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Conductance of Ag on Si (111): a two-dimensional percolation problem

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Abstract. It is well known that silver grows epitaxially on Si (111)- 7×7 . We deposited several monolayers of silver on silicon under UHV conditions ($p = 1 \times 10^{-10}$ mbar). Due to the perfect substrate we were able to observe electric conduction of one monolayer of silver. We made *in situ* measurements of the conductance during growth in the temperature range 50–130 K. We show that conduction starts at a critical coverage less than a monolayer, and the critical coverage decreases for increasing deposition temperature. We discuss a percolation model and several possible growth modes.

1. Introduction

Percolation problems have been treated by various authors with theoretical approaches [1–7], Monte Carlo simulations [8] and experimental investigations [9–14]. Several properties describing the systems follow a power law near the percolation threshold [6]. Most experiments deal with metallic grains in an isolating matrix with varying filling factors [9–12]. Usually these grains are not well defined in shape and size, so that these systems cannot be attached to different lattice types. Experiments that are comparable to Monte Carlo simulations require the use of spheres with uniform size arranged within a periodic lattice geometry. A simple and perfect model is given by atoms arranged on the sites of a crystal lattice, but in three dimensions it is difficult to realize different filling factors in such a crystal. Filling the necessary voids with other atoms would solve these problems, but the physics becomes more complicated. In two dimensions, however, it should be possible to fill an increasing fraction θ of surface sites with atoms until completion of a monolayer. Prerequisites for such an experiment are a perfect flat substrate (on an atomic scale) and a metal that grows epitaxially on this substrate. A substrate meeting these prerequisites in an almost perfect way is the Si(111)- 7×7 surface, which can be prepared and characterized easily under ultra-high-vacuum (UHV) conditions with average terrace widths of more than 150 nm [15]. A metal that is known to grow epitaxially on this substrate is silver [16]. At low temperatures it grows in (111) layers [17] with no intermixing with the substrate [18]. The geometry of the Ag(111) plane corresponds to a two-dimensional triangular-site percolation problem.

2. Experiment

All preparations and measurements were done in a UHV system (base pressure 1×10^{-10} mbar) as described elsewhere [19]. The substrates are squares of single-crystal

Si(111) wafers ($800 \Omega \text{ cm}$) with contact areas in the corners and notches on each side. The conductance is measured and evaluated according to van der Pauw [20]. The electrical contacts consist of 150 nm Mo deposited under high-vacuum conditions. After heating to 1400 K in UHV, the sample showed a bright 7×7 low-energy electron diffraction (LEED) pattern. With Auger electron spectroscopy (AES) no contamination of the surface, except for a small trace of carbon ($\sim 1\%$), could be detected. The Mo contacts were covered with 50 monolayers (ML) of Ag, which was deposited in UHV to ensure good contacts between the Mo and even the very thin films. The film thickness was monitored by a quartz microbalance, which was calibrated by comparing the measured temperature coefficient of the resistance of thick Ag films ($> 100 \text{ ML}$) with bulk literature data [21]. We measured at temperatures between 50–130 K.

3. Results

The conductance of the sample was measured during deposition. The silver was deposited at a rate of about 1/20 ML per minute under UHV conditions. A schematic picture of the electric connections is shown in figure 1. While a constant current flowed through the sample, the two-probe voltage U_2 , as well as the four-probe voltage U_4 , was measured. The two-probe voltage measured the voltage drop over the whole sample, including all contact resistances, whereas the four-probe voltage measured the resistance in the centre portion of the sample without contact resistances. Two typical curves are shown in figure 2. The starting value of the voltages is due to the substrate being illuminated by the silver evaporator. The silver film starts to conduct at a total coverage of about 0.9 ML, which is called the critical coverage θ_c . This is visible in a drastic decrease of both voltages. Similar curves have been measured for substrate temperatures from 50–130 K. For evaluation, the conductance of the film was determined and a critical coverage according to (1) was derived as described in the following section.

