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1997 J. Phys.: Condens. Matter 9 6643

(http://iopscience.iop.org/0953-8984/9/31/016)

J. Phys.: Condens. Matter **9** (1997) 6643–6650. Printed in the UK PII: S0953-8984(97)76820-6

The low-temperature resistivity and magnetoresistance of icosahedral quasicrystalline Al–Cu–Cr

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Received 31 July 1996, in final form 5 June 1997

Abstract. The temperature and magnetic field dependence of the resistivity of quasicrystalline $Al_{65}Cu_{20}Cr_{15}$ alloy in the temperature range 1.7 K to 115 K and for fields up to 6.5 T were measured. While the resistivity ($\rho(T)$) has a maximum at $T \simeq 45$ K, it decreases with increasing T for $T > 45$ K, as for other icosahedral quasicrystals. However, for $T < 45$ K, ρ decreases with decreasing T. The magnetoresistance (MR) is positive over the whole temperature range. We have tried to explain our observation using existing theories. The low-temperature resistivity $\rho(T)$ ($T < 45$ K), the resistivity maxima at $T \simeq 45$ K, and the positive MR can be explained by invoking antilocalization effects due to strong spin–orbit interactions. We have also tried to explore other approaches which give rise to positive MR.

1. Introduction

There is currently much interest in achieving an understanding of the electronic properties of icosahedral quasicrystalline material [1, 2]. This interest generally arises from the fact that stable icosahedral phases show electronic transport properties qualitatively different from any known so far for alloys. Quasicrystals have anomalously high electrical resistivity (ρ), and on annealing their resistivities increase. The resistivity $\rho(T)$ also has a unique temperature dependence. For $T > 50$ K, for most quasicrystals ρ decreases linearly as T increases. These are believed to be intrinsic properties of quasicrystals, and are believed to be due to either or both of the two characteristic features of the electronic structure: (1) the existence of a pseudogap in the density of states (DOS) at the Fermi energy E_F due to the Fermi surface–Brillouin zone boundary interaction; and (2) the tendency towards localization of the wave function of the electronic state near the pseudogap.

In a recent paper [3], we reported extensive structural and electrical transport studies on the icosahedral quasicrystal $Al_{65}Cu_{20}Cr_{15}$. In this paper we report on the low-temperature transport properties, in particular the resistivity $\rho(T)$ and magnetoresistance (MR) of the same quasicrystal below 100 K. The peculiar behaviour of the electronic transport properties at $T < 45$ K in icosahedral quasicrystals in general is discussed below. Low-temperature MR is a very useful probe for distinguishing electronic scattering processes. In the present work we have measured the MR in the temperature range 1.7 K to 115 K in fields up to 6.5 T. We have combined our MR data with the zero-field resistivity data, and have tried to see whether a self-consistent explanation can be given for both sets of data. It is interesting to

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note that the MR reported for icosahedral $Al_{63}Cu_{25}Fe_{12}$ by Klein *et al* [4, 5] was negative for $T \geqslant 30$ K, whereas the MR reported by Sahnoune *et al* [6] and Matsuo *et al* [7] was observed to be positive for $T \geqslant 30$ K, and in icosahedral $Al_{65}Cu_{20}Ru_{15}$ the MR was positive [8]. As we will see, in our case the MR is positive over the entire range of temperature and field studied by us. We have shown that it is possible to link this positive MR to the peculiar behaviour of the electronic conductivity for $T < 45$ K.

The experimental details of the alloy preparation, with structural and microstructural characterization, have been reported earlier [3]. The resistances of the alloy strips were measured by an a.c. (∼20 Hz) four-probe method [9]. The leads to the sample were attached with silver paste. The measuring current was ∼1 mA. The precision of the measurement was ∼±20 ppm, whereas the absolute accuracy limited by the sample geometry is ∼15%.

