Scattering times and mean free path in AlCuFe quasicrystalline thin films

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We present magnetoresistivity measurements on AlCuFe quasicrystalline thin films from which the temperature dependence of the inelastic scattering time (τ_{ie}) has been deduced down to 200 mK in the low field limit. We show that τ_{ie} presents a weak temperature dependence below 4 K ($1/\tau_{ie} \sim T^{0.87}$) which is in close agreement with the theory of Isawa predicting a crossover from a $T^{3/2}$ to a T regime. Saturation effects as well as a possible shift in dimensionality have also been investigated.

I. INTRODUCTION

Stable quasicrystals are known to present very unusual transport properties[1,2] such as very high-resistivity values which depend strongly on the composition and the structural quality, a low effective number of carriers and a reduced density of states at the Fermi level.[1,3] Among those, the very high-resistivity values, rising up to 10^7 $\mu\Omega$ cm at very low temperature in AlPdRe,[2] are maybe the most striking ones. Moreover there has been considerable interest in developing theoretical models for conduction in quasicrystals. Indeed such resistivity values should correspond to unrealistic mean free paths L_0 of a fraction of an Angstrom calculated in a classical way for metallic alloys; Phillips and Rabe[4] first proposed a structural model based on two building blocks to explain the very high resistivities. On the other hand, Fujiwara *et al.*[5] have shown that those values could be attributed to very strong anomalies in the density of states assuming that $L_0 \sim$ a few Å. Finally Mayou *et al.*[6] assumed that $L_0 \sim 20$ Å, corresponding to the distance separating equivalent sites in the structure, and they proposed that the conduction may be due to hopping between those sites. However no direct experimental evidence had been found so far to support any of those assumptions on the value of L_0.

Quantum interference theories[7,8] have been found to describe very well the magnetic-field dependence (up to 35 T in pulsed fields) and temperature dependence (up to 100 K) of the conductivity of both icosahedral phases[9,10] and their approximants.[11] These measurements gave direct information on the inelastic scattering time τ_{ie}; the analysis of $\sigma(T)$ is consistent with $\tau_{ie} \sim 1/T^2$ above 30 K and below 30 K a $T^{3/2}$ behavior has recently been deduced from the magnetoresistance.[10] However, the fitting procedure of both the temperature and magnetic-field dependence may be quite problematic due to the presence of a large set of physical parameters: the inelastic [$\tau_{ie}(T)$] and spin-orbit (τ_{so}) scattering times, the electron-electron screening factor (F), the diffusivity (D), and the effective Landé factor (g^*). Moreover, at low temperature, the magnetoresistance is dominated by electron-electron interactions in the high-field limit (see below) and it is then very difficult to extract the weak localization parameters [$\tau_{ie}(T)$, τ_{so}] from the data fits. Hence, no value for the inelastic scattering time below 2 K has been given so far. We present here low- (and high-) field measurements of the magnetoresistance performed on quasicrystalline thin films down to 200 mK. From those measurements we get a determination of the inelastic-scattering time at very low temperature and show that in this temperature range $\tau_{ie}(T)$ presents a weak temperature dependence [$1/\tau_{ie}(T) \sim T^{0.87}$]. One possible explanation is the presence of a crossover from a $T^{3/2}$ to a T regime as predicted by Isawa for screened Coulomb interactions in disordered metals.[12] From the crossover temperature, it is then possible to deduce the *elastic* scattering time in our AlCuFe quasicrystals. Other effects such as a change in dimensionality related to the small thickness of the films or a saturation effect are also investigated.

It has been shown by Lindqvist[13] that in amorphous metals it can be much more interesting to use the quantum interference theories in the low-field limit ($B/T<1$) where both the electron-electron interaction (EEI) and the weak localization (WL) contributions to the change in resistivity ρ can be written as $\Delta\rho/\rho = \rho\alpha B^2$ with

$$\alpha_{EEI} = 0.056(e^2/4\pi^2 h)F(2hD)^{-1/2}(g\mu_B)^2(kT)^{-3/2} \quad (1a)$$

and

$$\alpha_{WL} = (e^4/2\pi^2 h^3)(D\tau_{so})^{3/2} f(t) \ , \quad (1b)$$

where $f(t)$ generally is a complicated function of $t = \tau_{so}/4\tau_{ie}$, but it can be approximated for low enough temperatures ($t \ll 1$) by $f(t) \sim (1/96 t^{-3/2})$ and

$$\alpha_{WL} = 1.19 \times 10^{24} D^{3/2} \tau_{ie}(T)^{3/2} \ . \quad (2)$$

Knowing D, $\alpha_{WL}(T)$ is then directly proportional to $\tau_{ie}(T)^{3/2}$.

