Strong electron-electron interaction effects in highly resistive Al-Cu-Fe icosahedral phases

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We have analyzed the temperature and magnetic-field (up to 35 T in pulsed fields) dependences of the conductivity of pure Al-Cu-Fe quasicrystalline samples. The results of this analysis are consistent with predictions of weak-localization and electron-electron interaction theories. A maximum is observed in the high-temperature magnetoconductivity, which could be due to competing effects between these two contributions. The more resistive sample shows a peculiar low-temperature behavior, which could be attributed to band-structure effects.

The thermodynamically stable[1] Al-Cu-*M* (M=Fe,Ru,Os) icosahedral phases show remarkable structural quality[2] (very low density of defects, absence of phason strain) and may be obtained as single grains.[3] They are thus of great interest for the study of the specific properties of the quasicrystalline structure. The most salient feature is that they show very high resistivity values at 4 K: up to 10 000 $\mu\Omega$ cm for $Al_{62.5}Cu_{25}Fe_{12.5}$,[4] and even 30 000 $\mu\Omega$ cm for $Al_{65}Cu_{20}Ru_{15}$.[5] These values are associated with a reduced density of states at the Fermi level[4] which could be due to peculiar Bragg-like diffractions by analogy to Hume-Rothery rules. We report here on the first high-magnetic-field measurements (up to 35 T in pulsed magnetic fields) performed on pure Al-Cu-Fe quasicrystalline samples of high structural quality. A striking point is that, despite these very high resistivity values, the temperature and magnetic field dependences of the conductivity (σ) can be well described by quantum interference effects[6] (weak localization and electron-electron interactions) originally developed for disordered systems. However, our more resistive quasicrystalline sample presents a peculiar low-temperature behavior, which could be attributed to band-structure effects.

Master ingots of composition $Al_{63}Cu_{25}Fe_{12}$ and $Al_{62}Cu_{25.5}Fe_{12.5}$ were prepared by melting high-purity elements in an arc furnace under argon atmosphere. Thin ribbons (1mm×1 cm×30 μm) of pure icosahedral phase were then prepared by melt spinning. However. the as-quenched samples present structural defects and an additional cubic Al-Fe-type crystalline phase (~5%). The ribbons are thus subsequently annealed under vacuum for a few hours at 800 °C in order to obtain pure icosahedral phases of high structural quality. The purity and quality of the materials were confirmed by x-ray diffraction using the Cu Kα radiation as shown in Fig. 1.

The resistivity was measured using a classical four-probe method down to 300 mK in static magnetic fields up to 8 T between 1.8 and 110 K in pulsed magnetic fields up to 35 T. Both measurements give the same results in the low-field limit (< 8 T). The resistivity at 4 K depends strongly on the composition, ranging from 4300 $\mu\Omega$ cm in $Al_{63}Cu_{25}Fe_{12}$ to 7800 $\mu\Omega$ cm in $Al_{62}Cu_{25.5}Fe_{12.5}$. The variation of the conductivity ($\sigma = 1/\rho$) with temperature is shown in Fig. 2. This dependence can be well described by quantum interference effects[6] using a classical fitting procedure of the form $\Delta\sigma(T) = 3[a + (bT)^2]^{1/2} - bT + c\sqrt{T}$ (1) between 0.3 and 100 K. The two first terms refer to weak localization effects and a and b are related to spin-orbit ($\tau_{\mathrm{s.o.}}$) and inelastic scattering (τ_i) times, respectively,

$$a = (e^2/2\pi^2 h)^2 D\tau_{\mathrm{s.o.}}, \quad bT = (e^2/4\pi^2 h)\sqrt{D\tau_i} \; .$$

In the case of $Al_{63}Cu_{25}Fe_{12}$ we could estimate the diffusivity D by using the measured value of the density of states at the Fermi level (specific-heat measurements[4])

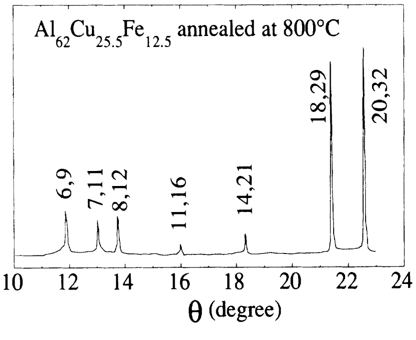


FIG. 1. X-ray diffraction pattern of i-$Al_{62}Cu_{25.5}Fe_{12.5}$ annealed at 800 °C for 3 h. (Indexing scheme of Ref. 16.)