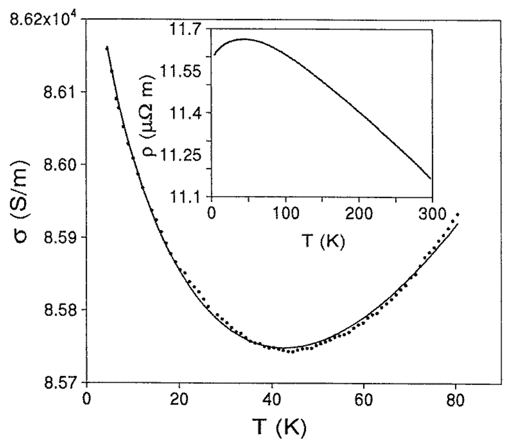

Figure 1. The conductivity σ of $Al_{65}Cu_{20}Cr_{15}$ quasicrystal as a function of the temperature. The solid line is the fit described in the text. Inset: the resistivity versus the temperature.

2. Results and discussion

In figure 1 we show the temperature dependence of the conductivity for $T \leqslant 80$ K (the solid line in figure 1 is a fit which we discuss below). In the inset of figure 1 we have shown the temperature dependence of the resistivity up to 300 K. In this paper we will attempt to achieve an understanding of the occurrence of a positive TCR in the low-temperature region ($T < 45$ K). It should be pointed out that for all good quality icosahedral quasicrystals, one observes a negative TCR for $T > 45$ K, but the behaviour below 45 K is not so clear-cut. For instance, for much-studied Al–Cu–Fe, for all of the alloy compositions studied, the TCR is negative at $T > 45$ K, but the TCR below 45 K can be positive or negative depending on the composition [6]. Even for those alloys for which the TCR remains negative below 45 K, there is a distinct change in the temperature dependence of ρ below 20–30 K. One of the explanations for the negative TCR for the quasicrystals uses the idea of quantum corrections to the conductivity (weak-localization and electron–electron interactions) [10, 11]. Whether such ideas can be applied to quasicrystals is debatable. But if we assume that such ideas are valid, then one can explain the negative TCR over the whole temperature range, including the region $T < 45$ K. A problem arises, however, with alloys which show a positive TCR at low temperatures and a negative TCR at higher temperatures. We show here that, staying within the framework of weak-localization ideas, it is possible to explain this behaviour. We substantiate our claim through magnetoresistance (MR) measurements, which we will discuss below. We show that the decrease in resistivity for $T < 45$ K causing a resistivity maximum at $T \sim 50$ K can be attributed to antilocalization effects caused by spin–orbit scattering [12]. The antilocalization effects can be destroyed by applying a magnetic field, giving rise to positive MR. This is in contrast to more common weak-localization effects, where one observes a rise in the resistivity with decrease in temperature [12], and this effect can be destroyed by application of a magnetic field, giving rise to negative MR.

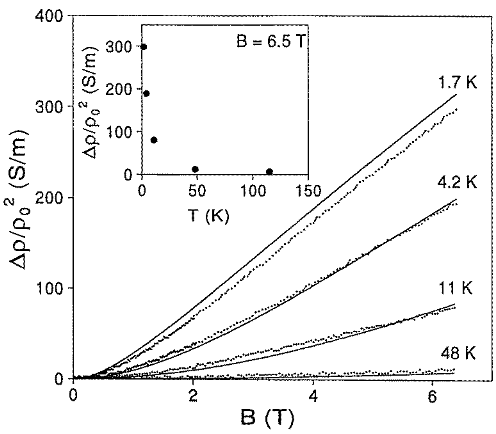

Figure 2. The magnetoresistance ($\Delta\rho/\rho_0{}^2 = -\Delta\sigma$) of $Al_{65}Cu_{20}Cr_{15}$ quasicrystal as a function of the magnetic field. The solid lines represent the fit described in the text. The inset shows the variation of the MR as a function of temperature at 6.5 T.

