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Conductivity of ultrathin Pb films during growth on Si(111) at low temperatures

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Abstract. The electronic properties of thin metallic films strongly depend on their structure. For the preparation of ultrathin films (0.8 to 12 ML) Pb was grown on a Si (111)-7 × 7 structure at temperatures below 25 K. Percolation as measured by DC conductance starts at 0.7 ML. Up to 4 ML the growing film is disordered. During continuation of the growth the film recrystallizes epitaxially and layer-by-layer growth connected with oscillations of conductivity and LEED intensities is observed even at 25 K. The defect structure as determined by SPA-LEED and the quantum size effect allow a quantitative description of the conductivity oscillations. The experiments show, that the electronic properties of ultrathin Pb films are clearly correlated with the structural properties of the film.

PACS. 73.61.At Metal and metallic alloys – 68.55.Jk Structure and morphology; thickness; crystalline orientation and texture – 81.40.Rs Electrical and magnetic properties

1 Introduction

Ultrathin films, that are films with a thickness of only a few monolayers, are already important for technological applications (see *e.g.* [1]), they are, however, especially attractive for fundamental reasons: the structure may be different due to the small thickness and due to the substrate and the electronic properties may be modified due to the narrow channel in the film. Although those issues have been addressed already many decades ago [2], clear answers are rare, since the requirements for well defined experimental situations are not so easy to meet. The system should be simple and well characterized, therefore the best realization would be a freestanding crystalline monolayer without substrate. Since such films are so far impossible to produce with metals, the next best solution is an epitaxial metallic film on a defect free and insulating substrate in ultra high vacuum. Among the many possibilities so far the best results have been obtained with metals on Si(111) and Si(100), since these surfaces may be well prepared step free over large distances with well-known superstructures. Silicon is an insulator at low temperatures, so that especially for samples with low doping level, temperatures below 50 K are sufficient in most cases. A disadvantage of silicon, however, is the formation of silicides with most metals. Therefore restriction to metals without silicide formation like Ag, Pb or In is favourable. Extensive experiments with Ag have shown, that it is possible to produce epitaxial monolayers and to characterize the conductance at low temperatures with DC conductance and magnetoconductance, demonstrating, that the electronic properties are dramatically modified by scattering processes, leading to weak and strong localization [3–5]. Experiments with Pb films have shown, that here the quantum size effect and the variation of band structure due to thickness are additionally very important, especially since the growth mode is close to layer-by-layer growth [6–9, 11]. New experiments with Pb have been promising since an improved control of surface structure during growth of ultrathin films provides better characterization of the films. The superconductivity enables additional insight into the electronic processes. Therefore the electronic properties of ultrathin Pb films have been studied in detail with DC conductance, magnetoconductance, and Hall effect at low temperatures during growth and after annealing. Here the DC conductance results are reported with special emphasis on the correlation of structural properties with the observed conductance.

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2 Experimental setup

The Si(111) samples size $15 \times 15 \times 0.3\,\text{mm}^3$ have been prepared in ultra high vacuum by flashing chemically oxidized samples up to 1500 K. Using well oriented samples ($< 0.1°$) and a proper cooling cycle (careful annealing around the temperature of superstructure transition at 800 °C) provided fairly step free surfaces (terraces> 100 nm), as checked with spot profile analysis of LEED (SPA-LEED) [15–17]. Using a He cryostat the sample has

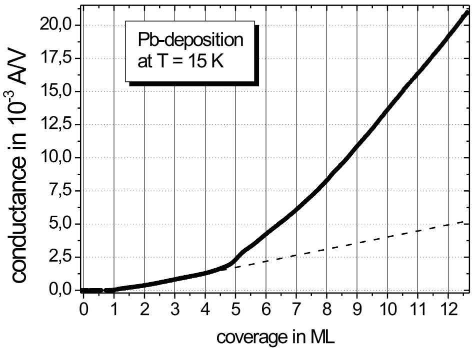

Fig. 1. Conductance of a Pb film during deposition onto insulating Si(111) 7×7 at 15 K. Up to 0.7 ML no conductance is observed. Up to 4.5 ML the linear increase points to bulk defect scattering (the conductance L is proportional to the thickness d). After recrystallization at about 5 ML the increase is steeper and more than linear.