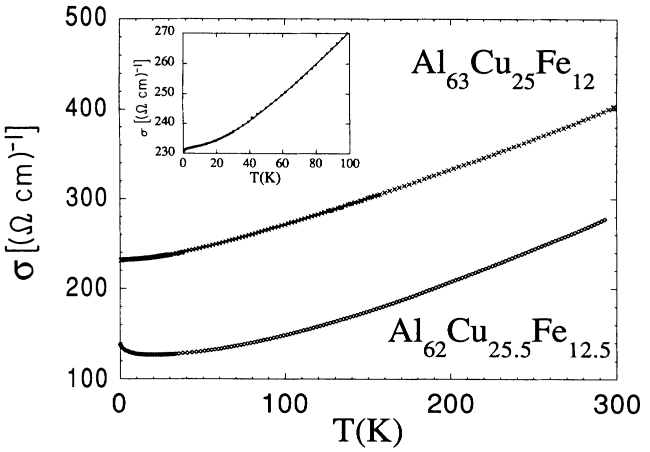


FIG. 2. Temperature dependence of the conductivity of i-Al$_{62}$Cu$_{25.5}$Fe$_{12.5}$ and i-Al$_{63}$Cu$_{25}$Fe$_{12}$. In the inset, the line is the fit from quantum interference effects (see text) for i-Al$_{63}$Cu$_{25}$Fe$_{12}$ between 0.3 and 100 K.

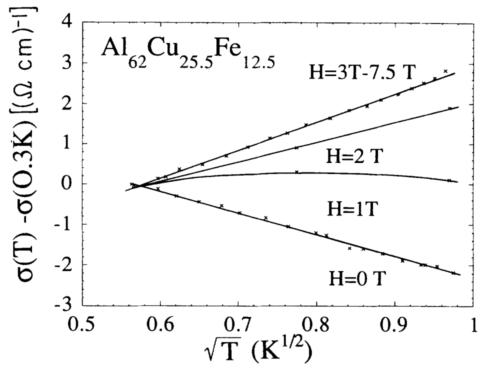


FIG. 3. Low-temperature conductivity as a function of $\sqrt{T}$ at different magnetic fields for i-Al$_{62}$Cu$_{25.5}$Fe$_{12.5}$.

to $D=\sigma/e^2N(E_F)\sim 0.3$ cm^2/s, and we then get $\tau_i\sim 2\times 10^{-9}$ T^{-2} s and $\tau_{\text{s.o.}}\sim 4\times 10^{-12}$ s. The value of $\tau_{\text{s.o.}}$ is typical of amorphous systems,[7] whereas τ_i is one order of magnitude higher than usually observed in these systems.

The $\sqrt{T}$ term in the conductivity is attributed to electron-electron interactions:

$$c=2.1(\tfrac{4}{3}-\tfrac{3}{2}\lambda F_\sigma)\sqrt{D}\ ,$$

where F_σ is a screening factor[6] $(0<F_\sigma<0.93)$ and λ a parameter introduced to take into account band-structure effects[8] (mass anisotropy and intervalley scattering effects). c is then usually positive in amorphous systems $(\lambda=1)$ and negative is heavily doped semiconductors on the metallic side of the metal insulator transition[7,8] $(\lambda>1)$. A negative value of the c coefficient is observed in the highly resistive Al$_{62}$Cu$_{25.5}$Fe$_{12.5}$ sample. Moreover, in that case, c is magnetic-field-dependent, as observed in heavily doped semiconductors[9] and predicted by electron-electron interactions theories[10]: $\Delta\sigma(T)=c(H)\sqrt{T}$, with $c(H)$ being negative at zero magnetic field and a positive constant at sufficiently high field such as $g^*\mu_BH>kT$ and $g^*\mu_BH>h/\tau_{\text{s.o.}}$ where g^* is the effective Landé factor ($g^*\sim 2$ in the free-electron limit but could be much higher in our system[3]). This peculiar low-temperature dependence of σ in the Al$_{62}$Cu$_{25.5}$Fe$_{12.5}$ samples is shown in Fig. 3 ($c>0$ and constant for $H>3$ T) and can be attributed to band-structure effects in this high-resistivity sample. On the other hand, the c coefficient is positive in Al$_{63}$Cu$_{25}$Fe$_{12}$. This change of sign from a negative to a positive value may be due to a decrease of the screening factor F_σ with decreasing resistivity and/or to a lower value of the band-structure parameter λ in this less resistive sample.

