Magnetoconductivity of quasicrystals in the insulating regime

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Magnetoconductivities of icosahedral $Al_{70.5}Pd_{21}Re_{8.5}$ alloys whose zero-field conductivity $\sigma(T)\rightarrow 0$ as $T\rightarrow 0$ K [$\sigma\sim 0.1\ (\Omega\,\text{cm})^{-1}$ at 0.45 K] have been measured. At low temperature and low field, both positive and negative contributions to magnetoconductivity are prominently present. The magnetoconductivity trend is distinctly different from those seen in disordered metals and insulators. The observed features are interpreted in light of anomalous diffusion of slowly decaying electron wave functions in insulating quasicrystals.

I. INTRODUCTION

It was reported several years ago that icosahedral (i)-AlCuRu (Ref. 1) and AlCuFe (Ref. 2) alloys were marginally metallic, or in proximity to a metal-insulator transition (MIT), their low-temperature conductivities were near Mott's minimum metallic conductivity. In the same reports, magnetoconductivities of these alloys were adequately analyzed in light of quantum corrections in the weakly localized regime, yielding inelastic electron lifetimes comparable to those found in metallic glasses.[3] Thus, i-AlCuRu and i-AlCuFe alloys are on the metallic side of the MIT. More recently, it is found that i-AlPdRe alloys, presumably of the same structural class as the mentioned i alloys, have conductivities (σ) about three orders of magnitude smaller than Mott's minimum metallic conductivity at low temperature [$\sigma(0.45\ \text{K})\sim 0.1\ (\Omega\,\text{cm})^{-1}$],[4] [$\sigma(4.2\ \text{K})\sim 0.5\ (\Omega\,\text{cm})^{-1}$].[5] The conductivity trend of i-AlPdRe is also distinctly different from those of metallic i alloys in that its $\sigma\rightarrow 0$ as $T\rightarrow 0$ K with $\sigma\sim T^{\beta}$, where $0.5<\beta<1$ below ~ 3 K. This finding points to the i-AlPdRe alloys being insulators. Information about the nature of electronic states that characterizes the insulating phase of these new i alloys may be gained by performing magnetoconductivity measurements. To date, many theoretical studies have been made about electronic states in quasicrystals,[6] the discovery of insulating i-AlPdRe provides a new system for experimental study. There are already indications that the electronic states in i-AlPdRe insulators are unusual, these include the carrier density determined from specific heat and Hall-effect measurements being at least two orders of magnitude higher than those seen in doped semiconductors that are insulators, and the insulating properties being greatly enhanced with improved structural order.[4] Results from studies of optical conductivities on i-AlPdRe and other i alloys as well as crystalline alloys have given some indications that quasiperiodicity favors carrier localization.[7] It is conceivable that the electronic wave functions which lead to the insulating state in i-AlPdRe may not decay exponentially in space, in view of the appreciable overlapping of wave functions due to the high carrier density. We report in this paper magnetoconductivity measurements of i-AlPdRe. Our data will be analyzed in light of theories applicable to insulators and quasicrystals. Implications of our results for electronic states in quasicrystals will be discussed.

II. EXPERIMENTAL PROCEDURE

Ingots of $Al_{70.5}Pd_{21}Re_{8.5}$ were made by melting appropriate combinations and quantities of high-purity elements sequentially in an arc furnace, under argon atmosphere. The procedure was carried out to maximize ingot homogeneity in alloying aluminum with high melting point refractory metals.[4] Samples cut from ingots were first annealed under highly purified conditions at 950 °C for 12 h and then at 600 °C for 2 h to obtain high-quality i phase with the aforementioned low conductivity trend, as described elsewhere.[4] Bar-shaped samples, in dimensions of $\sim 1\ \text{mm}\times 1.5\ \text{mm}\times 5\ \text{mm}$, were used in magnetoconductivity measurements in fields up to 5 T and over the temperature 0.45–20 K. Measurements were carried out both by ramping up and ramping down the field. High quality i-phase samples obtained by heat treating ingots only at 950 °C for 12 h, which showed insulating behavior in their conductivity trend but with a much higher conductivity [$\sigma(0.45\ \text{K})\sim 1\ (\Omega\,\text{cm})^{-1}$], were also measured. Thermal relaxation effects on conductivities of samples annealed under the given conditions, will be discussed elsewhere.

III. RESULTS AND DISCUSSION

Magnetoconductivity (MC) of the twice-annealed (950 °C/600 °C) i-AlPdRe samples, presented in the form $\Delta\sigma=\sigma(H,T)-\sigma(0,T)$ versus H, are shown in Fig. 1. Magnified data for $T=0.45$–2.0 K taken below 0.4 T are depicted in Fig. 2. Samples that were heat treated only at 905 °C show a similar trend, but the magnitude of $\Delta\sigma/\sigma(0,T)$ is noticeably different, as illustrated in Fig. 3. From the data it is clear that the trend in $\Delta\sigma$ of i-AlPdRe is prominently different from those of previous i alloys,[1,2,8] and it is also unknown in the metallic alloys studied.[3] The overall trend shown in Fig. 1 together with the low-field behavior detailed in Fig. 2 indicates there are two contributions to MC, one positive and one negative, which play competing roles at low T and small H.