Such an analysis may however be hard to do since the variations of the conductivity are very small in the low-field limit and it is then difficult to extract the $\alpha(T)$ term from the data. The preparation of quasicrystalline thin films in the AlCuFe system and the fabrication of small electrical circuits by photolithography provides samples with very high electrical resistance (about 1 kΩ) which can easily be used for such precise measurements.

II. SAMPLE PREPARATION

Al, Fe, and Cu layers were sputtered consecutively on $SrTiO_3$ substrates using a rf magnetron sputtering system. The most important parameter to control was the thickness of the different layers since pure quasicrystalline samples can only be obtained in a very narrow composition range around $Al_{62.5}Cu_{25}Fe_{12.5}$. The films were subsequently annealed in a quartz tube under high vacuum (10^{-6} Torr). More details about the preparation procedure have been given elsewhere.[14] X-ray-diffraction patterns performed on some of the samples confirmed the quasicrystalline structure of the films.[14] Several films 3000-Å thick were obtained with resistivity varying between 2000 and 3000 μΩ cm at room temperature. Those films also showed a strong negative temperature dependence of the resistivity with $1.5<\rho(4\ \mathrm{K})/\rho(300\ \mathrm{K})<2$ (see Fig. 1), in good agreement with results previously obtained on bulk samples. Small electrical circuits with electrical resistance around 1 kΩ at room temperature were fabricated from the thin films using conventional photolithography techniques.

III. EXPERIMENTAL RESULTS

The magnetic-field dependence of the resistivity was measured up to 8 T and between 4.2 K and 200 mK in a dilution refrigerator for two samples (sample no. 1: $\rho_{300\ \mathrm{K}}\sim 2000$ μΩ cm and sample no. 2: $\rho_{300\ \mathrm{K}}\sim 2700$ μΩ cm). The magnetoresistance was positive, around 8% at 200 mK and 8 T, and it was rapidly decreasing with temperature (Fig. 2, sample no. 2). This behavior is very similar to what has previously been observed in bulk samples. In particular, the magnetoresistance is around 2% at 8 T, 4.2 K which is very close to what has been observed by Sahnoune *et al.*[10] on samples of equivalent resistivity values (bulk $Al_{63.5}Cu_{24.5}Fe_{12}$, for instance). Due to the high electrical resistance of the circuits we were able to make high precision measurements from which it was easy to extract a B^2 behavior in the low-field limit (inset, Fig. 2, sample no. 2).

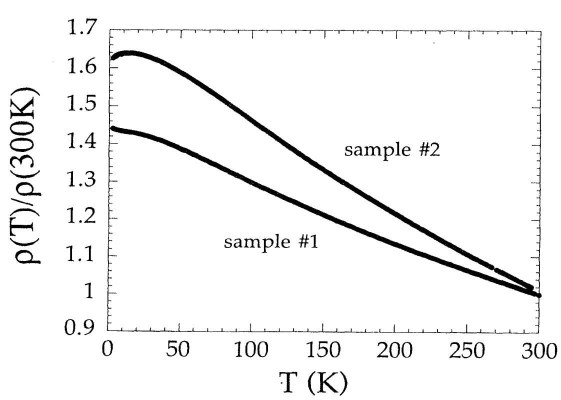

FIG. 1. Temperature dependence of the resistivity of AlCuFe quasicrystalline thin films, sample no. 1, $\rho_{300\ \mathrm{K}}\sim 2000$ μΩ cm; and sample no. 2, $\rho_{300\ \mathrm{K}}\sim 2700$ μΩ cm.

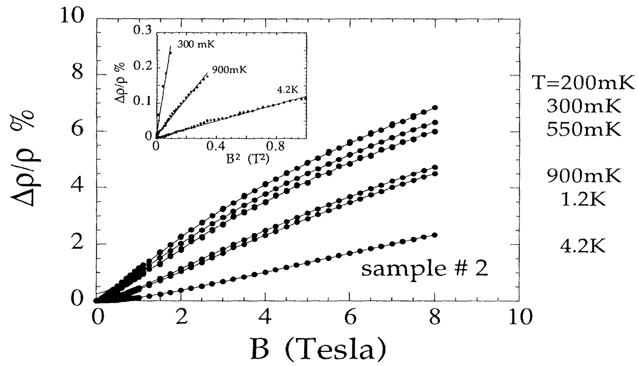

FIG. 2. Magnetoresistance (MR) of an AlCuFe quasicrystalline thin film; lines are guides for the eye. In the inset MR as a function of B^2 in the low-field limit. The solid lines are linear fits using equation $\Delta\rho/\rho=\rho\alpha(T)B^2$.

