering is strong, returning-loop effects can cause a positive MR and saturation of the MR at large enough fields, depending on the temperature and localization length.[9]

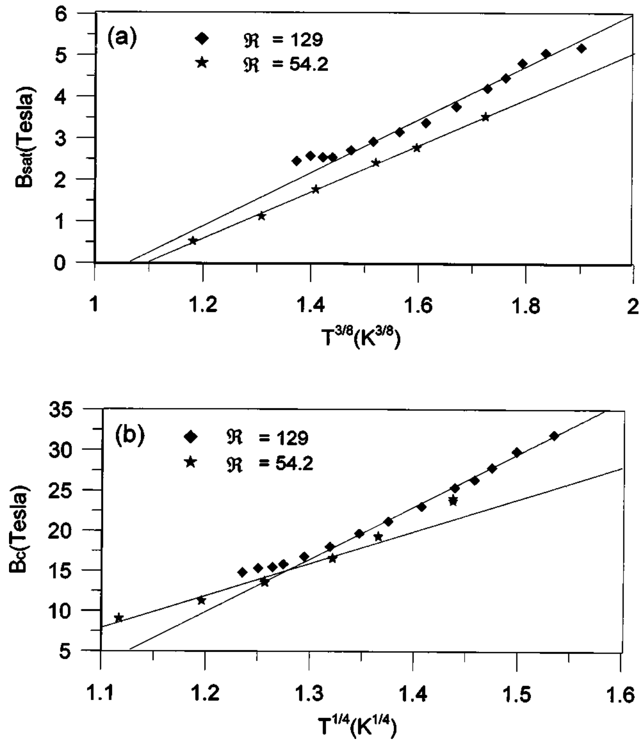

FIG. 5. (a) B_{sat} as a function of $T^{3/8}$ and (b) B_c as a function of $T^{1/4}$ for samples $\Re = 54.2$ and 129 in the Mott VRH regime.

Figures 5(a) and 5(b) show the extracted values of B_{sat} and B_c, respectively, versus $T^{3/8}$ and $T^{1/4}$ for samples $\Re = 54.2$ and 129. We can see that for sample $\Re = 54.2$, straight lines fits are observed, indicating that $B_{\mathrm{sat}} \propto T^{3/8}$ and $B_c \propto T^{1/4}$, confirming the Mott VRH behavior of the conductivity. For sample $\Re = 129$, both B_{sat} and B_c follow Mott VRH behaviors between 2.8 and 4.2 K, but start to deviate for $T < 2.8$ K. We note that deviations occur in the temperature regime where the C_{sat} exhibits a sharp decrease (see Fig. 3). The sharp decrease in C_{sat} was also observed in doped CdTe semiconductors and was found to correlate with the conduction via the states in the Coulomb gap.[22] Therefore, we try to fit the zero-conductivity data below 2.8 K with a modified ES VRH law, i.e.,

$$\sigma(T) = \sigma(0) + \sigma_0 \exp[-(T_{\mathrm{ES}}/T)^{1/2}]. \qquad (11)$$

The ES VRH law considers electron-electron interactions so that the density of states near the Fermi level is $N(E) \propto E^2$ instead of a constant in the Mott VRH law.[17] The extracted value of T_{ES} is about 11 K, which is also listed in Table I.

As seen in Fig. 1, at low temperatures ($T \leqslant 2.8$ K), $\sigma(T)$ for sample $\Re = 129$ can also be fitted well by Eq. (11). The χ^2 obtained for fitting the $\sigma(T)$ data below 2.8 K to either Eqs. (9) and (11) is very close and the obtained values of $\sigma(0)$ differ by about $\pm 0.5\%$. Thus the residual $\sigma(0)$ makes it difficult to determine whether $\sigma(T)$ should follow a Mott VRH or ES VRH law just from the zero-field conductivity. Then with the value of T_{ES} we fit the MR data below 2.8 K

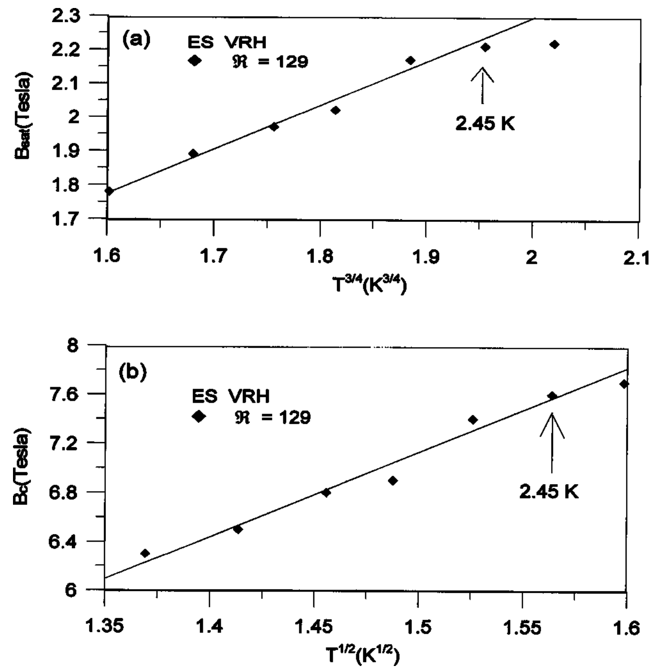

FIG. 6. (a) B_{sat} as a function of $T^{3/4}$ and (b) B_c as a function of $T^{1/2}$ for sample $\mathfrak{R}=129$ in the ES VRH regime.

with Eq. (8) for the Efros-Shklovskii (ES) VRH case. Here $(T/T_{\rm Mott})^{3/8}$ in B_{sat} is replaced by $(T/T_{\rm ES})^{3/4}$ and $(T/T_{\rm Mott})^{1/4}$ in B_c by $(T/T_{\rm ES})^{1/2}$. $\xi_C(0)$ for the ES VRH case is equal to $(T_{\rm ES}/T)^{1/2}$.

