

## Electrical conduction in low-resistivity (quasiamorphous) $\text{Ag}_{1-x}\text{Cu}_x$ alloys

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UHV-evaporated  $\text{Ag}_{1-x}\text{Cu}_x$  alloy films show a strong dependence of the crystallite sizes on the composition: In the middle of the concentration range, the mean grain size is smaller than 2 nm. The resistivity, however, is much lower than expected for such extremely-fine-grained materials ( $\rho < 9 \mu\Omega \text{ cm}$ ). The electrical transport parameters for these films were obtained from the thickness dependence of the conductivity without any *a priori* assumptions. It will be shown that the electrical transport in these alloys can be well understood as a limit of the reflection model for the electrical conductivity in polycrystalline metals [G. Reiss, J. Vancea, and H. Hoffmann, *Phys. Rev. Lett.* **56**, 2100 (1986)].

### I. INTRODUCTION

In the last few years, great efforts have been made in understanding structure and properties of amorphous metals. From a more traditional point of view, the well-ordered polycrystalline structure breaks down into a thoroughly disordered arrangement of constituent atoms like the structure of glass (metallic glasses). The transition between the two structural states should occur suddenly, without any crossing range. Recent High-Resolution Electron Microscopy (HREM) experiments on Fe-B alloys, regarded as standard amorphous materials, gave evidence for the existence of ordered microcrystalline regions with not more than 2–3 nm in diameter.<sup>1,2</sup> These new experiments show that the “amorphous state” should be rather looked at as a microcrystalline one, with a monotonic transition from the standard polycrystalline structure to the borderline case of metallic glasses (“amorphouslike”).

Consequently, other physical properties, like the electrical conductivity, should be explicable in the limits of conduction mechanisms of polycrystalline metals. Although  $\text{Ag}_{0.5}\text{Cu}_{0.5}$  alloys are classified in the literature as metallic glasses, very different values of their electrical resistivity have been reported. For films condensed in vacuum at 78 K substrate temperature, resistivities of  $175 \mu\Omega \text{ cm}$  ( $10^{-6}$  mbar) by Chopra *et al.*,<sup>3</sup>  $55 \mu\Omega \text{ cm}$  ( $10^{-9}$  mbar) by Reda *et al.*,<sup>4</sup> down to  $12 \mu\Omega \text{ cm}$  ( $10^{-8}$  mbar) by Mader *et al.*<sup>5</sup> and Mizutani and Yoshida<sup>6</sup> have been reported. Assuming a free-carrier density of one electron per atom, a rough estimation of the mean free path (MFP) of the conduction electrons (CE's) by means of Drude's formula gives 0.5 nm in the first case, 1.6 nm in the second case, and 7.5 nm in the third case. Therefore, dealing with the same structure and composition, scattering lengths associated with the amorphous as well as with the crystalline state can be suspected. No attempts have been reported up to now to determine the MFP in such alloys by an independent experiment as, for example, the investigation of the thickness dependence of the conductivity (size effect). Therefore it seems to be desirable to vary the crystallite size of these alloys continuously (for example, by varying

the composition) down to 2 nm and at the same time to evaluate the MFP of the CE by analyzing the thickness dependence of the conductivity. With a subsequent analysis of the electrical transport parameters related with the grain-boundary scattering mechanism, we try to understand the resistivity in the limit of the amorphous-like state.

### II. SURFACE AND GRAIN-BOUNDARY SCATTERING

Conduction electrons in a polycrystalline metal inevitably hit the grain boundaries. If the innercrystalline MFP is larger than the grain size, the additional resistivity due to the grain-boundary scattering cannot be neglected. The problem of the grain-boundary's resistivity does not seem to be definitely solved. The most used theoretical model relating to the scattering of electrons at grain boundaries was published in 1970 by Mayadas and Shatzkes.<sup>7</sup> The grain boundaries are represented by parallel planes ( $\delta$  functions) with average separation  $D$  (mean grain size) and reflection probability  $R$ . As long as the CE are not confined within one grain, i.e., as long as  $R$  is smaller than about 0.7, no band splitting will occur, so that it is possible to treat the problem using the effective-mass approximation and Boltzmann's formalism for the calculation of the conductivity. Therefore, these authors solved Boltzmann's equation by adding a correction to the background scattering time (Matthiessen's rule). The resulting conductivity is then given by

