Does the Quasicrystal AlCuFe Follow Ohm's Law?

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We present the first measurements of the electrical *I-V* characteristics of AlCuFe quasicrystalline thin films processed by solid state interdiffusion of magnetron sputtered Al, Cu, and Fe layers. Despite the peculiar electronic properties of quasicrystals, our results show that those samples follow perfectly Ohm's law for bias voltages which vary by 7 orders of magnitude.

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Since the discovery of the quasicrystalline structure, many papers have been devoted to the study of the structural and physical properties of this new state of condensed matter. Indeed, high structural quality samples in both AlCuFe [1,2], AlCuRu [3], AlPdMn [4], and recently AlPdRe [5] icosahedral phases present very peculiar electronic and magnetic properties such as very low electrical conductivity values, strong temperature and composition dependences of the conductivity, a reduced density of states at the Fermi level, and a diamagnetic behavior. On the theoretical side a few models have been proposed trying to explain those unusual properties [6–10] and especially the anomalously low conductivity values: 0.1 $(\Omega\,\mathrm{cm})^{-1}$ in AlPdRe at low temperature [5]. Hence one of the interesting experimental questions to verify is to see if this complicated structure will follow—or not—Ohm's law over a wide range of bias voltages. What are the electrical characteristics of the quasicrystalline structure?

Deviations from Ohm's law have been observed in other complicated structures. The diode is of course the best known case, but very peculiar *I-V* characteristics have also been observed in mesoscopic systems [11], AsGa/(AlGa)As heterostructures [12], granular Ag thin films [13], or Hg metallic wires embedded in porous substances [14]. In the latter case the authors suggested that the metallic filaments were actually not continuous but presented some breaks, and the voltage dependence of the conductivity was thus attributed to tunneling effects (in the presence of electron-electron interactions) through those breaks.

Actually a somewhat equivalent model has been proposed by Philips and Rabe to explain the unusually low conductivity of AlCu(Fe,Ru) quasicrystals [10]. Indeed those authors suggested that the transport properties of quasicrystals may be due to the existence of two main building blocks in the sample: an icosahedral one filling most of the volume whose properties would be similar to those of disordered systems and a highly resistive thin layered compound derived from the Al_7Cu_2Fe structure dispersed throughout the sample as a contaminating phase and surrounding the icosahedral one. In this case, the conductivity would be limited by tunneling effects through those insulating walls which would be equivalent to the breaks in the Hg wires. The voltage dependence of the conductivity for tunneling in the presence of electron-electron interaction effects (it has been shown that the temperature and magnetic field dependence of the conductivity of quasicrystals can be described by quantum interference effects, including strong electron-electron interaction effects [1]) has been calculated by Altshuler and Aronov [15], who predicted that the electrical conductance (ΔG) normalized to the conductance at zero electric field (G_0) is proportional to the square root of the bias voltage: $\Delta G/G_0 = k(D^{-3/2})(1/\nu_0)V^{1/2}$, where ν_0 is the density of states at the Fermi level, D is the diffusivity, and k is a numerical factor which will depend, in our case, on the random distribution of junctions in the sample. Those anomalies are expected to show up in the mV voltage range; however, the presence of a large number of junctions in the sample (if they exist) will split up the bias voltage, and thus we could only expect to see some deviations if the number of those junctions is less than 10 V/1 mV $\sim 10^4$ or if the size of the quasicrystalline domains is larger than about $L/10^4 \sim 1000$ Å (L is the length of the circuit $\sim$1 mm).

Generally speaking, the tunneling conductivity through an insulating interface will depend on the bias voltage if the density of states (DOS) presents some anomaly on an energy scale comparable to that of the bias voltage. As mentioned above the presence of electron-electron interactions will induce an anomaly in the DOS. Moreover, in the case of quasicrystals Janot and de Boissieu [7] proposed a model for the DOS based on a hierarchy of clusters also showing an asymmetric $(1 - \varepsilon/\varepsilon_F)^{1/2}$ anomaly close to the Fermi energy ε_F, and the calculated DOS of approximants presents very spiky structures with strong variations of the DOS on an energy scale of 10 meV or less [8]. Any of those anomalies in the DOS should lead to nonlinear effects in the *I-V* characteristics in the presence of tunneling interfaces at appropriate bias voltages.

