Transport Regimes in Tunable Gold Nanoconstrictions: Proposed Solution by Low-Frequency Noise Spectroscopy

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I. INTRODUCTION

Studying of nanoscale structures attracts considerable attention nowadays. The structures possess unique mechanical, chemical and electrical properties which differ from those of bulk materials. Because low-dimensional nanoscale devices are extremely sensitive to surface charges they can be applied in different scientific fields including chemical sensing and biosensing.

In conventional electronics there is a continuous tendency of shrinking down the sizes of active elements in electrical circuits. Cutting-edge technologies allow fabrication of reproducible sub-20 nm devices. However, further scaling down will inevitably lead to a breakdown of the laws of classical physics and to dominant role of quantum effects in charge transport processes. In this respect sub-nanometers objects such as molecular layers or even single molecules have to be considered as candidates for the role of functional elements. A possibility to use a single molecule as a molecular rectifier was first reported by Aviram and Ratner[1] in 1974. Nowadays applications of molecules cover a broad range of similar to semiconductor devices[2] including diodes, switches, etc. A huge variety of molecules and functional groups allows tailoring the molecular systems with predefined properties. Investigations of electrical properties of functional nanodevices, which could be operated in different transport regimes and whose properties can be modified by molecular layers[3] or even single molecules[4] are therefore especially important for the perspectives of applications in molecular electronics. Up to now, different approaches[5–8] are developed for characterization of molecular systems. Techniques, based on investigation of tunable metal nanoconstrictions are essentially important because their electrical properties are mainly determined by covering layers, including molecular ones and binding geometry.

In this contribution, we present analyses of the low-frequency noise behavior of bare-gold and benzene-1,4-dithiol (BDT) modified tunable metallic nanoconstrictions. Altering cross-section of the devices allows their operation in different transport regimes: diffusive, ballistic and tunneling. Changes of the dominant transport regime are revealed to be related with a change of the characteristic power dependence of the normalized flicker noise level S_R / R^2 on the resistance R. Modification of the sample surface by organic molecules results in a decrease of the normalized flicker noise level in the ballistic regime of sample conductance. We address this effect to an increased impact of the molecular layer to overall conductance of the system.

II. MATHERIALS AND METHODS

Samples studied are fabricated on the basis of rectangular shape stainless steel pieces (W × L × h = 1 cm × 5 cm × 0.25 mm). Fabrication procedure is performed as follows. At first substrates are cleaned in acetone and isopropanol to remove organic residuals from the surface. Then two layers of polyimide PI2611 are subsequently spin coated and hard-baked to create a passivation layer. Nanoconstriction pattern and contact pads are defined by means of e-beam lithography. Pattering is followed by metallization (5 nm of Ti and 60 nm of Au) and lift-off processes. Finally, polyimide layer is isotropically etched down to form freestanding gold nanoconstrictions. Characteristic sizes of fabricated devices in the narrowest parts are below 100 nm and can be further decreased in a controlled way.

All measurements are performed under a high vacuum (10^{-5} mbar) in a shielded environment. A homemade three point bending apparatus is used to tune the resistance of the sample in a controlled manner. Bending of the sample results in an elongation of the suspended nanoconstriction, narrowing of its cross-section and subsequent change of the resistance. Stabilization of the resistance is performed by a PC-controlled feedback system. After resistance stabilization, noise measurements are performed using homemade noise measurement setup. This setup[9] allows precise noise measurement in the frequency range 1 Hz - 100 kHz.

III. RESULTS AND DISCUSSION

Resistance tuning procedures with following noise spectra measurements are performed for a wide range of sample resistances including regions before (R = 10 Ohm - 12.9 kOhm), during and after (R = 12.9 kOhm - 10 MOhm) breaking of the constriction. In the case of bare gold nanoconstrictions noise follows exclusively $1/f$ behavior in all investigated range of sample resistances. Normalized flicker noise power spectral density S_R / R^2 has a power dependence on the resistance, which can be described by:

$$\frac{S_R}{R^2} \sim R^m \quad (1)$$

During the tuning process several regions with different value of the exponent m can be clearly resolved. At low resistances $m = 2$, while at higher resistances the value of the exponent decreases to $m = 1$. We attribute this effect to a change in the dominant transport regime in the nanoconstriction due to changes of the sample geometry while tuning. In fact, elongation of the gold nanoconstriction leads to shrinking down of its cross-section. When characteristic sizes of the constriction become comparable with the electron mean free path ($\lambda_e \approx 4$ nm for gold at room temperature[10]) the dominant mechanism of charge transport changes from diffusive to ballistic. Further elongation of the constriction results in its breaking and therefore transport is then defined by a quantum tunneling regime.

Noise spectra, measured for nanoconstrictions modified with BDT molecules demonstrate that flicker noise still remains a dominant component in noise spectra. However characteristic Lorentzian-shaped components can also be resolved. Noise behavior in diffusion and tunneling regions is similar to the one for samples without molecules. On the other hand, a pronounced difference is registered in the ballistic conductance region, where samples, modified with BDT show around one order of magnitude lower normalized flicker noise level in comparison to bare-gold structures. We explain this effect by an increased impact of the molecular conductance to overall conductance of the system in this regime.

Analytical model can be applied to describe behavior of the flicker noise in the diffusive regime of the sample conductance. For describing the flicker noise behavior of the system in ballistic and tunneling regimes we take into account changes of the nanoconstriction geometry due to elongation and Hooge's relation:

$$\frac{S_R}{R^2} = \frac{S_I}{I^2} = \frac{\alpha_H e}{\tau f I} \quad (2)$$

Here α_H is the dimensionless Hooge parameter, e is the elementary charge, τ is the characteristic time and I is the current. Obtained results are summarized in Table 1.

IV. CONCLUSION

Characterization of bare-gold and benzene-1,4-dithiol modified tunable cross-section nanoconstrictions is performed using low-frequency noise spectroscopy. Normalized flicker noise level demonstrates a characteristic power dependence on the nanoconstriction resistance with the exponent reflecting the dominant transport regime in the nanostructure. Noise behavior is well described by phenomenological models, developed considering changes of the system geometry while tuning. Samples modified by BDT molecules demonstrate lower normalized flicker level in comparison to bare gold samples and an additional characteristic Lorentzian noise component. We suggest that this effect takes place due to a significant contribution of molecules to the overall conductance of the system.

Table 1. Noise behavior in tunable gold nanoconstrictions

Dominant transport regime	Approximate resistance range [Ohm]	Characteristic flicker noise behavior
Diffusive	≤ 300	$S_R / R^2 \sim R^2$
Ballistic without molecules (BR1)	3000-12900	$S_R / R^2 \sim \sqrt{R}$
Ballistic with BDT molecules (BR2)	3000-12900	$S_R / R^2 \sim R$, *lower amplitude than in BR1 case*
Tunneling	> 12900	$S_R / R^2 \sim R / \ln R$

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