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Magnetoconductance related to change in density-of-states of decagonal quasicrystals—observation of a scaling behaviour

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Abstract

The magnetoresistance of fully oriented decagonal AlCuCo films has been carefully measured. From this, together with observations of the temperature dependence of the film resistivity and tunnelling experiments on a single quasicrystal of a similar material (AlNiCo), a scaling behaviour has been found. This behaviour strongly suggests that the drastic change of magnetoresistance occurs within an energy scale where a pseudogap appears in the materials.

1. Introduction

The question of how quasicrystalline order influences the electronic properties in quasicrystals has always been of central interest. While many unusual transport properties remain not clearly explained, the magnetoresistance (MR) is thought to be the only one which is well understood, in terms of the quantum interference effects (QIE) that are well known in disordered systems [1]. This conclusion is drawn on the basis of experimental data obtained predominantly for icosahedral samples. However, the not uncommon structural defects in i-phase samples make the above conclusion rather dubious because such defects may bring the behaviour closer to the amorphous rather than the quasicrystalline phase. The above inference seems to be supported by a very recent experiment which shows that, for stable icosahedral materials, the deviation of MR from QIE is enhanced with the increase of sample resistivity, i.e., with improvement of the quasicrystalline order [2]. But one still cannot use this against the applicability of QIE to quasicrystals due to the lack of knowledge of the QIE on the insulate side. In fact, applying the theory of QIE to the analysis of data from icosahedral samples is rather inappropriate because the theory predicts corrections to a metallic state [3], whereas i-phase samples are nearly insulators. Thus it would be interesting to make a detailed and accurate investigation on the MR of a quasicrystalline sample well within the metallic side and at the same time with the quasicrystalline order least perturbed by structural defects. Stable decagonal samples could be suitable candidates for this purpose.

In contrast to the icosahedral phase, the stable decagonal phase remains metallic down to low temperatures, as demonstrated by resistivity [4], Hall effect [5], thermopower [6], and optical conductivity [7] measurements. The success in growing fully oriented AlCuCo films [8] allows us to check the applicability of QIE well on the metallic side where the QIE theory is much more mature than on the insulate side. The MR of bulk decagonal $Al_{65}Cu_{15}Co_{20}$ samples has been reported already [9]. Here we show, by careful measurements of the MR and temperature dependence of the resistivity in the quasicrystalline plane of fully oriented decagonal AlCuCo films [8], that the theory of QIE cannot explain the data consistently in highly ordered samples. Instead, combining with the tunnelling results taken on single quasicrystals of a similar material (AlNiCo), there is strong evidence for a contribution to MR by the unique structure of the electronic states in quasicrystals. The MR is closely related to an unusual magnetic field-induced change of the density of states (DOS) at the Fermi level. It is clearly demonstrated that the drastic MR change occurs within an energy scale where a pseudogap occurs.

2. Experiment

The growth of the $Al_{71.4}Cu_{15.7}Co_{12.8}$ decagonal films was described in detail in [8]. The average size of the grains was about 600 nm and no trace of any second phase was detected by x-ray diffraction analysis. The rocking curve for the (00002) diffraction peak was as narrow as 0.11^0, comparable with that for epitaxial films. The accurate orientation of the ten-fold axis perpendicular to the film avoided any mixing of the properties in the quasicrystalline plane with those along the periodic direction. The large sample resistance and the standard four-leads pattern by photolithography guaranteed much greater accuracy of the conductivity calculation than for bulk samples. The temperature dependence of the resistivity was measured in the standard four-leads configuration, and the MR by a specially designed bridge circuit which gave high accuracy and stability. Details of tunnelling experiments were reported in [10].

