Fabrication of noble-metal nanoconstrictions and observation of conductance fluctuations

E. Scheer and H. v. Löhneysen

Physikalisches Institut, Universität Karlsruhe, Engesserstrasse 7, 76128 Karlsruhe, Germany

H. Hein

Kernforschungszentrum Karlsruhe, Institut für Mikrostrukturtechnik, Postfach 3640, 76021 Karlsruhe, Germany

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We describe the fabrication of planar metallic nanostructures with lateral dimensions down to about 80 nm and thicknesses varying from 15 to 50 nm. We use a lift-off process with a polymethylmethacrylate bilayer resist on silicon substrate and an electron beam writer system for microfabrication. The noble-metal layers are deposited by thermal evaporation. The geometries are point contacts, rings, and wires in two-lead configuration. We discuss measurements of the magnetoconductance at very low temperatures and in magnetic fields up to 4 T in dependence of the width of the samples. Here the electron transport is in the diffusive regime and the width dependence of the universal conductance fluctuations is investigated.

I. INTRODUCTION

Metallic systems with reduced geometry show new electronic phenomena evoked by quantum interference, e.g., fluctuations of the magnetoconductance of the order of a universal value e^2/h or oscillations of the magnetoconductance of ring structures.¹ These properties can be observed if the electron waves are able to interfere. This means that the dimensions of the sample must be of the order of magnitude of the phase coherence length l_ϕ or smaller.

For metals, this condition is fulfilled in small structures of lateral dimensions $L < 1 \mu\text{m}$ at very low temperatures $T < 1$ K. Because usually the elastic mean free path l_e (10 nm $< l_e < 50$ nm) is still smaller than the sample size L , the electronic transport is in the diffusive regime where the interference of diffusely scattered electron waves leads to universal conductance fluctuations (UCF), i.e., reproducible but stochastic variations of the electrical conductance as a function of the magnetic field or the electrochemical potential.²

The theoretically expected root-mean-square (rms) amplitude ΔG of the UCFs is about e^2/h and depends slightly on the dimensionality.² For one dimension $\Delta G_{1D} = 0.729 e^2/h$, for two dimensions $\Delta G_{2D} = 0.862 e^2/h$, and for three dimensions $\Delta G_{3D} = 1.088 e^2/h$ are calculated. These amplitudes are reduced in magnetic fields by a factor $2^{1/2}$ due to the suppression of the Cooperon channel. With the same assumptions as in weak (anti-) localization theory one obtains a further factor-of-2 reduction in amplitude due to the spin-orbit scattering in metals with large atomic number. Thus $\Delta G_{1D} = 0.26 e^2/h$ would be expected for gold wires, and $\Delta G_{2D} = 0.30 e^2/h$ for small two-dimensional gold films.

Experiments on long (one-dimensional) gold and silver samples^{3,4} show a rms amplitude of $0.12 e^2/h$ for two-lead measurements with large contact areas. This is a factor of 2 smaller than the expectation. The reduction is explained by the authors in terms of a reduced backscattering probability of electrons diffusing into the contact leads. For this contact geometry the fluctuations should be symmetric in the field.⁵ The characteristic fluctuation "period" B_c , the so-called cor-

relation field, is a measure for the area A surrounded by typical interfering electronic paths since $AB_c = \Phi_0$ with $\Phi_0 = h/e$,² the elementary flux quantum. Because all paths shorter than l_ϕ contribute to the interference pattern,⁶ B_c should depend on the geometry of the samples if the width and/or the length of the samples become smaller than l_ϕ . In addition, the energy range eV_m (V_m is the measuring voltage) around the Fermi energy where the electrons contributing to the transport come from, generates a new energy scale. This energy characterizes the Thouless length $L_T = (hD/eV_m)^{1/2}$. If L_T becomes shorter than l_ϕ , L_T limits the observability for interference of the electrons and determines the magnitude of the fluctuations.

The dependence of the characteristic features of the UCFs, B_c and rms amplitude, on the geometry are not yet understood completely. In particular, the influence of the contact geometry on the amplitude and the dependence of B_c on the width of the sample have not been investigated systematically up to now.

