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Highly anisotropic electronic structure in decagonal quasicrystals and approximants

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We present detailed *ab initio* calculations of the electronic structure in decagonal quasicrystals and approximants and in crystalline compounds in the Al-Cu-Co and Al-Mn systems. The observed anisotropy of the electronic structure is discussed in relation to the anomalous transport properties of the decagonal quasicrystals. Our results can explain the appearance of large transport anisotropies in the quasicrystalline and approximant phases. The effect of disorder on the anisotropies has been investigated. [S0163-1829(98)04533-0]

I. INTRODUCTION

The discovery of quasicrystalline alloys has stimulated numerous experimental investigations of physical properties intrinsically related to the quasiperiodicity of the atomic structure. Transport properties in particular are expected to be affected by the loss of translational invariance. For icosahedral quasicrystals it has been shown that the low-temperature resistivity reaches several Ω cm, decreases almost linearly with temperature, and increases with structural perfection.[1–3] Decagonal quasicrystals show in addition a strong anisotropy of the transport properties and the electrical conductivity may be described as metallic along the periodic direction and nonmetallic in the quasiperiodic planes.[4–6] Following the pioneering work of Kohomoto *et al.*[7] on quasiperiodic one-dimensional chains demonstrating that quasiperiodicity leads to ''critical'' eigenstates (with a power-law decay of the amplitudes) and a singular continuous spectrum there have been numerous attempts to relate the transport properties of real quasicrystals to the spikiness of the electronic density of states (DOS) and in particular to the existence of a pseudogap (=DOS minimum) at the Fermi level.[8–10]

Some recent results have demonstrated that the situation is in reality more complex. (a) Photoemission and optical conductivity experiments[11] have demonstrated that the pseudogap at the Fermi level characteristic for many icosahedral alloys is absent in decagonal alloys—in agreement with the most recent theoretical studies.[12,13] (b) Highly anisotropic transport properties have been reported also for some decagonal approximants [$Al_{13}Co(Fe)_4$, Al_3Mn].[14] In the Al-Mn approximant the nonmetallic behavior is even observed along the axis perpendicular to the plane with pentagonal atomic arrangements (= the ''pseudoquasiperiodic'' planes). The result (a) raises the problem of nonmetallic transport properties in the presence of a high number of carriers and (b) suggests that the anisotropic transport properties cannot be explained as an effect of the pseudoquasiperiodicity alone.

In this paper we present detailed *ab initio* calculations of the electronic structure and of the transport properties of model decagonal quasicrystals and approximants in the Al-Cu-Co and Al-Mn systems performed using the tight-binding linear-muffin-tin-orbital (TB-LMTO) (Refs. 15,16) technique. We use the Boltzmann-Bloch conductivity as a quantity for characterizing the anisotropy of the electronic structure. We show that in all systems the anisotropy of the transport properties with ''nonmetallic'' behavior along certain crystallographic planes or symmetry directions is related to a corresponding anisotropy in the electronic band structure. The band structure in turn depends quite sensitively on details of the atomic structure. Introducing atomic disorder leads to a pronounced enhancement of the Boltzmann-Bloch conductivity in the nonmetallic directions while the conductivity in the other directions is hardly affected.

II. STRUCTURAL MODELS

Our model structure for the decagonal Al-Cu-Co alloys is based on a Klotz-triangle tiling of the quasiperiodic plane[17] as proposed by Burkov.[18] We choose a variant which has a simple cut-and-projection representation in five-dimensional space—this facilitates the construction of rational approximants. Here we consider an orthorhombic approximant ($a = 32.07$ Å, $b = 37.78$ Å, $c = 4.18$ Å) with 352 atoms and the ''magic'' composition $Al_{\tau-1}Cu_{\tau'}Co_{\tau'}$, $\tau' = (2-\tau)/2$, i.e., $Al_{0.62}Cu_{0.19}Co_{0.19}$, where $\tau = (1+\sqrt{5})/2$ is the golden mean. The distribution of Cu and Co over the sites available for the transition-metal atoms is not determined by the available diffraction data, it has been chosen such as to optimize the agreement of the calculated electronic structure with the photoemission data.[11] The new Burkov-model for decagonal Al-Cu-Co leads to more realistic results for the electronic structure than earlier models based on a binary Penrose tiling.[19] Details of our structural model have been published elsewhere (see Ref. 12). In addition to the decagonal alloys we considered the crystalline intermetallic compounds $Al_{13}Co_4$ and Al_7Cu_2Co. The orthorhombic cell of $Al_{13}Co_4$ (a

