Magnetoconductivity of ultrathin epitaxial Ag films on Si(111) 7×7 at low temperatures

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Ultrathin epitaxial Ag films on Si(111) 7×7 have been shown to have at approximately 4 K a very low conductance, whereas at 100 K the metallic conductivity is evident. Therefore the magnetoconductance has been used to identify the different scattering mechanisms. The conductance and the magnetoconductance have been measured *in situ* with four-point probes in van der Pauw arrangement. For thicknesses from 1.8 to 20 ML different scattering mechanisms have been revealed in the temperature range from approximately 4 to 20 K and magnetic fields from −4 T to +4 T perpendicular to the sample surface. Whereas for films thicker than 3 ML the weak localization and antilocalization provide a complete description, the thinnest films show properties not yet described quantitatively by any theory.
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I. INTRODUCTION

The scattering of conduction electrons in metals is usually well described by phonons at high temperatures and by defects at low temperatures. For thin films the surface and the interface may act as effective scatterers. For thin films at low temperatures weak localization has been observed as quantum interference effect of the backscattered electrons.[1–3] Very thin films at low temperatures, however, cannot be described along these lines, since their conductance is too low for weak localization and shows some kind of metal-insulator transition for a temperature range from 100 to 15 K.[4] Therefore some change in scattering mechanism has to occur.

For a fundamental study of conduction mechanisms in thin films it is necessary to select a system as simple as possible. It should be a clean and elemental film with a constant thickness and well-ordered on a well ordered substrate with a characterization of the defects in the film. Therefore a clean single crystalline Ag film is here produced by epitaxy on a perfect Si(111) 7×7 in ultrahigh vacuum. This film is so far the best approximation to an ideal two-dimensional (2D) film.[5,6] After deposition of at least one monolayer at temperatures between 50 and 150 K the diffraction spots due to a Ag(111) film are visible with a half-width decreasing to about 4% during annealing up to room temperature. With a film of about 2 ML no superstructure spots of the substrate are visible up to annealing temperatures of about 200 K, pointing to a hole free film.[7,8] During warming up to room temperature the films up to a thickness of about 3 ML form holes, which is seen both in an irreversible decrease of conductance and in the appearance of superstructure spots of the Si(111) 7×7 substrate.[7,8,4]

At low temperatures Si is insulating, so that the conductance of the Ag film is easily measured with a four probe measurement in the van der Pauw arrangement.[4]

Magnetoconductance is a well-suited tool to study scattering mechanisms, since its sign, magnitude, and field dependence indicate different mechanisms. Low temperatures are required to avoid the smoothing effect of phonon scattering.

Earlier measurements have shown that even a temperature of 20 K is too high to isolate the different effects and to explain the transport phenomena in these metal films.[5,6] Therefore measurements at approximately 4 K have been performed to answer some of the remaining questions.

The measurements of the temperature-dependent resistance at low temperatures already show that there are several regions of different scattering mechanisms, depending on the thickness of the films.[4] Here the magnetoresistance of these films is reported to identify the mechanisms and to explain the experimental results.

II. EXPERIMENTAL SETUP

All measurements were performed in an UHV chamber with a base pressure of 1×10^{-10} mbar. Silicon wafers with a resistance of 5 kΩ cm at room temperature were used as substrate. Samples with a special 15×15 mm^2 van der Pauw shape have been laser cut from a 1-mm-thick wafer. The substrate was chemically cleaned and as a last step Mo contacts were evaporated onto the corners of each sample. After this procedure the substrate was mounted on the sample holder and transfered into the chamber with a 24-h bakeout afterwards. Finally the oxide was flashed off the substrate. All of the following steps were carried out *in situ* in ultrahigh vacuum.

Each silver film was evaporated onto a clean Si(111) 7×7 surface at a substrate temperature of about 15 K. The thickness of the film was controlled using a quartz crystal monitor. The calibration was done with the temperature dependance of the resistance of thick and well-annealed films.[4] When the desired thickness was reached the shutter and the radiation shields were closed and the sample was annealed up to the highest conductivity so that contribution of defects could be minimized. The effect of annealing is reported in a former paper.[4] All results presented in this paper were performed after this first annealing step, so that there is no contribution of any annealing effect left in the data.

