PHYSICAL REVIEW B VOLUME 41, NUMBER 13 1 MAY 1990

Anisotropic transport properties of a stable two-dimensional quasicrystal: $Al_{62}Si_3Cu_{20}Co_{15}$

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(Received 7 December 1989; revised manuscript received 16 January 1990)

The anisotropic nature of the transport properties in a two-dimensional quasicrystal has been observed for the first time. The resistivity depends linearly on temperature from 80 K up to 400 K for both the crystalline and quasicrystalline directions but with opposite temperature coefficients. The thermopower is positive in the crystalline direction and negative in the quasicrystalline plane. The results are discussed in connection with the anisotropy of band structure, scattering characteristics, and defects formation.

The surprising discovery of the quasicrystalline (QC) phase in a rapidly solidified Al-Mn alloy[1] has stimulated a great deal of efforts to investigate the properties of the new phase, though most of the work was devoted to the structural nature of the material. There were some measurements on transport properties,[2] but the conclusion about the relationship between the QC symmetry and the observed results became uncertain owing to the random QC orientations of the microdomains and the defects vastly existed in quenched samples. Recently, a mostly single icosahedral (I) phase was obtained in a thermodynamically stable state,[3] and the transport properties of some I phase were found to be distinctly different from those of glassy alloys, but similar to those of crystalline compounds.[4] Here we report our measurements of the anisotropic resistivity and thermopower on a new stable decagonal QC which is periodic in the tenfold direction and quasiperiodic in the plane perpendicular to it. As the new two-dimensional (2D) QC was obtained in almost a single domain state with a periodicity of about 0.8 nm along the tenfold axis, the present work provides the first experiment to compare the transport properties in a QC plane with that in the periodical direction in the same QC.

In an earlier work,[5] the $Al_{65}Cu_{20}Co_{15}$ decagonal QC was formed by slow solidification and the high degree of perfection of the QC was shown by the presence of many more diffraction spots in the electron-diffraction pattern than those for Al-Mn or Al-Fe QC, as well as by the sharp convergent-beam electron-diffraction pattern. Recently, QC of even better quality was obtained by substituting a little Si for Al ($Al_{62}Si_3Cu_{20}Co_{15}$).[6] The samples were found to be well formed prisms with a tenfold rotationary symmetry and proved to be of almost single domain over the whole sample which usually had a few tenths mm in diameter and several mm in length. The anisotropic resistivity was measured by both the standard four-leads method and the Montgomery method with measuring current working at 37 Hz. For one sample the results were checked by dc measurements. All the measurements gave consistent results. The thermopower of the samples was measured by a setup described in Ref. 7.

The anisotropic resistivity calculated by the Montgomery method for sample 1 is presented in Fig. 1. We see the positive temperature coefficient along the crystalline direction $\alpha_C > 0$ and the negative α_{QC} in the QC plane. The same results were obtained for all the measured samples. The anisotropy, i.e., the ratio of resistivity along the QC plane ρ_{QC} to that along the tenfold axis ρ_C, was in the range of 2–4, varying from sample to sample. It seemed to have a tendency that the lower the ρ_C, the higher the anisotropy, implying rather easier removal of defects in crystalline direction than in the QC direction, in agreement with the different growth philosophies of the crystalline and QC structures.[2] It is worth noting that, in spite of the sample and direction-dependent temperature coefficient of resistivity [$\alpha = (1/\rho)d\rho/dT$], the anisotropic band structure and the anisotropy in carrier properties shown by thermopower measurement reported here, as

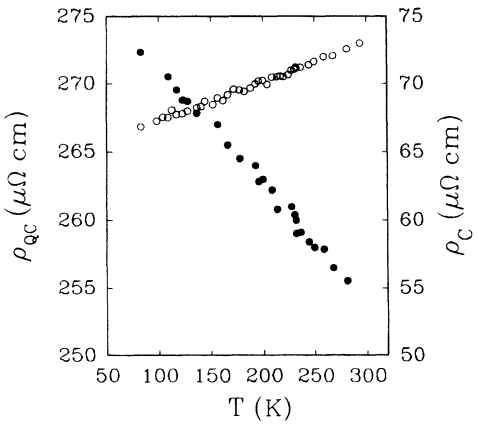

FIG. 1. Anisotropic resistivity and its T dependence of a thermodynamically stable $Al_{62}Si_3Cu_{20}Co_{15}$ 2D quasicrystal.