3. The temperature dependence of resistivity

Quantum interference gives rise to quantum corrections, weak localization (WL) and electron–electron interaction (EEI), affecting the dependence of the classical metallic conductivity on both temperature T and magnetic field H [3]. However, previous reports on QIE in quasicrystals all focused on MR. This is not so surprising when one notes that i-phase samples are almost insulators and, in contrast to metals, their classical resistivity is expected to be nonconstant at low temperatures, so it is difficult to say what the correction of QIE would be on the temperature dependence of resistivity. However, as mentioned above, the ground state of the stable decagonal phase is well on the metallic side, so it could be possible to find some fingerprints of QIE, if it exists, in the T-dependence of resistivity of decagonal samples. The high orientation accuracy of our film samples allows us to study the behaviour in the quasicrystalline plane with an accuracy which cannot be reached with bulk samples. Although the overall conductivity–temperature behaviour of decagonal AlCuCo films agrees well with bulk samples [4] (figure 1, inset), the present measurements were able to reveal a faint maximum around ∼15 K, while below ∼10 K the conductivity decreases with a fairly good logarithmic dependence (figure 1).

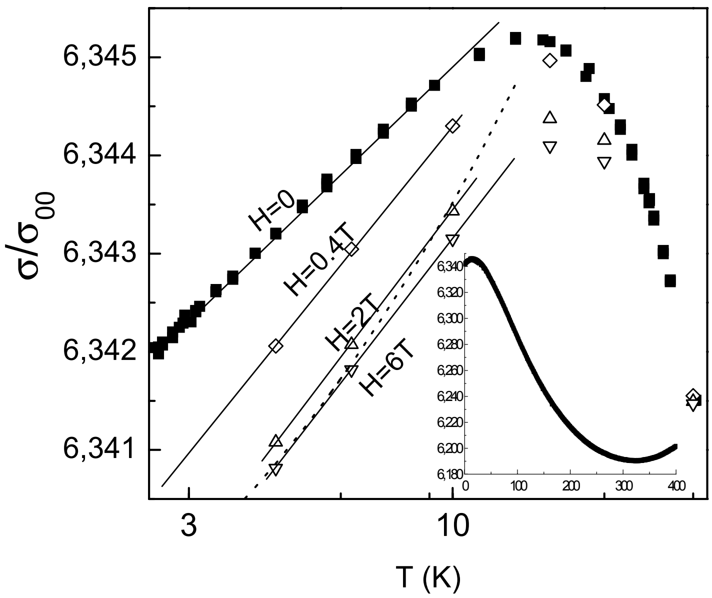

Figure 1. Dimensionless conductance σ/σ_{00} of an AlCuCo oriented film as a function of temperature in different magnetic fields: $H = 0$, 0.4, 2 and 6 T. The conductance decreases below ~15 K, and follows a logarithmic temperature dependence below ~10 K. The low temperature anomaly is enhanced and the slope increases with H. The behaviour contradicts both the weak localization which demands no anomaly with H, and the 3D electron–electron interaction effect which demands a $T^{1/2}$ dependence (the upward dashed curve in the figure for $H = 6$ T).

A logarithmic change is expected in the 2D WL, 2D EEI, and the Kondo effect. Both AlCuCo and AlNiCo decagonal quasicrystals show similar magnetic behaviour [11], i.e., very weak diamagnetism and independence of temperature below 300 K [12]. Therefore, the Kondo effect in these materials can be ruled out. This is in agreement with the results that the d-band is fully occupied and impurity magnetic moments do not exist in Al rich alloys [13, 14]

Although both WL and EEI were considered in the analysis of MR in icosahedral samples [1, 2], only the former was used to fit the MR data in a previous study on a bulk decagonal sample [9]. Here we can probe further and investigate the temperature dependence of both resistivity and MR. If EEI can be totally neglected, the logarithmic T-dependence in figure 1 immediately suggests that 2D WL is the underlying mechanism. However, the 2D WL correction $\Delta\sigma$ in a zero magnetic field predicts an exact logarithmic behaviour only in two extremes: when the spin-orbit scattering time is much longer than the inelastic scattering time, $\tau_{so} \gg \tau_i$, or in the opposite extreme when $\tau_{so} \ll \tau_i$. The T-dependent correction changes sign in the two cases, the conductivity decreases with decreasing temperature in the former extreme but increases in the latter [15]. Thus the conductivity behaviour in zero field in figure 1 indicates that $\tau_{so} \gg \tau_i$, i.e., there is negligible spin-orbit scattering, which would lead to negative MR according to WL theory. However, the experimental data is just the opposite, as can be seen in figure 2. Positive MR was also observed in bulk decagonal samples [9].