$$\sigma_{\infty}/\sigma_0 = f(l_0/D, R) < 1 \quad (1)$$

with  $\sigma_{\infty}$  the conductivity of the polycrystalline metal (infinitely thick) including grain-boundary scattering,  $\sigma_0$  the conductivity of the same metal with background scattering only, and  $l_0$  the background scattering length. As the authors stated in the original paper and also as pointed out by other authors,<sup>8,9</sup> in fact an “effective intrinsic MFP” was defined for the whole polycrystal, given by

$$l_g \simeq (\sigma_{\infty}/\sigma_0)l_0 \simeq f(l_0/D, R)l_0, \quad (2)$$

$$l_g < l_0.$$

The conductivity of the polycrystal is derived to

$$\sigma_{\infty} \simeq (mv_F/e^2)n_0l_g = kl_g, \quad (3)$$

i.e., the product  $\rho_{\infty}l_g$  is a material constant, given mainly by the density of free carriers.

In the next step the thickness dependence of the conductivity was calculated by means of Fuch's formalism<sup>10</sup> using the corrected scattering time. Consequently, the size effect  $S$  will be suppressed by grain-boundary scattering:

$$S \simeq S(d/l_g, p) \quad (4)$$

with  $d$  the film thickness and  $p$  the specularity parameter. In the limit of very strong grain-boundary scattering, no size effect should be observed.

Recently, this problem was questioned with very accurate *in situ* measurements of the thickness-dependent conductivity of a great number of metals and alloys, made by the group in Regensburg.<sup>11-15</sup> In the view of Mayadas-Shatzkes model, the experiments gave rather astonishing results: Even for very-fine-grained metals ( $D \simeq 2-3$  nm) and large resistivities (100  $\mu\Omega$  cm) an important size effect was observed. Already at this stage it will be clear that the results cannot be explained by the Mayadas-Shatzkes theory, because this size effect is a direct manifestation of a rather large electronic MFP. By analyzing the experimental data with a modified Fuchs theory (model of Namba<sup>16</sup>), such large MFP's are determined. These values of the MFP cannot be directly correlated with the grain size and only partly with the resistivity. The scattering length responsible for the size effect is rather the innercrystalline (background) MFP. This produces a serious problem for the understanding of the conductivity in polycrystalline metals. Particularly, it seems to be impossible to define an intrinsic MFP for the whole polycrystal. A reflection model of electrons at the grain-boundary potentials (with transmission  $T$ ) was proposed to solve this dilemma.<sup>11,12</sup> The model is roughly outlined in Fig. 1.

A fraction  $T$  of electrons passes through the grain-boundary potential by remaining in the same  $k$  state (tunneling effect). After  $N = l_{\infty}/D$  potentials, the transition  $k$  to  $k'$  occurs due to background scattering. The fraction  $R = 1 - T$  of electrons will be elastically reflected at each grain boundary and will not contribute effectively to the

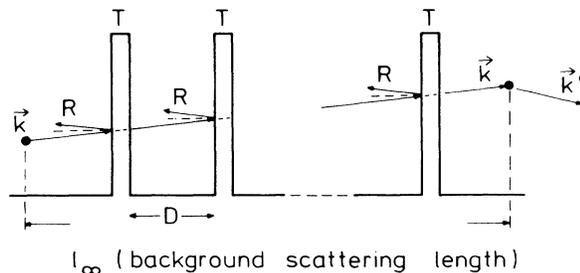


FIG. 1. Reflection model for the CE at grain boundaries.