Details concerning the experimental setup and preparation of the samples can be found elsewhere.[4,9] The indicated temperature has been measured at the base of the cryostat. The temperature at the sample has been measured directly only

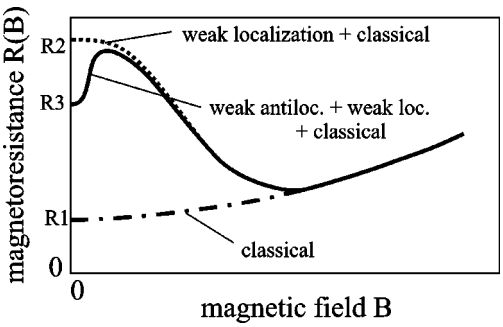

FIG. 1. Schematic representation of resistance change with magnetic field at low temperatures: the classical residual resistance R_1 increases with magnetic field (dash-dotted curve), the resistance R_2, which is increased due to weak localization, decreases down to the classical curve (dotted curve), the again lower resistance R_3 due to antilocalization increases at low magnetic fields to the curve of weak localization (solid curve).

for dummy samples, since a low-temperature thermocouple would not stand the flashing. Therefore the sample temperature may differ from the indicated temperature by a few degrees.

All measurements have been performed with increasing and decreasing, positive and negative magnetic fields. Therefore any different values at zero field are due to drift during cycling the field.

III. THEORETICAL BACKGROUND

Conductivity in metals is known to be described by the classical Drude formula. A decrease of the inelastic scattering by reducing of the temperature and/or an increase of elastic scattering in thin metal films may lead to interference effects with an increase of the resistance of the films. These effects are called weak localization (localization of conduction electrons by constructive interference of backscattered electrons), and weak antilocalization (antilocalization of conduction electrons caused by destructive interference due to spin-orbit scattering).[1–3] Both of them can be removed by applying high magnetic fields. The magnetoresistance of metals has therefore three contributions, which are qualitatively shown in Fig. 1. In the classical description the low-temperature residual resistance R_1 is increased by a magnetic field B by

$$R = R_1(1+\mu^2 B^2) \tag{1}$$

with μ the mobility of the carriers. For sufficiently high temperatures and not too thin films (in our experiments for films with at least 10 ML thickness) this equation describes the magnetoresistance completely (dash-dotted curve in Fig. 1). For thinner films the residual resistance is increased up to R_2 in Fig. 1 due to an increased backscattering (weak localization).[1–3] Since this interference effect is removed by a magnetic field due to a phase change of the electron wave functions, a negative magnetoresistance is observed (dotted curve in Fig. 1). Spin-orbit scattering reduces again the weak localization to a lower resistance R_3, which is only observed here for temperatures lower than about 20 K. Already a small magnetic field removes this effect. Therefore the solid curve of Fig. 1 shows first an increase and then a decrease of the resistance for a combination of weak antilocalization and localization.

Hikami, Larkin, and Nagaoka found a quantitative description of the conductance change due to the magnetic field B, when both weak antilocalization and localization are important:[10]

$$\Delta L = L(B) - L(0) = -L_{00}\left\{\Psi\left(\frac{1}{2}+\frac{B_1}{B}\right)+\ln\left(\frac{B}{B_1}\right) - \frac{3}{2}\left[\Psi\left(\frac{1}{2}+\frac{B_2}{B}\right)+\ln\left(\frac{B}{B_2}\right)\right] + \frac{1}{2}\left[\Psi\left(\frac{1}{2}+\frac{B_3}{B}\right)+\ln\left(\frac{B}{B_3}\right)\right]\right. . \tag{2}$$

In this equation $\Psi(x)$ is the digamma function. The characteristic magnetic fields B_1, B_2, and B_3 are defined as

$$B_1 = B_0 + B_{so} + B_s , \tag{3}$$

$$B_2 = B_i + \tfrac{4}{3}B_{so} + \tfrac{2}{3}B_s , \tag{4}$$

$$B_3 = B_i + 2B_s \tag{5}$$

with

$$B_n = \frac{\hbar}{4eD\tau_n}, \quad n = 0,i,so,s . \tag{6}$$

The indices 0, *i*, *so*, and *s* stand for elastic, inelastic, spin-orbit, and spin-flip scattering, respectively. For all fits presented in this paper it was assumed that there are no magnetic particles in the films ($\tau_S = \infty$) and that the elastic scattering time τ_0 can be estimated by using the Drude formula. The inelastic scattering time τ_i and the spin-orbit scattering time τ_{so} are free parameters in the fit.

