

## Nonmetallic conductivity of epitaxial monolayers of Ag at low temperatures

M. Henzler, T. Lüer, and A. Burdach

*Institut für Festkörperphysik, Universität Hannover, Appelstraße 2, D-30167 Hannover, Germany*

(Received 8 July 1997; revised manuscript received 3 December 1997)

Epitaxial metallic monolayers are models for two-dimensional conduction. They provide well-ordered monatomic films with an atom distance as in the bulk. Measurements of the dc conductivity of epitaxial Ag films on a clean Si(111)  $7\times 7$  substrate at about 100 K reveal that the conductance for a thickness down to nearly a monolayer is well described by a simple Drude model with a mean free path given by the film thickness. For lower temperatures (down to nearly 4 K) the conductance of very thin films is reversibly decreased by orders of magnitude. Whereas annealing increases the mean free path for thick films up to twice the thickness, the films with a thickness of less than 2 monolayers show no annealing effect. The results may be discussed with models of amorphous or granular films. The lack of agreement with theoretically predicted temperature dependences may be due to the special structure of epitaxial films. [S0163-1829(98)01639-7]

### I. INTRODUCTION

The ideal two-dimensional (2D) metallic system would be a perfectly periodic arrangement of metal atoms in a plane. The distances between the atoms should be as in the bulk, so that an overlap of the wave functions would enable plane waves. No substrate should disturb the electrons in the layer. At a sufficiently low temperature it should be possible to study the ideal 2D electronic conduction. The conductance of thin metallic films has been studied already over decades. Thin metallic films have been deposited onto glass under bad vacuum conditions, so that both cleanliness and crystallinity have been not well defined. Here the resistance increase has been studied due to surface scattering and small grain sizes as a function of film thickness, temperature, and parameters of production like annealing or oxidation as reported in Ref. 1. In these experiments usually many monolayers had to be deposited, until a continuous, percolating film is found. Some experiments have already been performed in ultrahigh vacuum with a film of Pt on Si.<sup>6</sup> Here a percolation is obtained already in the monolayer range. Since, however, a polycrystalline silicide with varying stoichiometry and only short-range order is formed, the film is far from an ideal 2D system.

Ag films on Ge (001) have been studied after deposition in ultrahigh vacuum.<sup>2</sup> They report that a pseudomorphic monolayer is formed with granular growth on top of it. Conductance could be measured after transfer through air into a cryostat. Morphology, cleanliness, and thickness were not exactly controllable. Nevertheless the measured dependences of two-point resistance change with temperature, and electric and magnetic field could be clearly attributed to 2D conductivity within the regime of weak localization as predicted by theory<sup>3,4</sup> and a surprising incomplete transition to superconductivity at 1.6 K. In a more recent experiment<sup>5</sup> Ag was also deposited in ultrahigh vacuum onto Ge(100). They report a disordered film, which recrystallizes at room temperature to a polycrystalline film with more or less separated grains with (110) orientation. Although rough conductance measurements could be performed *in situ*, the careful experiments at He temperatures were only possible for film thicknesses be-

tween 10 and 30 ML and after transfer through air for preparation of contacts in high vacuum. The conductance measurements are interpreted with a granular structure and an interface, which may produce incomplete superconductivity. In both experiments the incomplete characterization of the film, the lack of good epitaxy, the transfer through air into high vacuum for contact production, and the measurement in a cryostat were limitations for a straightforward interpretation.

The best experimental realization for a 2D metallic film is so far given by epitaxial films grown in ultrahigh vacuum on insulating substrates and *in situ* measurement at low temperatures. Many metals form highly ordered superstructures or grow epitaxially on semiconductors (which are insulating at low temperatures) (see, e.g., Ref. 7). In the superstructures usually the distance of the metal atoms is considerably larger than in the bulk metal. For example, the well-known superstructure of Ag in the Si(111)  $\sqrt{3}\times\sqrt{3}$  Ag structure forms a periodic monolayer. Since the distance between Ag atoms in this monolayer, however, is given by the Si substrate, it is 30% larger than in bulk Ag. Therefore the formation of a 2D metal is not very likely. Those superstructures, however, may form bands of surface states, which may have metallic properties also. This is a quite different conduction mechanism, which has been discussed already some twenty years ago.<sup>8</sup> Recent experiments show that this type of conduction may be realized in superstructures on Si.<sup>9</sup> Here low-temperature measurements have not yet been performed, so a comparison is so far only partially possible. Therefore the measurements of the resistance of epitaxial films are of immediate interest for the study of an ideal 2D metallic system.<sup>10-12</sup> Since the temperature for those earlier experiments was not lower than 70 and about 20 K, respectively, at least the really low temperature was missing for the ideal system. Nevertheless these experiments have shown several 2D features, which will be discussed later.

The resistance of clean metals is usually given by an increase proportional to the temperature and a residual resistance due to defects, which dominates the resistance at low temperatures. Very early a resistance increase due to surface scattering is found, as soon as the dimensions of the sample

(thickness of film or diameter of wire) gets smaller than the mean free path of the electrons.<sup>13</sup> Amorphous and granular films show peculiar effects like positive or negative temperature coefficients of the resistance. Elaborate theoretical models provide detailed explanations.<sup>3,14</sup> For very thin continuous films and at low temperatures also quantization effects have been detected (weak localization).<sup>15,16</sup> The effects are described by strong elastic scattering at defects and at surfaces. Epitaxial thin films should have lower defect densities and therefore a higher mobility, so that quantization effects due to thickness should be seen more clearly. Experiments of epitaxial Ag films on Si(111)  $7 \times 7$  have shown a fairly low conductance even at 20 K, which could be described by interface scattering and weak localization for thicknesses of 5 to 100 ML (monolayers).<sup>11,17</sup> For thicknesses down to 1.5 ML a dramatic increase of resistivity was observed, which requires for description some kind of strong localization. A percolation limit close to 1 ML with a weak dependence on deposition temperature could be detected.<sup>12</sup> Although the effects have been in part quite dramatic, it has been obvious that with a lower temperature for deposition and measurement the effects should be much clearer, so that the chances for a clarification of the conductance mechanisms for the different thicknesses should be high.