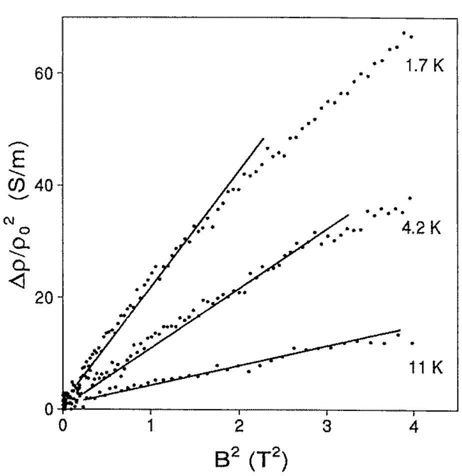

Figure 3. $\Delta\rho/\rho_0{}^2$ versus B^2 for $Al_{65}Cu_{20}Cr_{15}$ quasicrystal. The solid lines are a guide to the B^2-dependence at low field.

Longitudinal MR data, $\Delta\rho/\rho_0$ versus B ($\Delta\rho/\rho_0 = (\rho(B,T) - \rho(0,T))/\rho(0,T)$), measured up to 6.5 T and at different temperatures from 1.7 K to 115 K, are shown in figure 2 (the solid lines through the data are derived from a fit which will be discussed below; we have plotted the data as $\Delta\rho/\rho_0{}^2$ which is $=-\Delta\sigma$.) In figure 2 we have not shown the data obtained at 115 K, for clarity. The MR at 115 K for fields up to 6.5 T was positive. In the inset of figure 2, the MR at 6.5 T as a function of temperature is shown. It can be seen that the MR of the sample is positive over the whole range of temperature and field. For all temperatures, we observe the MR varying as B^2 for low fields, and at higher fields the variation is slower than B^2. The magnetoresistance continues to increase for fields up to 6.5 T, without saturation (see figure 3). Generally, in the presence of spin–orbit interaction, the MR is often positive at low field and then becomes negative at higher field, after passing through a maximum [12]. The position of the peak depends on the strength of the spin–orbit interaction. Even if the maximum occurs in the MR of our sample because of the spin–orbit interaction, the maximum will be at very high field. We discuss this particular aspect below.

In the following, we will explore whether both the conductivity, $\sigma(T)$, and magnetoconductivity data can be self-consistently explained using the idea of anti-localization. Fukuyama and Hoshino have calculated the quantum correction to the conductivity in the presence of both spin–orbit interaction and inelastic scattering. Using an expression for the conductivity given by Fukuyama and Hoshino [13], Matsuo *et al* [7] have obtained an expression for the change in conductivity $\delta\sigma(T)$ (see equation (1) in reference [7]).

In their expression, the four parameters ($A = \sqrt{3}e^2/(2\pi^2\hbar v_f\sqrt{\tau_\epsilon\tau_{so}})$, $t = \tau_{so}/4\tau_i$, $t_1 = 3\tau_\epsilon/\tau_{so}$, and $t_2 = 3\tau_\epsilon/\tau_i$) are related to the three characteristic relaxation times τ_i (inelastic), τ_{so} (spin–orbit) and τ_ϵ (elastic). We are working in the regime where $\tau_\epsilon \ll \tau_{so}, \tau_i$. As a result, both t_1 and $t_2 \ll 1$. One can therefore see that the main temperature dependence arises from the first term and the fourth term of their expression for $\delta\sigma(T)$. The other terms are essentially constants. The temperature dependence arises because of terms like $\sim 1/(v_f\sqrt{\tau_i\tau_\epsilon})$. This varies as $\sim 1/L_\phi$ and gives the requisite temperature dependence to $\delta\sigma(T)$. Whether $\delta\sigma(T)$ is positive or negative depends on the total contribution arising from these two terms. When the spin–orbit scattering is very weak, such that $\tau_{so}{}^{-1} \ll \tau_i{}^{-1}$, and $t_1 \ll t_2$, the fourth term in the equation varies as $(3\pi/2)\sqrt{\tau_{so}/\tau_i}$. When the spin–orbit scattering is very strong, such that $\tau_{so}{}^{-1} \gg \tau_i{}^{-1}$, $t_1 \gg t_2$, this term reduces to a constant. The first term always varies as $\sim -(\pi/2)\sqrt{\tau_{so}/\tau_i}$, irrespectively of the relative values of t_1 and t_2 as long as $t_2 \ll 1$. One can therefore see that when $\tau_{so}{}^{-1} \ll \tau_i{}^{-1}$, the dominant temperature-dependent part of $\delta\sigma(T)$ is approximately equal to

$$(A/\pi)(\pi\sqrt{\tau_{so}/\tau_i}) \approx \text{constant} \times \frac{e^2}{\hbar v_f\sqrt{\tau_\epsilon\tau_i}} \approx \text{constant} \times \left[\frac{e^2}{\hbar L_\phi}\right].$$