We will report on measurements of the magnetoconductance, so-called "magnetofingerprints," of gold samples in two-lead configuration with short and wide leads and typical sample dimensions between 80 nm and $5 \mu\text{m}$ where the geometry of both samples and leads determines B_c .

II. SAMPLE FABRICATION

The present route of preparation is aimed at producing the nanostructures and the macroscopic contact pads simultaneously within one single step for exposure, development, metallization and lift-off in order to avoid overlay and interface problems.

Undoped 4-in.-diam silicon wafers are used as substrates. The wafers are exposed to 100% hexamethyldisilazane (HMDS) vapor at 130 °C in a vacuum oven to improve the adhesion to the resist system. The resist system consists of a double layer of polymethylmethacrylate (PMMA), where as first layer PMMA with a molecular mass of 600 kg/mol and a solids content of 3% dissolved in chlorobenzene is spin

coated to a thickness of 200 nm. In order to remove residual solvent a prebake for 30 min at 170 °C is performed. A higher molecular mass of 950 kg/mol with a lower solids content of 2% is used for the toplayer. With a dynamic dispense of the toplayer PMMA no thinning out of the first layer is observed. After a second prebake step with the same parameters a total resist thickness of about 300 nm is measured.

For the exposure a commercial electron beam lithography machine EBP55-HS (Leica-Cambridge) is used. It is a high-speed vector scan machine with a maximum acceleration voltage of 100 kV, which results in a low percentage of back-scattered electrons and supports the exposure of patterns with lateral dimensions $L < 1 \mu\text{m}$. The e-beam operates with a LaB_6 flat tip cathode with an original $20 \mu\text{m}$ diameter truncation. This improves the cathode lifetime but limits the brightness; consequently very small spot diameter (30–50 nm) and beam current $< 1\text{--}4 \text{ nA}$ are difficult to establish. Both the coarse contact leads and pads of about 1 mm^2 size and the nanostructures with dimensions down to 80 nm are exposed in the same run but with different e-beam parameters. For the coarse structures, a beam step size of 250 nm with a current of about 200 nA and a spot size of 300 nm diam is used. The exposure dose for these structures is $700 \mu\text{C}/\text{cm}^2$. For the fine structures the parameters are: beam step size 20 nm; current 2 nA; nominal spot size 40 nm; and exposure dose $1300 \mu\text{C}/\text{cm}^2$. After the exposure the PMMA is developed in a bath of 75% isopropanol (IPA) and 25% isobutylmethyl ketone (MIBK) at 30 °C for about 5 s and rinsed for around 1 min in pure IPA. The bilayer resist system together with the slightly enhanced developing temperature cause an undercut profile of the resist edges suitable for the lift-off following later in the manufacturing process.

Because the adhesion of gold and silver on the silicon wafer covered by its native oxide layer is rather poor, a primary treatment of the developed substrate is necessary before depositing the metal film. Therefore the substrate is etched in a solution of 80% de-ionized (di) water with 25% HF at room temperature for 15 s, rinsed in di-water for 30 s, and then coated with *n*-decane alcohol in order to avoid reoxidization of the cleaned surface. Then the substrate is immediately mounted in the lock chamber of an ultrahigh vacuum (UHV) system with exposure to air for less than 5 min. The alcohol evaporates during pumping down of the system in about 15 min without residue. At a pressure of 10^{-8} hPa a 1.3-nm thin germanium seeded layer is e-gun evaporated. Then the substrate is cooled down to about -20 °C and the noble-metal layer with a thickness of 15–50 nm is thermally evaporated at rates of 0.01–0.02 nm/s. The evaporation at reduced substrate temperatures leads to a slightly worse adhesion of the metal film to the substrate but together with the low evaporation rate a much better quality of the film is achieved, i.e., smaller grain size, mechanical stability, and smoothness, which is favorable for the lift-off process. Finally, the lift-off is performed in acetone at room temperature for 2 h. Short ultrasonic treatment for a few seconds improves the sample quality.

FIG. 1. SEM picture of (a) sample D1-2, (b) sample D4-2. Magnification as indicated by the bars.