Therefore new experiments have been performed down to about 15 K for deposition and nearly 4 K for measurement. Especially for film thicknesses up to 2 ML the conductance in the temperature range from 4 to 100 K brings evidence for a conduction mechanism, which is not described by weak localization, variable range hopping or by a simple activation process. The annealing experiments show clear differences between thin and thicker films. The unexpected percolation results at the lowest temperatures again point to a different conduction mechanism to describe the apparent high critical coverage for the start of percolation.

The results point to a conduction mechanism that may depend on the special structure of epitaxial films.

## II. EXPERIMENTAL SETUP

A special sample holder has been constructed to combine very different requirements: In the same position the sample had to be heated in UHV up to 1000 °C for cleaning and annealing of the silicon sample and to be cooled down to about 4 K for conductance measurements. For that purpose the sample is mounted on a He-cooled copper sample holder with sapphire platelets for electrical insulation and limited thermal coupling, so that during heating by the  $e$ -beam from the backside the holder will not be heated appreciably. The sapphire guarantees the low temperature after heating. The sample is completely surrounded by two He cooled Cu shields with windows, which are opened only during Ag deposition and LEED observation (Fig. 1). With the windows open (for LEED observation and Ag deposition) and closed (for conductance measurements) the lowest temperature at the sample is about 15 and about 4 K, respectively.

The square shaped Si samples with preevaporated Mo contacts in van der Pauw geometry were protected during mounting with a chemical oxide, which was removed in UHV at a pressure of less than  $2 \times 10^{-10}$  mbar after bake-out of the system and an annealing of the sample at 600 °C

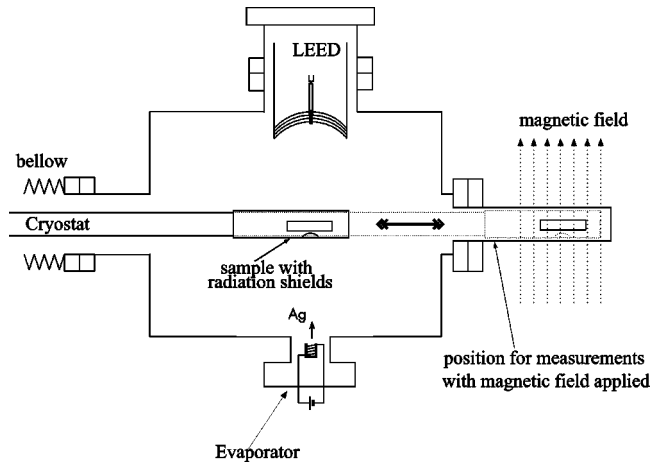


FIG. 1. The experimental setup for sample preparation (15 to 1500 K) and measurement (around 4 K) *in situ*. The magnet has been used for magnetoconductance experiments of similar Ag films to be reported later.

for several hours by flashing. The well annealed Si(111)  $7 \times 7$  surface was the substrate for deposition of Ag at temperatures between about 15 and 100 K. The thickness of the deposited film was monitored with a quartz microbalance. The calibration was possible via the reversible resistance change of a thick and annealed film during heating as described previously.<sup>11,12</sup> The film is even for monolayer thicknesses well ordered like a (111) plane of a Ag bulk crystal. It is epitaxial with a small rotational mosaic disorder of  $\pm 3^\circ$  and a grain size of about 15 nm.<sup>12</sup> Therefore here a monolayer is given by a (111) plane of a Ag crystal with  $1.4 \times 10^{15}$  atoms/cm<sup>2</sup>. The conductance has been averaged over several measurements each in the various van der Pauw arrangements (rotation of the contact connections). The homogeneity of the surface conductance of the sample was checked by measurement of the van der Pauw geometry factor. For evaluation only samples with a constant geometry factor close to one and therefore good homogeneity have been used. The temperature has been measured with a He vapor pressure thermometer and a carbon glass resistor in the base plate of the sample holder. With the radiation shields closed the sample did not show a different temperature as measured with a differential thermocouple between a dummy sample and the base of the cryostat. For the samples for measurement no thermocouple could be used, since the samples had to be flashed to high temperatures in the UHV. Therefore the temperature of the sample may have been a few degrees higher than the base of the cryostat, depending of the actual mounting of the sample. In the figures the measured temperature at the cryostat is always given.