Thus the temperature dependence of σ has a positive coefficient in the absence of strong spin–orbit scattering. However, when $\tau_{so}{}^{-1} \gg \tau_i{}^{-1}$, the temperature-dependent part of $\delta\sigma(T)$ is approximately equal to

$$(A/\pi)(-\pi/2)\sqrt{\tau_{so}/\tau_i} \approx \text{constant} \times \left[\frac{-e^2}{\hbar L_\phi}\right].$$

In this case, σ has a negative temperature coefficient.

The changeover of σ from a negative temperature coefficient at low temperature ($T < 40$ K) to a positive temperature coefficient at higher temperature ($T > 50$ K) therefore signifies a crossover from a spin–orbit-scattering-dominated ($\tau_{so}{}^{-1} \gg \tau_i{}^{-1}$) region at low temperature to an inelastic-scattering-dominated ($\tau_i{}^{-1} \gg \tau_{so}{}^{-1}$) region at higher temperatures. This is explained schematically in figure 4. With this understanding, we now fit the complete equation (equation (1) of reference [7]) to our data. We obtain the following

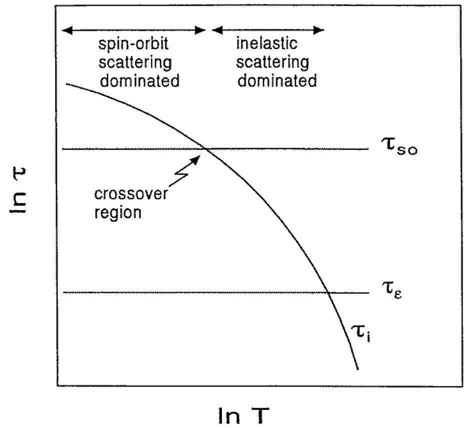

Figure 4. A schematic diagram showing the crossover from a spin–orbit-scattering-dominated region to an inelastic-scattering-dominated region. (τ_{so} (the spin–orbit scattering time) and τ_ϵ (the elastic scattering time) are temperature independent, and τ_i (the inelastic scattering time) is temperature dependent.)

fitting parameters : $v_f \tau_\epsilon = 1.8$ Å, $\tau_\epsilon/\tau_{so} = 4.45 \times 10^{-4}$, $\tau_i/\tau_{so} = (1/4)(180/T)^{1.38}$ (T is in degrees Kelvin), and $e^2 DN(E_F) \approx 1.89 \times 10^5$ S m^{-1}. With these values of the parameters, we will now see whether the MR data can be explained.

The MR ($\Delta\rho/{\rho_0}^2 = -\Delta\sigma$) in the presence of spin–orbit scattering and the Zeeman splitting of the spin bands [13] is given by Baxter *et al* (see equation (2.1) in reference [14]). The equation contains the different characteristic timescales involved through the formula $B_x = \hbar/(4eD\tau_x)$, where τ_x stands for the scattering times τ_i (inelastic) and τ_{so} (spin–orbit). The function $f_3(x)$ in the equation has been derived by Kawabata [15], and the compact summation formula (used by us) is given by Baxter *et al* (see equation (2.4) in reference [14]). In the low-field limit, the factor $\gamma \ll 1$ ($\gamma = (3g^*\mu_B B/(8eDB_{so}))^2$), and the sign of $\Delta\rho/{\rho_0}^2$ are determined by the relative strength of the terms $f_3(B/B_i)$ and $f_3(B/B_2)$ (see equation (2.2) in reference [14]). When the spin–orbit scattering is weak ($\tau_{so} \gg \tau_i$), $B_{so} \ll B_i$ and $B_2 \approx B_i$. One then obtains a negative MR (or a positive $\Delta\sigma$). In the presence of strong spin–orbit scattering ($\tau_{so} \ll \tau_i$), $B_{so} \gg B_i$, and $B_2 \simeq 4B_{so}/3$. In this case,