current. The Drude formula for polycrystalline metals should be consequently corrected to

$$\sigma_{\infty} = [(mv_F/e^2)n_0]l_{\infty}G(l_{\infty}, D, T) = kl_{\infty}G(l_{\infty}, D, T) \quad (5)$$

instead of Eq. (3), where the function  $G(l_{\infty}, D, T)$  gives the correction due to the grain-boundary scattering. Now, the product  $l_{\infty}G(l_{\infty}, D, T)$  cannot be regarded as an "effective intrinsic MFP." Using the transfer matrix approach<sup>17</sup> for transferring the solutions of the Schrödinger equation from the grain  $n$  to the grain  $n+1$ , the total transmission [function  $G(l_{\infty}, D, T)$  in Eq. (5)] for an array of  $l_{\infty}/D$  potentials was calculated.<sup>18</sup> For  $T > 0.6$ , the function  $G(l_{\infty}, D, T)$  is given in the first approximation by a power law:

$$G(l_{\infty}, D, T) \simeq T^{l_{\infty}/D} \quad (6)$$

in good agreement with the result found empirically from the size-effect experiments.<sup>12</sup>

As already outlined in the foregoing discussion, this treatment of the grain-boundary resistivity is limited by three conditions.

Firstly, the grains must not be strongly decoupled, i.e., the transmission probability  $T$  must not be too small. Otherwise band splitting and phonon-assisted grain-to-grain tunneling would govern the resistivity. This, however, occurs only if the potential barrier of the grain boundaries is remarkably larger than the Fermi level, i.e., for  $T < 0.3$ . This value should be accepted as a limit for the validity of the model outlined before.

Secondly, the splitting of "inner grain bands" due to the finite number  $N$  of atoms within one grain should be smaller than  $k\theta$  ( $\theta$  being the temperature); otherwise no really extended wave function can be defined and the transmission parameter concept is not valid. The splitting  $\Delta E$  of levels within one grain can be estimated to be

$$\Delta E \simeq [Ng(E)]^{-1},$$

where  $g(E)$  is the density of states at the Fermi level per atom. For the example of Cu we get in the approximation of the free-electron gas  $g(E) \simeq 0.21$  eV<sup>-1</sup> per atom. Consequently, even for extremely small grains ( $D = 2$  nm,  $N \simeq 10^3$  atoms) the upper value of this splitting will not exceed 5 meV. This value will be lowered by the noncubic statistical form of the grains and by the transmission from grain to grain. Therefore, at least for room temperature the condition  $\Delta E < k\theta$  is always fulfilled, so that the effective-mass approximation remains valid.

Finally, the potentials of grains and grain boundaries must not overlap too strongly. This leads to the requirement that the mean grain size  $D$  must be much larger than the screening length. In metallic films this condition will always be fulfilled due to the small screening length of about 0.1 nm.

If these conditions are fulfilled, a kind of effective density of conduction electrons will be introduced formally by Eq. (5) (the true density of free carriers is surely the same as in the single crystal) as long as a constant Fermi energy is accepted. The product  $(\rho l)_{\infty}$  is no longer a material constant, but strongly dependent on the grain-boundary scattering.

This model does not suppose imperatively a very short MFP for "amorphous" materials and does not lead to unphysical MFP's (shorter than the lattice constant) as for instance in the case of very-high-resistive metallic glasses. Consequently, we try to understand the conductivity of the low-resistive "amorphouslike" Ag-Cu alloys from this point of view.

### III. THE STRUCTURE OF THE FILMS

The films were prepared by evaporation from two sources in UHV ( $10^{-9}$  mbar during evaporation) onto Corning glass substrates held at room temperature. By variation of the evaporation rates, alloys in the whole concentration range were obtained. The evaporation rates were monitored by computer-controlled quartz-oscillator balance with a relative error of less than  $5 \times 10^{-4}$ . Due to the special construction of the evaporation sources, very constant (better than 1%) evaporation rates could be held for at least one hour. Details of the experimental apparatus have been described in Ref. 12. During evaporation, resistance-versus-thickness data were obtained with a relative error less than  $2 \times 10^{-3}$ . Finally, the films (with standard thickness of 50 nm) were removed from the substrate and prepared for transmission electron microscopy and electron diffraction. A typical bright field transmission picture with the corresponding diffraction rings of a  $\text{Ag}_{0.55}\text{Cu}_{0.45}$  film is given in Fig. 2.