## III. RESULTS

### A. Percolation threshold

Ag has been deposited at about 15 K onto the clean Si(111)  $7 \times 7$  surface. The dc conductance measured during deposition is shown in Fig. 2. The constant conductance up to the increase at 1.4 ML is due to illumination of the substrate by the evaporator. Extrapolating the increase of conductance to zero in a log-log plot yields the critical coverage

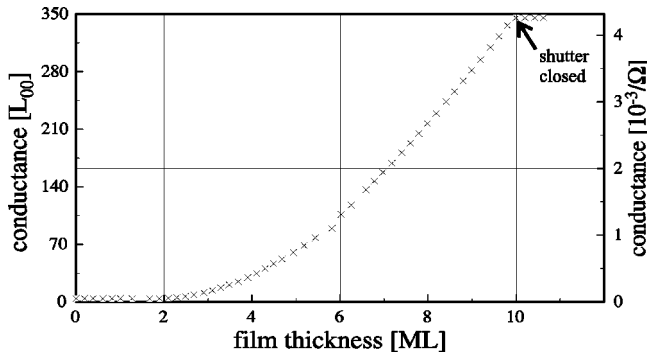


FIG. 2. Conductance of a growing Ag film during deposition at about 15 K. The conductance unit on the left side  $L_{00} = e^2/2\pi\hbar$  is the quantum unit of conductance as used for the description of weak localization (Ref. 4).

$\Theta_c$ , as shown in detail in Ref. 12. The critical coverage as derived by extrapolation to the onset of conductance depends strongly on the temperature of deposition. In Fig. 3 the present data together with the former data<sup>12</sup> reveal a strong increase of the critical coverage with decreasing deposition temperature. This increase at low temperatures is unexpected, since the nucleation probability should be higher and therefore the formation of 3D islands less likely, which yields a lower percolation limit at lower temperatures.

### B. Annealing effects

Continuous monolayers may not be produced at room temperature. After deposition at low temperatures the films are continuous, they may, however, be disordered. Therefore the best films with respect to a brilliant LEED pattern and a high conductivity are obtained after low-temperature deposition and annealing.<sup>18,19</sup> Increasing the temperature after deposition the conductance always increases up to an optimum temperature, where a maximum and with further temperature increase a decrease of conductance was observed due to irreversible breaking up into 3D islands. This optimum annealing temperature increases with increasing film thickness.

The conductance during first heating up to the optimum temperature and the reversible changes during further cool-

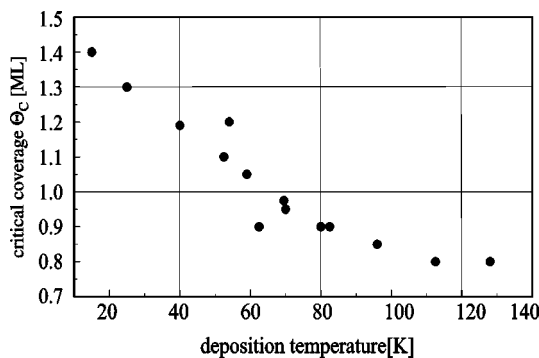


FIG. 3. Critical coverage  $\Theta_c$  for percolation of Ag films on Si(111)  $7 \times 7$  vs temperature during deposition. The coverage 1 corresponds to a complete (111) plane of bulk Ag. The critical coverage has been derived from extrapolation to zero conduction in a log-log plot of excess conduction vs coverage minus critical coverage.

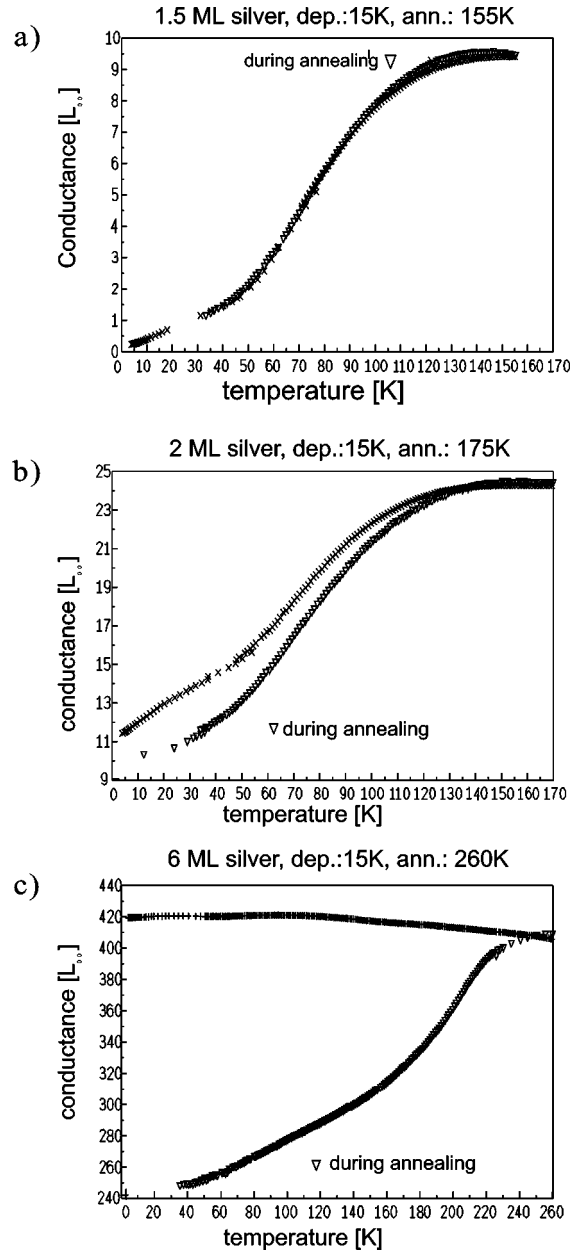


FIG. 4. Conductance of Ag films during first annealing from the deposition temperature at 15 K to the temperature of maximum conductance and subsequent cooling: (a) coverage  $\Theta = 1.5$  ML, (b)  $\Theta = 2$  ML, (c)  $\Theta = 6$  ML. All subsequent heating and cooling cycles followed the cooling curve. Coverage, deposition temperature, and maximum annealing temperature are given at the top of each figure.

ing and heating cycles is shown in Fig. 4. Whereas the increase during the first heating for thick films is irreversible due to annealing of defects in the film, the very thin films show a completely reversible change of the conductance. This difference is obviously due to the different conduction mechanisms in the films with small thicknesses.