It is thus possible to get $\tau_{ie}(T)$ from $\alpha(T)$ using Eqs. (1a), (1b), and (2) assuming that the condition $t\ll 1$ is satisfied. The value of the diffusivity D can be estimated using the Einstein equation for the Boltzmann conductivity $\sigma_B=e^2N(E_F)D$. Despite very strong variations of the conductivity, it has been observed that the density of states at the Fermi level (deduced from specific-heat measurements[1,3]) is almost constant in all the measured samples: $N(E_F)\sim 0.14-0.15$ st/eV at. On the other hand, we assumed that all the interference effects are destroyed at 300 K and that σ_B is close to $\sigma(300\ \mathrm{K})$ and we get $D=0.29\ \mathrm{cm}^2/\mathrm{s}$ for sample no. 2 and $D=0.37\ \mathrm{cm}^2/\mathrm{s}$ for sample no. 1. The calculated values of $\tau_{ie}(T)$ deduced from $\alpha_{WL}(T)=\alpha(T)-\alpha_{EEI}(T)$ are represented in Fig. 3 for both samples. In this temperature range our data are consistent with a power-law variation $1/\tau_{ie}(T)=kT^{0.87}$ for both samples with $k=4.5\times 10^{10}$ for sample 1 and $k=1.8\times 10^{10}$ for sample 2. Note that in this tempera-

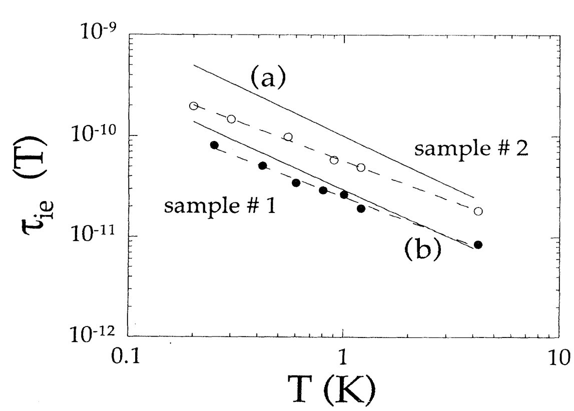

FIG. 3. Temperature dependence of the inelastic scattering time of quasicrystalline thin films calculated using Eqs. (1) and (2), see text. (a) and (b) Solid lines represent the values expected for electron-electron interactions in 2D for $a<L_T$ (a) and $L_T<a<L_{ie}$ (b), see text.

ture range, $\tau_{ie}(T) > 10$ ps and by taking $\tau_{so} = 2$ ps (Ref. 10) we have $t = \tau_{so}/4\tau_{ie}(T) < \frac{1}{20}$ showing that our assumption of $t \ll 1$ was correct.

IV. DISCUSSION

Generally speaking $\tau_{ie}(T)$ can be described by a combination of a saturation term (A_0), electron-electron scattering (τ_{ee}), and electron-phonon (τ_{ep}) scattering times:

$$1/\tau_{ie}(T) = A_0 + 1/\tau_{ee}(T) + 1/\tau_{ep}(T) \ . \quad (3)$$

The temperature dependence of those inelastic scattering times has been investigated by several groups who predicted $1/\tau \sim \tau^p$ with $2 < p < 4$ for electron-phonon interactions[15] and $p = \frac{3}{2}$ or 2 for electron-electron interactions.[16,17] An exponent $p \sim 2$ has been deduced from the temperature dependence of the conductivity of AlCuFe quasicrystals above 50 K,[9] and $p \sim \frac{3}{2}$ has recently been obtained from the analysis of the magnetoconductivity between 2 and 30 K by Sahnoune *et al.*[10] in agreement with those theories.