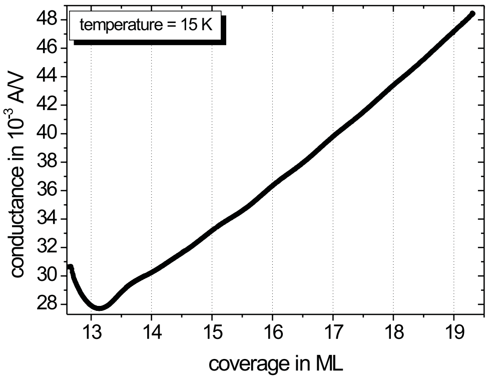

Fig. 2. Conductance of a Pb film during deposition at 15 K onto a film of 12.8 ML, which had been annealed at about 160, to reduce all kinds of defects.

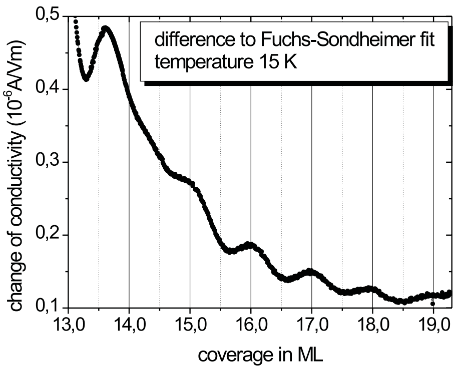

Fig. 3. Difference of the conductance in Figure 2 to an expected conductance as calculated with a constant surface scattering using the Fuchs-Sondheimer equation with fitting parameters for high coverages.

been cooled to 15 K during deposition of Pb and, after closing of the cooled shutter, to 4 K for measurement. Pb has been deposited by thermal evaporation at a rate of 0.1 to 0.2 ML/min. The calibration was taken from the oscillation of conductance and LEED intensities and monitored by a quartz microbalance. For the electric measurements the samples had four predeposited Mo contacts in the corners of the (111) square for a v.d.Pauw measurement, providing directly the 2D conductance per square [13]. The conductance is converted into bulk conductivity (A/Vm) using the known average thickness, when appropriate for a plot. The temperature has been measured with a Pt100 resistance thermometer and a Si diode at the bottom of the cryostat, which was calibrated against a Si diode glued on a dummy Si sample in the measuring position. The superconductivity of bulk Pb on top of the Si sample provided a calibration point. More details for the measurements are found in [11,12].

3 Results

The films used in this study have been deposited onto the Si(111) 7×7 structure, since extensive structural investigations are available for that surface [16,17]. In these papers, the following surface and bulk defects of the Pb films have been characterized quantitatively by SPA-LEED: atomic steps, grain boundaries, bulk disorder and the surface roughness and surface correlation length during epitaxial layer-by-layer growth. Percolation started at 0.7 ML. Further depositions up to 12 ML yielded first a linear increase up to 4 to 6 ML and then a step-like increase with an increasing slope after the recrystallization (Fig. 1). The step-like increase occurs at the coverage where the oscillations of the LEED intensities start, which depends on the deposition temperature and probably also on the defect structure after annealing. Here the increase occurs at about 5 ML (deposition temperature is 15 K). A shift of the steep increase to about 4 ML is observed for a deposition around 70 K in accordance with the onset of LEED intensity oscillations at this temperature.