### C. Temperature dependence

More information about the conduction mechanism is derived from the reversible resistance changes with temperature after annealing. Whereas a thick film shows the well

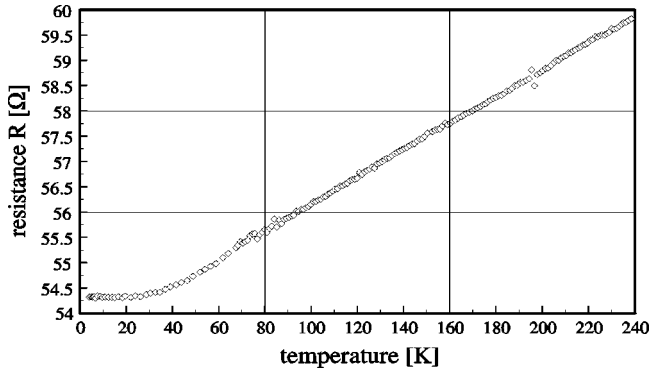


FIG. 5. Resistance vs temperature for a well annealed thick Ag film (10 ML). The residual resistance at low temperatures and the linear increase with higher temperatures reproduce the classical behavior.

known bulk behavior with a constant resistance at low temperatures due to distortions in the film and with a resistance increase proportional to the temperature with the bulk coefficient (Fig. 5), films with thicknesses between 5 and 7 ML thickness indicate weak localization with a maximum conductance at a temperature, which increases with decreasing film thickness (Fig. 6). Another indicator for weak localization is the negative magnetoresistance. First results are shown in Fig. 7 with antilocalization and localization at low and higher magnetic fields, respectively. More results will be reported in a forthcoming paper.

A completely different behavior is seen for films with a thickness less than 2 ML (Fig. 8). The conductance increases in the temperature range from about 4 to 160 K monotonously. The conductance is not affected by annealing, unless the maximum temperature of here 160 K is surpassed, which brings down the conductance irreversibly due to breaking up of the film into 3D islands.

#### D. Thickness dependence

The bulk conductivity of Ag films deposited at about 15 K, annealed to the temperature of maximum conductance and measured at about 4 K is shown in Fig. 9. For comparison also the conductance during deposition at 90 K is shown (taken from Ref. 12).

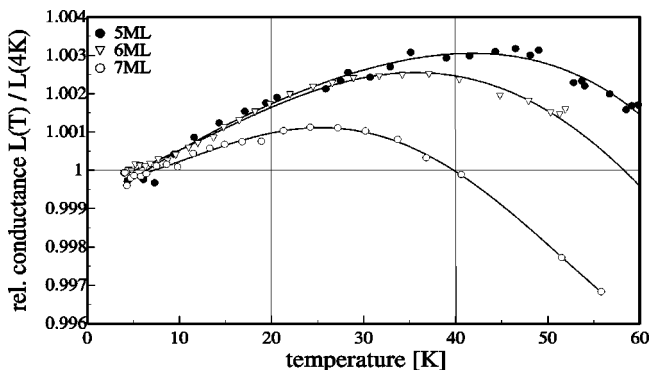


FIG. 6. Conductance vs temperature for well annealed Ag films with intermediate thickness (5 to 7 ML). The decrease at low temperatures is due to weak localization.

The results for thicknesses over 3 ML are well described within a Drude model using the bulk electron concentration of Ag and a mean free path equal to the thickness of the film up to twice the value. The corresponding mean free path is shown on the right side, the expected conductivity using just the film thickness as a mean free path is given as a dashed line in the figure ( $\sigma \propto d$ ).

The very thin films show a distinctly lower conductance due to a different conduction mechanism, which depends on both the temperature and the film thickness, not, however, on any annealing of the film up to the temperature of maximum conductance. The data of the curve “90 K” are taken from the measurement during deposition and therefore without annealing. After annealing they would coincide with the curve “4 K” for thicknesses over 4 ML.

## IV. DISCUSSION

### A. Percolation threshold

The usual percolation models assume a constant conductance per occupied site or per connection between occupied sites. The application of such a model requires here the assumption that a single atom (or a bridge between neighboring atoms) may be represented by a resistor with a fixed value and an arrangement of several atoms may be described as an interconnection of several resistors of the same value as for the single atom. Since the experimental percolation limit is close to the theoretical limit, the units of the model have to be single atoms or at most islands with only a few atoms. The plotting of the experimental data using the models of percolation of single units provides well-defined critical coverages and a power of increase of the conductance compatible with the models.<sup>12,21</sup> Since no other models are available, the discussion uses this model, as if a single atom could be easily represented by a fixed resistor in the network of lattice sites.