The "mean crystallite size" is smaller than 2 nm. "Crystallites" of these dimensions should be interpreted rather as domains with different density due to the ordered or disordered structure.<sup>19</sup> The diffraction rings in Fig. 2, however, correspond rather to a very small grained fcc polycrystal. We consequently called this structure "ultramicrocrystalline" or "amorphouslike."

The dependence of the crystallite size on the composition of the alloys is given in Fig. 3. A very large variation can be observed, with a wide minimum between 40–80 at. % Cu. In this concentration range, where commonly the amorphous structure was observed, we find "mean crystallite sizes" of at most 2 nm. Figure 4 shows the dependence of the lattice constant on the alloy composition as determined from the diffraction rings. As expected for a substitution crystal (Vegard's rule) a linear dependence can be observed. Full miscibility of Ag and Cu in

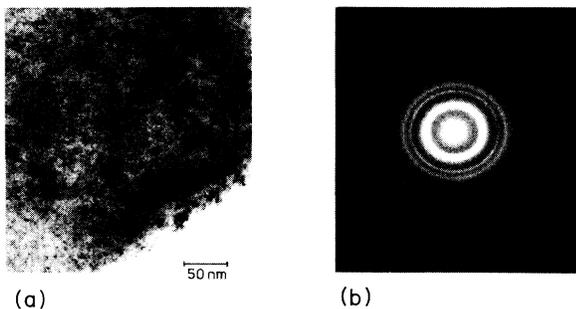


FIG. 2. TEM—pictures of a 50-nm-thick  $\text{Ag}_{0.55}\text{Cu}_{0.45}$  film. (a) Bright field transmission; (b) diffraction patterns.

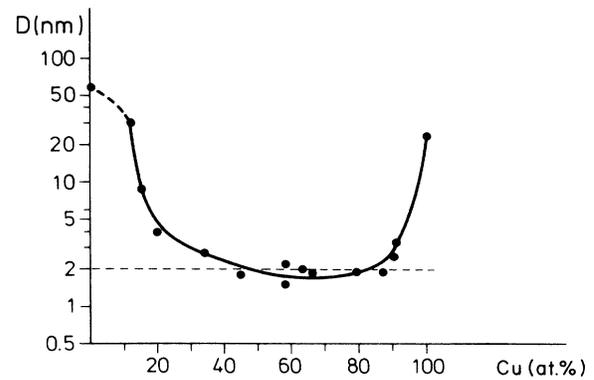


FIG. 3. The dependence of the "mean grain size" on the alloy composition.

evaporated films was observed in the entire composition range by all other authors,<sup>3–5</sup> although the phase diagram of this alloy<sup>20</sup> permits a maximal miscibility of about 10%. The decomposition temperature of the metastable fcc binary alloy is greater than 400 K.<sup>21</sup>

### IV. ELECTRICAL TRANSPORT PARAMETER

An example of the measured thickness dependence of the conductivity for a  $\text{Ag}_{0.2}\text{Cu}_{0.8}$  alloy is shown in Fig. 5 in a  $\rho d$ -versus- $d$  plot ( $\rho$  represents resistivity,  $d$  represents film thickness). The mean grain size for this film amounts to 2 nm. Above a thickness of 20 nm, the plot shows a linear behavior, as demanded by the well-known Sondheimer approximation:<sup>22</sup>

$$\rho d = \rho_{\infty} d + \frac{3}{8} \rho_{\infty} l_{\infty} (1-p). \quad (7)$$

The resistivity shows an important size effect; otherwise, this linear approximation should intercept the ordinate in the origin. As demonstrated by the authors<sup>23,24</sup> reasonable transport parameters can be obtained only by close fitting of the theory below this linear thickness range. This is practicable only with a four-parameter fit, by introducing the macroscopic surface roughness in the Fuchs theory (model of Namba<sup>16</sup>). The thickness dependence of the conductivity is then given by

