Evidence for Unconventional Electronic Transport in Quasicrystals

D. Mayou, C. Berger, F. Cyrot-Lackmann, T. Klein, and P. Lanco
Laboratoire d'Etudes des Propriétés Electroniques des Solides, Centre National de la Recherche Scientifique,
BP 166, 38042 Grenoble Cedex 9, France

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The conductivity σ of a series of AlCuFe and AlPdMn quasicrystals and approximant phases can be written $\sigma(T)=\sigma_{4\mathrm{K}}+\delta\sigma(T)$. The $\sigma_{4\mathrm{K}}$ term is very low, decreases with improvement of structural quality, and varies strongly with composition. $\delta\sigma(T)$ is independent of the sample and increases anomalously with temperature up to 1000 K, with $\sigma_{1000\mathrm{K}}/\sigma_{4\mathrm{K}}$ greater than 10. The results are interpreted by electronic hopping between structural entities separated by about 30 Å. Within a band picture, this process would correspond to interband transitions.

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The AlCuFe, AlCuRu, and AlPdMn systems make accessible very high structural quality quasicrystalline (QC) phases [1], and also allow us to get very close approximants in the case of AlCuFe [2]. For these QC phases, anomalous transport [3–6] and optical properties [7] were measured, with high resistivity values up to 30000 $\mu\Omega$ cm, close to a metal-insulator transition. A low density of states at the Fermi level, a small number of carriers, strong electron interactions, and a diamagnetic behavior also are indications of the proximity of this transition. The temperature and magnetic field dependences of conductivity at low temperature are well described [8] by quantum interference effects [9], which was at first surprising in view of the strong resistivities. Further, the increase of resistivity due to removing defects seems to be a common feature of these systems, and has not yet received satisfactory explanation. Moreover, we note that from published results in AlCuFe [5,10], the drastic changes of resistivity with concentration and structural quality are observed for samples having all the same density of states, within a few percent.

On the theoretical side, except for the structural model of Phillips and Rabe [11], electronic properties have been studied by similarity with Hume-Rothery alloys [12]. In particular the existence of a pseudogap near the Fermi level due to diffraction by Bragg planes seems determined, and has been confirmed by *ab initio* calculations on approximant phases [13,14]. The scattering by d states was also shown to be important [15]. The band structure due to the strong interaction between the Fermi surface and the Brillouin zone is peculiar: In particular it was suggested that the properties could vary rapidly with the Fermi energy on a scale of a tenth of an eV [5,13] and that the band structure could be very sensitive to temperature via the variation of the Debye-Waller factor [16]. However, these theories do not fully take into account the QC character of the systems. Studies on tight-binding (1D and 2D) models [13,17] have shown that the eigenstates are critical. They are mainly localized around local environments with reappearances in similar arrangements of the structure, but the envelope of the wave function decreases like $1/r^{\alpha}$. Sire and co-workers [18] have shown that the propagation of a wave packet in the 2D lattice is then neither ballistic nor diffusive. This bad propagation is consistent with flat bands (i.e., small group velocities) observed in band structure calculations of approximants. In 3D critical states could also exist [19]. Clearly these effects can only be seen experimentally in systems where the order is realized on a sufficiently long scale.

In this paper we will first present new results of conductivity σ on quasicrystals as well as approximant phases. We then interpret our results by an unconventional electronic transport, due to scattering induced hopping between structural entities separated by about 30 Å.

We produced icosahedral samples of about twenty different compositions close to $\mathrm{Al_{63}Cu_{25}Fe_{12}}$ (Cu concentration in the range 24–25.5 at.%; Fe concentration in the range 12–13 at.%) by melt spinning and subsequent annealing as described elsewhere [3]: The ribbons annealed at 800 °C are pure icosahedral phase (*i* phase) of high structural quality, whereas by annealing at 600 °C defects are retained in the pure *i* phase. We also measured two slightly different compositions of the rhombohedral approximant R phase ($\mathrm{Al_{61.8}Cu_{26}Fe_{11.2}}$ and $\mathrm{Al_{61.5}Cu_{26.5}Fe_{11}}$) elaborated and characterized at the Centre d'Etudes de Chimie Metallurgique laboratory [20]. An *i* phase was obtained at the same composition ($\mathrm{Al_{61.8}Cu_{26}Fe_{11.2}}$) by using the transformation of the approximant into an *i* phase above 700 °C.

The resistivity measurements were performed by using a four probe method down to 300 mK and up to 1000 K.