The growth of Ag on Si(111)  $7 \times 7$  at room temperature is given by 3D island growth.<sup>20</sup> The thermodynamic equilibrium obviously favors the forming of islands, which are larger the higher the temperature is (Volmer-Weber growth). Therefore a percolation at room temperature with a dc conductivity in the film is observed only after deposition of many monolayers. With lowering of the temperature for deposition (and also for measurement) the mobility of the atoms decreases and the nucleation density increases accordingly.<sup>22,23</sup> For sufficiently low temperatures the surface mobility of the Ag atoms after deposition should be negligible. When all atoms stay at the lattice site, where they arrive, a percolation limit for random arrangement in a triangular lattice is expected at the theoretically predicted value of  $\Theta_c = 0.5$  in the first level.<sup>21</sup> The experimentally observed higher value of  $\Theta_c = 0.8$  by conductance<sup>12</sup> and STM (Ref. 24) has been explained by a start of second-layer growth before completion of the first layer or by preferred nucleation on the triangles of the Si(111)  $7 \times 7$  unit meshes.<sup>12,24</sup> Some mobility of the Ag atoms is needed to reach those nucleation sites. With further lowering of the deposition temperature it is expected that due to further reduction of the atom mobility after deposition the percolation limit is closer to the theoretical value. Also processes like the “downward funneling”<sup>26</sup> would not increase the critical coverage, they would rather

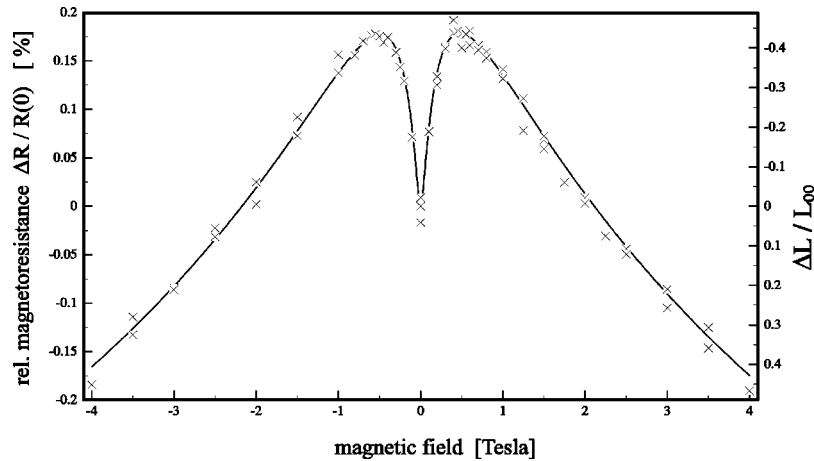


FIG. 7. Resistance vs magnetic field strength for a well annealed Ag film with a thickness of 5 ML. The increase at low fields and the decrease with higher fields resembles weak antilocalization and localization, respectively.

reduce the probability of growth in the second level and therefore reduce the critical coverage. The experimental result of Fig. 3 is therefore in direct contradiction to the expected result.

Two models may explain the result: the first one ascribes the high percolation limit to a growth mode with a high nonthermal mobility, so that the formation of 3D islands is more likely at very low temperatures. Since no calculations are so far known to us, only speculations are possible. Here it would have to be assumed that the arriving atom with its kinetic energy of several eV (binding energy to substrate) has to excite substrate phonons for accommodation to the temperature of the substrate. The lower the temperature the less likely should be the excitation of phonons. Therefore the Ag atom may lose at very low temperatures only a fraction of its kinetic energy per strike and may perform with decreasing temperature more and more hops until an accommodation is completed. This type of “hopping” is known for He accommodation in the field ion microscope.<sup>25</sup> MD calculations for metal atoms, however, cannot reproduce such nonthermal mobility.<sup>26</sup> Such a hopping mechanism would favor 3D island growth at very low temperatures. So far no structural studies for this special problem have been performed. Since available MD studies do not favor such a

mechanism, it will not be considered here further. The other model ascribes the higher percolation limit to a different conduction mechanism for very thin films at very low temperatures.

The thermal mobility of atoms is so low that the film may show more disorder. Simulations with molecular dynamics using classical mechanics for the damping mechanism indicate that an Ag atom should lose its kinetic energy immediately, so that it rests at the lattice site of impingement.<sup>26</sup> Annealing experiments after deposition just over the percolation limit [1.5 ML in Fig. 4(a)] show that the low conductance is not produced by a disorder, which may be annealed afterwards. It is therefore unlikely that a structural disorder produced by low temperature deposition is responsible for the high percolation limit. The reversibility of the conductance in Fig. 8 favors the following model: the apparent high percolation limit at low deposition temperatures is not due to a special growth mode at low temperatures; it rather should be due to a strong temperature dependence of the conductance of the very thin films. The simple percolation model with a constant resistance per site is therefore not applicable. The arrangement of single atoms should provide an overall conductance, which at low temperatures is quite different from the arrangement of fixed conductances at the sites of the atoms.

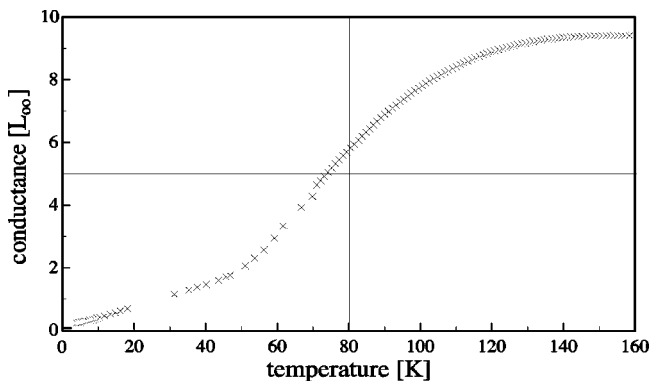


FIG. 8. Conductance vs temperature for a thin Ag film (1.5 ML) deposited at about 15 K. The conductance changes are completely reversible. The conductance at the low temperatures is well below the limit of weak localization.