However Isawa[12] suggested that a $T^{3/2}$ as well as a T term should be observed in the temperature dependence of $1/\tau_{ee}$ with a crossover between those two regimes at some temperature T_c given by

$$T_c = 1.24 \times 10^{-12}/\tau_0 \ , \quad (4)$$

where τ_0 is the *elastic* scattering time. This qualitative behavior is in good agreement with the low-temperature dependence of $1/\tau_{ie}$ deduced from our experiments and the previous higher-temperature data obtained by Sahnoune *et al.*[10] [Fig. 4 for sample no. 2 and bulk $Al_{63.5}Cu_{24.5}Fe_{12}$ (Ref. 11) which present very close resistivity values $\sim 2700\ \mu\Omega$ cm at room temperature and similar magnetoresistivity curves at 4.2 K]. In this case the crossover temperature would be around 4 K leading to an elastic scattering time of $\tau_0 \sim 4 \times 10^{-13}$ s and an elastic mean free path of $L_0 = (D\tau_0)^{1/2} \sim 20 \pm 5$ Å. Applying this theory here, the experimental determination of the elastic mean free path in quasicrystals appears to be quite large and is in support of the model recently presented by Mayou *et al.*[6] to describe the unusual properties of quasicrystals. A very important feature here is that this value of L_0 is not related to the presence of defects in the samples but is directly related to structural characteristics of the quasicrystal. Indeed, it has been shown that the structure consists of clusters separated by about 30 Å built by embedded shells and a model based on such hierarchy of clusters has actually been developed by Janot *et al.*[18] to describe in a general way the physical properties of quasicrystals.

In the theory of Isawa, the inelastic scattering time can be written as

$$\tau_{12} = AT^{3/2} + BT \quad (5)$$

with $A = 3.2(h\tau_0)^{1/2}k^{3/2}/(E_F\tau_0)^2$ and $B = 1.3kh/(E_F\tau_0)^2$ where E_F is the Fermi energy and k the Boltzmann constant. These values are equal to the experimental ones for $E_F\tau_0/h \sim 7$ or taking $\tau_0 = 10^{-13}$ s for $E_F \sim 50$ meV. This surprisingly small value of E_F can however be related to the very peculiar band structure of the quasicrystalline structure.[19] Moreover a comparable value of $E_F \sim 100$ meV has been estimated by Pierce *et al.*[20] from thermopower measurements in agreement with the value of the density of carriers inferred from Hall measurements.

An alternative explanation of the results would be to consider our temperature dependence as an approximation of a more complex behavior over this temperature range indicating that the inelastic scattering time is actually saturating as T tends towards zero. The dashed line in Fig. 4 presents a fit to a $A_0 + A_1T^{3/2}$ law of the data. This fit gives a saturation value of $\tau_{ie}(0) \sim 200$ ps; such saturation could be attributed to (i) decoupling of the electron gas from the thermal bath at very low temperature, (ii) spin-flip scattering of conduction electrons off magnetic impurities (a few ppm of magnetic impurities

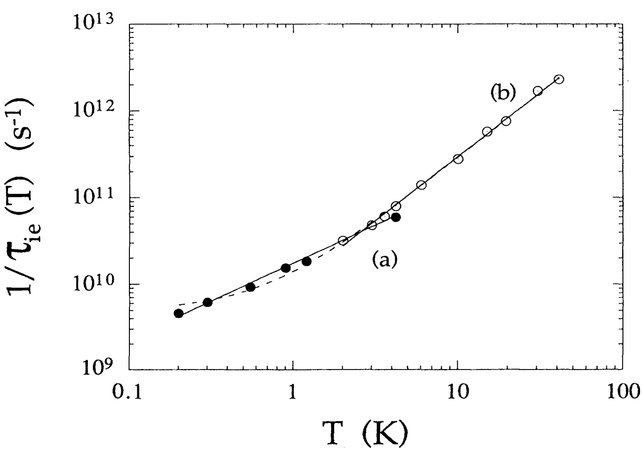

FIG. 4. Temperature dependence of $1/\tau_{ie}$ between 200 mK and 30 K (solid circles, our data; open circles, data from Ref. 11). The solid lines are (a) 1.9×10^{10} $T^{0.87}$ and (b) 1.0×10^{10} $T^{1.47}$, the dashed line is a fit to a $A_0 + A_1T^{1.5}$ law.