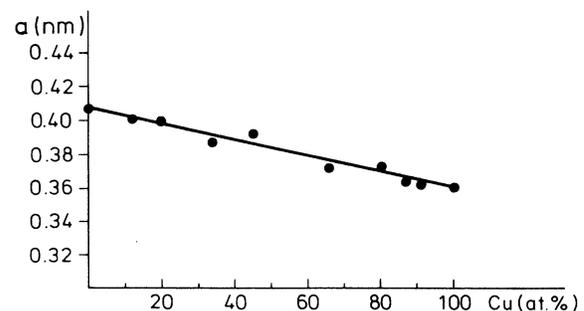


FIG. 4. Lattice constant of  $\text{Ag}_{1-x}\text{Cu}_x$  films vs composition.

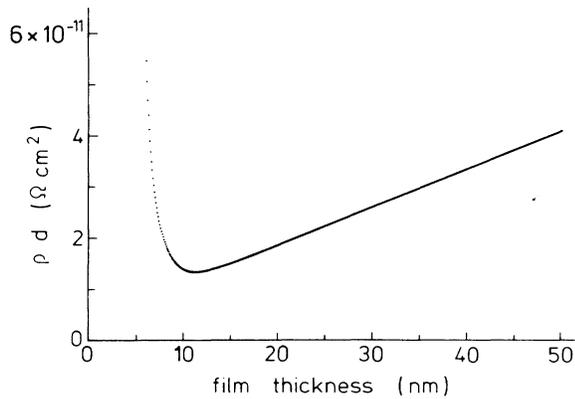


FIG. 5.  $\rho d$ -vs- $d$  plot of a  $\text{Ag}_{0.2}\text{Cu}_{0.8}$  alloy.  $\rho$ , resistivity;  $d$ , film thickness.

$$\langle \sigma(d) \rangle = \sigma_{\infty} \frac{L}{d} \left[ \int \frac{\sigma_{\infty}}{\sigma(d(x)) d(x)} dx \right]^{-1} \quad (8)$$

with  $\langle \sigma(d) \rangle$  the measured conductivity at the mean thickness  $d$  and  $L$  the film length. The surface roughness effect in (8) is given by a size-dependence fluctuation of the film thickness (in first approximation a one-dimensional sinusoidal fluctuation)  $d(x)$ :

$$d(x) = d + h \sin(2\pi/sx) \quad (9)$$

with  $h$  the surface roughness and  $s$  the surface roughness wavelength. The local conductivity in (8) is given by the Fuchs integral  $\sigma(d(x))$ .<sup>10</sup>

A typical example of the fitting results is given in Fig. 6, for a  $\text{Ag}_{0.2}\text{Cu}_{0.8}$  film, with a mean grain size of 2 nm. The difference between theory and experiment is smaller than 0.5% above a film thickness of 14 nm. The relative errors for the fit parameters are then typically as follows:<sup>24</sup>  $\Delta\sigma_{\infty}/\sigma_{\infty}$  and

$$\Delta h/h = 1\%, \quad \Delta l_{\infty}/l_{\infty} = 15\%, \quad \Delta p/p = 40\%.$$

For the discussion of the conductivity in Ag-Cu alloys,

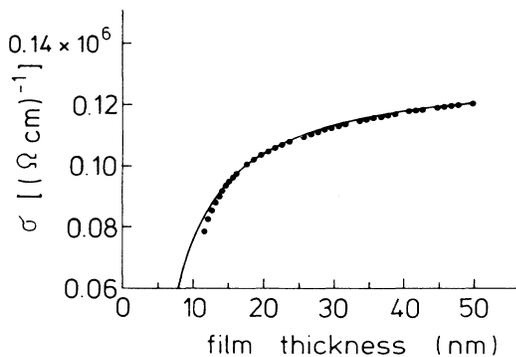


FIG. 6. Fitting example for the thickness dependence of the conductivity of a  $\text{Ag}_{0.2}\text{Cu}_{0.8}$  alloy. . . . ., experiment; —, fitted Namba model with the parameters:  $\sigma_{\infty} = 13 \times 10^4$  ( $\Omega \text{ cm}^{-1}$ ),  $l_{\infty} = 12.3$  nm,  $p = 0.2$ ,  $h = 4.8$  nm.

the dependence of the conductivity and MFP on the composition will be pointed out in the following. The problem of surface scattering in thin metal films was already discussed in Ref. 25.

