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The generation of the electrical oscillations in a contact of an AFM probe to an individual Au nanoparticle in a SiO_2/Si film

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Abstract. We report on the experimental observation of the electrical oscillations in an oscillating loop connected to a contact of a conductive probe of an atomic force microscope to a tunnel-transparent (≈ 5 nm thick) SiO_2/Si(001) film with embedded Au nanoparticles. The oscillations were attributed to the negative differential resistance of the probe-to-sample contact originating from the resonant electron tunnelling between the probe and the Si substrate via the quantum confined electron energy states in small (~1 nm in size) Au nanoparticles. This observation demonstrates the prospects to build an oscillator nanoelectronic device based on an individual nanometer-sized metal nanoparticle.

1. Introduction

Metal nanoparticles (NPs) embedded into dielectric films have attracted much attention in recent years due to a variety of potential applications in non-volatile memory, plasmonics, nonlinear optics, etc. [1]. Earlier, we have demonstrated the current-voltage (*I–V*) curves of a contact between a conductive probe of an atomic force microscope (AFM) and a tunnel transparent (3 to 5 nm thick) SiO_2/Si(001) film with embedded Au NPs deposited by Pulsed Laser Deposition (PLD) to manifest the negative differential resistance (NDR) attributed to the resonant electron tunneling via the quantum confined electron states in small enough (~1 nm in thickness) Au NPs [2]. It is well known that once a nonlinear electrical element with NDR is available, one can build an oscillator by combining this NDR element with an oscillating loop. In Ref. [3], we have reported an experimental observation of the electrical oscillations in an oscillating loop connected to a contact of a conductive AFM (CAFM) probe to a tunnel-transparent yttria stabilized zirconia (YSZ) film with embedded Au NPs. The excitation of the electrical oscillations was attributed to NDR of the CAFM probe-to-sample contact originating from the resonant electron tunneling via the quantum confined electron states in small Au NPs. This result demonstrated experimentally the prospects to build an oscillator nanoelectronic device based on an individual metal NP.

In the present paper, we report on the experimental observation of the electrical oscillations in the oscillating loop connected to the CAFM probe contact to SiO_2/Si(001) film with embedded Au NPs.

2. Experimental details

The SiO_2 films with embedded Au NPs were fabricated using self-assembling during the plasma-enhanced oxidation [4] of the amorphous Au–Si mixture (1:6) films of ≈1.5 nm in thickness deposited by PLD at 300 K onto the n^+-Si(001) substrates covered by the native oxide. The Au NPs of 5–7 nm in the lateral size, according to Transmission Electron Microscopy (TEM) data [4], and of 1–1.5 nm in thickness were arranged almost in single sheet inside ≈3.5 nm thick SiO_2/Si film (Fig. 1).

The CAFM investigations were carried out in Ultra High Vacuum (UHV) environment (the base residual gas pressure was $<10^{-10}$ Torr) at 300 K using Omicron® UHV AFM/STM LF1 atomic force/scanning tunneling microscope (AFM/STM) installed into Omicron® MultiProbe™ RM UHV system. The schematic representation of the CAFM experiment is shown in Figure 1. The sample surface was scanned across in Contact Mode by a conductive AFM probe (NT-MDT® NSG-11 DLC diamond-like film coated probes were used). A bias voltage V_g was applied to the n^+-Si substrate relative to the AFM probe and the electric current between the probe and the substrate I_t was measured as a function of the probe coordinates in the sample surface plane (x, y). In another measurement mode, the I–V curves of the probe-to-sample contact were measured in every point of the AFM scan.

In order to observe the oscillations of I_t due to NDR of the probe-to-sample contact, an oscillating loop consisting of a capacitor C and a coil L was connected between the sample and the bias voltage source (Fig. 1). The free oscillation frequency of the oscillating loop f_0 was chosen to be ≈24.3 kHz, because the bandpass of the STM preamplifier (I–V converter) was limited to 30 kHz at the factory in order to suppress the high frequency noise. The waveforms of the probe current $I_t(t)$, where t is the time, were recorded by Agilent 3000A oscilloscope connected to the STM preamplifier output.