The obtained values of B_{sat} and B_c are plotted, respectively, against $T^{3/4}$ and $T^{1/2}$ between 1.8 K and ~2.8 K, and are shown in Fig. 6. It is clearly seen that B_{sat} and B_c at low temperatures do follow the ES VRH law rather than the Mott VRH law. The crossover temperature from the Mott VRH conduction to ES VRH conduction is determined to be around 2.45 K.

The values of C_{sat} are extracted from theoretical fits. C_{sat} as a function of T for samples $\mathfrak{R}=54.2$ and 129 is shown in Fig. 7. In the Mott VRH regime, the value of C_{sat} for the sample $\mathfrak{R}=129$ is much larger than that for the sample $\mathfrak{R}=54.2$, but the temperature dependence of C_{sat} in both sample is similar. For the sample $\mathfrak{R}=129$ in the ES VRH regime, the value of C_{sat} drops very rapidly with decreasing temperature and is almost equal to that for the sample $\mathfrak{R}=54.2$ at 1.9 K.

The temperature dependence of C_{sat} (or negative MR) has been studied theoretically by several authors. Schrimacher[23] and Raikh and Wessels[24] predicted a significant increase in C_{sat} with decreasing temperature in either Mott or ES VRH conduction. This apparently disagrees with the experimental data observed here. Agrinskaya, Korub, and Shamshur predicted a decrease in C_{sat} with decreasing temperature in the Coulomb-gap regime,[22] and an increase in C_{sat} in the Mott VRH regime. Their predictions are partially correct because our results (see Fig. 7) do show that in the ES VRH regime, the C_{sat} values decrease with decreasing temperatures, but so do the C_{sat} values in the Mott VRH regime, although the temperature dependence is much weaker. The inconsistence among these theoretical predictions suggests that the temperature dependence of C_{sat} (or negative MR) is still not well understood and deserves further experimental and theoretical investigations.

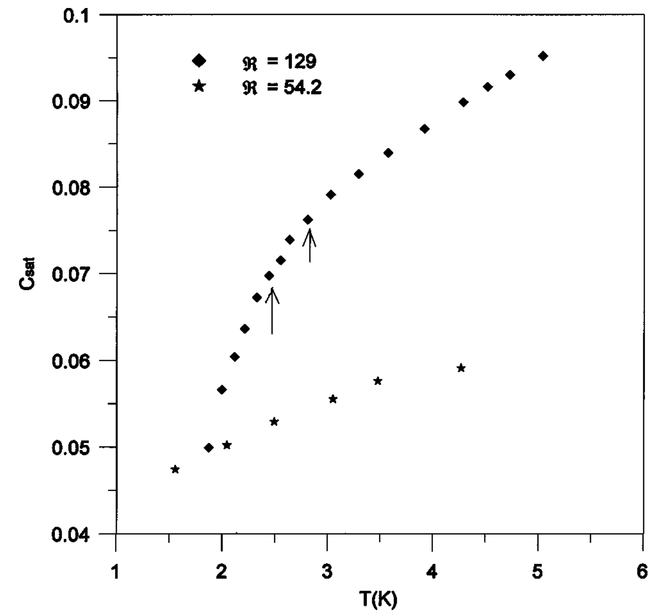

FIG. 7. Extracted values of C_{sat} as a function of temperature T for samples $\mathfrak{R}=54.2$ and 129. The crossover region is between the two arrows marked in the figure.

V. CONCLUSION

(1) We are able to explain well MR of Al-Pd-Re QC's at liquid-helium temperatures except for the MR data in the high field regime by means of the MR theories of the FI and WFS.

(2) Combining the conductivity and MR data with MR theories, we found that there is a crossover from Mott VRH conduction to ES VRH conduction at liquid-helium temperatures in the $Al_{70}Pd_{22.5}Re_{7.5}$ QC with $\mathfrak{R}=129$. This suggests the existence of the Coulomb gap at the Fermi level in highly resistive Al-Pd-Re QC's. Our results are different from the results reported by Srinivas *et al.*[25] in which the hopping conduction (1.5–8 K) in Al-Pd-Re QC's with $\mathfrak{R}\geqslant 45$ follows the ES VRH law. We found that their analysis based on the Eqs. (3) and (7) obtained in the low-field limit cannot be justified here.

(3) At low temperatures the rapid decrease in the magnitude of the negative MR in highly resistive QC's might be due to the conduction via the states in the Coulomb gap as observed in doped CdTe semiconductors.[22,26]

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