Figure 7 shows the conductivity of infinitely thick films ( $\sigma_{\infty}$ ) evaluated from the fitting calculations as a function of the alloy composition. The conductivity decreases, approaches a minimum in the middle of the concentration range, and increases again. The alloys show a relative low resistivity, at most  $9 \mu\Omega \text{ cm}$  in the minimum. This is still lower than the values of  $12 \mu\Omega \text{ cm}$  reported by Mizutani<sup>6</sup> for  $\text{Ag}_{0.5}\text{Cu}_{0.5}$  amorphous films, although the grain sizes in our films are of the same order of magnitude as the commonly observed ordered regions in amorphous structures. Mizutani prepared the films by flash-evaporation onto cold substrates. This suggests that the ordered regions are more distorted as in our case. The relative low sensitivity of the conductivity to the "grain size" suggests a low scattering power of the "grain boundaries" in these extremely small dimensions.

The dependence of the MFP (responsible for the measured size effect) on the composition of the alloys is given in Fig. 8. The MFP of the pure metals shows values larger than 25 nm. Similar to the resistivity, a minimum in the middle of concentration range was observed. The behavior of the MFP shown in Fig. 8 can be well understood to be typical in substitution crystals. Ag and Cu have different ionic radii of 0.113 and 0.096 nm, respectively;<sup>26</sup> therefore the fcc lattice of the metastable alloy will be locally distorted and will act as a statistical scatterer on the conduction electrons. The amount of scatterers has a maximum for  $\text{Ag}_{0.5}\text{Cu}_{0.5}$  alloys. On the other hand, the two elements are both monovalent with the same electronic structure. This explains why the MFP is not drastically reduced in the middle of the concentration range. The minimal value is only 10 nm; this is much larger than expected for metallic glasses. The dimensions of the ordered regions in these alloys seems to have no influence on the MFP, although they are of the same order as observed in commonly accepted metallic glasses. An ordered region of about five lattice constants seems to be enough for the conduction electrons to form Bloch states. In such ultramicrocrystalline structures, CE's can move therefore as in a single crystal.

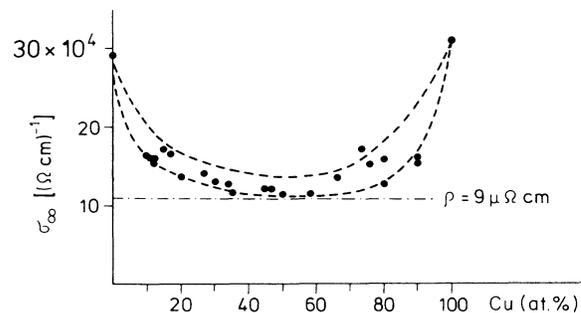


FIG. 7. The dependence of the conductivity ( $\sigma_{\infty}$ ) on the alloy composition.

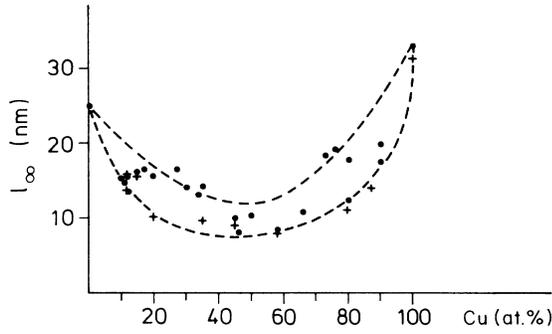


FIG. 8. The dependence of the MFP ( $l_\infty$ ) on the alloy composition.