III. MEASUREMENTS

After the preparation, the structures are inspected in a scanning electron microscope (SEM) with a magnification of 50 000, the metal layer thickness is checked by mechanical step height measurement, and the wafer is cut into chips of $12 \text{ mm} \times 12 \text{ mm}$ size each carrying an array of 4×4 samples with the same contact geometry but different samples on a mesoscopic scale (Fig. 1). Electrical contacts to the sample are accomplished by spring contact pins which are pressed onto the pads. The mesoscopic geometries of the samples are as follows: point contacts with width w from 80 nm up to 560 nm and wires with width from 80 to 340 nm and length L from 100 to 400 nm are investigated. In addition a ring structure with line width $w = 150 \text{ nm}$, inner diameter 750 nm, outer diameter 1050 nm and a long wire with length $L = 91 \mu\text{m}$, width $w = 220 \text{ nm}$ are designed. The typical classical resistance of the samples at low temperatures is between 15 and 100Ω for the short wires, point contacts, and rings, and around 1300Ω for the long wire. From the residual resistivity ratio $\text{RRR} = R(T = 300 \text{ K})/R(T \rightarrow 0) \approx 1 + l_{\text{ph}}/l_e$ and the phonon scattering length of gold at room temperature⁷ we deduce elastic mean free paths from 23 to 27 nm. With this value we calculate the electronic diffusion constant $D = v_F l_e / 3 \approx 0.011 \text{ m}^2/\text{s}$.

The measurements of the magnetoconductance are per-

FIG. 2. Magnetoresistance (magnetofingerprint) of sample D3 for $-2\text{ T} \leq B \leq 2\text{ T}$.

formed with the sample mounted in the mixing chamber of a $^3\text{He}/^4\text{He}$ dilution refrigerator. In order to avoid eddy-current heating when sweeping the field the mixing chamber is made of araldite. The magnetoresistance is measured with an ac-resistance bridge with excitation currents of about $3\text{ }\mu\text{A}$. Typical voltage changes corresponding to the UCF signals are some nanovolts. Lowest measuring temperatures are approximately 30 mK .

IV. RESULTS AND DISCUSSION

Figure 2 shows the magnetoresistance of the gold sample D3 with $L=400\text{ nm}$, $w=340\text{ nm}$ and $d=18\text{ nm}$ for magnetic fields $-2\text{ T} \leq B \leq 2\text{ T}$ (Dimensions, measurement parameters, and results for all investigated samples are listed in Table I). The absolute resistance at $T=30\text{ mK}$ is $21.44\text{ }\Omega$. Because the samples are prepared in the one-step process described above, the leads and the samples themselves have the same thickness and therefore the same sheet resistance $R_{\square}=3.1\text{ }\Omega$. This results in an offset resistance R_o in series with the sample resistance R_s . From the geometry of sample and leads and the measured resistances we can estimate the size of R_o as $16.1\text{ }\Omega$. It is not clear to what amount R_o should contribute to the UCFs. Therefore, in the following discussion of the rms amplitudes we assume that R_o does not fluctuate at all. We scale the measured resistance fluctuations ΔR by the sample resistance $R_s=R-R_o$ to obtain the conductance fluctuations $\Delta G=-\Delta R/R_s^2$.

TABLE I. Dimensions, measuring parameters, and results for gold samples C4, D1-2, D2-2, D3, D4, and D4-2. The meanings of the symbols are: L : sample length, w : width, d : thickness, R : measured resistance, R_o : offset resistance as discussed in the text, R_s : sample resistance $R-R_o$, T : measuring temperature, I_m : measuring current, V_m : measuring voltage at R_s , ΔG : rms amplitude of the UCF, B_c : correlation field of the UCF, K : relative correlation between magnetofingerprint for positive and negative applied magnetic fields.

Sample	L (nm)	w (nm)	d (nm)	R (Ω)	R_o (Ω)	R_s (Ω)	T (mK)	I_m (μA)	V_m (μV)	ΔG (e^2/h)	B_c (mT)	K (%)
C4	91000	220	18	1307	8	1299	30	0.1	130	0.0002	...	86
D1-2	400	160	18	24.5	14.4	10.1	140	3	30	0.19	27	78
D2-2	400	240	18	22.4	14.4	8.0	25	3	24	0.24	18	75
D3	400	340	18	21.4	16.1	5.3	30	4	21	0.27	12	92
D4	4700	150 (ring)	18	91.5	8.0	83.5	30	3	250	0.01	25	92
D4-2	4700	150 (ring)	18	83.0	7.5	75.5	30	3	227	0.01	27	91

FIG. 3. Symmetric part $[\Delta G(B) + \Delta G(-B)]/2$ (right-hand trace) and anti-symmetric part $[\Delta G(B) - \Delta G(-B)]/2$ (left-hand trace) of the conductance fluctuations of Fig. 2. The resistance fluctuations have been scaled to obtain conductance fluctuations by $\Delta G = -\Delta R/R_s^2$ with $R_s=5.3\text{ }\Omega$.