Before recrystallization the conductance increases linearly with thickness, pointing to constant and low bulk mobility due to bulk defect scattering. Here the surface and interface scattering is less important. After recrystallization the conductance is increased due to crystalline order. Now the interface scattering is obviously the dominant scattering process, so that the average conductivity increases with thickness. The surface scattering should be less important, since the oscillation of conductance, as described below, requires a variation of surface scattering. The elastic scattering time has been derived from magnetoconductance in reference [12], Figure 5. The constant low value for the disordered films (up to 5 ML) confirms the bulk scattering, the linear increase after recrystallization points to the dominance of the interface (and surface) scattering.

The roughness scattering at the surface is more clearly seen for deposition of Pb at low temperatures (15 K) onto well annealed films, so that the starting condition is a film with low bulk and low surface scattering, the interface scattering, however, should not be changed, since the mean free path is still close to the film thickness [12] (Fig. 2).

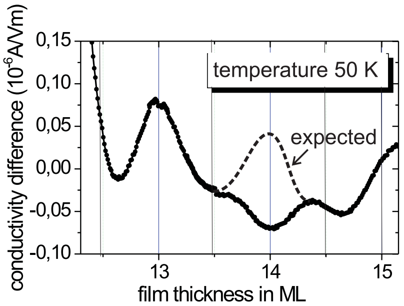

Fig. 4. Changes of the conductivity of a Pb film during deposition at 50 K onto an annealed Pb film (thickness 12 ML) after subtraction of the expected conductance due to a surface scattering independent of thickness. The deposition temperature has been selected to improve the layer-by-layer growth, so that the quantum size effect around 14 ML is more clearly seen. The dashed curve indicates the expected variation with roughness scattering and without quantum size effect.

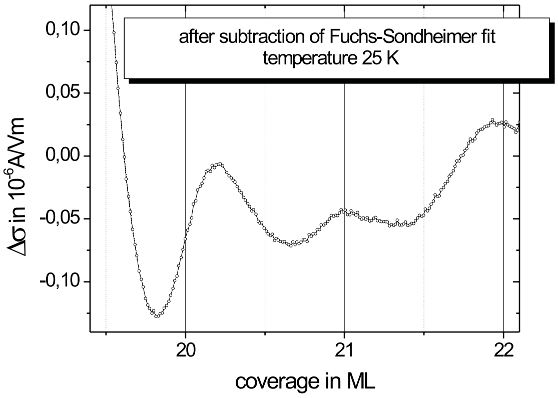

Fig. 5. Same as Figure 3 and 4 after starting deposition at 25 K on top of 18.5 ML after annealing. The reduced maximum at 21 ML shows, that all multiples of 7 ML show a reduction of conductance due to opening of a new scattering channel with a new subband.

The strong decrease during the first half monolayer is due to the growth of separated islands at the surface, which produce an increase of roughness and therefore a diffuse scattering of electrons at the surface. Here the effect of additional surface scattering is larger than the expected increase of conductance due to increasing thickness, so that at first the conductance decreases. Only during further deposition the conductance increases with thickness. The oscillating roughness during the layer-by-layer growth (as derived from the LEED intensity oscillations [16,17]) is reproduced in a small oscillation of conductance. This oscillation is more clearly seen after subtraction of a smooth curve as calculated with a constant surface scattering (using the Fuchs-Sondheimer equation with a constant roughness parameter to fit the changes for high coverages) (Fig. 3). The layer-by layer growth has not been observed for growth of Ag at low temperatures onto annealed Ag films, where the roughness increases monotonically with the square root of the thickness [18]. At a higher temperature (50 K for less bulk defects and larger terraces during growth) and after subtractions of effects from an averaged surface scattering (as described above) the effects of roughness (integer value for monolayers of coverage) and of quantum size effect (film thickness in multiples of 7 monolayers) are more clearly seen (Fig. 4).

Due to roughness scattering maxima of the conductance are expected for completion of each monolayer, as seen in Figure 3. Nevertheless the maximum of conductance for a thickness of 14 ML is missing. This effect has been observed for all multiples of 7 ML, which points to the quantum size effect [6–10]. At multiples of 7 integer multiples of the half Fermi wave length fit exactly into the film thickness. Therefore a new subband is shifted below the Fermi level and filled, so that a new scattering channel reduces the conductance at these integer values of film thickness [10]. Now the expected maximum of conductance due to small roughness of the complete top layer is reduced or even converted into a minimum (Figs. 3, 4 and 5). In Figure 5 the appearance of the same effect at 21 ML is shown.