On the other hand, material specific models have been developed trying to explain the unusual properties of quasicrystals. We did already mention the model proposed by Janot and de Boissieu [7] as well as the *ab initio* calculations by Fujiwara, Yamamoto, and Trambly de Laissardiere [8]. Other models have been proposed based on the similarity to Hume-Rothery alloys

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[6] or in terms of electronic hopping between structural entities separated by about 20 Å [9]. Those models are assuming that the linear response approximation is valid and thus that Ohm's law will be satisfied. Hence, measurements of the *I-V* characteristics of quasicrystals will give strong indications about the validity of the structural model proposed by Philips and Rabe [10] or else support the existence of a more material specific model. These can be done with high precision for a wide range of bias voltages on thin films of quasicrystals as small electrical circuits can be fabricated photolithographically yielding very high electrical resistances ($\sim 1\ k\Omega$).

Al, Fe, and Cu layers were sputtered consecutively on $SrTiO_3$ substrates using a rf magnetron sputtering system. The substrate was fixed to the sample holder with grease so that the temperature did not rise above 100 °C during the sputtering. The base pressure was around 10^{-7} Torr, and an internal liquid nitrogen cold trap prevented contamination during the deposition of the film. A few mTorr of high purity argon was used as sputtering gas. The most important point to control was the thickness of the different layers, i.e., the composition of the film. Indeed, pure quasicrystalline samples can only be obtained in a very narrow composition range around $Al_{62.5}Cu_{25}Fe_{12.5}$ [16], corresponding to a 7.0/2.0/1.0 thickness ratio for the Al, Cu, and Fe layers, respectively. Films were made by keeping the Al layer thickness at 2100 Å and varying slightly the Cu and Fe layers thickness ($590 < \text{Cu} < 630$ Å, $290 < \text{Fe} < 320$ Å) until the correct composition was found. To ensure good thickness homogeneity the sample holder was oscillated back and forth at 0.5 Hz (i.e., 30°) below the target during the deposition. The films were subsequently annealed in a quartz tube under high vacuum (10^{-6} Torr) first at 350 °C overnight and then at 600 °C for 2 h. More details about the preparation procedure have been given elsewhere [17]. It has been shown in bulk materials that the resistivity of quasicrystals are very sensitive to the structural quality and the purity of the samples. We thus made, as a first characterization, resistivity measurements of our films. Several samples were obtained with resistivities varying from 2000 to 3000 $\mu\Omega$ cm at room temperature. Those films also presented strong negative temperature dependences of the resistivity similar to those observed in bulk quasicrystalline samples, $1.5 < \rho(4\text{ K})/\rho(300\text{ K}) < 2$ leading to resistivities at 4 K between 3000 and 6000 $\mu\Omega$ cm (see, for instance, Fig. 1). Such resistivities are, within less than a factor of 2, comparable to the best AlCuFe bulk samples. Note that a very small amount of contaminating crystalline phase leads to resistivities less than 1000 $\mu\Omega$ cm. Resistivity measurements are thus a very good characterization technique, and the differences between our samples and the best bulk ones are possibly due to some trapped defects during the fabrication of the film. X-ray diffraction patterns [17] performed on some of the samples confirmed the quasicrystalline structure of the films.

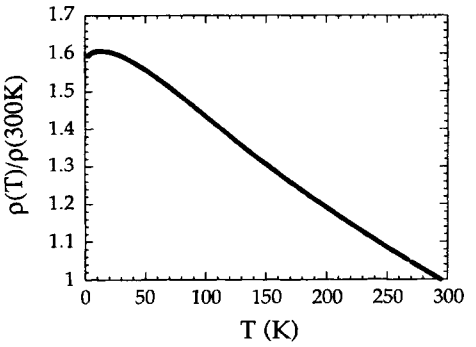

FIG. 1. Temperature dependences of the resistivity normalized at 300 K of an AlCuFe quasicrystalline thin films, $\rho_{300\text{ K}} \sim 2700\ \mu\Omega$ cm.