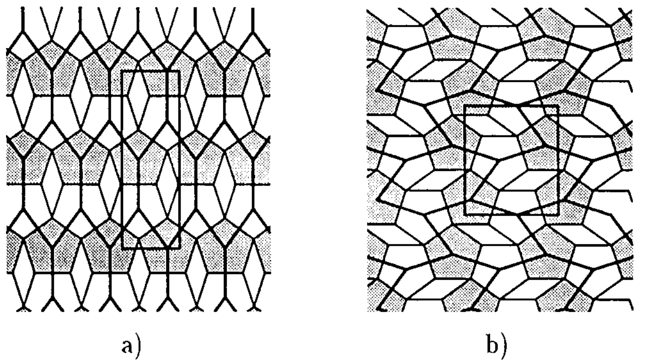

FIG. 1. Tiling of the pseudoquasiperiodic planes of the R and T phases with squashed hexagons (H tiles) and decomposition into Hiragas pentagonal columnar clusters. The shading of the pentagons indicate the twofold screw-symmetry relationship between columnar clusters decorating pairs of edge-sharing pentagons.

$=14.0$ Å, $b=11.84$ Å, $c=7.72$ Å) contains 92 atoms, we used the idealized atomic coordinates proposed by Widom.[20] This structure is characterized by pentagonal atomic arrangements in the plane perpendicular to the b-axis, similar to the decagonal phase. Al_7Cu_2Co has a composition very similar to that of the decagonal phase, but its tetragonal structure (space group $P4/mnc$, $a=b=6.315$ Å, $c=14.74$ Å, 40 atoms/cell) (Ref. 21) is not directly related to the decagonal phase.

For decagonal Al-Mn and Al-Pd-Mn a close structural relationship between the decagonal phase and crystalline Al_3Mn (Taylor's phase or T phase,[22] orthorhombic, $a=12.56$ Å, $b=14.77$ Å, $c=12.43$ Å) and the Robinson (or R) phase $Al_{60}Ni_4Mn_{11}$ (Ref. 23) (orthorhombic, $a=7.76$ Å, $b=23.90$ Å, $c=12.43$ Å) has been established.[13,24] In both cases the "pseudoquasiperiodic" plane is perpendicular to the c axis. While both R and T phases can be described in terms of tiling of a single hexagonal (H) supercell, in the decagonal phase there are two more supercells in the form of a pentagonal star (P) and a decagon (D). All DPH tilings may be decomposed into pentagonal clusters proposed by Hiraga *et al.*[24] Figure 1 shows the arrangement of the H tiles and their decomposition into Hiragas pentagonal clusters for both phases. For the idealized structures of the R and T phases based on H tiling and preserving the pentagonal clusters, we derive a composition of 20.5 at. % Mn, further structural details may be found in Ref. 13.

III. ANISOTROPIC ELECTRONIC STRUCTURE

Figure 2 shows the total and partial electronic densities of states (DOS) calculated for decagonal Al-Cu-Co using 32 **k** points in the irreducible zone. The main characteristics are a parabolic Al-DOS with a broad structure-induced (and sp-d hybridization enhanced) minimum at the Fermi level superposed by Co and Cu bands peaked at -0.7 eV and -3.6 eV below E_F, respectively. d-band positions and the high value of the total DOS at the Fermi level are well confirmed by photoemission[11] and soft-x-ray spectra.[25] Trambly de Laissardière and Fujiwara[26] have published calculations of the electronic structure of a smaller approximant with composition $Al_{66}Cu_{30}Co_{14}$ based on the earlier version of the Burkov model,[19] predicting a low density of states at the Fermi level.