Since WL alone cannot properly explain the T-dependence of resistivity and MR, one would like to see a comprehensive QIE explanation that includes both WL and EEI.

As mentioned above, in a 2D system both WL and EEI contribute to the $\ln(T)$ dependence of conductance. One can separate the two effects in the following way [15]. The quantum correction in zero field is composed of WL and EEI contributions, and the slope

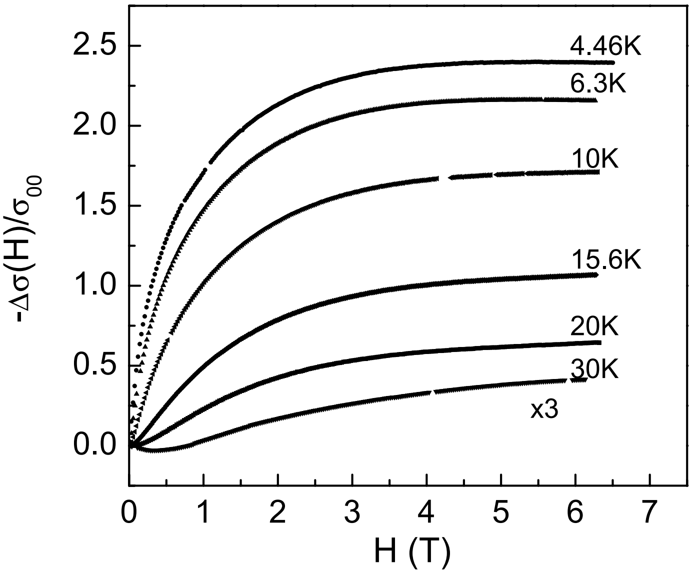

Figure 2. Dimensionless magnetoconductance of an oriented AlCuCo film as a function of magnetic field with the field perpendicular to the film. A striking feature is that the magnetoconductance becomes saturated at high fields. Small positive magnetoconductance emerges above ~ 20 K at low fields which may represent the actual weak localization effect.

of dimensionless conductance against T on a ln scale gives $\alpha p + 1 - F$, where $-p$ is the exponent for the T-dependence of τ_i, F is a measure of screening, $\alpha = 1$ when spin-orbit scattering can be ignored and $\alpha = -1/2$ when $\tau_i \gg \tau_{so}$. A magnetic field H suppresses the T-dependence of the WL so in a high field only the EEI remains, i.e., in high fields the slope becomes $1 - F$. From the data shown in figure 1 we see that $F \sim -1.9$ and $\alpha p \sim -0.75$. In other words, we require a negative F and a negative α to fit the data.

Before discussing the consequence of this separation for the MR analysis, we should note several relevant points.

First, the 2D condition for EEI would be fulfilled if the film thickness $t < (\hbar D_c/k_B T)^{1/2}$, where D_c is the electron diffusion constant along the ten-fold axis. For a film of $t = 300$ nm and at 10 K, this would require $D_c > 1200\ \mathrm{cm^2\ s^{-1}}$, a value too large to be true [9]. Therefore, the system is very likely 3D with respect to EEI [3]. The logarithmic behaviour then implies that the contribution of EEI is not important in the present case, but we have already shown that total omission of EEI leads to a contradiction. What we should keep in mind here is that we have omitted the dimensional criterion during the quantitative separation of EEI and WL. Moreover, the logarithmic dependence of conductance in high fields of 6 T (figure 1) strongly suggests that EEI is effectively 2D. These suggest that the QIE explanation might be questionable.