well as by Hall effects,[8] the Mooij correlation[9] roughly holds for all the data of our samples (Fig. 2). The continuous crossover of α from a positive to a negative value could indicate that α is insensitive to the structural difference of QC and crystalline phases, and depends only on the disorder of a given sample at a given direction. We may also plot the absolute temperature coefficient of resistivity $\alpha_a = d\rho/dT$ against ρ_0, the resistivity extrapolated to $T=0$. The results are shown in Fig. 3. We see that α_a is approximately independent of ρ_0 in the positive range of α_a, which means the contributions of electron-phonon scattering and static disorder are roughly additive for the periodic direction. But when α_a becomes negative, it depends markedly on ρ_0, implying the breakdown of Matthiessen's rule. That α_a could be so scattered without the loss of QC symmetry seems to support Sokoloff's conclusion, i.e., the scattering of electrons by the potential due to the almost-periodic lattice proposed for QC does not contribute any resistivity, and the observed resistivity can be accounted for by defects in the perfect Penrose tiling.[10]

However, if the defects are the dominant factor deciding the resistivity and its T dependence, we should have observed some crossover in between the positive and negative α_a either by changing the disorder or by sweeping the temperature. The Mooij correlation is often accounted for by the competition of classical Boltzmann transport and the effects stemming from incipient localization of electrons: quantum interference effect (QIE), phonon-assisted tunneling, etc.[11] For a given metallic system, a specific mechanism manifests itself in a definite range of temperature and disorder, showing a characteristic temperature dependence of resistivity.[11] But for the present stable decagonal QC, we failed to find any trace of such transition. Instead, in all measured temperature range the resistivity keeps a satisfactory linear dependence on temperature for both positive and negative α.[12] We had extended the measurements up to 400 K for one sample, but we still could not find any tendency to saturation, though the temperature had, probably, surpassed the Debye temperature.[13] The failure to find a crossover region then seems in favor of a picture that the difference in α_a is intrinsic to this anisotropic material. The phenomenon certainly needs to be further studied.

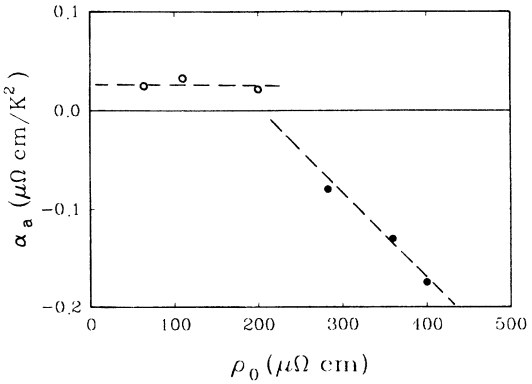

FIG. 3. The absolute temperature coefficient of resistivity tends to become a sample-independent value with decreasing disorder; ○, periodic direction; ●, QC plane.

While the vast amount of defects in QC complicate the analysis of resistivity data, they certainly simplify the treatment of thermopower because we do not worry about the contribution of phonon drag. Figure 4 shows the results for one of our samples. The thermopower S was measured for several samples which gave similar results. For both QC and crystalline directions, $|S|$ is only a few μV per degree at room temperature, and is roughly proportional to temperature, just as for amorphous transition metal alloys.[11] A striking contrast is the opposite sign of S_C, the thermopower along tenfold axis, and S_{QC}, the thermopower in the QC plane. S can be calculated by the Mott formula

$$S = -\frac{\pi k_B^2 T}{3|e|E_F}\xi, \quad \xi = \left.\frac{d\ln\sigma(E)}{d\ln E}\right|_{E=E_F}.$$

The contribution to ξ can be separated to a term depending on band structure and a term related to scattering

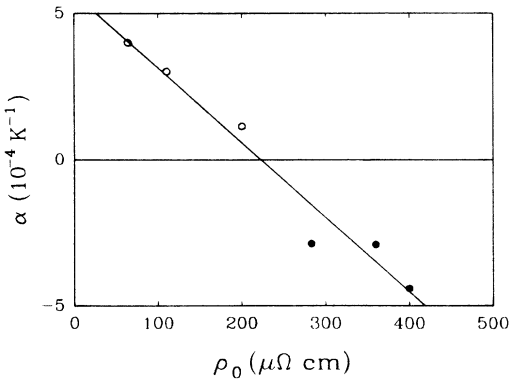

FIG. 2. The measurement of resistivity for several samples show a tendency obeying the Mooij correlation. ○, periodic direction; ●, QC plane.