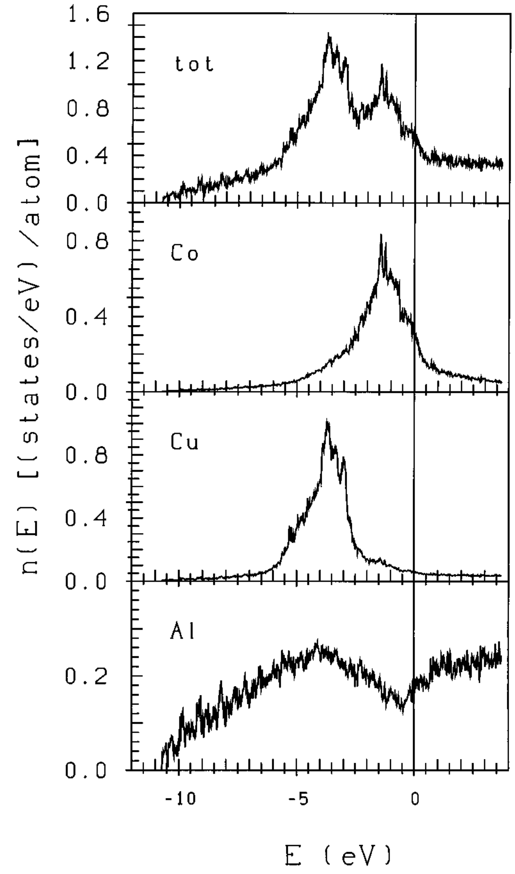

FIG. 2. Electronic density of states for decagonal $Al_{0.62}Cu_{0.19}Co_{0.19}$. From top to bottom: total DOS and partial Co, Cu, and Al DOS.

The difference between both sets of calculations can be attributed to the structural models. Both crystalline compounds show stronger Al-sp-TM-d hybridization and Cu and Co-d bands shifted to higher binding energies so that the pseudogap in the Al band is no longer covered up by a high Co-d DOS. The pseudogap is deepest in Al_7Cu_2Co. Details have been published elsewhere.[12]

The structure of bands $E_n(\mathbf{k})$ in a system with many atoms in the unit cell is very complicated. We use the Boltzmann-Bloch conductivity as a convenient quantity for characterizing the anisotropy of the electronic structure. Within the Boltzmann-Bloch theory the conductivity σ can be expressed as[8]

$$\sigma_{\alpha\beta}=\frac{e^2}{\Omega_0}\tau\sum_{n,\mathbf{k}} v_{n\alpha}(\mathbf{k})v_{n\beta}(\mathbf{k})\left(-\frac{\partial f}{\partial E_n(\mathbf{k})}\right), \tag{1}$$

where τ is the relaxation time, Ω_0 is volume per atom, and f is the Fermi-Dirac function. The band velocities are calculated in terms of the gradients of the dispersion relations of the electronic eigenstates[27] as

$$v_{n\alpha}(\mathbf{k})=\frac{1}{\hbar}\frac{\partial E_n(\mathbf{k})}{\partial k_\alpha}. \tag{2}$$

At zero temperature the conductivity σ may be understood as a function of the position of the Fermi level E_F, $\sigma=\sigma(E_F)$. The application of Boltzmann-Bloch theory to quasiperiodic systems has been criticized because the scaling properties of critical eigenstates lead to the expectation that the propagation of the electrons is not ballistic in quasiperiodic in contrast to periodic systems.[9,10] However, so far the criticality of the eigenstates has been established only for simplified model quasicrystals, and the appearance of the anomalous transport properties in crystalline systems suggests that criticality of the eigenstates cannot be the decisive factor. As noted above we understand the Boltzmann-Bloch conductivity as a convenient quantity for characterizing the anisotropy of the electronic structure. However, as we shall see later the anisotropy of the transport properties with nonmetallic behavior along certain crystallographic planes or symmetry directions is related to a corresponding anisotropy in the electronic band structure. Although we are interested in anisotropies of the electronic structure and the absolute value of the zero temperature conductivity is not of our primary interest, the realistic assumption about the value of the relaxation time $\tau \sim 10^{-15}$ s provides values of conductivities well comparable with experimental values. The low-temperature resistivity of d-$Al_{0.65}Cu_{0.15}Co_{0.20}$ measured in the periodic direction[4] is 32 $\mu\Omega$ cm, the corresponding value calculated for the Burkov model of d-AlCuCo from the Boltzmann-Bloch formula (1),(2) is 25 $\mu\Omega$ cm.