Second, the system can be considered as 2D with respect to WL if $t < (D_c \tau_i)^{1/2}$. Taking $D_c \sim 10\ \mathrm{cm^2\ s^{-1}}$ [9], and $t = 300$ nm, we need $\tau_i > 10^{-10}$s at 10 K. From the relative change in conductivity we may estimate the elastic relaxation time to be $\tau_e \sim 6 \times 10^{-14}$ s with a renormalized Fermi velocity of $\sim 2 \times 10^7$ cm s^{-1}, which is not unreasonable.

Third, although the screening factor F is considered to be positive for normal metals, it can be negative for superconductors or other systems with large electron–phonon coupling. Negative values of F were also found for 2D systems [16], but it should be remarked that no negative F was used in previous studies on MR in quasicrystals [1]. This negative F directly indicates a negative MR due to EEI [3], contrary to the previous studies. If the negative F comes from large electron–phonon coupling in decagonal quasicrystals, we would expect a negative value also in icosahedral samples since there is no reason why the coupling should be stronger in the decagonal phase than in the icosahedral one. However, former studies of QIE were only limited to MR in icosahedral samples [1].

Fourth, the negative α together with the logarithmic behaviour in zero field in figure 1 indicates that we are in the strong spin-orbit scattering limit, but we should be wary of assuming this limit for the present decagonal samples. The spin-orbit scattering rate depends on both the number and the properties of the impurity centres. Heavy atoms and/or atoms with unfilled d-shells give strong contributions. These cases do not apply to our samples since Al, Cu, and Co are light atoms and the d-orbit is fully occupied in the alloy. In addition, it is only natural to expect very few random impurity scattering centres in a well-ordered AlCuCo d-phase sample. As we shall see in the following section, the strong spin-orbit scattering limit is not consistent with the MR behaviour of the sample.

4. Magnetoresistance

The MR curves of the AlCuCo film as a function of transverse magnetic field, H, at several temperatures are shown in figure 2. Two striking features immediately emerge. First, the MR in most cases is positive, but above $\sim$20 K a negative term appears at low H. Secondly, the MR shows some tendency to saturation at high H. For example, $\Delta\sigma(H)$ normalized to $\sigma_{00} = e^2/2\pi^2\hbar$ varies within less than 1% in the range of $\sim$ 3.8 T to $\sim$ 6.5 T for the 4.46 K curve.

The appearance of a negative MR around 30 K itself is a great challenge to the explanation of QIE. In the analysis of QIE, only WL is important at low H, and this negative MR means that the spin-orbit scattering time is comparable to the inelastic scattering time in the temperature range concerned [15, 17], which is in contrast to the expectation of strong spin-orbit scattering as deduced in the last section. If such a weak spin-orbit scattering time is used as a parameter, no fitting can reproduce the saturation of MR at high H because in this case MR due to WL decreases with $\ln(H)$ while MR due to EEI depends on H^2 [3]. Furthermore, to compensate for the decrease of MR at high H, EEI should give a positive MR, which also is contrary to the result of the last section. One cannot expect the negative MR to come from EEI because if EEI dominates WL at low H, their different field dependences will guarantee that no sign change of MR occurs at high H. It is worth noting that a similar negative MR also exists in a very recent study on icosahedral samples, which was treated as an open question there [2].

It is interesting to see how far we can go when we make a quantitative comparison of the behaviour of the QC films with QIE theory. The results in the last section allow us to subtract the EEI contribution from the data. Taking $F \sim -1.9$, we find: (1) the EEI contribution to MR is negative instead of positive; (2) the EEI constitutes only $\sim$ 10% of the total MR at 4.46 K and decreases to $\sim$ 2% at 20 K. That is, the T-dependence of MR is enhanced when the EEI correction is subtracted, and the discussion below is not much influenced by whether the EEI correction is subtracted or not.