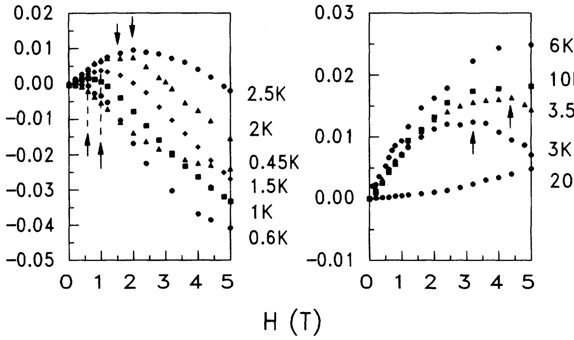

FIG. 1. Isothermal magnetoconductivity for $Al_{70.5}Pd_{21}Re_{8.5}$ annealed at 950 °C for 12 h followed by an additional anneal at 600 °C for 2 h. Arrows indicate the "crossover field" where $\Delta\sigma$ is a maximum. The graph is split into two parts, low-temperature portion, and high-temperature portion, for greater clarity.

Moreover, $\Delta\sigma$ is noticed to have nonlinear dependence on H at low H (Fig. 2). Competition between the two contributions is most conspicuous below 4.2 K in fields up to 5 T, as witnessed in the change of sign in $d(\Delta\sigma)/dH$, and at $T<3$ K the change of sign in $\Delta\sigma$. The maximum in $\Delta\sigma$ shifts to a higher field as the temperature is increased. Although in principle, $\sigma(T)/\sigma(0)\gg 1$ at low T would indicate the nonapplicability of weak-localization theory, one may contend that since the magnitude of $\Delta\sigma/\sigma(0,T)<1$ quantum corrections may still be relevant as far as MC is concerned. Therefore, we also attempted to fit the data to contributions due to quantum interference and various electron-electron-interaction effects in the metallic regime, by considering a wide range of electron-dephasing times including those obtained for previous i alloys[1,2,8] and metallic glasses.[3] In addition, different signs of the electron-electron-interaction parameters were also attempted. Our effort was found to be of total futility. The fits had failed to account for the robust features in the $\Delta\sigma$ trend,

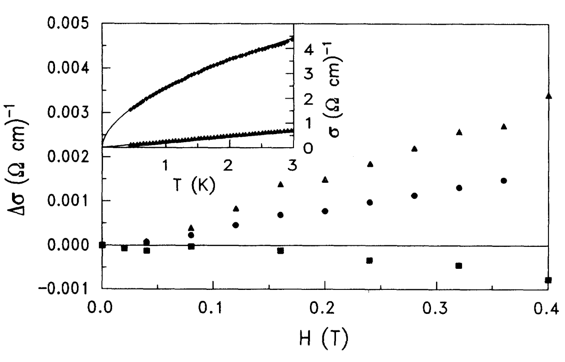

FIG. 2. Low-field magnetoconductivity data for $Al_{70.5}Pd_{21}Re_{8.5}$ annealed at 950 °C for 12 h followed by an additional anneal at 600 °C for 2 h, taken at 0.45 K (■), 1 K (●), and 2K (▲). Inset: Low-temperature σ for $Al_{70.5}Pd_{21}Re_{8.5}$ annealed at 950 °C for 12 h $\sigma(0.45\ K)=1.5\ (\Omega\,cm)^{-1}$, $\sigma\sim T^{0.51}$ (upper curve), and for samples with additional annealing at 600 °C for 2 h $\sigma(0.45\ K)=0.12\ (\Omega\,cm)^{-1}$, $\sigma\sim T$ (lower curve). Lines are drawn to show the indicated power-law dependence.

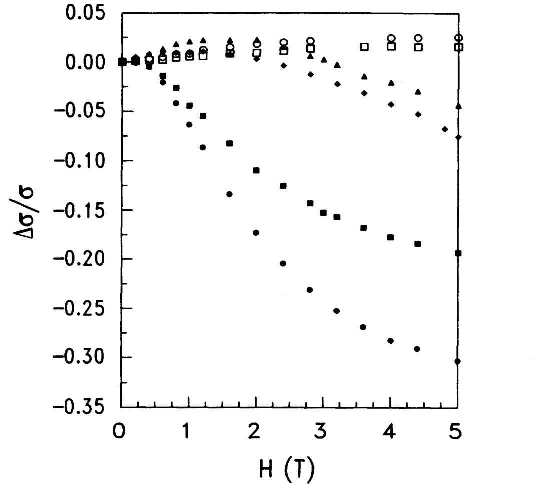

FIG. 3. $\Delta\sigma/\sigma(0,T)$ for $Al_{70.5}Pd_{21}Re_{8.5}$ annealed at 950 °C for 12 h taken at 0.45 K (■), 2 K (♦), and 6 K (□); and for samples with an additional anneal at 600 °C for 2 h taken at 0.45 K (●), 2 K (▲), and 6 K (○).

particularly in the rapid change of curvature it exhibits.