$$\left[\frac{\Delta\rho}{{\rho_0}^2}\right] \approx \frac{e^2}{2\pi^2\hbar}\sqrt{eB/\hbar}\left[\frac{1}{2} f_3\left[\frac{B}{B_i}\right] - \frac{3}{2} f_3\left[\frac{3B}{4B_{so}}\right]\right]. \qquad (1)$$

Since $f_3(x) \sim x^{3/2}$ for $x \ll 1$, we find that for $B_i \ll B_{so}$, $\Delta\rho/{\rho_0}^2$ is positive and is $\propto B^2$ as observed by us (see figure 3). Using the parameters from the fit of the $\sigma(T)$ data, we have fitted our MR data to equation (2.1) of reference [14]. The fit to the MR data required two more parameters: $D\tau_{so}$ and $g^*\tau_{so}$, both of which are constants. The fits to the MR data are shown as solid lines in figure 2. The fits at different temperatures are satisfactory. We obtained from the fit $D\tau_{so} = 3.9 \times 10^{-16}$ cm^2 and $g^*\tau_{so} = 4.0 \times 10^{-13}$ s. It is interesting to note that the temperature dependence of MR has no free parameter. The parameters are essentially the same as used in the fit to the $\sigma(T)$ data. The temperature dependence of $\sigma(T)$ and that of the MR are therefore self-consistent. From the values of $D\tau_{so}$ obtained from the fit, we obtained $B_{so} = 44.5$ T. Since the value of B used by us is $\approx(1/8)B_{so}$, we do not expect to see any saturation or downturn of $\Delta\rho/{\rho_0}^2$ in our experiment. At higher temperatures, ${\tau_i}^{-1}$ becomes greater than ${\tau_{so}}^{-1}$; it is therefore expected that one may see a positive MR (small though it may be) at higher temperatures. Our conclusion from the above analysis of $\sigma(T)$ and MR is that a self-consistent evaluation of the data can be performed, invoking the presence of strong spin–orbit interaction.

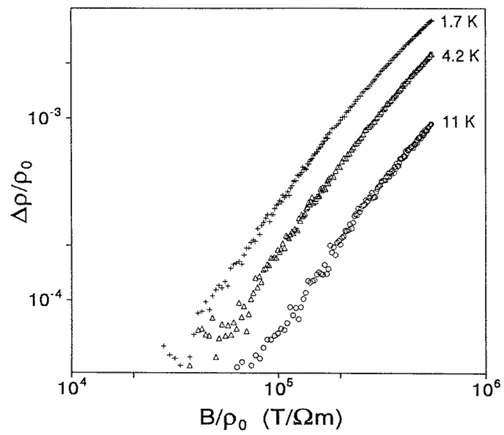

Figure 5. $\Delta\rho/\rho_0$ versus B/ρ_0 for $Al_{65}Cu_{20}Cr_{15}$ for $T = 1.7$ K, 4.2 K, and 11 K.

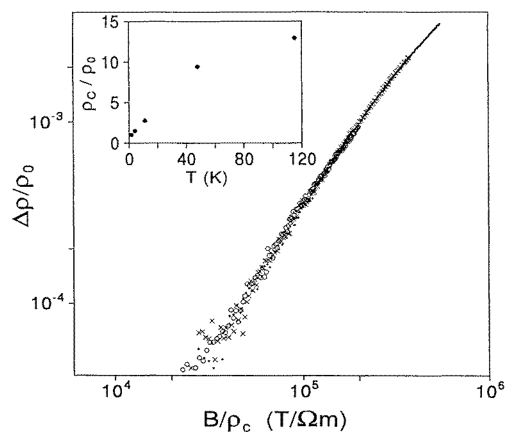

Figure 6. $\Delta\rho/\rho_0$ versus B/ρ_c for $Al_{65}Cu_{20}Cr_{15}$ quasicrystal for $T = 1.7$ K (•), 4.2 K (×), and 11 K (○). Inset: ρ_c/ρ_0 versus the temperature.