After subtracting the EEI contribution, we will see how the data fit with the WL theory. In fact, since WL is a weak field theory, people believe that more reliable parameters are found in the low field limit where $\Delta\sigma(H) \propto H^2$. In this field range we have [15]:

$$48\frac{-\Delta\sigma(H)}{\sigma_{00}} = \left[\left(\frac{1}{H_i}\right)^2 - \left(\frac{3/2}{H_i + 4/3H_{so}}\right)^2\right]H^2 \tag{1}$$

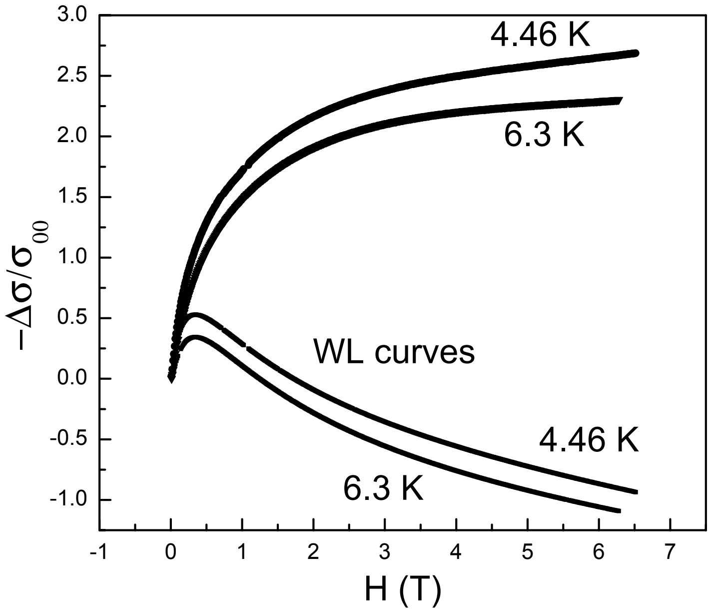

Figure 3. Comparison of the measured magnetoconductance (after subtracting the contribution from electron–electron interaction) with weak localization theory based on parameters taken at zero magnetic field.

or

$$48\frac{-\Delta\sigma(H)}{\sigma_{00}}H^{-2} = \left[\left(\frac{1}{H_i}\right)^2 - \left(\frac{3/2}{H_i + 4/3H_{so}}\right)^2\right] \tag{2}$$

where H_i is a characteristic field related to inelastic scattering and H_{so} is the corresponding parameter due to spin-orbit scattering. This is a field region not always experimentally accessible, especially at the low temperature end since it needs $H \ll H_i$. Although our data points were very dense, we still take the following measure to ensure the reliability of the results. We plot the magnetoconductance (MC) as a function of H^2 with the data below, from say, $\sim$ 0.2 to 0.3 T, and then fit the data by a polynomial. The coefficient of the linear term should give the right side quantity in equation (2). By plotting this quantity as a function of T, we find it crosses zero at about 20.5 K. This gives $H_{so} = 0.549H_i(20.5\text{ K})$ at this temperature. Since H_{so} is T-independent and $H_i = \alpha T^{1.5}$, as obtained in the last section, we have $H_{so} = 1.6H_i(10\text{ K}) = 5.4H_i(4.46\text{ K})$. The low field data at 4.46 K give $H_i(4.46\text{ K}) = 1.06 \times 10^{-2}$ T and $H_{so} = 5.72 \times 10^{-2}$ T.

What does this result mean? First, it means there is no case where τ_i is much greater than τ_{so}. Since H_{so} and H_i are comparable, we should not expect a logarithmic T-dependence in this range [15], in contrast to our results which show logarithmic behaviour (figure 1).

Secondly, since we are in the range of comparable H_{so} and H_i, a positive MR throughout the wide field range cannot be ensured. Indeed, using the parameters obtained at low fields, a drastic discrepancy occurs between WL theory and the measured results when H is increased (figure 3). This discrepancy is certainly not caused by EEI as the latter has already been subtracted from the data.