IV. RESULTS AND DISCUSSION

Figures 3(a)–3(c) show the energy-dependent conductivities $\sigma_{\alpha\beta}(E)$ scaled by the relaxation time τ calculated for d-$Al_{0.62}Cu_{0.19}Co_{0.19}$ (a), $Al_{13}Co_4$ (b), and Al_7Cu_2Co (c) along the directions of the Cartesian axes. The Fermi level is located at $E=0$. The energy dependence of the conductivity shows its behavior (in a rigid-band approximation) if the Fermi level is shifted. For the decagonal phase we find very low conductivities in the quasiperiodic (x,y) plane, for all energies in the range of Co and Cu-d bands. The conductivity along the decagonal axis is much higher and strongly energy dependent. At the actual position of the Fermi level the transport anisotropy is $\sigma_{zz}/\sigma_{xx} \sim 50$. If the chemical decoration of the tiling is changed, the conductivity in the quasiperiodic plane is hardly affected, but the peak close to the Fermi energy in the conductivity along the periodic direction changes in position and amplitude. Altogether the anisotropy ratio varies between $\sigma_p/\sigma_q \sim 20$ and $\sigma_p/\sigma_q \sim 150$. We assume that the origin of the experimentally observed anisotropic transport properties is in a strongly anisotropic band structure. The electronic eigenstates show only very weak dispersion along directions within the quasiperiodic planes and much stronger dispersion in the periodic direction. If we scale the conductivities with the square of linear dimensions we find that $l_\alpha^2 \sigma_{\alpha\alpha}(E)$ is almost isotropic. In order to simulate thermal disorder we generated disordered configurations by shifting the atoms by random displacement vectors with a Gaussian distribution. Here we report the conductivities calculated for a standard deviation of $\Delta r = 0.15$ Å (for comparison the shortest nearest-neighbor distance is ~ 2.5 Å). We find that the conductivity in the quasiperiodic plane increases by a factor of 5 at the Fermi-energy of d-AlCuCo (and even more at lower energies), while that along the periodic direction is hardly affected [see Fig. 3(a)] and the transport anisotropy is reduced to $\sigma_{zz}/\sigma_{xx} \sim 10$. Martin *et al.*[4] found for d-$Al_{65}Cu_{15}Co_{20}$ anisotropies of $\sigma_{zz}/\sigma_{xx} \sim 10$ at low temperature and $\sigma_{zz}/\sigma_{xx} \sim 8$ at room temperature. Considering the small differences in composition and certain degree of disorder that is unavoidable in the laboratory-made samples, the agreement is satisfactory.

Earlier calculations by Trambly de Laissardière and Fujiwara[26] based on a smaller approximant (110 atoms/cell) of a different structural model[19] and a slightly different composition led to a higher resistivity along the periodic direction ($\rho_p = 69$ $\mu\Omega$ cm) and a considerably smaller anisotropy ratio (for the perfectly ordered model) of $\sigma_{zz}/\sigma_{xx} \sim 5$. Hence the electronic structure calculated for the older variant of the Burkov model is less anisotropic than that of the new models,[18] it also agrees better with spectroscopic data.