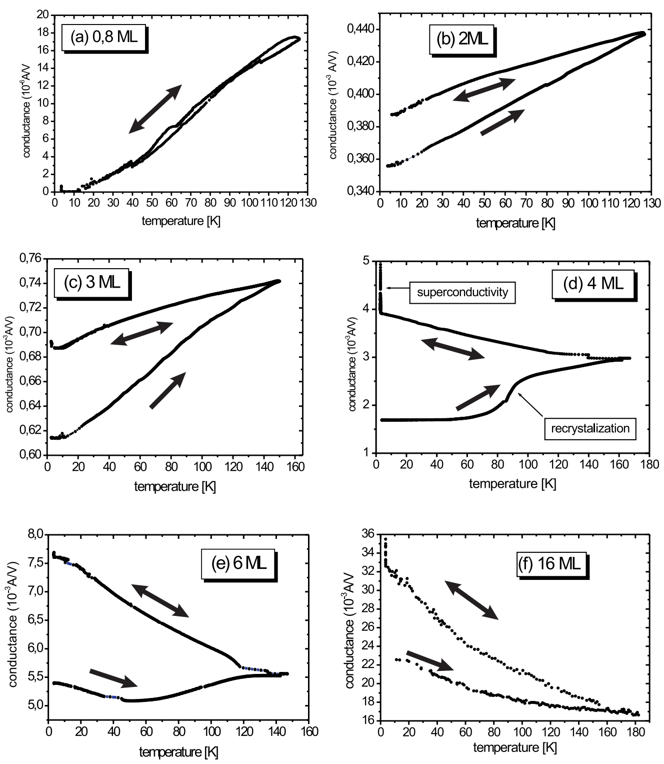

Fig. 6. Conductance of Pb films during the first annealing and for the following reversible changes with temperature. For higher temperatures irreversible decrease of conductance is observed due to the irreversible formation of multilayer islands (not shown in this figure).

For many films the conductance has been measured during annealing and the following cooling, which showed the irreversible (during the first heating) and then the reversible changes with temperature (during cooling and all further heating). The irreversible changes are due to annealing of defects in the bulk and at the surface during the first rise of temperature. Some results are shown in Figure 6.