However, we must also point out that a semi-quantitative agreement of the theories with the data is not a guarantee that this is the only explanation. We do not rule out any alternative explanation, and we are also aware of such explanations. For instance, below, we point out another alternative explanation. (We should clarify that we are not contradicting ourselves. We are simply noting the possibility of different explanations which are difficult to rule out categorically.) The almost quadratic dependence of the MR on B is particularly intriguing. Suppose we take a radically different viewpoint, and argue that, in quasicrystals, quantum transport theories are not valid, and that we have a system which has a MR with a dominant contribution coming from band motion. In that case one would expect Kohler's rule to be valid, with [16, 17]

$$\Delta\rho/\rho_0 = f(B/\rho_0) \approx \text{constant} \times (B/\rho_0)^2 \tag{2}$$

for all temperatures, where ρ_0 is the zero-field resistivity at a given temperature. In figure 5 we have plotted $\Delta\rho/\rho_0$ versus B/ρ_0 at different temperatures on a log–log plot. For $\Delta\rho/\rho_0 \leqslant 4 \times 10^{-5}$, we reach the limit of resolution of our experiment. We find that $\Delta\rho \propto B^n$ with $n \simeq 1.7$ to 1.8. But the temperature independence of the $\Delta\rho/\rho_0$ versus B/ρ_0 curves, expected on the basis of equation (2), is not found. We then modified equation (2) given above, and tried the empirical form [17]

$$\Delta\rho/\rho_0 = \text{constant} \times (B/\rho_c)^n. \tag{3}$$

In this equation ρ_c is a temperature-dependent parameter, which is not the same as ρ_0. In other words, the constant of equation (2) is temperature dependent, and we absorb this into a temperature-dependent parameter, ρ_c. We are attempting to see whether at each temperature we can define ρ_c in such a manner that $\Delta\rho/\rho_0$ versus B/ρ_c is independent of temperature. We find that it is indeed possible to find such forms of ρ_c. We have shown this in figure 6. We find that $n = 1.75$. ρ_c obtained by such procedure has a distinct temperature dependence, as shown in the inset of figure 6. We have taken the data at the lowest temperature (1.7 K) as a reference, so $\rho_c \equiv \rho_0$ at $T = 1.7$ K. Therefore the temperature dependence as shown in figure 6 represents the temperature dependence of $\rho_c/\rho_0(1.7\ \text{K})$. This implies that if we want to explain this MR using a Kohler-type contribution, we need to impose some modification to explain the temperature dependence of ρ_c. The validity of equation (2) presupposes a number of factors which are not strictly valid even for simple metals. The temperature-independent constant of equation (2) arises if the carrier concentration and the anisotropy of the scattering (arising from the anisotropy of the Fermi surface and the effective mass) are temperature independent [16]. If either or both of them are temperature dependent, then equation (2) will show a deviation. We suspect that the origin of the temperature dependence as parametrized through equation (3) can arise if the anisotropy of the scattering process over the Fermi surface decreases as the temperature increases. This however is only a speculation. The main problem of this approach is that the resistivity and MR cannot be explained within one theory.

To summarize, we have measured the low-temperature resistivity and magnetoresistivity of the icosahedral quasicrystal $Al_{65}Cu_{20}Cr_{15}$. We find that, while at higher temperature, ρ decreases as T increases, ρ reaches a maximum at $T \simeq 50$ K, and, for $T < 50$ K, ρ decreases as T decreases. The MR is positive for all T in the range $115\ \text{K} \geqslant T \geqslant 1.7$ K. We were able to give a self-consistent explanation for the resistivity behaviour at low temperature and the positive MR by invoking strong spin–orbit interaction. We have also provided a plausible alternative explanation for the positive MR.

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