The temperature dependence of conductivity $\sigma(T)$, shown in the inset of Fig. 1 for different AlCuFe samples, presents a very surprising feature: The $\sigma(T)$ curves are *parallel* for all the samples measured (note all measured samples are not presented here, but they all behave the same). Thus $\sigma(T)$ can be written: $\sigma(T)=\sigma_{4\mathrm{K}}+\delta\sigma(T)$. This means that the already well known low conductivity values [3–5] (down to 100 $\Omega^{-1}\mathrm{cm}^{-1}$ in $\mathrm{Al_{62.5}Cu_{25}Fe_{12.5}}$ annealed at 800 °C, corresponding to high resistivities up to 10000 $\mu\Omega$ cm) are due to the $\sigma_{4\mathrm{K}}$ term. As shown in the inset of Fig. 1, it is the decrease of the $\sigma_{4\mathrm{K}}$ term that accounts for the conductivity decrease when structural defects are removed, which is in striking contrast to the normal behavior of metals, as already pointed out [3,5].

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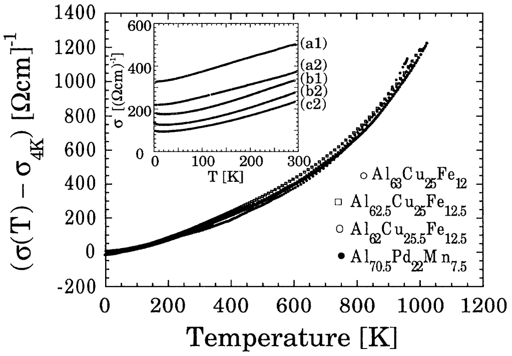

FIG. 1. Temperature dependence part of the conductivity: $\delta\sigma(T) = \sigma(T) - \sigma_{4K}$ up to 1000 K for $Al_{63}Cu_{25}Fe_{12}$, $Al_{62}Cu_{25.5}Fe_{12.5}$, $Al_{62.5}Cu_{25}Fe_{12.5}$, and $Al_{70.5}Pd_{22}Mn_{10.5}$. Inset: Conductivity $\sigma(T)$ for AlCuFe samples. (a) $Al_{63}Cu_{25}Fe_{12}$, (b) $Al_{62}Cu_{25.5}Fe_{12.5}$, (c) $Al_{62.5}Cu_{25}Fe_{12.5}$, annealed at (1) 600 °C, (2) 800 °C.

While σ_{4K} depends strongly on composition and structural quality, $\delta\sigma(T)$ is almost independent of the sample as seen in Fig. 1. At low temperature $\delta\sigma(T)$ is weak and of the same order of magnitude as in amorphous systems. Furthermore the $\sigma(T)$ curves for all the high quality AlCuFe *i* phases remain almost parallel up to 1000 K. The same temperature dependence is also observed in the high quality AlPdMn *i* phases as exemplified in Fig. 1 for $Al_{70.5}Pd_{22}Mn_{7.5}$. Such additive conductivities are just opposite to the classical Matthiessen rule for impurities dissolved in a metal, for which the resistivities add $[\rho(T) = \rho_{\text{impurity}} + \delta\rho(T)]$, this latter rule coming from the additivity of the inverse scattering time τ and from ρ proportional to $1/\tau$.

We also find (see Fig. 2) the remarkable fact that the very same $\sigma(T)$ is measured in *i*- and approximant *R*-$Al_{61.8}Cu_{26}Fe_{11.2}$ and *R*-$Al_{61.5}Cu_{26.5}Fe_{11}$ phases. On the contrary the crystalline ω-$Al_{70}Cu_{20}Fe_{10}$ phase of close composition presents a $\sigma(T)$ typical of a metal with both a positive temperature coefficient and a low resistivity value: $\rho_{4K} \sim 20$ $\mu\Omega$ cm [21].