The fitting is even worse when we try to use the high field data to obtain the parameters.

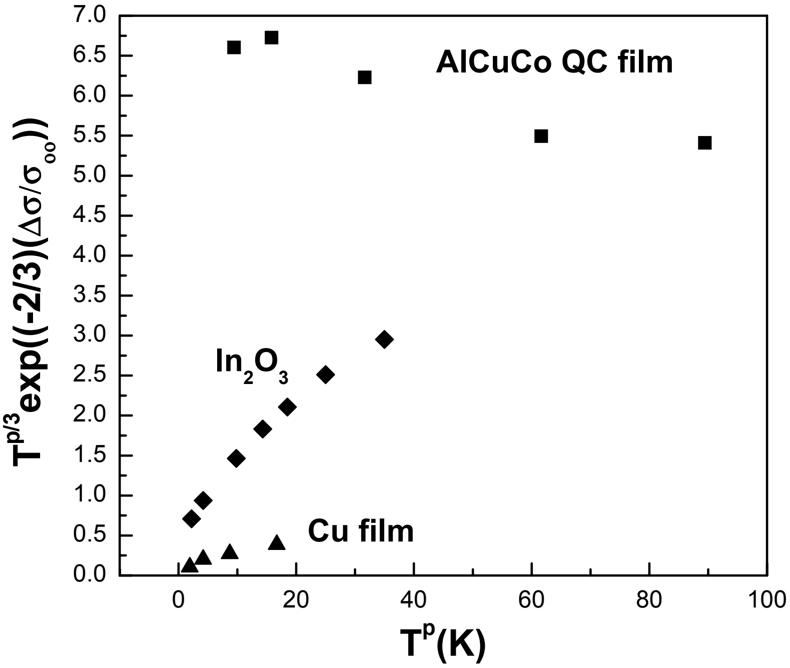

Figure 4. Points plotted according to equation (6), showing the difference between the AlCuCo decagonal quasicrystalline film and other disordered films.

From WL theory we expect

$$\frac{-\Delta\sigma(H)}{\sigma_{00}} = \frac{1}{2}\ln\frac{H}{H_i} - \frac{3}{2}\ln\frac{H}{H_i + 4/3H_{so}}, \qquad H \gg H_i, H_{so} \tag{3}$$

Although we cannot find a logarithmic dependence throughout the entire high field range, the data between ~ 1 T and ~ 2 T roughly follow a logarithmic behaviour. From equation (3) we find for the intercept at $H = 1$ T that

$$\frac{2}{3}\frac{-\Delta\sigma(H)}{\sigma_{00}} = \ln\left(\frac{4/3H_{so}}{H_i^{1/3}} + H_i^{2/3}\right). \tag{4}$$

This quantity changes sign at about 26 K, which gives

$$H_{so} = \frac{3}{4}[H_i^{1/3}(26\ \mathrm{K}) - H_i(26\ \mathrm{K})]. \tag{5}$$

Using this condition to derive the H_i value at, say, 4.46 K from equation (4), we find that H_i should be negative for the temperature! Equation (4) can be written in the form

$$x = \alpha^{2/3}y + \frac{4/3H_{so}}{\alpha^{1/3}} \tag{6}$$

with $x = T^{p/3}\exp(-2\Delta\sigma(H)/3\sigma_{00})$ and $y = T^p$.

If we plot x as a function of y, we find the slope is negative (figure 4)! For comparison, data for Cu film [18] and In_2O_{3-x} film [19] are also shown in the figure. Negative slope means that the T-dependence of MR is substantially greater than that expected by WL. It is interesting to note here that the low field limit of $-48H^{-2}\Delta\sigma(H)/\sigma_{00}$, rather than following equation (2), exponentially changes with T (see figure 5).