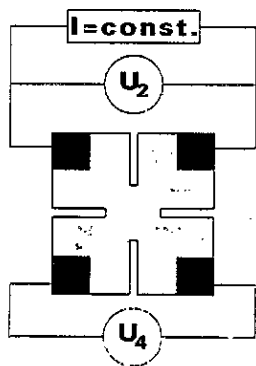


Figure 1. Experimental set-up for the conductance measurements. The size of the sample is $15 \times 15 \text{ mm}^2$. The black areas in the corners are the contact areas.

4. Discussion

4.1. Derivation of the silver film conductance

The advantages of the silicon substrate have already been discussed, but for electric measurements silicon has one handicap: it is not an insulator. So the measured voltages

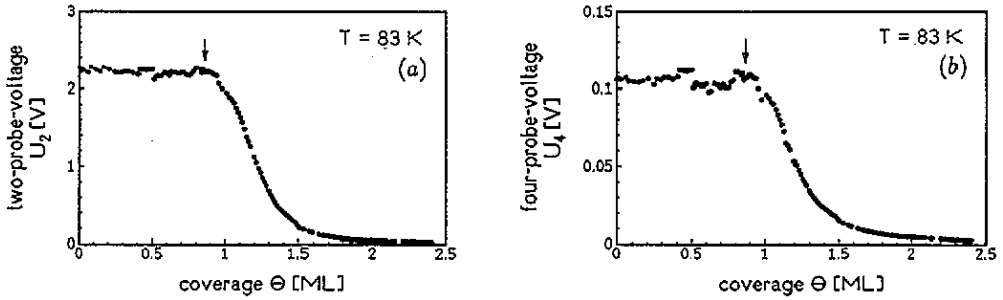


Figure 2. The measured two- and four-probe voltages for a current of $1 \mu\text{A}$ (figure 2(a) and (b) respectively). Both voltages are measured simultaneously *in situ* during the silver deposition at 83 K (rate: 1/20 ML per minute). The onset of silver conductance is indicated with an arrow.

contain contributions from two different paths: the silicon wafer conducts parallel to the silver film. Both planes are connected internally and externally via different contact resistances. A schematic picture of the parallel planes with the electric connections is shown in figure 3. With the help of the circuit diagram the silver resistance A_g is calculated from the measured voltages U_2 and U_4 . The values of the silicon resistance, S_i , and of its contact resistances, H , are derived from the same voltages for coverages smaller than the critical coverage. For those small coverages the silver does not contribute to the conductance. The values of S_i and H are assumed to be constant for all coverages. It is therefore possible to calculate unambiguously the two unknown resistances A_g and M from the two voltages U_2 and U_4 . The constancy of the conductance up to more than half a monolayer of silver coverage is proof that there is no contribution of the space charge layer of the substrate to the conductance. The silver conductance ($= 1/\text{silver resistance}$) derived from the results in figure 2 is shown in figure 4. It is given in units of $L_{00} = e^2/2\pi^2\hbar \approx 1/(81 \text{ k}\Omega)$.

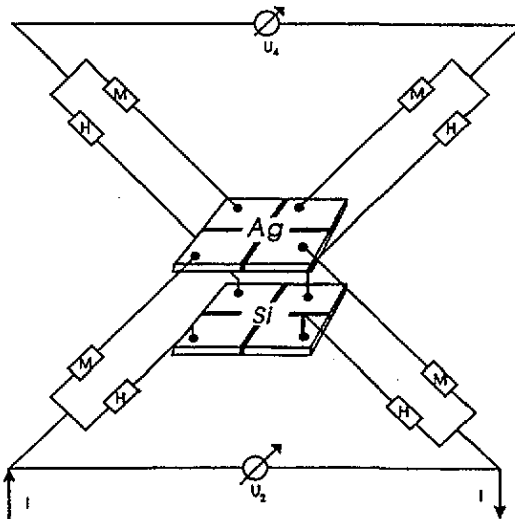


Figure 3. Schematic representation of resistances and measuring circuit. The silver resistance A_g and the metal contact resistances M are calculated from the measured voltages U_2 and U_4 . The silicon resistance S_i and its contact resistances H are derived from the same voltages as for the uncovered silicon.