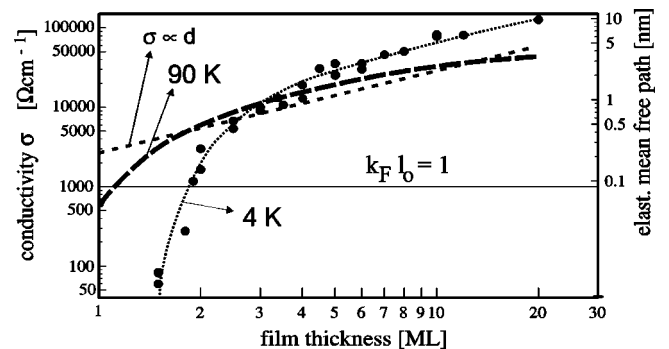


FIG. 9. Conductance of Ag films at about 4 K for coverages from 1.5 to 20 ML after optimum annealing. The dotted line is just a guide for the eye. For comparison the measured conductance during deposition at 90 K (no annealing) is also shown (taken from Ref. 8). The calculated line  $\sigma \propto d$  yields the expected conductivity for bulk Ag with the mean free path equal to the film thickness.

### B. Annealing effects and thickness dependence

Deposition at temperatures less than 100 K produces continuous and conducting films after deposition of about a monolayer. Annealing improves the structural order as seen in the LEED pattern.<sup>18</sup> Although the local order still may be improved, the film breaks up into islands, when a specific temperature (depending on thickness) is surpassed. The surpassing of this maximum temperature is seen both in the LEED pattern (reappearance or intensity increase of substrate spots) and in the conductance (irreversible decrease). Even in the temperature range up to the optimum temperature the annealing effect depends strongly on film thickness.

Films with a thickness of at least 10 ML show the well-known bulk behavior: a strong annealing effect and afterwards the resistance proportional to temperature over a residual resistance due to defects. For less than 2 ML thickness the conductance increase is completely reversible, although some structural annealing is seen in the LEED pattern. The conductance in the very thin layers is therefore not described by defect and phonon scattering even after inclusion of surface and interface scattering. The conduction mechanisms are better discussed with the thickness dependence at 4 K.

As with the former data from measurements at 20 K the present data in Fig. 9 ( $T=4$  K) may be described roughly with two regions.

The line indexed with  $\sigma \propto d$  is calculated with the bulk carrier concentration and a mean free path equal to the thickness  $d$ . For thicknesses over 2.5 ML the mean free path is well described by the thickness  $d$  of the film. The experiments with relatively thick films<sup>27,28</sup> have shown that the surface of the annealed film is nearly perfectly specular for the conduction electrons, since the surface scattering is drastically increased by a low-temperature deposition of additional Ag monolayers, which are rough due to deposition at temperatures below 200 K. So the mean free path not larger than twice the thickness of the film is caused mainly by the interface. It may be the corrugation given by the  $7 \times 7$  superstructure or by a disordered arrangement of the Si adatoms of the superstructure after the formation of the Ag film. Further experiments could reveal the reason in detail.

The strong decline for thicknesses less than 2.5 ML points to a different conduction mechanism. The conductivity crosses the line  $k_F l_D = 1$ , which indicates the metal insulator transition according to the Ioffe-Regel criterion.<sup>29</sup> Simultaneously the 2D conductance is lower than the quantum conductance  $L_{00} = e^2/2\pi\hbar$ . It may be therefore described by a metal-insulator transition. Its special conduction mechanism may be especially derived from the temperature dependence, as discussed in the following subsection.

### C. Models of conduction

The variations of the conductance with temperature and magnetic field are the best indicators for the type of conduction. The present results of temperature dependence will therefore be discussed in this respect.

The temperature dependence of the resistance of thick films (more than about 10 ML) is well described in classical terms: residual resistance at low temperatures and increase proportional to the temperature (Fig. 5). Due to the low measuring temperature the films of intermediate thickness (5 to 7

ML) show a well developed conductance maximum as described by weak localization (Fig. 6). For the low-temperature region a dependence of the conductance with  $\log(T)$  is expected. Since we have a small uncertainty in the temperature scale and the shape of a plot versus  $\log(T)$  depends on the amount of the uncertainty, we cannot check the dependence with  $\log(T)$  unambiguously. The magnetoconductance, however, clearly shows all features of weak localization. The classical negative magnetoconductance is outside the available range of 4 T. As seen in Fig. 7 the positive magnetoconductance of weak localization dominates the changes for fields over 0.5 T. For small magnetic fields the negative magnetoconductance of weak antilocalization due to spin-orbit coupling is clearly visible, which was not yet separable at higher temperatures in the former experiments.<sup>17</sup> A detailed discussion of magnetoconductance is planned to be presented in a forthcoming paper.