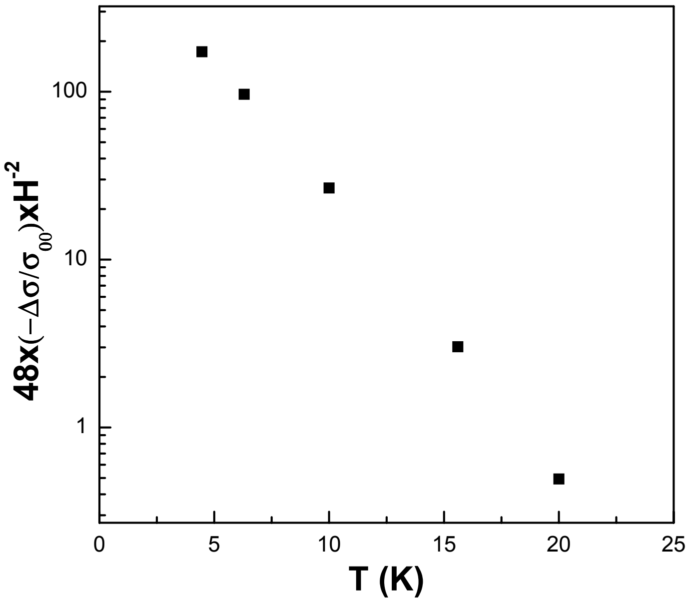

Figure 5. The quantity $-48(\Delta\sigma(H)/\sigma_{00})H^{-2}$ as a function of T follows an exponential law rather than equation (2).

The above discussion definitively demonstrates that QIE cannot explain the behaviour of our QC films in a consistent way, and suggests that the previous QIE studies on quasicrystals are misleading.

5. A scaling behaviour

Now we show that the observed MR could be related to an unusual H-induced change in the density of states (DOS) at the Fermi level of the decagonal quasicrystals. First we notice that, by taking the high field values of $\Delta\sigma_s$ where the functional dependence on H becomes ineffective, it decays on an energy scale of $\sim$1 meV when we express T in units of eV. A very surprising discovery is that $\Delta\sigma_s$ satisfactorily scales with the pseudogap, Δ, observed in tunnelling experiments on similar material at high H and ultralow T [10] (figure 6). This is really unexpected since it is difficult to imagine how the pseudogap and MR should be put together. We propose that the MR behaviour in our quasicrystals is determined by two factors: the contribution by a single carrier and the number of carriers which take part in the contribution. It is natural to think that the latter factor should depend on the pseudogap. Therefore, the scaling behaviour of the pseudogap with MR is indicative that only the carriers left in the gap give important contribution to MR. However, at present it would be premature to give a functional dependence because that would require a detailed knowledge of how the carriers are activated across the gap and how each carrier contributes to MR.

Further strong support to the above picture is that the tunnelling resistance at zero bias shows an H-dependence very similar to that of the MR [10]. We have also measured the tunnelling resistance as a function of H at 25 mK for different junction biases. The results are shown in figure 7. A remarkable result is the existence of a 'threshold magnetic field' H_{th} above which the tunnelling resistance is very insensitive to changes in H. The threshold H_{th}