Around $B=0$ one observes a sharp minimum due to weak antilocalization. A fit to the 2D theory yields $l_e=19\text{ nm}$ in rather good agreement with the value deduced from the RRR. In addition to this anomaly which is subtracted in the following analysis one observes fluctuations of the conductance which are symmetric in the field, as theory predicts. We use the symmetry in order to estimate the experimental noise. The left trace in the Fig. 3 shows the antisymmetric part $[\Delta G(B) - \Delta G(-B)]/2$ which should represent the experimental noise and the right trace includes the symmetric part $[\Delta G(B) + \Delta G(-B)]/2$, i.e., the UCF signal and the symmetric part of the noise. Figure 4 shows the autocorrelation functions of the antisymmetric part, $C_{AA}(\Delta B)$ together with the autocorrelation functions of the magnetofingerprints for negative field and positive field, $C_{nn}(\Delta B)$ and $C_{pp}(\Delta B)$, and the cross-correlation function of the traces with negative and positive field $C_{np}(\Delta B)$. The symmetric and antisymmetric parts of the noise should have the same size, which is indeed roughly the case as checked by a detailed analysis. We obtain the rms amplitude of the experimental noise by $[C_{AA}(0)]^{1/2} \approx 1\text{ nV}$. The value $C_{np}(\Delta B=0)$ gives the rms amplitude of the fluctuations $\Delta G=[C_{np}(0)]^{1/2}=0.27\text{ }e^2/h$, which is a little smaller than the theoretical value $\Delta G_{2D}=0.3\text{ }e^2/h$ and larger by a factor 2.3 than the observations^{3,4} for samples with wide leads. The half-width of the maximum of

FIG. 4. Correlation functions C_{np} , C_{nn} , C_{pp} , and C_{AA} , of the conductance fluctuations of Fig. 3. The half-width of $C_{np}(\Delta B)$ indicates the correlation field $B_c = 12$ mT. Symbols indicate calculated correlation functions (in intervals of 5 mT). Lines are guide to the eye.

$C_{np}(\Delta B)$ gives the correlation field B_c . In this sample we obtain $B_c = 12$ mT. The last column of Table I contains the correlation $K = C_{np}(0)/[C_{nn}(0)C_{pp}(0)]^{1/2}$, which is a measure for the reproducibility of the fingerprints and is in our examples always better than 75%. The reproducibility is also seen in the overall agreement $C_{nn}(\Delta B)$, $C_{pp}(\Delta B)$, and $C_{np}(\Delta B)$ in Fig. 4.

For samples much shorter and narrower than the minimum of L_T and l_φ , B_c should be nearly independent of the length and the width because the interference patterns are characterized by electrons diffusing into the contact pads. For samples with $w < l_\varphi$ and $L \geq \min(l_\varphi, L_T)$, B_c should be inversely proportional to w : $B_c = h/[e w \min(l_\varphi, L_T)]$. This is observed for samples D1-2, D2-2, and D3 with $w = 160$, 240, and 340 nm, $B_c = 27$, 18, and 12 mT, respectively, although $L = 400$ nm is smaller than $\min(l_\varphi, L_T)$. With these values we obtain $\min(l_\varphi, L_T) \approx 1 \mu\text{m}$. For sample D3 the measuring voltage was 21 μV . Thus we deduce $L_T \approx 1.4 \mu\text{m}$ in consistence with the value obtained from the fluctuations.