For thicknesses of less than about 4 ML the conductance has a positive temperature coefficient in the whole available temperature region. For the film with a thickness of 1.5 ML the conductance increases from 4 to 150 K already by a factor of 30. There is no clear distinction for films from both sides of the metal-insulator transition as seen from the thickness dependence. It looks like a smooth transition. In the literature are several experiments with thin films and models for an increasing conductance with temperature (“negative temperature coefficient for resistance”).<sup>1,14</sup> As long as the mean free path is considerably larger than the atom distance, the weak localization describes the effects perfectly, as is here also the case for films with a thickness of more than 3 ML. In amorphous films an excitation out of localized states over the mobility edge may provide an activated conductivity with an energy barrier given by the distance from the Fermi energy to the mobility edge. A conduction within a band of localized states may yield variable range hopping with  $R(T) \propto \exp(T_0/T)^{1/3}$  according to Mott.<sup>30</sup>

The model has been modified by including different interaction by Efros<sup>31</sup> and Pollack.<sup>32</sup> Here the above equation for variable range hopping is varied by different powers for  $T$  from 0 to 0.5. It has been also tried to use the intrinsic conduction with a gap as in a semiconductor.<sup>33</sup> All these models predict an increase of conductance with temperature. A quantitative fit, however, is not obtained even for the lowest-temperature region, where a superposition of different mechanisms should be less likely. Even adding a reasonable amount of uncertainty to the indicated temperature does not yield a straight line in the appropriate plots. The lack of agreement is demonstrated with the variable range hopping in Fig 10.

For granular systems also other mechanisms have been discussed.<sup>34–36</sup> The transition from one grain to the next one may be blocked by the energy of charging up a grain (Coulomb blockade with energy  $E = e^2/2C$ ). A granular film may be considered as a network of isolated grains. When the tunneling conductance is high, the weak localization regime is found. For a tunneling conductance lower than the quantum conductance  $L_{00}$  the conductance will be activated by the Coulomb energy and additionally possibly by a barrier between the grains. Experiments with oxidized grains of copper show an activated change of conductivity.<sup>35</sup>

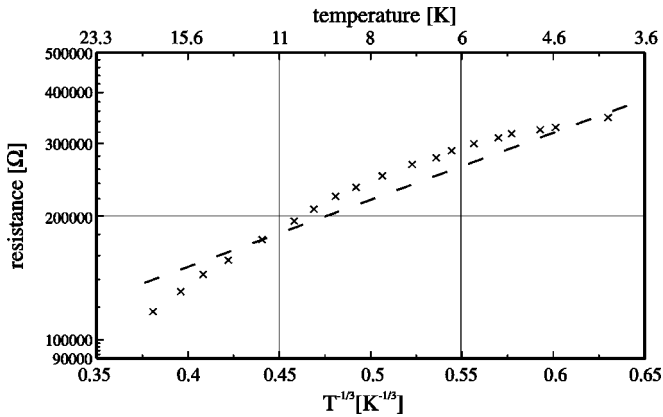


FIG. 10. Data of Fig. 8 as a plot to check for a dependence as given by the variable range hopping:  $R(T) \propto \exp(T_0/T)^{1/3}$ .

Our epitaxial films cannot easily be described by an amorphous or a granular system. The film consists of grains with a size of about 15 nm. Defects within the grains produce only a small broadening of high-order diffraction spots in the LEED pattern, so that the grains should be fairly perfect.<sup>37</sup> The grain boundaries are small-angle grain boundaries within 3 degrees of in-plane rotation. They should provide a good metallic contact, since they consist of a series of dislocations, which are 10–20 atomic distances apart. Since the substrate spots are invisible even after a deposition of 2–3 ML, there cannot be an appreciable distance between the grains. Therefore the LEED pattern indicates that the film is well ordered and without holes or gaps between the grains, so that the small-angle grain boundaries are the only substantial defect.

The conduction around 90 K is metallic for thicknesses down to nearly 1 ML. The high-temperature value for the film with 1.5 ML in Fig. 8 agrees completely with the value from the Drude approximation in Fig. 9. Even with a thickness of 1 ML the conductivity of the film deviates only by a factor of 4 from the simple Drude model (see Fig. 9). The metallic conduction points to an atom arrangement similar to a bulk metal, so that wave functions overlap as in the metal. Only during cooling to 4 K the conductance decreases reversibly to a different type of conduction mechanism. Therefore only at low temperatures a localization mechanism has to come into effect. The very thin films may have a nearly random perturbation by the  $7 \times 7$  structure of the substrate, which may be shielded only by multiple layers. A localiza-

tion as in a granular system would require gaps between the grains. Those gaps would have to be several nm wide to yield an appreciable tunneling resistance for continuum states on both sides of the gap. Due to different sizes and the small rotational disorder the states may be localized and therefore the coupling hindered at low temperatures. For such a mechanism an activated conduction would be expected, which is not observed.

Therefore so far no model can provide a satisfactory agreement with the observed temperature dependence, although the present system provides more information on the structure of the film than in most former experiments.

## V. CONCLUSION

Epitaxial metallic monolayers on an insulating substrate should be the ideal models for a 2D metal with atomic structure. The semiconductor quantum wells are truly two dimensional; they, however, exceed atomic dimensions by far. Another example might be the conduction in surface state bands of silicon, as recently revealed by Hasegawa and co-workers.<sup>9</sup> Here, however, so far measurements of temperature dependence and of magnetoconductance are missing. The epitaxial monolayers have the advantage that their structure may be easily measured by diffraction and by microscopy. Simultaneously a wide range of structural modifications is possible with variation of thickness, temperature of deposition, and annealing. The present experiments show that the very thin films are metallic down to 100 K. It is not clear, however, which mechanism decreases the conductance around 4 to 20 K. Therefore new experiments of structure determination at low temperature are in progress. Simultaneously, however, it is highly desirable that theoretical investigations reveal special features of low-temperature crystal growth and show new possibilities of conduction mechanisms, which may then be checked experimentally in detail.