In view of the insulating phase in a metal-based quasicrystal, perhaps some new physics can be gleaned from the new data on the i phase by comparing them with MC of metals and insulators. In disordered metals, the contribution from self-crossing diffusion paths (returning loops) of electrons, gives rise to weakly localized behavior. On the other hand, the presence of spin-orbit scattering in the system, which has a dephasing effect on the quantum interference mentioned, can lead to antilocalization behavior which enhances conductivity. It follows that negative MC can be expected once spin-orbit scattering is mitigated by an applied field, as observed.[8] For insulating materials, a new mechanism of MC has been proposed.[9] In the variable range hopping (VRH) regime, MC is believed to be dominated by the quantum interference between different Feynman paths along which spatial overlap between pairs of sites may occur, a phenomenon known as "oriented-path interference." Calculations based on the new model suggest that MC is positive in the VRH regime due to the dephasing of those Feynman paths, which, in the absence of a magnetic field, interfere destructively to give zero overlap between sites. It is also expected that oriented-path interference is insensitive to spin-orbit scattering due to the small momentum transfer involved. Meanwhile, the returning-loop effect, proposed for the metallic regime, which can give rise to negative MC in the presence of spin-orbit scattering, is negligible when the magnetic length $L_H\gg$ the localization length ξ in the VRH regime. Another criterion, r (hopping length) $\gg\xi$, is also known for the negligibility of the returning-loop effect.[10] Based on these two criteria, negative MC would not be favored in insulators. The issue of observing negative MC in the insulating regime remains controversial on both experimental and theoretical fronts,[11] but it is in general agreed that directed-path interference is important in insulators. The insulating state of i-AlPdRe is peculiar in the sense that despite its diffusion constant $D\sim 0.001\ cm^2\,s^{-1}$ [obtained from $D \sim \sigma / N(E_f)$] being smaller than those found in some insulating doped semiconductors with comparable σ and smaller carrier density, it is not in the VRH regime.[4] The latter point can be seen from the nonpositive curvature of $\sigma(T)$ (inset, Fig. 2). However, this peculiarity may be explained by the nonexponential decay of wave functions mentioned in Ref. 4. Indeed, there has been computational work that shows power-law decay wave functions in model two-dimensional (2D) quasicrystals.[6,12] It is further proposed in one of the theories that, due to the peculiar nature of the wave functions, the group velocity of carriers will tend to zero for large spatial extent.[12] Thus, rather than the hopping mechanism of transport, quasicrystals conduct by the anomalous diffusion of carriers. Both conduction mechanisms result in $\sigma \rightarrow 0$ and $T \rightarrow 0$. To underline another difference between the two types of insulators, we compare the isothermal plots of $\Delta\sigma/\sigma$ versus H for the two groups of *i*-AlPdRe samples mentioned, whose low-T σ values differ by one order of magnitude (Fig. 3). It is inferred from these plots that both positive and negative components of $\Delta\sigma/\sigma$ for the low-σ samples increase quite substantially relative to the high-σ samples. In comparison, $\Delta\sigma/\sigma$ in VRH is scaled to some positive power of ξ[9] and it is thus expected to decrease with decreasing σ.