4.2. Percolation

Percolation theory predicts for the conductance G near the percolation threshold (= critical coverage θ_c), a behaviour following a power law

$$G(\theta) \propto (\theta - \theta_c)^t \quad \text{for } \theta > \theta_c \quad (1)$$

with an exponent t depending on dimension. Accordingly, the critical coverage θ_c has been derived using all available data. To check for shortcomings of the evaluation procedures, the measured values of U_2 and U_4 have been used directly for the determination of θ_c . All data are shown in figure 5. To check the validity range, the silver conductance as shown in figure 4 is presented in a log-log plot against the reduced coverage $(\theta - \theta_c)/\theta_c$ in figure 6. Values following a power law $(\theta - \theta_c)^t$ give a straight line. The silver conductances near the critical coverage show large error bars in the logarithmic plot, because their magnitude is close to the noise amplitude. These points are nevertheless important for the determination of the critical exponent, because the power law is only expected close to the critical coverage. The conductance can be fitted with a straight line up to $(\theta - \theta_c)/\theta_c \approx 0.2$. For higher coverages a deviation from the power law is visible. The values for the power-law exponent t scatter strongly (from $t = 0.5$ to $t = 3$), with large error bars (in some cases up to 100%), but they show no dependence on deposition temperature. Averaging, the following value is derived:

$$t = 1.36 \pm 0.25. \quad (2)$$

Percolation theory gives for two dimensions a value $t \approx 1.3$ [3], and for a triangular lattice $\theta_c = 0.5$ [6]. The value of the critical coverage will be discussed later. The agreement between the theoretically predicted exponent and the value found experimentally indicates that we found a percolation threshold in two dimensions, as expected from the geometry of the problem.

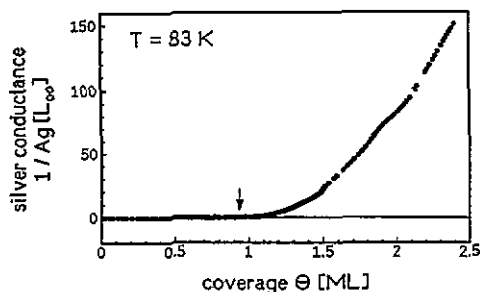


Figure 4. The silver conductance (=1/silver resistance) determined from the measured voltages shown in figure 2. The conductance is given in units of $L_{00} = e^2/2\pi^2\hbar \approx 1/(81 \text{ k}\Omega)$.

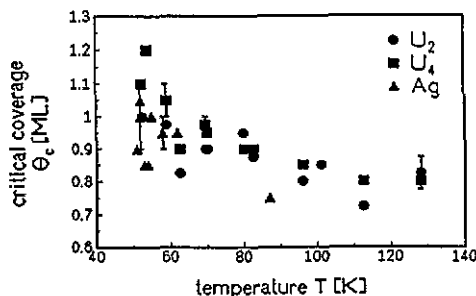


Figure 5. The temperature dependence of the critical coverage θ_c , i.e. the coverage at which the silver conduction starts.

The deviations from the power law for coverages higher than $(\theta - \theta_c)/\theta_c \approx 0.2$ can be understood in terms of effective medium theory (EMT) [5]. This theory predicts $\sigma \propto \theta^2 - \theta_c^2$. As shown in figure 7, our measurements can be fitted with this theory. It is well known that EMT overestimates the critical coverage θ_c [8]. This feature can be found in our fits,

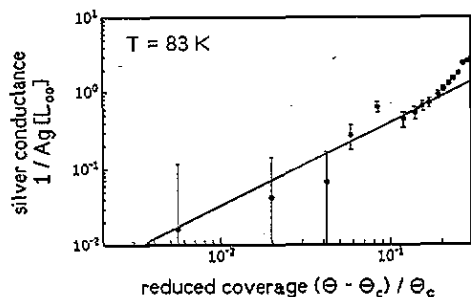


Figure 6. A log-log plot of silver conductance against reduced coverage. The straight line indicates the best fit for a power law $G(\theta) \propto (\theta - \theta_c)^t$ with $\theta_c = 0.9$ ML and $t = 1.23$ (data from figure 4).