## ACKNOWLEDGMENTS

The authors acknowledge fruitful discussions with W. Apel, H.U. Everts, and R. Haug at the University of Hannover, and F. Komori at the University of Tokyo. Support by the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

<sup>1</sup>K.L. Chopra, *Thin Film Phenomena* (Krieger, Malabar, 1985).

<sup>2</sup>N.J. Burns, J.R. Lince, R.S. Williams, and P.M. Chaikin, *Solid State Commun.* **51**, 865 (1984).

<sup>3</sup>E. Abrahams, P.W. Anderson, D.C. Licciardello, and T.V. Ramakrishnan, *Phys. Rev. Lett.* **42**, 673 (1979).

<sup>4</sup>P.W. Anderson, E. Abrahams, and T.V. Ramakrishnan, *Phys. Rev. Lett.* **43**, 718 (1979).

<sup>5</sup>A. Iradj-zad and M. Hardiman, *Solid State Commun.* **83**, 467 (1992).

<sup>6</sup>R.S. Markiewicz and L.A. Harris, *Phys. Rev. Lett.* **46**, 1149 (1981).

<sup>7</sup>V.G. Lifshits, A.A. Saranin, and A.V. Zotov, *Surface Phases on*

*Silicon: Preparation, Structures and Properties* (Wiley, Chichester, 1994).

<sup>8</sup>M. Henzler, in *Surface Physics of Crystalline Material*, edited by J. Blakely (Academic Press, New York, 1975).

<sup>9</sup>X. Tong, S. Hasegawa, and S. Ino, *Phys. Rev. B* **55**, 1310 (1997).

<sup>10</sup>M. Jalochowski and E. Bauer, *Phys. Rev. B* **37**, 8622 (1988); **38**, 5272 (1988).

<sup>11</sup>R. Schad, S. Heun, T. Heidenblut, and M. Henzler, *Phys. Rev. B* **45**, 11 430 (1992).

<sup>12</sup>S. Heun, J. Bange, R. Schad, and M. Henzler, *J. Phys.: Condens Matter* **5**, 2913 (1993).

<sup>13</sup>K. Fuchs, *Proc. Cambridge Philos. Soc.* **34**, 100 (1938).

- <sup>14</sup>N.F. Mott, *Conduction in Non-Crystalline Materials* (Oxford University Press, Oxford, 1987).
- <sup>15</sup>N. Mott, *Conduction in Noncrystalline Material* (Clarendon Press, Oxford, 1987).
- <sup>16</sup>G. Bergmann, Phys. Rep. **107**, 1 (1984).
- <sup>17</sup>R. Schad, S. Heun, T. Heidenblut, and M. Henzler, Appl. Phys. A: Solids Surf. **55**, 231 (1992).
- <sup>18</sup>S. Heun, *Fortschr.-Ber. VDI Reihe 9* No. 159. (VDI-Verlag, Düsseldorf, 1993).
- <sup>19</sup>D. Thielking, Ph.D. thesis, Hannover, 1993.
- <sup>20</sup>S. Tosch and H. Neddermeyer, Phys. Rev. Lett. **61**, 349 (1988).
- <sup>21</sup>D. Stauffer and A. Aharony, *Introduction to Percolation Theory* (Taylor and Francis, London, 1991).
- <sup>22</sup>J.A. Venables, G.D.T. Spiller, M. Hanbücken, Rep. Prog. Phys. **47**, 399 (1984).
- <sup>23</sup>S. Stoyanov and D. Kashchiev, in *Current Topics in Material Science*, edited by E. Kaldis (North-Holland, Amsterdam, 1981), Vol. 7, Chap. 2 .
- <sup>24</sup>G. Meyer and K.H. Rieder, Appl. Phys. Lett. **64**, 3560 (1994).
- <sup>25</sup>E.W. Mueller and T.T. Tsong, *Field Ion Microscopy: Principles and Applications* (American Elsevier, New York, 1969).
- <sup>26</sup>J.W. Evans, D.E. Sanders, P.A. Thiel, and A.E. DePristo, Phys. Rev. B **41**, 5410 (1990).
- <sup>27</sup>E.Z. Luo, J. Wollschläger, F. Wegner, and M. Henzler, Appl. Phys. A: Mater. Sci. Process. **60**, 19 (1995).
- <sup>28</sup>E.Z. Luo, S. Heun, M. Kennedy, J. Wollschläger, and M. Henzler, Phys. Rev. B **49**, 4858 (1994).
- <sup>29</sup>A.F. Ioffe and A.R. Regel, Prog. Semicond. **4**, 237 (1960).
- <sup>30</sup>N.F. Mott and E.A. Davis, *Electronic Processes in Non-Crystalline Materials* (Clarendon Press, New York, 1971).
- <sup>31</sup>A.L. Efros and B.I. Shklovskii, J. Phys. C **8**, L49 (1975).
- <sup>32</sup>M. Pollack, Philos. Mag. B **42**, 781 (1980).
- <sup>33</sup>R.J. Hodgkinson, J. Phys.: Condens. Matter **2**, 6563 (1990).
- <sup>34</sup>S. Kobayashi and F. Komori, Prog. Theor. Phys. Suppl. **84**, 224 (1985).
- <sup>35</sup>R. Yamada, S. Katsumoto, F. Komori, and S. Kobayashi, J. Phys. Soc. Jpn. **62**, 2229 (1993).
- <sup>36</sup>F. Komori (private communication).
- <sup>37</sup>M. Henzler, Surf. Sci. **357**, 809 (1996).