We emphasize that $\sigma(T)$ at high temperature (i.e., $T \gtrsim \Theta_D/3$, with $\Theta_D \sim 400$ K [10]) is very anomalous also for QC as for approximant phases. Indeed $\sigma(T)$ *increases* up to the highest temperature of measurement (1000 K). As a comparison, for heavily doped semiconductors, which have about the same order of σ values, $\sigma(T)$ decreases at high temperature when all the impurity levels are ionized. Further, as shown in the inset of Fig. 2, the T dependence of σ is *not* simply thermally activated: The conductivity fits either with $\sigma(T) = \alpha + \beta T^3$ or with $\sigma(T) = A \exp(BT)$. The parallelism of the $\sigma(T)$ curve is also difficult to understand with a classical thermally activated mechanism. Furthermore, σ_{4K} varies strongly with composition or structural quality whereas the density of states is nearly constant and is much higher than that of a semiconductor [3]. In disordered metallic alloys, $\sigma(T)$ increases with temperature, but more slowly above $\Theta_D/3$ [9]; finally in a weak localization scheme (see next), once all the interference effects are destroyed, one also recovers a normal metallic conductivity.

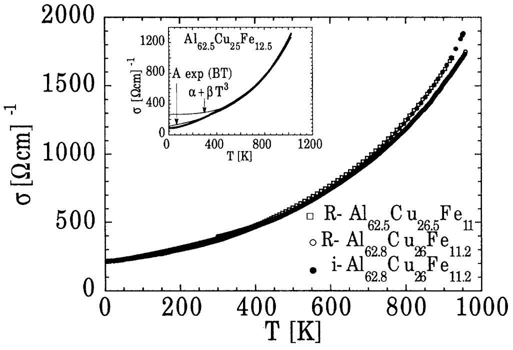

FIG. 2. Conductivity $\sigma(T)$ for *i* and *R*-$Al_{62.8}Cu_{26}Fe_{11.2}$, and *R*-$Al_{62.5}Cu_{26.5}Fe_{11}$. Inset: $\sigma(T)$ fits for *i*-$Al_{62.5}Cu_{25}Fe_{12.5}$, with $\alpha = 268$ $(\Omega\,\text{cm})^{-1}$, $\beta = 9.4 \times 10^{-7}$ $(\Omega\,\text{cm})^{-1}\text{T}^{-3}$; $A = 114$ $(\Omega\,\text{cm})^{-1}$, $B = 2.3 \times 10^{-3}\ \text{T}^{-1}$.

This leads us to conclude that two effects may dominate the transport properties depending on the temperature range. First below $T \sim 100$–200 K, the temperature and magnetic field dependences $\sigma(T,H)$ can be well described [8] by quantum interference effects (QIE) [9]. At higher temperature, QIE are expected to be destroyed; as a matter of fact, the $\sigma(T,H)$ curves cannot be fitted by QIE theories above ~ 100–200 K. The $\sigma(T)$ behavior, as well as changes in temperature dependences of Hall effect [5,22,23] and thermoelectric power [5], suggests that a second and unusual mechanism dominates for $T \gtrsim 100$–200 K which we now discuss.

In a classical band scheme the electronic conductivity is analyzed via the Bloch-Boltzmann theory. The basic assumption is that the charge carriers have a ballistic propagation between two scattering events and that after a few collisions the propagation becomes diffusive. We cannot exclude such a scheme, but the theoretical works mentioned in the introduction suggest that the nature of states and their propagation are different in quasiperiodic structures or periodic ones on length scale of several unit cells. Thus in the following, we assume that the propagation of wave packets on a length scale l is diffusive if $l > L_0$ (L_0 is the mean free path) but is unconventional, i.e., nonballistic, if $l < L_0$. We emphasize that the systems studied here have a high structural quality and a strong chemical order observed on length scales of several 10^2 Å [24]. Then it is reasonable to assume that the mean free path L_0 is of several interatomic distances and that the propagation on length scale $l < L_0$ is sensitive to the quasiperiodic order. This means that the quasicrystals considered here are quite different from amorphous metals in which the mean free path is of the order of the interatomic distance. The Hall effect which is also rather peculiar is discussed in more detail elsewhere [23] and is not contradictory with the explanation developed in the following, since its dependence on the Fe concentration suggests R_H is dominated by sp-d hybridization effects.

In the diffusive regime ($l > L_0$) quantum interference effects should occur as in any disordered systems. Indeed, QIE theories [9,22] are developed for electrons that are elastically scattered many times by impurities with a mean time τ_0 between collisions and which phase coherence is destroyed in times $\tau_i(T)$ (inelastic scattering by phonons) and $\tau_T(T) \approx \hbar/kT$ (two electron terms), with $\tau_i(T)$, $\tau_T(T) > \tau_0$. In i-AlCuFe, the agreement between QIE theories and experiments below 100 K [8] indicates many elastic collisions and thus a diffusive motion on the inelastic time scales $t > \tau_i$ (100 K), τ_T(100 K), i.e., $t > \tau_0 \sim 10^{-13}$ s or equivalently on length scale $l > L_0 \sim 20$–30 Å [$L_0 = (3D\tau_0)^{1/2}$ where D is the diffusivity [3]]. Note that in agreement with the very good long range order of our QC samples we thus find a fairly long scattering time τ_0 which is to be compared with a scattering time of typically $\tau \sim 10^{-15}$ s in amorphous metals.