For the orthorhombic approximant $Al_{13}Co_4$ the conductivities are very similar to those in the decagonal phase [see Fig. 3(b)], with $\sigma_{zz} \gg \sigma_{yy} \sim \sigma_{xx}$ in a wide energy range and a transport anisotropy of $\sigma_{zz}/\sigma_{xx} \sim 8$ close to the Fermi level, increasing steeply to $\sigma_{zz}/\sigma_{xx} \sim 20$ only 0.2–0.3 eV below E_F (note that for consistency we have oriented the b axis along the z direction). In contrast to our model decagonal alloy where we find a large transport anisotropy for a wide range of energies, the anisotropic behavior of $Al_{13}Co_4$ compound depends quite sensitively on the precise position of the Fermi level. Experimentally Volkov and Poon[14] reported for orthorhombic Al-Co compounds with compositions varying between 23.2 and 25.8 at. % Co (to be compared with 23.5 at. % in $Al_{13}Co_4$) an anisotropy of ~ 30. The tetragonal Al_7Cu_2Co [see Fig. 3(c)] compound, on the other hand, shows essentially isotropic transport properties, with conductivities that are in all directions of the same order as in the metallic directions of the quasiperiodic and approximant phases.

The electronic density of states of all Al-Mn phases is strongly structured around the Fermi level (see Fig. 4). In particular we find a sharp peak immediately at E_F and a very deep DOS minimum at $E \sim 0.8$ eV. These details depend very sensitively on details of the chemical decoration—particularly on that of the sites not belonging to the pentagonal columnar clusters whose decoration is hence less subject to symmetry constraints. Interchanging the occupation of two sites with Al and Mn, respectively, such as to enhance the Mn-Mn coordination (and increasing the concentration to 23.1 at. %) leads for both R and T phases to very deep pseudogaps only a few tenths of an eV above E_F [see the results labeled $R(T)$-$V2$ in Fig. 4]. Close inspection of the band structure shows a strong dispersion of eigenstates along k_z (the periodic direction) and much weaker dispersion in the pseudoquasiperiodic plane. In both phases a real gap of ~ 50 meV is found in the quasiperiodic plane for $k_z \sim 0$. For the R-$V2$ phase the gap closes near the boundary of the Brillouin zone as one moves away form $k_z = 0$ to $k_z = 0.5$, in the T-$V2$ phase the gap persists but its position shifts because of the dispersion of the eigenstates. Hence only a pseudogap appears in the total DOS. For icosahedral quasicrystals and approximants (i-AlPdRe,i-AlCuFe) similar very sharp and deep pseudogaps have been detected by tunneling spectroscopy.[28]

For the conductivities we find a strongly structured $\sigma_{\alpha\beta}(E)$ with very low values for all three Cartesian directions, for both $V2$-type compounds—so far in agreement