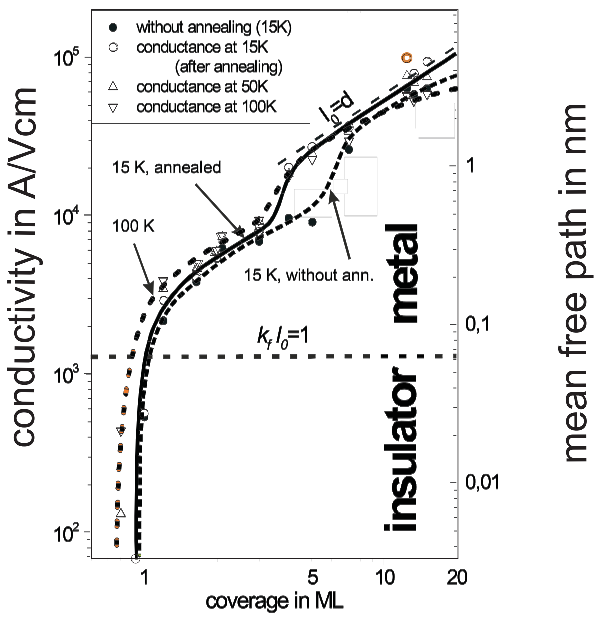

Fig. 7. Summary of conductivity of Pb films immediately after deposition at 15 K (filled circles), after annealing and cooling to 15 K (open circles), at 50 K (triangles with tip up) and at 100 K (triangles with tip down). The symbols are explained in the inset. The Ioffe-Regel criterion (with the Fermi vector $k_F = 1.6\times10^{10}$ m^{-1} and the mean free path l_0) $k_F l_0 = 1$ marks the limit between metal and insulator. The lines connect identical symbols, to see the effect of treatment and temperature (narrow dashed line: before annealing, solid line: after annealing and cooling to 15 K, widely spaced dashed line: at 100 K). The annealed films with $d > 5$ ML are well described in a model with the mean free path l_0 equal to the film thickness, which is reasonable for dominant interface scattering.

For disordered films up to 3 ML the conductance always increases with temperature, whereas the thicker films showed, with increasing temperature, an increasing resistance proportional to temperature after annealing like bulk Pb, but with a much larger residual resistance at low temperatures. The conductance of the thinnest film (0.8 ML) increases reversibly by a factor of 50 between 4 and 120 K. The superconductivity for annealed films with $d > 4$ ML could be directly seen at temperatures close to the bulk temperature (more results in [12]). All films showed the superconducting fluctuations, which allows a determination of the transition temperature down to 1 K with a fitting of the magnetoconductance [12]. A summary of conductance measurements for freshly deposited and for annealed films at 15 K and for films at 100 K is shown in Figure 7. For clarity reasons rather than conductance the average conductivity is shown, so that the scattering mechanisms are more obvious.

4 Discussion

Due to the insulating substrate and the well characterized structure and structural defects of the epitaxial films [17] the different scattering processes are well correlated with structural data.

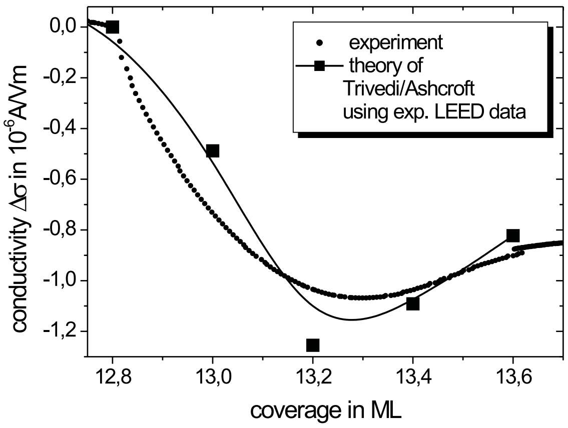

Fig. 8. Conductivity data out of Figure 2 together with calculated data using experimental roughness data from [16,17] and the theory of [14].

For annealed films with a thickness of more than 5 ML the conductivity increases linearly with thickness (Fig. 7). Therefore the interface scattering is the dominating effect, whereas bulk scattering is negligible. Surface scattering is also negligible, since an additional half monolayer increases the resistivity by the amount expected for deposition onto a step free surface. This is demonstrated in Figure 8.

Here the conductance change has been calculated using the theory of Trivedi-Ashcroft (using roughness and quantum size effect, not, however, a correlation length) [14] and the roughness data from LEED investigations [16,17]. The good correspondence shows that the annealed surface has to be rather specular, so that the additional roughness (as observed with LEED during epitaxy of half a monolayer) predicts correctly the observed decrease in conductance. Obviously the inclusion of the quantum size effect into the theory is important. If a theory, using just surface roughness and no quantum size effect (like the theory of Kaser and Gerlach, using roughness and correlation length [19]) is used, the agreement is not convincing. Therefore both roughness and quantum size effect have to be included. The quantum size effect is even more prominent in Figure 4; unfortunately corresponding calculations of the expected conductance variations have not been possible due to the lack of corresponding roughness data for this experiment.

For films with a thickness around 1 monolayer the films behave like an insulating film; the mean free path in a Drude model would yield distances less than atomic distances. The increase of conductance with temperature may be due to an activation process. It was not possible, however, to derive an activation energy from an appropriate fit. The Joffe-Regel-criterion (Fermi vector times mean free path $k_F \times l_0 = 1$) [20] indicates a possible limit between metal and insulator.