As a consequence, the average velocity between two scattering events $v = L_0/\tau_0$ is $v = 3D/L_0 \gtrsim 0.3 \times 10^7$ cm/s which is typically a few percent of the Fermi velocity in aluminum ($v_F \sim 2 \times 10^8$ cm/s) and is more comparable to thermal velocities of electrons and holes in silicon at room temperature ($v_{\rm th} \sim 10^7$ cm/s).

The observed variations of σ up to 100–200 K, attributed to QIE, are only a fraction of σ [$\delta\sigma(T = 0$–200 K) ~ 40 $(\Omega\,{\rm cm})^{-1}$]. This shows that these systems are *not* near an Anderson transition. Indeed, from scaling theory of localization [25], QIE are expected to be important if the ratio $R = g/g_c$ (conductance of a cube with size L over the universal value g_c) is of order of unity. Using the conductivity σ_B of the cube, $\sigma_B = g/L_0$, and the Mott minimum metallic conductivity, $\sigma_{\rm Mott} = g_c/a \sim 200$ Ω^{-1} cm^{-1} where a is of order 3 Å or less [26], we get $R = g/g_c = (\sigma_B/\sigma_{\rm Mott})\,(L_0/a)$. The conductivity σ_B is obtained when quantum interferences are destroyed and is greater than the measured σ. Thus we find $R > (\sigma/\sigma_{\rm Mott})(L_0/a) \gg 1$. This means that these QC are not near the Anderson transition.

As QIE are small, it is tempting to analyze transport properties of approximants with the Bloch-Boltzmann approach, but this may be erroneous. Indeed 1D results [13,17] show that band widths vary as $\Delta\varepsilon \sim 1/L^{\alpha}$ (L is the size of the cell and $\alpha > 1$); thus wave packet velocities vary as $v \sim (1/L^{\alpha-1})$ tending to zero for large approximants. For 2D systems, the results of Sire and coworkers [18] also give a vanishing group velocity as L increases. Experimental results indicate also small velocities as presented above. Thus, the current due to the propagation of a wave packet (within the same band) between two scattering events is small, and in the Bloch-Boltzmann approach this current is the only source of diffusivity. However, an interband transition between two wave packets $\varphi(\mathbf{r}) = U_{n\mathbf{k}}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}g(r)$ and $\psi(\mathbf{r}) = U_{m\mathbf{k}'}(\mathbf{r})e^{i\mathbf{k}'\cdot\mathbf{r}}g'(r)$ (g and g' are two slowly varying envelopes) leads also to a displacement of charge if the centers of gravity of $U_{m\mathbf{k}'}(\mathbf{r})$ and $U_{n\mathbf{k}}(\mathbf{r})$, in the unit cell, are different. In approximants, by analogy with critical states of QC we expect that $U_{n\mathbf{k}}(\mathbf{r})$ is mainly localized around some local entities with reappearances in similar environments in the unit cell. When $U_{n\mathbf{k}}(\mathbf{r})$ and $U_{m\mathbf{k}'}(\mathbf{r})$ are nonzero in the same region of the unit cell, a local modification of the Hamiltonian by structural defects creates a coupling and allows a transition $U_{n\mathbf{k}}(\mathbf{r}) \rightarrow U_{m\mathbf{k}'}(\mathbf{r})$, which corresponds to an important charge displacement. Because the interband transitions are due to scattering, this mechanism leads to a diffusivity *increasing* with scattering. Finally we note that if several bands cross the Fermi level [13,14] elastic interband transitions are possible, which means that elastic scattering induces hopping and increases the conductivity. This is in contrast with the thermally activated hopping that occurs in impurity bands of semiconductors. We find (see below) that experiments are consistent with a mechanism of this kind, and no difference is expected between QC and good approximants containing few defects.