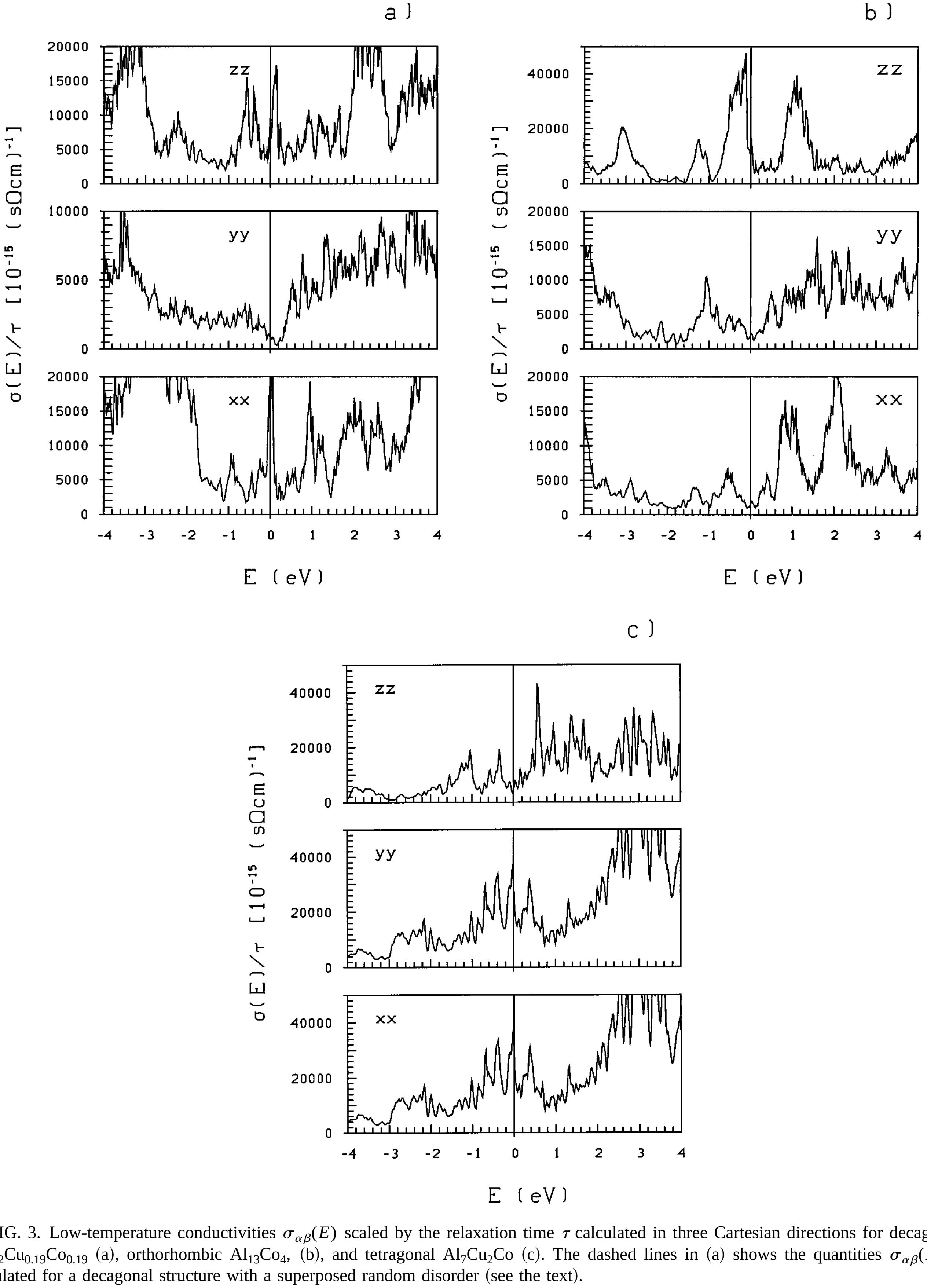

FIG. 3. Low-temperature conductivities $\sigma_{\alpha\beta}(E)$ scaled by the relaxation time τ calculated in three Cartesian directions for decagonal $Al_{0.62}Cu_{0.19}Co_{0.19}$ (a), orthorhombic $Al_{13}Co_4$, (b), and tetragonal Al_7Cu_2Co (c). The dashed lines in (a) shows the quantities $\sigma_{\alpha\beta}(E)/\tau$ calculated for a decagonal structure with a superposed random disorder (see the text).

with the results of Volkov and Poon[14] (see Fig. 5). Most remarkably, for both components the conductivity in one direction goes to zero at an energy ~0.2 eV (R-$V2$), respectively ~0.4 (T-$V2$) eV above the Fermi level. For the R phase this direction is along the b edge of the cell, for the T phase this is the a axis. Note that in both cases this is the