The magnetic-field dependence of the conductivity is presented in Fig. 4 for both samples. The order of magnitude of $\Delta\sigma(H)$ is comparable to that observed in amorphous systems[7] and $\Delta\sigma(H)$ can be analyzed by nonmagnetic weak localization (including Zeeman spin splitting)[11] and electron-electron interaction[12] theories. Indeed, we have shown in a previous paper[4] that the Al$_{63}$Cu$_{25}$Fe$_{12}$ sample shows a diamagnetic contribution at high field (1–2 T, 4–300 K) in agreement with values published on other compositions[13] and our preliminary susceptibility measurements on Al$_{62}$Cu$_{25.5}$Fe$_{12.5}$. Thus we do not expect to find magnetic contributions to $\Delta\sigma(H)$. Figure 5 presents schematically the contributions to the magnetoconductivity due to weak-

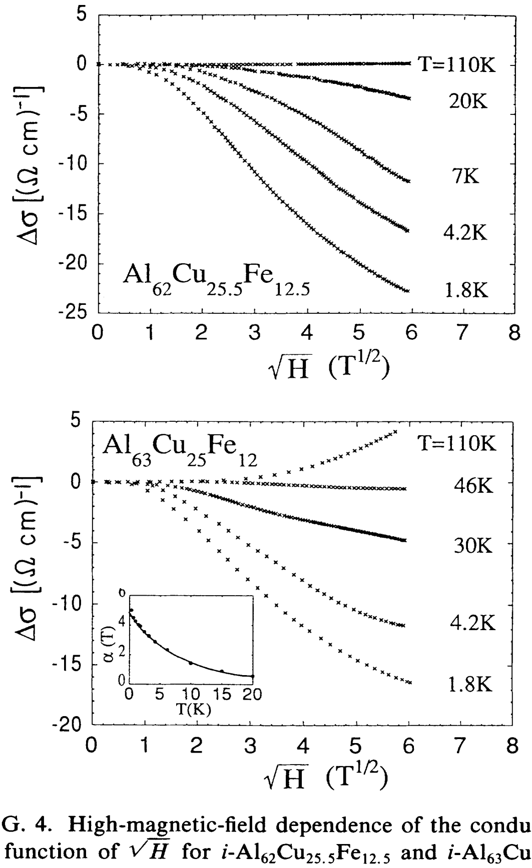
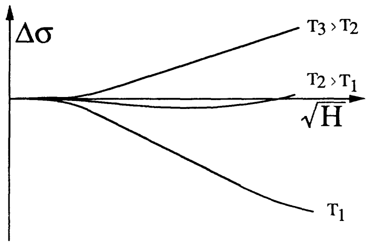


FIG. 4. High-magnetic-field dependence of the conductivity as a function of $\sqrt{H}$ for i-Al$_{62}$Cu$_{25.5}$Fe$_{12.5}$ and i-Al$_{63}$Cu$_{25}$Fe$_{12}$. In the inset: temperature dependence of the slope $\alpha(T)=d(\Delta\sigma(H))/d(\sqrt{H}\,)$ for i-Al$_{63}$Cu$_{25}$Fe$_{12}$ (the line is a guide for the eyes).

(a) weak-localisation part



(b) electron-electron interaction part

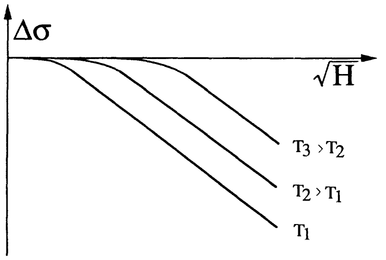


FIG. 5. Schematical contributions to the magnetoconductivity from weak-localization (a) and electron-electron (b) interactions.

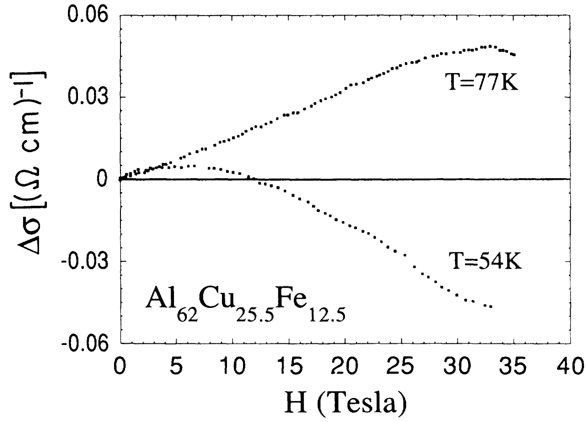


FIG. 6. High-temperature magnetoconductivity in *i*-$Al_{62}Cu_{25.5}Fe_{12.5}$ showing a maximum as a function of the applied field.

localization and electron-electron-interaction effects: there is a magnetic-field range where both theories predict a $\sqrt{H}$ dependence of the conductivity. This behavior can be clearly seen in Fig. 4 with a slight saturation at low temperature and high field. Moreover, the slope $d(\Delta\sigma)/d(\sqrt{H})$ is temperature dependent.