We now develop an interpretation of the experimental conductivity assuming that its variation is due to the scattering of eigenstates by slight imperfections of the structure. We consider scattering by impurities or defects (characteristic time $\tau_{\rm imp}$) which is the only one present at 4 K, and scattering depending on temperature (characteristic time $\tau_{\rm ph}$). Phonons, phasons [27], or variation of the average crystal potential due to the Debye-Waller factor may contribute to $\tau_{\rm ph}$. We expect the resulting scattering time τ to be given by $1/\tau = 1/\tau_{\rm imp} + 1/\tau_{\rm ph}$; the measured σ is a function of τ. Then $\sigma_{4{\rm K}}$ depends on $\tau_{\rm imp}$ and $\delta\sigma(T)$ only on $\tau_{\rm ph}$, since experimentally $\delta\sigma(T)$ is independent of structural quality. Thus $\sigma(\tau) = \sigma_{4{\rm K}}(\tau_{\rm imp}) + \delta\sigma(\tau_{\rm ph})$, where σ, $\sigma_{4{\rm K}}$, and $\delta\sigma$ are *a priori* unknown and different functions. An obvious solution is $\sigma(\tau) = \sigma_0 + a/\tau$ and it is straightforward to show that it is the only one. As σ is positive, we expect σ_0, a positive. This formula is then consistent with a σ decrease when τ increases, i.e., when structural quality improves or when temperature is lowered.

From the Einstein formula $\sigma = e^2 n(E_F)D$, with $D = L_0^2(\tau)/3\tau$ where $L_0(\tau)$ is the mean free path (i.e., the displacement between two scattering events) and $n(E_F)$ the nearly constant DOS in i AlCuFe, we get $\sigma = \alpha/\tau$ if $L_0(\tau)$ is nearly independent of τ. This is realized if scattering induces hopping between eigenstates which do not, or badly, propagate and which centers of gravity have a characteristic separation L_0. This corresponds to the interband transition scheme discussed above. More generally if hopping is possible between states separated by l_α with a probability per unit time $1/\tau_\alpha$ we get $D = \sum_\alpha l_\alpha^2/3\tau_\alpha$. We expect $1/\tau_\alpha = 1/\tau_{\alpha_{\rm ph}} + 1/\tau_{\alpha_{\rm imp}}$ and thus $\sigma = \sigma_{\rm imp} + \sigma_{\rm ph}$ in agreement with experiment.

If we suppose that current does not correspond to the interband transition described above but is due to the propagation of a wave packet between two scattering events, $L_0(\tau)$ nearly constant could also be explained.

Assuming the nonballistic law [18] $L_0(\tau) = A\tau^{\beta}$, we find that $L_0(\tau)$ is nearly constant if $\beta \sim 0$. However, the value $\beta \sim 0$ is obtained in the limits of the parameter range for the Hamiltonian in a 2D calculation, therefore we are more convinced by an interband picture.

It is interesting to note that modeling of AlCuFe and AlPdMn QC and approximants use similar structural entities about 30 Å apart and connected with one another [24]. Our estimated mean free path $L_0 \sim 20$–30 Å is consistent with these structural models if eigenstates near E_F are centered on these entities. The similar behavior of σ in AlCuFe and AlPdMn appears also more natural if hopping is the relevant mechanism.

We now briefly discuss optical conductivity. As one cannot define free electrons near E_F, we do not expect a Drude peak in the optical conductivity in agreement with experiment [7]. Further interband transitions between filled and empty states, which are possible at all energies in large approximants (see above), can explain the nonzero $\mathrm{Re}[\sigma(\omega)]$ at low frequencies.

To conclude, the measured conductivity σ of a series of AlCuFe and AlPdMn icosahedral and approximant phases can be written $\sigma(T) = \sigma_{4\mathrm{K}} + \delta\sigma(T)$, where $\sigma_{4\mathrm{K}}$ is very low, *decreases* with improvement of structural quality, and varies rapidly with composition. $\delta\sigma(T)$ is almost independent of the sample, and increases strongly up to 1000 K, with a dependence that cannot be ascribed to a thermally activated process. The high structural order of the samples and the observed quantum interference effects below $T \lesssim 100$–200 K are consistent with an electronic mean free path of the order of a few interatomic distances ($L_0 \sim 20$–30 Å, or a scattering time $\tau_0 \sim 10^{-13}$ s at low T). As a consequence, the average velocity between two scattering events $v = L_0/\tau_0$ is only a few percent of v_F in Al, and the QC conductivity is far from an Anderson transition. We propose that the electronic transport is due to hopping between eigenstates of the perfect structure which are centered on structural entities separated by about 30 Å. The coupling between two eigenstates is then due to a local modification of the Hamiltonian in regions where both eigenstates are nonzero. Within a band picture, this process would correspond to interband transitions. This mechanism could similarly describe QC and approximant phases. The nonballistic propagation of wave packets in the perfect structure could also lead to peculiar effects and will be discussed elsewhere [28].