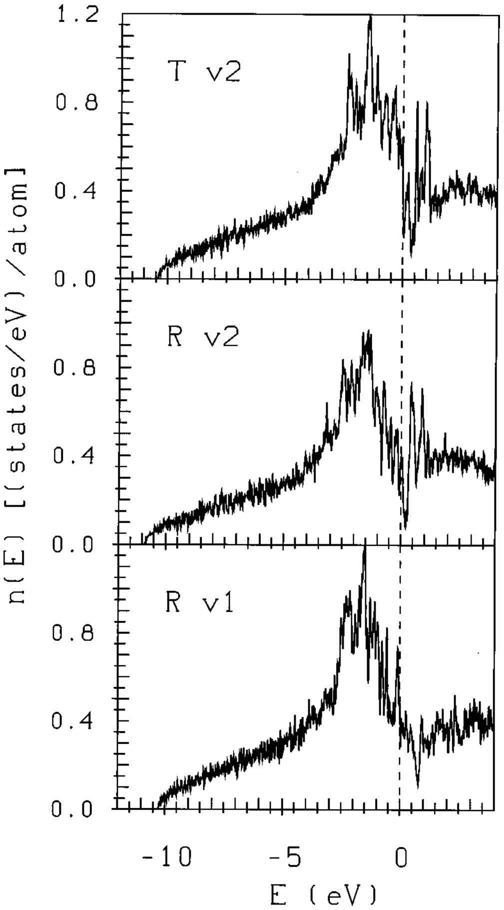

FIG. 4. Electronic densities of states for the Al-Mn *R* and *T* phases [(a) *R*, version 1, (b) *R*, version 2, (c) *T*, version 2).

direction along which the edge sharing pentagons are aligned (see Fig. 1).

V. SUMMARY AND CONCLUSION

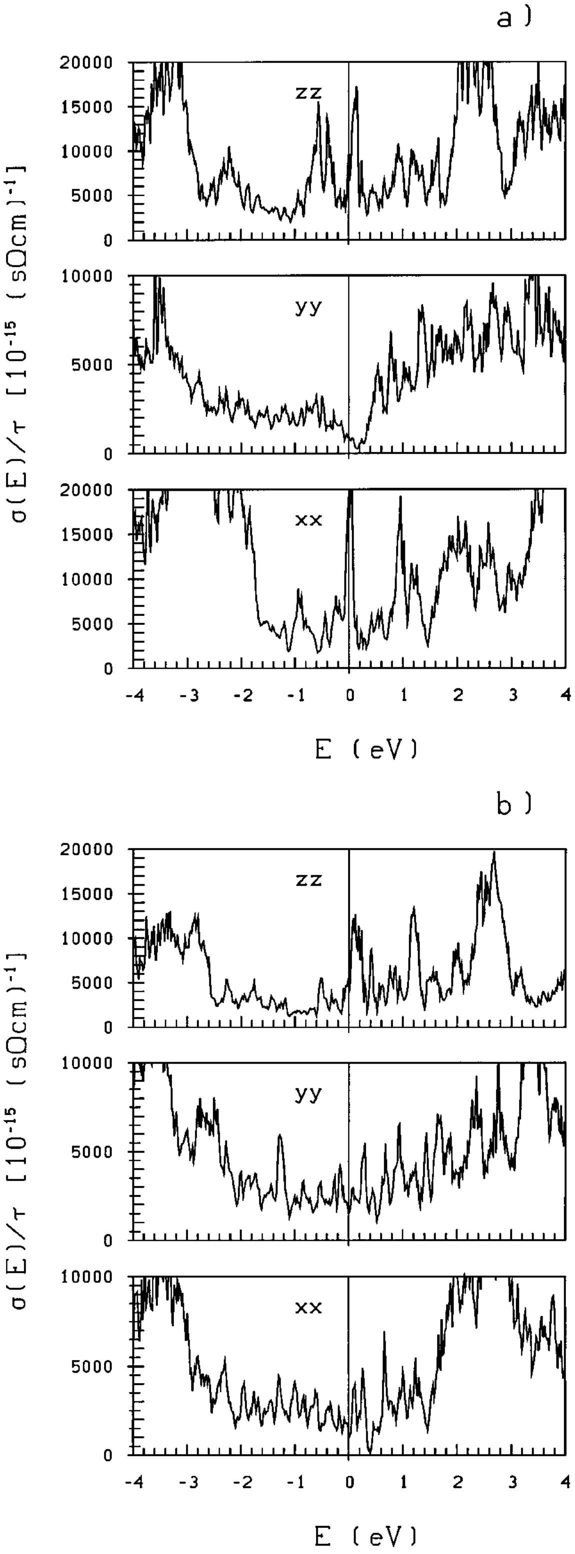

FIG. 5. Low-temperature conductivities $\sigma_{\alpha\beta}(E)$ scaled by the relaxation time τ calculated in three Cartesian directions for the *R* and *T* phases (both version 2), corresponding to the densities of states shown in Fig. 4.