For the ring sample we also observe aperiodic conductance fluctuations (Fig. 5) with amplitude $\Delta G = 0.01 e^2/h$ and $B_c = 25$ mT. B_c is in agreement with the result for sample D1-2 with nearly the same width. Because of the relatively high measuring voltage $V_m = 250 \mu\text{V}$, the Thouless length $L_T = (hD/eV_m)^{1/2} \approx 430$ nm in this case is much smaller. For narrow samples ($w \ll L_T$) longer than L_T the magnetofingerprint should be an incoherent superposition of $N = L/L_T$ independent samples with same width and thus same B_c . Therefore the amplitude of the UCF should be reduced by $\Delta G/\Delta G_0 = (L_T/L)^{3/2} \approx 0.03$ (Ref. 6) for this sample with $L = L_{\text{eff}} \approx 4.7 \mu\text{m}$ (leads about 1.7 μm each, half-perimeter about 1.3 μm). This yields an unaveraged rms amplitude $\Delta G_0 \approx 0.33 e^2/h$ which is again somewhat higher than the theoretical value.

We do not observe periodic Aharonov–Bohm oscillations for the ring sample expected for multiples of $B = 6$ mT, presumably because of the following two reasons: (1) the aspect ratio between the large linewidth w of 150 nm and the outer

FIG. 5. Symmetric part $[\Delta G(B) + \Delta G(-B)]/2$ (right-hand trace) and anti-symmetric part $[\Delta G(B) - \Delta G(-B)]/2$ (left-hand trace) of the conductance fluctuations of sample D4.

ring diameter d is unfavorable. w should be less than 50 nm;⁷ (2) the amplitude of the oscillations is damped with $\exp(-\pi d/L_T)$ because interference only occurs for electrons which surround one ring branch without losing their phase coherence.

Another interesting observation is made in the magnetoconductance of sample D4. Figure 6 shows two magnetofingerprints of sample D4-2 (same geometry as D4, but different $R = 81.5 \Omega$). The traces have been recorded on two different days without warming up the sample, but switching short cuts open and closed in the leads. The cross correlations of the positive and negative parts of each trace are better than 90%, but the patterns have changed slightly from the first measurement (upper curve) to the second measurement (lower curve). For low fields $|B| < 1.7$ T the pattern is nearly unchanged, for higher fields characteristic deviations occur, e.g., at $|B| \approx 1.85$ T. This means that the changing of some single scattering centers does not change the whole fluctuation pattern, presumably because the number of trans-

FIG. 6. Comparison of two magnetofingerprints of sample D4-2 with same measurement parameters. Traces are offset for clarity.

port channels in our sample is still very large, so that most of the electron scattering paths remain unaffected.

For completeness we mention the result for the long wire C4 with $L=91\text{ }\mu\text{m}$: We observe fluctuations of the magnetoconductance with amplitude $\Delta G=0.0002\text{ }e^2/h$ in qualitative agreement with the expected reduction by a factor ≈ 1900 for $L>l_\varphi$ and $L>L_T\approx 600\text{ nm}$. A reliable result for B_c cannot be obtained because the scatter of the data is too large for the small amplitude of ΔG .

V. CONCLUSIONS

We have presented a process to fabricate noble-metal nanostructures with lateral dimensions down to 80 nm and contact lead dimensions up to 1 mm simultaneously. A bilayer resist system was exposed with 100 kV acceleration voltage. For metallization thermal evaporation of noble metal followed by lift-off is used.

Our preliminary investigations of quantum interference effects in these gold samples are reported. Conductance fluctuations have been observed for all investigated samples with dimensions up to several micrometers. From the absolute resistances and the residual resistance ratio RRR we deduce an offset resistance around $10\text{ }\Omega$ due to the two-lead contact geometry with large areas of the thin metal layer. We interpret our data in terms of a non-fluctuating offset resistance. For short samples we obtain rms amplitudes of $\Delta G\approx 0.24\text{ }e^2/h$. With our experimental parameters the length $L_T=(\hbar D/eV_m)^{1/2}$ is of the same order of magnitude or smaller than the phase coherence length l_φ and therefore influences the interference paths contributing to the fluctua-

tion patterns. We have investigated the dependence of the typical "period" B_c of the fluctuations on the width of the samples. For short samples with $L\approx L_T$ we find that B_c is inversely proportional to the width in agreement with the picture that all electron paths within the phase coherence area contribute to the fluctuation pattern. For a very long sample with $L\gg L_T, l_\varphi$ we also observe reproducible fluctuations with very small amplitude.

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