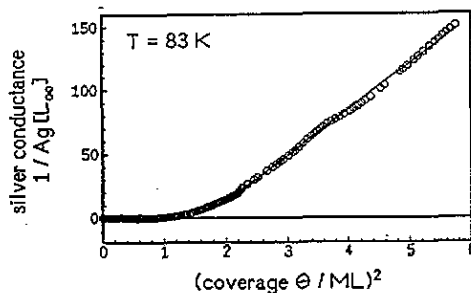


Figure 7. The silver conductance against the square of the coverage. The straight line indicates the best fit for a quadratic dependence of the conductance from the coverage for high coverages.

too. So the correct explanation for the higher coverage measurements ($(\theta - \theta_c)/\theta_c > 0.2$) is given by EMT. This interpretation is supported by a Monte Carlo simulation [8]: in this paper the conductivity σ for different filling factors θ is simulated on a triangular lattice. In agreement with our measurements they found a power law for $(\theta - \theta_c)/\theta_c < 0.2$ with an exponent $t = 1.23 \pm 0.04$. For higher coverages the Monte Carlo values did not fit to a power law, but they did fit with EMT.

As already mentioned percolation theory predicts for the two-dimensional triangular-site percolation a critical coverage of $\theta_c = 0.5$. In our case the measured values lie in a range from 1 ML at 50 K to 0.75 ML at 110 K. This temperature dependence points to a difference in atomic arrangement close to the coverage θ_c . Two models may describe this effect. The lower the temperature the more disordered the film should be; therefore, a higher coverage is needed at lower temperatures to provide a measurable conductance. This model may be disregarded since the LEED pattern shows, for all temperatures, an epitaxial structure with no dramatic increase of defects at lower temperatures [17]. A lower carrier mobility due to localization of the electrons should not influence the critical coverage.

A second model may ask for a temperature-dependent growth mode. It is reported for metals such as Cu [22], Pt [23] and Ag [24] that, for a given temperature range, diffusing single atoms may not cross a step edge due to a potential barrier. If this temperature limit (which is not yet determined for a monolayer Ag on Si) is around 80 K, a monolayer will completely spread on Si for all higher temperatures. For lower temperatures, however, a higher total coverage is needed for the same coverage in the first layer. This effect could easily describe the temperature dependence. A direct structural measurement to prove the validity of this model is still missing.

On the other hand, the model does not describe why more than half a monolayer is needed for percolation. The theoretical model requires a random deposition onto lattice sites, with no mobility afterwards. For heteroepitaxial growth with the silver lattice constant, a surface diffusion definitely occurs so that epitaxial islands are formed. Their diameter is of the order of 10 nm [17]. The larger the islands (with non-wetting edges), the higher the coverage in the first level has to be for percolation. Special measurements (like *in situ* low-temperature scanning tunnelling microscopy) are needed to check those considerations. Also open is the question of whether a single-atom chain, or a chain with a width of only a few atoms, may provide a conducting path between epitaxial islands. Such an effect would also increase the critical coverage.

5. Conclusion

We showed that, using a very smooth substrate, electric DC conduction of less than a monolayer of silver is possible. The onset of conduction can be described within the framework of percolation theory. A more detailed understanding of the atomic processes on the surface would require more structural information about the atomic arrangement, obtained either with diffraction methods, like spot-profile analysis of LEED, or with microscopic methods, like scanning tunnelling microscopy, which have to be done *in situ* at low temperatures.

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