Our results for the Al-Mn-*d* approximants may be summarized as follows. (a) The close structural relationship with the *d* phase leads to a very spiky structure of the DOS in the vicinity of E_F; the details of the structure depend quite sensitively on minor aspects of the chemical decoration. (b) Averaged over an energy range of even a very few tenths of an eV, the conductivity is essentially isotropic, despite the structural similarity with the decagonal phase. (c) At the energy where the deepest pseudogap in the DOS is found, a true conductivity gap of ~ 50 meV exists in the direction along which the pentagonal columnar clusters are joined directly together. If the Fermi edge falls at this energy, the conductivity along this direction will decrease steeply in this direction. Again the existence of the conductivity depends sensitively on the chemical decoration—it is absent in version $V1$ of the *R* phase, in spite of a very similar structure of the DOS. The existence of a conductivity gap in a single crystallographic direction agrees with the experimental results of Volkov and Poon—but there is no agreement in the directions along which this effect is observed. The reasons for the discrepancy have to be found in the details of the crystalline structure. Our calculations have been performed on idealized models with a composition very close to $Al_{13}Mn_4$. Volkov and Poon[14] give a composition of $Al_{10.6}Mn_4$ for a structure described as identical to the Hiragas[24] $Al_{11.7}Mn_4$ phase. A number of atomic sites in the Hiragas structure have a fractional or mixed occupation. Hence a number of open problems concerning the precise structure of the $Al_{11}Mn_4$ approximant phase will have to be clarified before we can proceed to a more detailed comparison between theory and experiment.

However, it remains remarkable that extremely narrow gaps in the electronic density of states and *unidirectional* conductivity can exist in decagonal approximants. Together, our results for the decagonal Al-Cu-Co quasicrystals and approximants and for the decagonal Al-Mn approximants shed light on two facts of the electronic transport properties of quasicrystals. (a) The results for d-Al-CuCo quasicrystals emphasize the important role of the widely different length scales in the periodic and quasiperiodic directions leading to strong differences in the dispersion of the electronic eigenstates and to the characteristic scaling behavior in the electronic conductivities. (b) The results for the d-Al-Cu-Co and d-Al-Mn approximants emphasize the important role of the local atomic structure. For a certain local order the electronic spectrum show very strong structures (spikes, gaps), much stronger than usually observed even in the quasiperiodic limit. For certain directions and energies this can even cause the appearance of conductivity gap of the order of ~ 50 meV and hence to an insulating behavior in the limit $T \rightarrow 0$ K. From the analysis of the local densities of states, the dispersion relations and of the participation ratio we suspect that the structure in the spectrum and the existence of a vanishing conductivity is related to a special character of the electronic eigenstates (resonant properties, large coherence lengths)—however, this remains to be explained in more detail. In this context it would be extremely helpful if the experimental transport investigations were supplemented by spectroscopic investigations with very high resolution.

However, one should also not forget to emphasize the limitations of the Boltzmann-approach. Several attempts have been made to go beyond the quasiclassical description, based either on a multichannel Landauer formula for the conductivity[29,30] or using the knowledge of the electronic eigenstates (and not only of the dispersion relations as in Boltzmann theory) within the framework of Kubo-Greenwood theory.[10,31] So far such calculations are restricted to simple model systems, with the exception of the work of Fujiwara *et al.*[10] Fujiwara *et al.* analyzed the electronic diffusivity in the quasiperiodic plane of approximants to d-Al-Cu-Co, with particular emphasis on the scaling behavior, but stopped short of a calculation of the anisotropy of the transport properties. A Kubo-Greenwood calculation for the systems discussed in the present study is under way. Results will be published in due course.[32]

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