IV. RESULTS

A. Thickness dependence of the magnetoresistance at low temperatures

Measurements of the temperature-dependent resistance of thin Ag films have shown that there is a classical metallic behavior present for films with a thickness of more than about 10 ML. Films of a lower thickness show at low temperatures a resistance increase, that can be explained by weak localization.[4] Figure 2 shows the magnetoresistance of a 6-ML thick Ag film at a temperature of 4 K. The experimental results are represented by the crosses, the solid line shows the fit by using the theory of Hikami[10] for these results, which provides a perfect fit with the fitting parameters τ_i and τ_{so}. When a magnetic field is applied the effect of weak localization is destroyed, the resistance is decreasing with increasing strength of the magnetic field. For this thickness and this temperature there is no contribution of weak antilocalization clearly visible.

There is a similar behavior observed for film thicknesses between 2 and 5 ML. The results and the fits [with Eq. (2)] are shown in Fig. 3. Again, the magnetoresistance is symmetric for positive and negative magnetic fields so that there only the results for positive magnetic fields are shown. Here

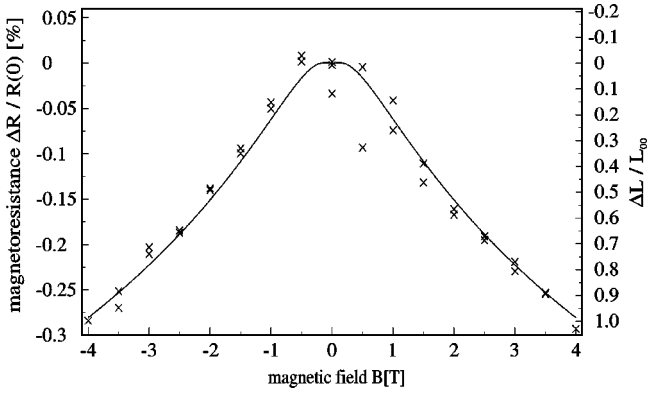

FIG. 2. Magnetoresistance of a 6-ML-thick Ag film for positive and negative magnetic field. The conductance unit on the left side $L_{00}=e^2/2\pi^2\hbar$ is the quantum unit of conductance as used for the description of weak localization (Ref. 3). The starting resistance is 240 Ω/square.

the experimental results are well described with contributions of weak localization and weak antilocalization. The contribution of weak antilocalization is clearly indicated by the increasing resistance with increase of a low magnetic field.

For a thickness of less than 2 ML the results are already qualitatively different. Figure 4 shows the experimental results for thicknesses of 1.8 and 1.9 ML, each with the best possible fit with Hikami's theory, although the applicability of the theory is questionable due to the high film resistance (its value is beyond the limit given by the Ioffe-Regel criterion[4]). Already the elastic scattering time τ_o should no more be derived with the Drude formula (which would yield a value close to 10^{-16} sec). There are two remarkable differences compared with the results of films of higher thickness: the resistance does not increase for low magnetic fields, pointing to a change in the transport mechanism. Additionally it is no longer possible to obtain a satisfactory fit of the data with the Hikami theory. The extremely high value for the time τ_{so} (see Fig. 6). and the extremely low value for τ_o (from the no more applicable Drude formula) require a model with a different scattering mechanism.

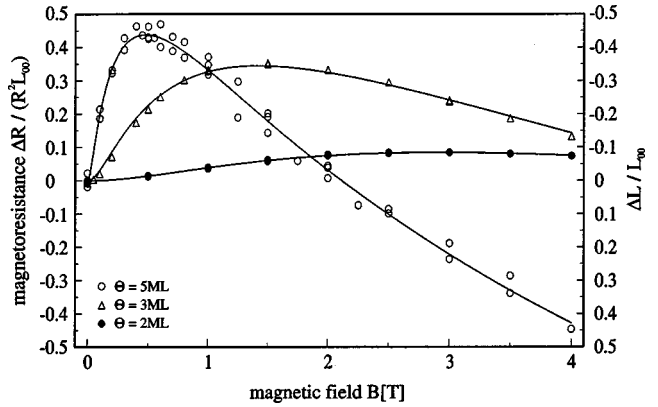

FIG. 3. Magnetoresistance of Ag films with different thicknesses in the weak localization regime at $T=4$ K. Due to the symmetry the resistances at negative fields have been plotted together with the resistances at positive fields. The starting resistances for 5, 3, and 2 ML are 325, 1500, and 12 000 Ω/square, respectively.