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[1] Y. Calvayrac *et al.*, J. Phys. (Paris) **51**, 417 (1990); C. A. Guryan, A. I. Goldman, and P. W. Stephen, Phys. Rev. Lett. **62**, 2409 (1989).

[2] M. Audier *et al.*, Philos. Mag. **6**, 1375 (1991); S. Ebalard and F. Spaepen, J. Mater. Res. **5**, 62 (1990).

[3] T. Klein, C. Berger, D. Mayou, and F. Cyrot-Lackmann, Phys. Rev. Lett. **66**, 2907 (1991).

[4] B. D. Biggs, Y. Li, and S. J. Poon, Phys. Rev. B **43**, 8747 (1991); U. Mizutani, Y. Sakabe, and T. Matsuda, J. Phys. Condens. Matter **2**, 6153 (1990).

[5] S. J. Poon, Adv. Phys. **41**, 303 (1992).

[6] P. Lanco, T. Klein, C. Berger, F. Cyrot-Lackmann, G. Fourcaudot, and A. Sulpice, Europhys. Lett. **18**, 227 (1992).

[7] C. C. Homes *et al.*, Phys. Rev. Lett. **67**, 2694 (1991); E. Bustarret *et al.* (to be published).

[8] T. Klein *et al.*, Phys. Rev. B **45**, 2049 (1992); A. Sahnoune, J. O. Ström-Olsen, and A. Zaluska, Phys. Rev. B **46**, 10629 (1992).

[9] M. A. Howson and B. L. Gallagher, Phys. Rep. **170**, 265 (1988).

[10] K. Wang *et al.*, J. Phys. (Paris) **2**, 1553 (1992).

[11] J. C. Phillips and K. Rabe, Phys. Rev. Lett. **66**, 923 (1991); J. C. Phillips, Solid State Commun. **83**, 379 (1992).

[12] J. Friedel and F. Dénoyer, C. R. Acad. Sci. (Paris) **305**, 171 (1987); A. P. Smith and N. W. Ashcroft, Phys. Rev. Lett. **59**, 1365 (1987); S. Burkov, T. Timusk, and N. W. Ashcroft, J. Phys. Condens. Matter **4**, 9447 (1992).

[13] T. Fujiwara and H. Tsunetsugu, *Quasicrystals: The State of the Art*, edited by D. P. DiVincenzo and P. Steinhart (World Scientific, Singapore, 1991), p. 343.

[14] J. Hafner and M. Krajči, Phys. Rev. Lett. **68**, 2321 (1992).

[15] G. Trambly de Laissardiére, D. Mayou, and D. Nguyen Manh, Europhys. Lett. **21**, 25 (1993).

[16] N. W. Ashcroft (private communication).

[17] M. Kohmoto, L. P. Kadanoff, and C. Tang, Phys. Rev. Lett. **50**, 1870 (1983).

[18] B. Passaro, C. Sire, and V. Benza, Phys. Rev. B **46**, 13751 (1992).

[19] K. Niizeki and T. Akamatsu, J. Phys. Condensed Matter **2**, 2759 (1990).

[20] D. Gratias *et al.*, J. Non-Cryst. Solids **153 & 154**, 482 (1993).

[21] C. Berger *et al.* (to be published).

[22] R. Haberkern, P. Lindqvist, and G. Fritsch, in *Proceedings of the Fourth International Conference on Quasicrystals* (Ref. [18]); P. Lindqvist *et al.*, Phys. Rev. B (to be published).

[23] P. Lindqvist, C. Berger, T. Klein, P. Lanco, and F. Cyrot-Lackmann (to be published).

[24] Y. Calvayrac and D. Gratias (private communication).

[25] E. Abrahams *et al.*, Phys. Rev. Lett. **42**, 673 (1979).

[26] N. F. Mott, *Metal-Insulator Transition* (Taylor & Francis, London, 1990), 2nd ed.

[27] G. Coddens, R. Bellissent, Y. Calvayrac, and J. P. Ambroise, Europhys. Lett. **16**, 271 (1991).

[28] C. Sire and D. Mayou (to be published).