For photolithography purposes, mask patterns were generated by a computer and pictures of those masks were taken on ultraflat transparent photographic plates. The pattern of the mask was generated on the film using conventional photolithography techniques. The unprotected part of the film was then chemically etched. The picture of the circuit used for our measurements as well as the drawing of the mask are represented in Fig. 2. This circuit consists of a very thin line (20 μm × 3000 Å) having an electrical resistance of about 1 kΩ at room temperature for a resistivity of 2700 $\mu\Omega$ cm. The temperature dependence of the resistivity normalized to 300 K of this circuit is represented in Fig. 1.

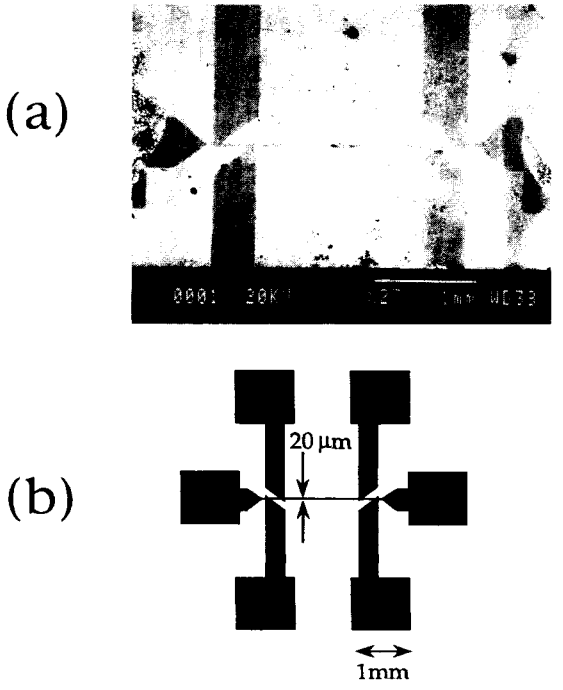

FIG. 2. Scanning electron microscope picture of an AlCuFe circuit (a) and drawing of the mask used for the fabrication of the circuit (b).

Silver paint was used to ensure a good electrical contact between the film and the small contact pins of the resistance probe. The circuit was cooled down to 4.2 K and the voltage was measured for currents ranging from 1 nA to 10 mA in both positive (V_+) and negative (V_-) directions. The results are represented in Fig. 3. They show that our quasicrystalline samples follow perfectly Ohm's law for 7 orders of magnitude change in the bias voltage ranging from 1 μV to 10 V. Neither a significant difference between V_+ and V_- nor any evidence for a voltage threshold was observed (inset, Fig. 3). The derivative of the I-V curve has also been measured by adding a small ac modulation dI to the dc current and measuring the corresponding change in voltage dV across the sample. We did not observe any voltage dependence of the conductivity up to a bias voltage of 10 V ($\Delta G/G_0 < 2 \times 10^{-4}$), suggesting that there are no tunneling interfaces in our samples (for which $\Delta G/G_0 \sim V^{1/2}$ is expected, as shown above). This indicates that the model proposed by Philips and Rabe may not be applicable here and that the unusual properties of quasicrystals should be explained by a model directly related to their peculiar structure [6–9].

As pointed out by Shockley [18], Ohm's law is actually only a linear approximation that is expected to fail at high enough electric fields. Deviations from Ohm's law have indeed been observed in Ge above 10^2 V/cm and interpreted in terms of increase in the electron energy and scattering by photons. This critical field corresponds, however, only to our maximum field, and even for the highest bias voltages we are thus probably still in the low-field limit. We could also have expected to find some nonlinearity related to energy (i.e., voltage) dependence of the conductivity predicted by Fujiwara, Yamamoto, and Trambly de Laissardiere [8] (on an energy scale of about 10^{-2} eV), but the energy change of the electrons due to our measuring electric fields E is only of the order of $\Delta\varepsilon \sim eEL_0$, where L_0 is the electronic mean free path, following Mayou *et al.* [9] for $L_0 \sim 20$ Å, we get $\Delta\varepsilon \sim 10^{-5}$ eV.