### V. DISCUSSION

As stated by the reflection model, the electrons in Ag-Cu alloys must unavoidably tunnel through one up to ten potential barriers of the grain boundaries. The conductivity should be determined by the number of electrons which pass (remaining in the same  $k$  state) these grain-boundary potentials included in one MFP. This is given by the function  $G(l_\infty, D, T)$  in Eq. (5). Using the simple form of this function [Eq. (6)], the dependence of the transmission  $T$  on the alloy composition can be roughly estimated. The values of  $v_F$  and  $n_0$ , which depend on the composition were calculated as

$$\frac{1}{v_F(\text{Ag}_{1-x}\text{Cu}_x)} = \frac{x}{v_F(\text{Cu})} + \frac{1-x}{v_F(\text{Ag})}, \quad (10)$$

$$n_0(\text{Ag}_{1-x}\text{Cu}_x) = \frac{4}{[a(\text{Ag}_{1-x}\text{Cu}_x)]^3},$$

with  $a$  being the lattice constant of the alloys as given in Fig. 4. Figure 9 shows the results together with the estimated heights of the potential barriers; these were calculated by averaging of the quantum-mechanical transmission probability of a square-well barrier over the assumed

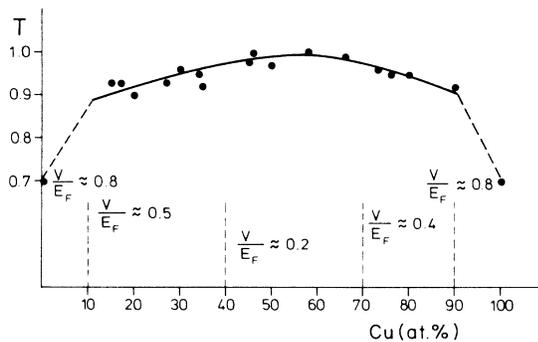


FIG. 9. The transmission probability of the grain-boundary potentials vs the composition of alloys.  $V/E_F$ , the relative height of the potential barrier to the Fermi energy.

width range 0.5–1 nm of the grain boundaries.

For the pure metals, the transmission coefficient is 0.7; this corresponds to the universal value found for all clean metals condensed in UHV at 300 K substrate temperature.<sup>12</sup> The transmission increases rapidly by alloying up to 0.9 and reaches  $T=1$  in the middle of the concentration range. This behavior can be well understood if we keep in mind that the effective height of the potential barrier is given by the difference between the inner potentials of crystallites and grain boundaries. By alloying, the disorder inside the crystallites increases (the MFP will be shorter). Consequently, the height  $V$  of the barrier decreases rapidly from  $V/E_F \approx 0.8$  (corresponding to  $T=0.7$ ) to  $V/E_F \approx 0.4-0.5$  (corresponding to  $T \approx 0.9$ ). The following decrease of the potential barrier to  $V/E_F \approx 0.2$  (corresponding to  $T \approx 1$ ) in the middle of concentration range can be caused by two effects: (i) further increasing disorder inside the crystallites due to alloying and (ii) below 2 nm, the spatial extensions of crystallite and of the grain boundary are of the same order of magnitude. A smearing effect between grain-boundary and intercrystalline potential could be presumed; this smearing, however, cannot be caused by overlapping screening potentials. As just mentioned, the screening length in metals is typically below 0.1 nm and depends only on the density of free charge carriers. The full disappearance of the grain boundary's scattering potential is more likely caused by the overlap of the electron's wave packet over a number of grains: Dealing with  $s$ -type conduction electrons, the wave number  $k_F$  can be regarded as a good quantum number even in amorphous materials. Therefore, the wave packet should have a spatial extension much larger than  $\lambda_F = 2\pi(|\mathbf{k}_F|)^{-1}$ . With  $(|\mathbf{k}_F|)_{\text{Cu}} \approx 1.36 \times 10^{10} \text{ m}^{-1}$  we get  $\lambda_F \approx 0.46 \text{ nm}$ . Therefore, if the grain sizes reach a value below 2 nm it is very likely that the electrons themselves smear over more than one grain and give rise to the apparent disappearance of the grain-boundary scattering.