For films between 1 and 5 ML the linear increase in Figure 1 points to a constant bulk conductivity with bulk defects due to the disordered state, here the conductance of the first monolayer has to be neglected to obtain the linear increase beyond 1 monolayer. This may be attributed to interface scattering. The effects of band structure of the films on conductivity are seen in the comparison of Hall data with calculated band structure and calculated Hall effect, which will be reported in a forthcoming publication [21]. The observed superconductivity, which varies with the structure and thickness of the film is reported in reference [12].

5 Conclusion

Ultrathin epitaxial Pb films show quite a variety of order and defect structure depending on substrate, deposition and annealing parameters. The conductance measurements show that the conductance of thick and annealed films depends on interface scattering. Surface roughness scattering is quantitatively controlled by deposition conditions. Quantum size effect is especially important for multiples of 7 ML, which has also been seen in several other experiments. For very thin films (less than 5 ML) the bulk disorder is decisive and the interface scattering is the most important factor. Here a metal insulator transition is observed, although no simple model is available so far. Due to the rich structural variants, which may be well characterized and reproduced by proper procedures, the Pb films are good candidates for a fundamental description of properties of 2D conduction in ultrathin metallic films.

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References

1. M.K. Weldon, K.T. Queeney, J. Eng, K. Raghavachari, Y.J. Chabal, Surf. Sci. **500**, 859 (2002)
2. E.H. Sondheimer, Adv. Phys. **1**, 1 (1952)
3. M. Henzler, T. Lüer, A. Burdach, Phys. Rev. B **58**, 10046 (1998)
4. M. Henzler, T. Lüer, J. Heitmann, Phys. Rev. B **59**, 2383 (1999)
5. S. Heun, J. Bange, R. Schad, M. Henzler, J. Phys. Cond. Matt. **5**, 2913 (1993)
6. M. Jalochowski, E. Bauer, Phys. Rev. B **38**, 5272 (1988)
7. M. Jalochowski, H. Knoppe, G. Lilienkamp, E. Bauer, Phys. Rev. B **46**, 4693 (1992)
8. M. Jalochowski, E. Bauer, H. Knoppe, G. Lilienkamp, Phys. Rev. B **45**, 13607 (1992)
9. M. Jalochowski, M. Hoffman, E. Bauer, Phys. Rev. Lett. **76**, 4227 (1996)
10. V. Yeh, L. Berbil-Bautista, C.Z. Wang, K.M. Ho, M.C. Tringides, Phys. Rev. Lett. **85**, 5158 (2000)
11. O. Pfennigstorf, K. Lang, H.-L. Günter, M. Henzler, Appl. Surf. Sci. **162**, 537 (2000)
12. O. Pfennigstorf, A. Petkova, H.L. Günter, M. Henzler, Phys. Rev. B **65**, 45412 (2002)
13. L.J. van der Pauw, Philips Research Rep. **13**, 1 (1958)
14. N. Trivedi, N.W. Ashcroft, Phys. Rev. B **38**, 12298 (1988)
15. M. Horn-von Hoegen, Z. Kristallogr. **214**, 1 (1999)
16. A. Petkova, J. Wollschläger, H.L. Günter, M. Henzler, Surf. Sci. **482-485**, 922 (2001)
17. A. Petkova, J. Wollschläger, H.L. Günter, M. Henzler, Surf. Sci. **471**, 11 (2001)
18. J. Wollschläger, E.Z. Luo, M. Henzler, Phys. Rev. B **44**, 13031 (1991)
19. A. Kaser, E. Gerlach, F. Zeitschrift, Phys. B **97**, 139 (1995)
20. A.R. Regel, A.F. Joffe, Prog. Semicond. **4**, 237 (1960)
21. I. Vilfan, M. Henzler, O. Pfennigstorf, H. Pfnür, Phys. Rev. B (in press)