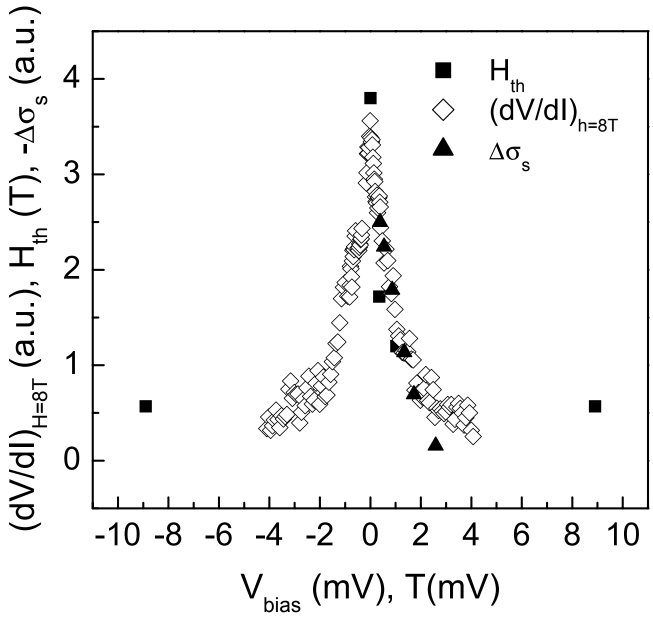

Figure 6. A remarkable scaling behaviour between the temperature dependence of the saturated magnetoconductance $\Delta\sigma_s$, the threshold magnetic field H_{th} (see text) and the tunnelling resistance $(dV/dI)_{H=8T}$. The dominant change in magntoconductance occurs on an energy scale within the gap where the DOS is substantially influenced by the magnetic field.

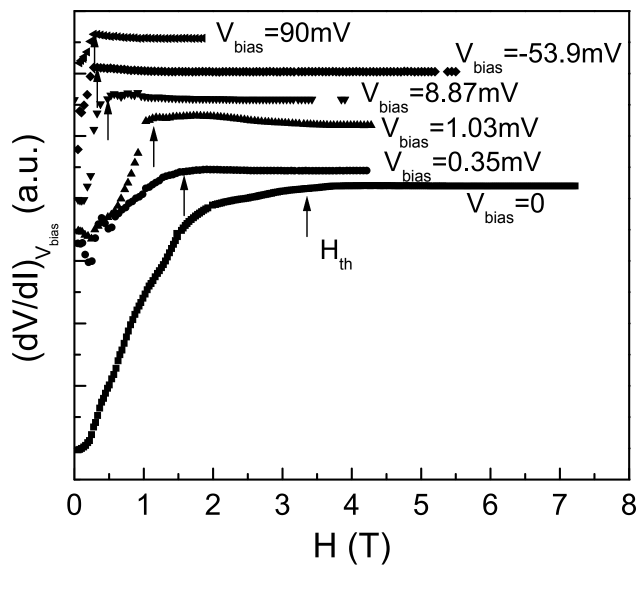

Figure 7. Tunnelling resistance as a function of magnetic field at 25 mK for different dc junction biases. The most fascinating feature is the existence of a threshold magnetic field above which the tunnelling resistance becomes insensitive to the field. The curves are shifted upward as the bias value increases.

is located at about 3.6–3.8 T for $V_{bias} = 0$. This behaviour is in good agreement with the MR curve of 4.46 K in which MR reaches a saturation value at about the same H (figure 2). Also, H_{th} manifests itself more clearly with the increase of dc bias on the junction. The existence of such a threshold field is really unusual and has never been predicted in any previous theoretical models of MR or single electron tunnelling. If we plot H_{th} as a function of the dc bias V, again we find a satisfactory scaling behaviour with the pseudogap as well as with $\Delta\sigma_s$, as shown in figure 6. All these results provide consistent evidence that the drastic change in MR occurs in the energy range of a pseudogap where the DOS is most sensitive to the applied magnetic field. In other words, the MR observed in high quality AlCuCo decagonal samples could be closely related to the H-induced DOS change rather than to the WL determined by different scattering processes. This picture is consistent with the very recent report that the electron states are in a high degree of extended character in this kind of decagonal quasicrystals [20].

6. Summary

In summary, by simultaneous measurements of the temperature and magnetic field dependences of the resistivity of fully oriented decagonal films we have been able to make detailed quantitative comparisons of our data with those predicted by QIE theory. The results show that QIE cannot explain the behaviour satisfactorily.

Combining the data on MR and tunnelling experiments on decagonal quasicrystals, we have observed a striking scaling behaviour with regard to the DOS pseudogap, saturated MR, and a newly discovered magnetic field threshold. This scaling behaviour strongly suggests that the MR is closely related to the carriers within the pseudogap.

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