Referring to the foregoing discussion, it is possible to understand the existence of amorphous metals with low resistivity and large MFP. The opposite case is also possible: amorphous metals with high resistivity and relative large MFP. Very high resistivity [and also negative temperature coefficients of resistivity (TCR)] is commonly obtained by addition of a third component which suppresses the crystallite size into the so-called "amorphous" region. The potential of the barriers then could increase due to the segregation of this third component into the grain boundary. The same mechanism can be expected due to the oxidation. As shown before, an unphysical MFP shorter than the lattice constant (or next-nearest-neighbor distance) is not imperatively necessary even in this case. The true MFP can be determined independently by the analysis of the size effect for each individual alloy.

The reflection model can explain the temperature dependence of the resistivity in a quite different way as done by the present theories. The function  $G(l_\infty, D, T)$  is temperature dependent, and therefore

$$\frac{d\sigma_\infty}{d\theta} = k \left[ \frac{\partial l_\infty}{\partial \theta} G(l_\infty, D, T) + l_\infty \frac{\partial G(l_\infty, D, T)}{\partial \theta} \right] \quad (11)$$

with

$$\frac{\partial l_{\infty}}{\partial \theta} < 0 \quad \text{and} \quad \frac{\partial G(l_{\infty}, D, T)}{\partial \theta} > 0.$$

Consequently, the temperature coefficient of the resistivity (TCR) can be positive, zero, or negative for certain critical values of  $l_{\infty}/D$  and  $T$ . The negative TCR in amorphous metals can be explained in this way, especially for high-resistive metals.

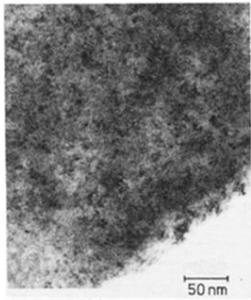
Note, that for a MFP shorter than the lattice constant (an unavoidable result if only the MFP should be responsible for the resistivity) the commonly used Faber-Ziman theory<sup>27</sup> for amorphous metals is not valid.

In the case of the Ag-Cu alloys the temperature dependence was fitted very accurately (relative error smaller than 0.4%) without making use of the function  $G(l_{\infty}, D, T)$ . This is not the case for other materials, where values in the range  $0.3 < T < 1$  are necessary. The temperature dependence of the polycrystalline metals will be discussed in detail elsewhere.<sup>28</sup>

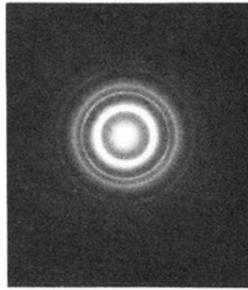
## VI. CONCLUSION

Down to the limit of very small crystallites (ordered regions of about 2 nm), the conductivity of amorphouslike metals can be understood by a grain-boundary reflection model of the conduction electrons. The MFP is not influenced by the size of microcrystals but merely by the disorder (scattering power) inside of these. Consequently, no unphysical short MFP needs to be accepted in high-resistive materials. Moreover, the observed thickness dependence of the resistivity can be explained independent of his absolute value. The existence of amorphouslike metals with low resistivity can be interpreted well as a limit of the reflection model. Negative TCR of high-resistive alloys can be understood avoiding simultaneously the short unphysical MFP. For very small dimensions of the crystallites and large values of the MFP, the resistivity will drastically depend on the site where scatterers are built in: in the crystallites or in the grain boundaries. This explains the large differences in the resistivity measured even for the same material.

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(a)



(b)

FIG. 2. TEM—pictures of a 50-nm-thick  $\text{Ag}_{0.55}\text{Cu}_{0.45}$  film.  
(a) Bright field transmission; (b) diffraction patterns.