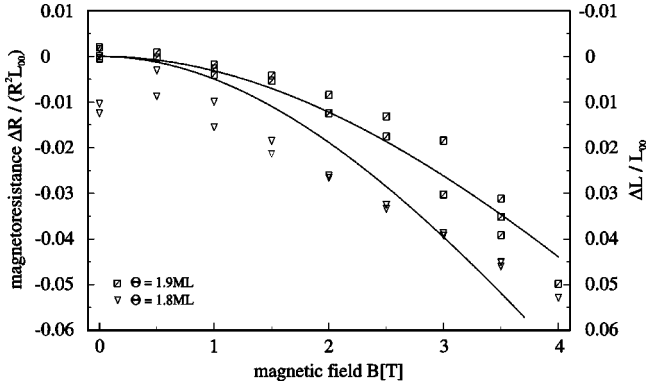

FIG. 4. Magnetoresistance of two Ag films with thicknesses of 1.8 and 1.9 ML at approximately $T=4$ K. The starting resistances for 1.8 and 1.9 ML are 85 and 19 kΩ/square, respectively.

B. Temperature dependence of the magnetoresistance of a Ag film with a thickness of 3 ML

The investigation of the magnetoresistance of thin epitaxial silver films at low temperatures (approximately 4 K) leads to some results that differ from the results of the former investigations at higher temperatures (approximately 20 K). Therefore the temperature dependence of the magnetoresistance of a film with a thickness of 3 ML has been measured (Fig. 5). The contribution of antilocalization decreases with increasing temperature until finally only contributions of weak localization can be seen from the magnetoresistance, which agrees completely with earlier measurements.[6] Therefore the differences are only due to the temperature and not due to any difference in the film.

V. DISCUSSION

A. Films with a thickness from 2.5 to 10 ML at 4 K

The conductivity in these films is well described as metallic with an elastic mean free path given by the thickness of the film (up to twice the value).[4] The temperature dependance shows that inelastic scattering is smaller by more than one order of magnitude. Therefore the model of weak localization as described quantitatively by Hikami, Larkin, Nagaoka is applicable. Since magnetic scattering is negligible for Ag and Si, only the elastic, inelastic and spin-orbit scat-

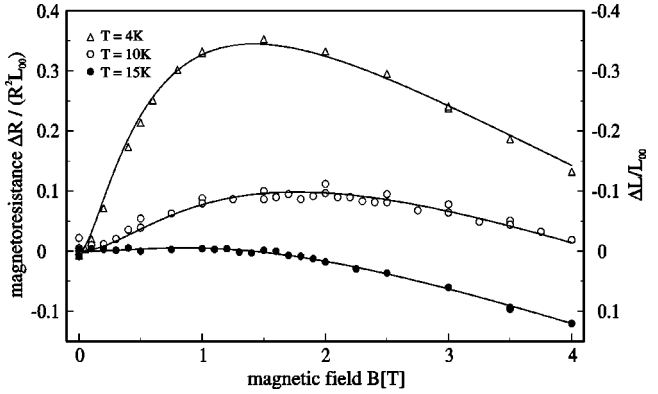

FIG. 5. Magnetoresistance of Ag films with a thickness of 3 ML measured at different temperatures. The starting resistances for $T=4$, 10, and 15 K are 1.5, 2.3, and 2.5 kΩ/square, respectively.