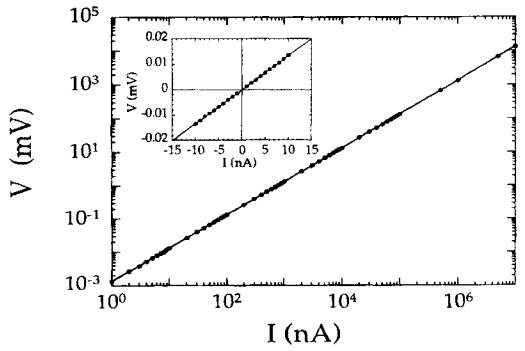

FIG. 3. V versus I characteristics of an AlCuFe quasicrystalline thin film cooled down to 4.2 K showing that this sample follows perfectly Ohm's law for 7 orders of magnitude bias voltage change; the slope of the log-log plot is equal to 1.0. In the inset, V versus I characteristic in the low electric field limit.

Besides those "intrinsic" deviations, other secondary effects may also occur at high electric fields such as heating or interband transition induced by the field. A rough estimate for the electric field necessary to induce such a transition is given by $eEh\upsilon_F = (\varepsilon_{gap})^2$ where ε_{gap} is the energy gap (~20 meV in quasicrystals according to Ref. [8]) and υ_F the Fermi velocity. We can estimate the Fermi velocity from the diffusivity $D = \upsilon_F L_0/3 = 0.3$ cm^2/s, following Mayou *et al.* [9] for $L_0 \sim 20$ Å, we get $\upsilon_F \sim 5 \times 10^6$ cm/s. An electric field of $E \sim 10^5$ V/cm would thus be necessary to induce those transitions in quasicrystals, and with our measuring fields we would only be sensitive to gaps smaller than about 1 meV.

Finally, we note that deviations from Ohm's law have been observed in metallic thin films due to either (i) electrical breakdown of insulating junctions between metallic grains as the voltage is increased (as observed in ultrathin Ag layers [13]) or (ii) local fusing of the links between grains due to Joule heating [19]. We could also have expected to find such effects due to the granular morphology of our films. Indeed scanning electron microscope pictures reveal that our films are composed of small grains ranging from about 500 to about 5000 Å in diameter. The absence of any deviations from Ohm's law thus shows that grain boundaries do not affect the electronic properties of our samples.

Despite all their peculiar electronic properties, we have shown here that the electrical behavior of AlCuFe quasicrystalline thin films is perfectly linear for a 7 orders of magnitude bias voltage change with no evidence for any voltage threshold or tunneling effects through insulating interfaces, if they exist, between quasicrystalline domains. Those measurements thus support the existence of a material specific model for the electronic properties of quasicrystals. We feel that the Ohmic behavior observed in our films is probably common to all quasicrystals.

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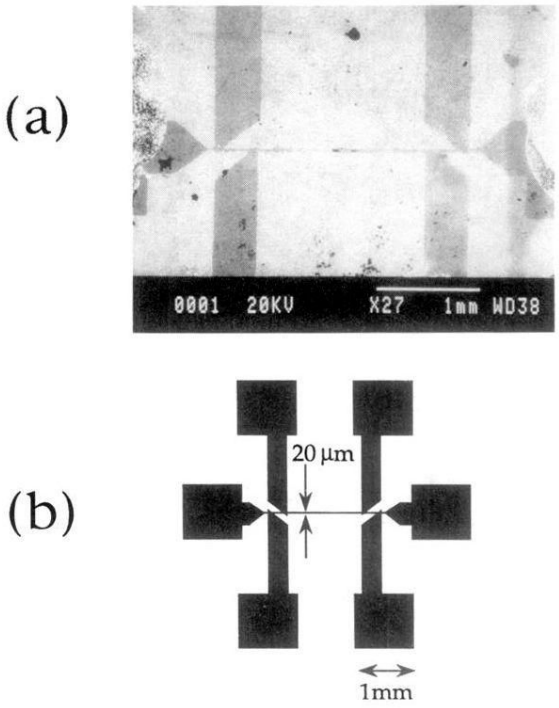

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