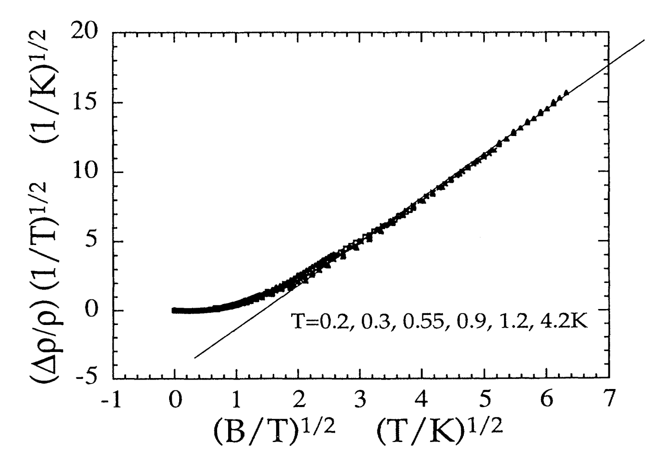

FIG. 5. $(\Delta\rho/\rho)(T)^{-1/2}$ as a function of $(B/T)^{1/2}$ showing a universal curve characteristic of electron-electron interaction effects.

can significantly reduce the effect of localization leading to a saturation), or (iii) zero-point motion of the scattering centers.

Finally, because the films are thin (~ 3000 Å) the results could also suggest a shift in dimensionality from a three-dimensional (3D) to a 2D regime. In 2D, $1/\tau_{ee}$ is usually expected to show a logarithmic singularity $1/\tau_{ee} \sim T \ln T$.[21] However, Altschuler *et al.*[17] have shown that this singularity can be wiped out for very thin films [$a \ll L_T = (hD/kT)^{1/2}$, where a is the thickness of the film and L_T is the thermal coherence length] and they get the following expressions for $1/\tau_{ee}$:

$$1/\tau_{ee} = \ln(C)/2CkT/h \quad \text{with } C = \pi a \sigma_B h/e^2 \quad \text{for } a < L_T \qquad (6a)$$

and

$$1/\tau_{ee} = \ln(CDh/kTa^2)/4CkT/h \quad \text{for } L_T < a < L_{ie} = (D\tau_{ie})^{1/2} . \qquad (6b)$$

In our case, at 0.2 K, $L_T \sim 400$ Å and the dephasing length $L_{ie} \sim 900$ Å are getting close to the thickness of the film and some effects related in particular to the granularity of the films could be expected (a scanning electronic microscopy analysis revealed that the films consist mainly of grains ranging from about 500 to 5000 Å in diameter). For $a = 500$ Å and $\sigma_B = 370$ $(\Omega\,\text{cm})^{-1}$ both Eqs. (6a) and (6b) give values which are quite close to the experimental data (see Fig. 3). Note however that in the high-field limit the magnetoresistance is proportional to $\sqrt{B}$ down to the lowest temperature (straight line on Fig. 5) which is a typical 3D behavior and hence the 2D aspect is probably not dominant here.

Figure 5 actually presents a plot of $(\Delta\rho/\rho)(T)^{-1/2}$ as a function of $(B/T)^{1/2}$ for all our temperatures from 4.2 K to 200 mK and in fields up to 8 T. Another interesting point here is that all the curves collapse into a unique curve emphasizing the fact that the magnetoresistance is dominated by electron-electron interaction effects in this temperature and magnetic field range. Indeed the electron-electron interaction contribution to the magnetoresistance is given by[9]

$$(\Delta\rho/\rho)_{\text{EEI}} = \rho(e^2/4\pi^2 h) F (kT/2hD)^{1/2} g_3(g^* \mu_B B/kT)$$

and following this equation $(\Delta\rho/\rho)(1/T^{1/2})$ is expected to be a function of B/T only, as observed experimentally.

Finally, we note that $1/\tau_{ie} \sim T$ has been recently observed in other highly resistive disordered systems[22] and such a temperature dependence has also been suggested by Belitz *et al.*[23] for metallic systems close to the metal-insulator transition.

V. SUMMARY

We measured the magnetic field dependence of the resistivity in AlCuFe quasicrystalline thin films. The analysis of the magnetoconductivity in the low-field limit leads to $1/\tau_{ie}(T) \sim T$. A crossover from a $T^{3/2}$ to a T regime has been suggested by Isawa, and this is in agreement with our results. Applying this theory here we get from the crossover temperature an estimate of the elastic mean free path in AlCuFe quasicrystals: $L_0 \sim 30 \pm 5$ Å. Some effect due to saturation or a shift in dimensionality can however not be completely excluded.

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