Although it is not clear how one may apply the model of positive MC in the VRH regime to *i*-AlPdRe, the insulating transport behavior noted at low T must render some kind of forward-path interference effect, but not necessarily that of directed path as in VRH, quite favorable in quasicrystals. However, in contrast to VRH insulators, the relatively large spatial extent of nonexponential wave functions in quasicrystals suggests that one should also consider the returning-loop effect and thus spin-orbit scattering effect on MC, which gives rise to negative MC. High spin-orbit scattering rates have been referred to in *i* alloys.[1,2,8] In light of this picture, one can explain qualitatively the MC behavior observed in *i*-AlPdRe, based on the competition of positive and negative MC's. The change of sign in $d(\Delta\sigma)/dH$ then simply reflects the growing importance of the returning-loop effect at increasing field. The "crossover field" H_c, which marks the position of the maximum in $\Delta\sigma$ is noted in Fig. 1. H_c can be seen to increase with T. This trend may be understood in terms of the interplay of magnetic length and the phase-coherence length L of power-law wave functions, where L is the characteristic length scale in the anomalous diffusion model.[12] Within the model, L is the length size of the "unit cell" inside which the wave functions are localized, and anomalous diffusion of carriers leads to $\sigma \sim L^{\alpha}$ with $\alpha < 0$. At $T=0$, $L \rightarrow \infty$, and L decreases as T increases, which gives rise to increasing σ. Thus, the returning-loop effect is important inside L, and at increasing T the spatial extent ($\sim L$) of the effect diminishes. Meanwhile, if L_H is not much larger than L, the spin-orbit scattering effect will give rise to negative MC. Assuming that H_c is qualitatively given by the condition $L_{H_c} \simeq sL$ (s is of order unity), then L_{H_c} decreases as T increases. As a result, one finds the crossover field H_c to increase with T since $L_H \sim H^{-1/2}$. Extrapolating the proposed scenario further, one may even derive the temperature dependence of L from the data to be $L \sim T^{-0.8}$, which yields a conductivity exponent $\alpha \sim -1.2$, since $\sigma \sim T$ below 3 K for the more insulating *i*-AlPdRe samples. The phase-coherence length at 1 K is estimated to be of the order of 400 Å. But much of these are too speculative for our present goal which is to feature unusual magnetoconductivity data from insulating *i* alloys. Clearly, quantum interference effects in quasicrystals need to be investigated. So far, a preliminary study has been made for a 2D Penrose lattice.[13]

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[1] B. D. Biggs, S. J. Poon, and N. R. Munirathnam, Phys. Rev. Lett. **65**, 2700 (1990).

[2] T. Klein, C. Berger, D. Mayou, and F. Cyrot-Lackmann, Phys. Rev. Lett. **66**, 2907 (1991); T. Klein, H. Rakoto, C. Berger, G. Fourcaudot, and F. Cyrot-Lackmann, Phys. Rev. B **45**, 2046 (1992).

[3] P. A. Lee, and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985); M. A. Howson and B. L. Gallagher, Phys. Rep. **170**, 265 (1988).

[4] F. S. Pierce, S. J. Poon, and Q. Guo, Science **261**, 737 (1993); F. S. Pierce, Q. Guo, and S. J. Poon, Phys. Rev. Lett. **73**, 2220 (1994).

[5] H. Akiyama, Y. Honda, T. Hashimoto, K. Edagawa, and S. Takeuchi, Jpn. J. Appl. Phys. **32**, L1003 (1993) [σ(4.2 K)$\sim$10 (Ω cm)$^{-1}$]; Y. Honda, K. Edagawa, A. Yoshika, T. Hashimoto, and S. Takeuchi, *ibid.* **33**, 4929 (1994) [σ(4.2 K)$\sim$0.5 (Ω cm)$^{-2}$].

[6] T. Fujiwara and H. Tsunetsugu, in *Quasicrystals: The State of the Art,* edited by D. P. DiVincenzo and P. J. Steinhardt (World Scientific, Singapore, 1991), p. 343; H. Tsunetsugu, T. Fujiwara, K. Ueda, and T. Tokihiro, Phys. Rev. B **43**, 8879 (1991), and references cited therein.

[7] D. N. Basov, T. Timusk, F. Barakat, J. Greedan, and B. Grushko, Phys. Rev. Lett. **72**, 1937 (1994); D. N. Basov, F. S. Pierce, P. Volkov, S. J. Poon, and T. Timusk, Phys. Rev. Lett. **73**, 1865 (1994).

[8] A. Sahnoune, J. O. Strom-Olsen, and A. Zaluska, Phys. Rev. B **46**, 10 629 (1992); A. Sahnoune, J. O. Strom-Olsen, Z. Altounian, C. C. Homes, T. Timusk, and X. Wu, J. Non-Cryst. Solids **153&154**, 343 (1993); R. Haberkern, G. Fritsch, and J. Schilling, Z. Phys. B **92**, 383 (1993).

[9] For a review, see B. I. Shklovskii and B. Z. Spivak, in *Hopping Transport in Solids,* edited by M. Pollak and B. I. Shklovskii (North-Holland, Amsterdam, 1991), p. 271.

[10] Y. Shapir and Z. Ovadyahu, Phys. Rev. B **40**, 12 441 (1989).

[11] Y. Meir and O. Entin-Wohlman, Phys. Rev. Lett. **70**, 1988 (1993), and references cited therein.

[12] C. Sire, B. Passaro, and V. Benza, J. Non-Cryst. Solids **153&154**, 420 (1993); (private communication).

[13] F. Nori and Q. Niu, in *Quasicrystals and Incommensurate Structures in Condensed Matter, Proceedings of the Third International Meeting on Quasicrystals,* edited by M. Jose Yacaman, D. Romeu, V. Castano, and A. Gomez (World Scientific, Singapore, 1990), p. 434.