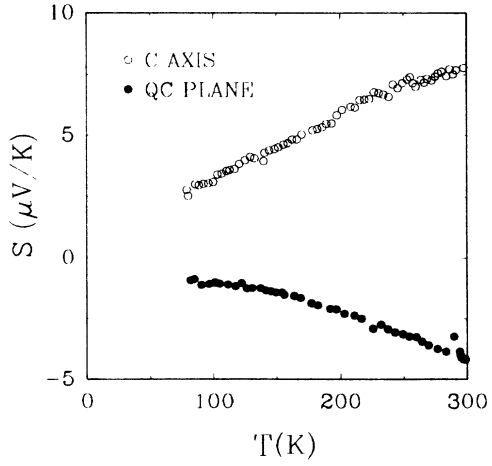

FIG. 4. Anisotropic thermopower of the $Al_{62}Si_3Cu_{20}Co_{15}$ 2D quasicrystal.

mechanisms. We feel that the difference in the sign of S_C and S_{QC} is probably determined by the band term rather than by the scattering term. The scattering term for nonmagnetic samples[14] is often discussed in the Ziman-Faber model or the Mott *s-d* scattering model. The application of the Ziman-Faber diffraction model to highly disordered alloys is questionable[15] and this model requires the sign of S to be opposite to α, in contradiction to our results. If the *s-d* scattering dominates, because Co is a late transition metal and the energy derivative of the density of states (DOS) of *d* band at E_F is negative,[16] we expect $\xi > 0$ and $S < 0$ in both directions because the disorder makes the scattering nearly isotropic.[11] It is possible that some anisotropic spin flip contributes to the scattering term, which for the elements as Fe, Ni, Co, etc., makes the thermopower negative in the QC plane.[17] However, like other scattering terms, this term should be sample dependent, which has not been proved in our experiments. Thus, the anisotropy of thermopower is possibly caused by the anisotropy of band structure.

It is interesting to note that the deviation from linear T dependence of thermopower for the present 2D QC sample can be fairly accounted for by the electron-phonon enhancement effect.[11] Following Kaiser,[18] the effect can be expressed by

$$\frac{S}{T} = \frac{S_0}{T}[1+\lambda(T)] + 2\alpha\lambda(T) ,$$

where S_0 is the bare thermopower, $\lambda(T)$ is the electron-phonon mass enhancement parameter, and $2\alpha\lambda$ comes from the velocity and relaxation-time renormalization. What is special here is the following: While the electron-phonon enhancement effect along the crystalline direction is similar to that often observed in amorphous transition metals,[11] the effect along the QC plane is rather unusual (Fig. 5). A possible explanation, by comparing the two curves in Fig. 5, consists in the contribution of $2\alpha\lambda$. From Kaiser's expression we see that the $2\alpha\lambda$ term is additive to the mass enhancement effect when S_0 is positive, but subtractive when S_0 is negative. The curve for the QC plane implies the dominant role of velocity and relaxation-time enhancement at high temperatures and the tendency of a takeover by mass enhancement at lower temperatures.[19] If the explanation is reasonable, the present results provide the first example to show the existence of velocity and relaxation time enhancement effect.

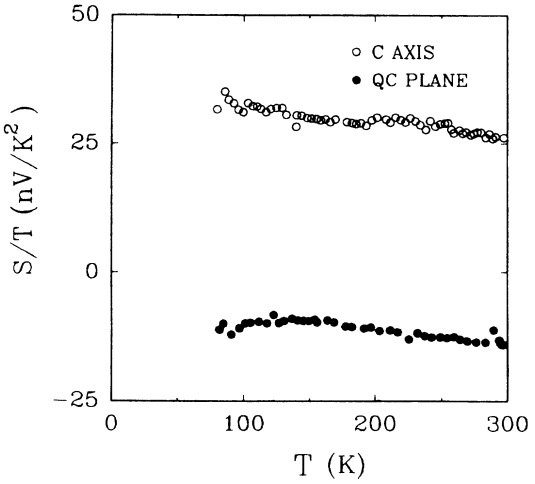

FIG. 5. Anisotropic behavior of S/T, showing possibly the effect of mass enhancement as well as the velocity and relaxation-time renormalization of electrons due to electron-phonon interaction.

In summary, we have shown, for the first time, the striking anisotropic transport properties of a stable 2D quasicrystal. Whether this anisotropy originates from the difference in QC and crystalline band structure still needs to be further clarified.

The authors thank Professor K. K. Fung for his interest in this work.

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[12]Very recently, when we measured the magnetoresistance, we found the linearity to be held to ~40 K below which the resistivity became T independent for both directions.

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[14]Preliminary measurement shows that the material is paramagnetic above ~80 K. However, there may be some 2D magnetism in the QC plane as shown by the Hall effect and magnetoresistance. See Ref. 8, and L. X. He *et al.* (unpublished).

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