For the weak-localization part of the magnetoconductivity (MC) this temperature dependence of the slope can be attributed to Zeeman-spin-splitting effects as already observed by Lindqvist and Rapp in amorphous Cu-Ti.[14] Indeed, these authors have shown by numerical calculations that for low-diffusivity alloys ($D<1$ cm^2/s) Zeeman-splitting effects dominate the weak-localization part of the MC at low temperature. There is then a magnetic-field range where $\Delta\sigma(H)=-A(T)\sqrt{H}$ with a theoretical value of $A(T=0K)=1.4/\sqrt{D}$ where D is the diffusivity.

For the electron-electron interactions part, Lee and Ramakrishnan[12] have shown that for $H \gg kT/g\mu_B$, $\Delta\sigma(H)=-B\sqrt{H}$ with $B=1.8F_\sigma/\sqrt{D}$ [F_σ is the screening factor, see $\sigma(T)$]. From the values of $D\sim 0.3$ cm^2/s and $F_\sigma \sim 0.65$ deduced from $\sigma(T)$ (with $\lambda=1$) in $Al_{63}Cu_{25}Fe_{12}$ we get $A(T=0\text{ K})\sim 2.5\ (\Omega\,\text{cm})^{-1}\ \text{T}^{1/2}$ and $B\sim 2.2\ (\Omega\ \text{cm})^{-1}\text{T}^{1/2}$. Assuming that these two contributions are additive we have $\Delta\sigma(H)=-[A(T)+B]\sqrt{H}$ with a theoretical value of $A(T=0K)+B=4.7\ (\Omega\,\text{cm})^{-1}\text{T}^{1/2}$. This value is in good agreement with the measured value of $5\ (\Omega\,\text{cm})^{-1}\text{T}^{1/2}$ at 0.3 K.

Figure 6 presents the magnetoconductivity at high temperature for $Al_{62}Cu_{25.5}Fe_{12.5}$. We can observe a rather unexpected feature with the MC being positive at low field and negative at higher field (the same behavior is observed in $Al_{63}Cu_{25}Fe_{12}$). Such a behavior can be attributed to competing effects between weak-localization and electron-electron interactions. Indeed, at high temperature ($\tau_i/\tau_{\text{s.o.}}<0.1$) inelastic scattering destroys the Zeeman-splitting effects and the weak-localization part of the magnetoconductivity becomes positive (Fig. 5, temperature T_3). On the other hand, the electron-electron interactions part remains negative but is shifted to higher fields (Fig. 5, $H>kT/g\mu_B$). Thus, competing effects between these two contributions could lead to the magnetoconductivity being positive at low field (weak-localization effects) and negative at higher fields (electron-electron interaction effects). This effect is usually not observed in disordered systems[7] for which electron-electron interactions are negligible at these temperatures. The MC behavior of our Al-Cu-Fe icosahedral phases is thus a remarkable feature that can be understood by strong electron-electron interaction effects if one assumes no magnetic contributions. Strong electron-electron interaction contributions at high temperature have also been observed in other high resistivity samples such as Si based amorphous alloys[15] or heavily doped semiconductors.[9] At higher temperature the electron-electron contribution is shifted to very high fields and we get a positive magnetoconductivity characteristic of weak-localization effects (Fig. 4). For a more detailed analysis of the MC curves we must take care of the fact that these theories are perturbation developments theoretically valid only in the $k_F l \gg 1$ limit, whereas for our high-resistivity samples $k_F l \sim 1$.

In conclusion, the Al-Cu-Fe alloys allowed us for the first time to obtain very high structural quality icosahedral phases. These samples show very high resistivity values at 4 K for 4600 $\mu\Omega$ cm and 7800 $\mu\Omega$ cm for $Al_{63}Cu_{25}Fe_{12}$ and $Al_{62}Cu_{25.5}Fe_{12.5}$, respectively. Despite these high resistivities the temperature and magnetic-field dependence of the conductivity can be analyzed through quantum interference theories. The magnetic-field dependence of the conductivity at 50 K can be attributed to strong electron-electron interactions, and the more resistive sample presents a peculiar low-temperature behavior, which could be due to band-structure effects, as those observed in heavily doped semiconductors close to the metal-insulator transition.

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