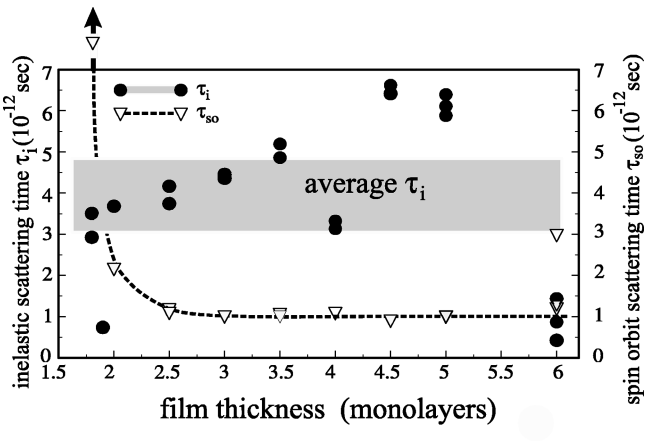

FIG. 6. Inelastic scattering time τ_i and spin-orbit scattering time τ_{so} vs Ag film thickness at approximately 4 K.

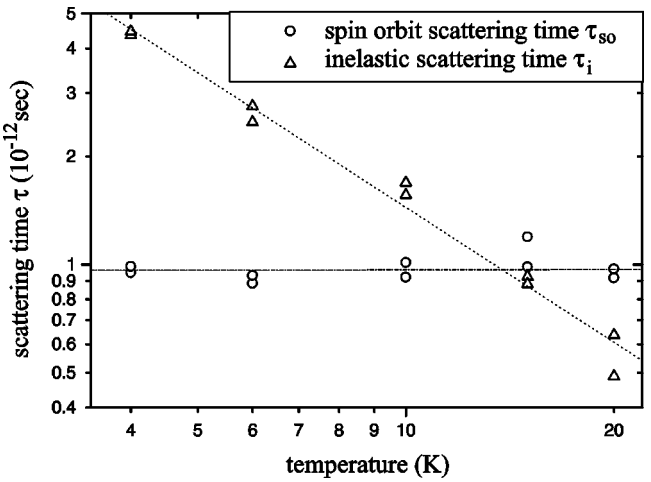

FIG. 7. Temperature dependence of the inelastic scattering time τ_i and of the spin-orbit scattering time τ_{so} for an Ag film with $d=3$ ML.

tering times (τ_o, τ_i, and τ_{so}, respsectively) are relevant. The elastic time τ_o is taken from the conductivity, since its dominates the conductance ($\tau_o < 2\times10^{-15}$ sec). A separation of the other two relevant scattering times is now possible, since both localization and antilocalization are clearly distinguished from increase and the following decrease of the resistance with increasing magnetic-field strength. The good fit with these two parameters shows the applicability of the theory and the independant determination of these scattering times (Figs. 2–5). A summary is shown in Fig. 6. For thicknesses of at least 2.5 ML the spin-orbit time τ_{so} is independent of the thickness, pointing to a uniform conduction mechanism as described within the Hikami theory. The spin-orbit scattering is obviously a bulk effect due to the independence of thickness, it is not related to the interface. The inelastic time is approximately independent of thickness (here the actual sample temperature—dependent on the thermal coupling between sample and cryostat—and the film preparation may produce some scatter of the data), whereas the elastic time τ_0 varies in this thickness range by nearly an order of magnitude. A direct correlation with the conductance is not evident. Since the ratio τ_i/τ_{so} is larger than or close to unity, antilocalization is easily separated. Schad *et al.*[6] and Bergmann and Horriar-Esser[11] reported a dependance of the spin-orbit-scattering time on elastic scattering time (or film thickness). Schad performed the experiments at higher temperatures of approximately 20 K for different thicknesses. Since at these temperatures only a very small contribution of spin-orbit scattering is left in the magnetoresistance, the present experiments have a much smaller error bar. In the experiments of Bergmann and Horriar-Esser highly disordered and rather thick (8.3 nm) films had been deposited onto glass and the resistivity was altered by partial annealing. Since we used only epitaxial and well-annealed films, the films are not directly comparable. It would be interesting to study to what extent the kind of disorder (besides resistivity) has a direct impact on spin-orbit scattering.

In the same way it is difficult to compare the absolute values for τ_{so} that were published by other authors[15,11] and the values we determined from our experimental results, $\tau_{so}\sim 9.5\times10^{-13}$ sec, since so far only our experiments show data from epitaxial and well-ordered films.

The inelastic scattering has contributions both from electron-phonon and electron-electron scattering.[16,17] They are not separated by a measurement of the temperature dependance of conductivity; rather, the magnetoconductance is necessary. For strong magnetic fields the weak localization due to electron-phonon scattering is removed, that due to electron-electron scattering, however, is not removed. Therefore the temperature dependance of the conductance of a 6-ML film has been measured both at zero and at maximum (4 T) magnetic field. Unfortunately at that field strength the magnetoconductance was still dependent on the field strenght. Therefore a contribution due to electron-electron scattering could not be separated.

B. Temperature dependence of τ_i

The results of Fig. 5 have been evaluated with respect to the relevant scattering times (Fig. 7). The spin-orbit scattering time τ_{so} is independent of temperature, as expected. Li *et al.*[15] published similar results for observations on non epitaxial silver films on glass substrate.

The inelastic scattering time decreases with increasing temperature with a power of -1.25, when the temperature scale is used as measured at the base of the cryostat. When the temperature of the sample is assumed to be a few degrees higher, the power increases up to about -2.0. Theory predicts exponents between -1 and -2, depending on the mechanism.[18] In the temperature range of measurement obviously several mechanisms contribute. A separation would be possible with a higher accuracy of the measurement of the temperature and an expansion of the temperature range to lower temperatures. Those experiments are in progress.

Other authors published values between -1.0 and -2.6 for the exponent.[12–15] Again different procedures for the production of the film as crystallinity, roughness, grain boundaries, voids and any other defects may be important besides resistivity for details of the scattering mechanisms. So far only our results refer to epitaxial and well-annealed films.

C. Films with a thickness of less than 2 ML

The conductivity already pointed to a different conduction mechanism: a ''mean free path'' much less than atomic dis-

tances and a negative temperature coefficient for the resistivity up to 100 K and the high absolute values for the resistance indicated, that weak localization is no more appropriate for description.[4] Some kind of strong localization has to be assumed. The magnetoconductance shows qualitative and quantitative differences. There is no increase of resistance for low magnetic fields. When the Hikami theory is nevertheless used, a very strong increase of the spin-orbit scattering time would be the result (Fig. 6). Therefore also the magnetoconductance gives evidence, that the conduction mechanism is well outside the regime of weak localization. Unfortunately there is no theory of magnetoconductance in the regime of strong localization available. The new data represent clearly a regime beyond weak localization, since the resistance is too high and the magnetoconductance cannot be described within weak localization.

Only speculations are so far possible concerning the physical mechanism responsible for the changes in the scattering mechanism. The ideal two-dimensional system even with a small defect concentration should turn from a metal into an insulator with decreasing temperature. An epitaxial monolayer on an insulating substrate may be so far the closest realization of an ideal two-dimensional system. The defects are the domain boundaries of the rotational mosaic structure and maybe also the corrugation due to the underlying Si(111) 7×7 structure. Even at 100 K in the metallic state the mean free path is given by the thickness, which is approximately 0.23 nm. Therefore there is indeed a strong scattering. With lowering of the temperature the conduction may change to strong localization, so that the Hikami theory of weak localization is no more applicable. The result, that already a few monolayers return to weak localization, may be interpreted with the prediction, that only true two-dimensional systems may turn into an insulator, since the second and third monolayer provide detours into the third dimension. Those speculations may be checked at least in part with measurements at lower temperatures and with a more detailed study of the film defects at low temperatures.

New insight into the conduction mechanism for strong localization would also reqire a theory that makes use of the well-documented structural properties. The role of the small-angle grain boundaries would be an interesting point, since they are well studied and the most relevant defect in the film.[8] Another support may be possible with experiments in the submonolayer regime and a theory, which describes percolation with a quantum-mechanical approach.

VI. CONCLUSION

Thin epitaxial films are closest to an ideal metallic 2D system with the electron density close to 10^{15} cm^{-2}. Here the atomic structure and defects like roughness of surface and interface are much more important than with semiconductor structures, where the space charge layer smoothens out a lot of inhomogeneities. Therefore the present experiments with *in situ* studies of magnetoconductance of epitaxial metallic monolayers may open the field for studies of well defined films with a low density of defects and a high density of electrons in a film, which is produced, studied, and controlled with respect to defects in the same ultrahigh vacuum.

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