Capturing the waveform $I_t(t)$ at the moment, when the excitation of the oscillations takes place, was the key issue of the experiment. Indeed, the following conditions should be satisfied for the electrical oscillations in the oscillating loop to occur:

- the AFM probe should be positioned just above an Au NP and the NDR should be present in the I–V curve of the probe-to-sample contact;
- the bias voltage V_g should be within the NDR region in the I–V curve.

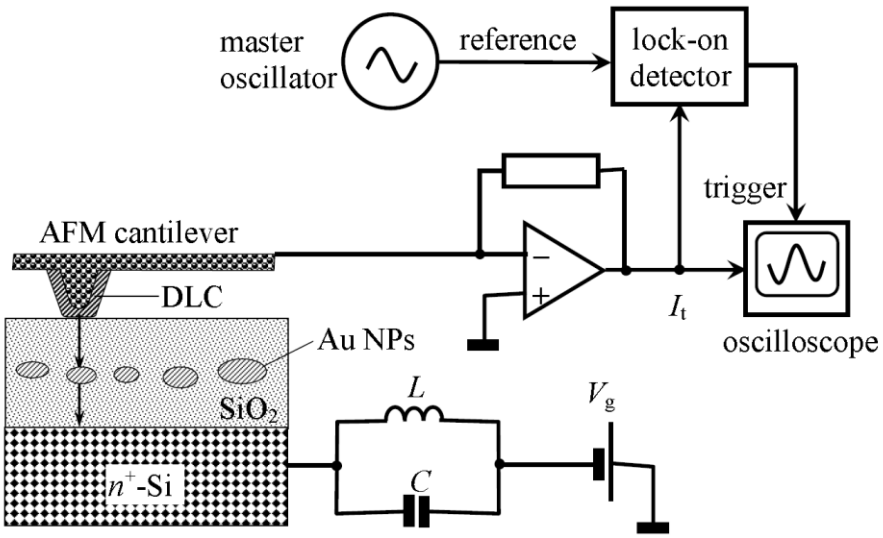

Figure 1. Schematic representation of the CAFM experiment.

These conditions are difficult to satisfy simultaneously because of the creep of the AFM piezo scanner. The AFM probe having been positioned onto a NP drifts away very quickly. Therefore, in order to record the waveforms of the electrical oscillations in the oscillation loop, the procedure described earlier in [3] was applied. The *I–V* curves were recorded in every point of the AFM frame. The limits of the V_g sweep were set from 0 to 8 V in order to ensure the NDR region in the *I–V* curves to be passed through. Stanford Research® SR-530 lock-in detector has been connected to the user's "I_t" output of Omicron® AFM/STM control electronics in parallel to the oscilloscope (Fig. 1). The reference signal for the lock-in detector has been provided by a high precision digital generator serving as a master oscillator. The frequency of the reference signal was set equal to the free oscillation frequency of the oscillating loop f_0. The output of the lock-in detector was connected to the trigger input of the oscilloscope. In the course of scanning, once the AFM probe encounters a NP and V_g reaches the beginning of the NDR region, the excitation of the electrical oscillations begins in the oscillating loop and the sinusoidal component with the frequency f_0 appears in the spectrum of the probe current I_t. Hence, the output signal of the lock-in detector increases with increasing oscillations' amplitude at the frequency f_0. Once the output signal of the lock-in detector reaches a preset threshold of the oscilloscope trigger action, the waveform $I_t(t)$ is captured.

3. Results and discussion

An AFM image and current one, i. e. a map of $I_t(x, y)$ at given V_g of a SiO_2:NP-Au/Si film are shown in Figures 2a and 2b, respectively. The spots of increased I_t (hereafter referred to as the current channels) in Figure 2b were related to the electron tunneling between the AFM probe coating and the n^+-Si substrate via the Au NPs [2] as shown in Figure 1.

The sizes of the current channels in Figure 2b ranged from ≈30 to ≈100 nm, whereas the lateral sizes of the Au NPs were 5 to 7 nm, according to the TEM data [4], as it has been already mentioned above. This disagreement can be explained as follows. In general, the sizes of the CAFM images of the metal NPs inside the dielectric films are determined by the diameter of the contact area between the AFM probe tip and the sample surface D_p and do not depend on the size of the NPs themselves [5]. In turn, D_p depends on the tip curvature radius R_p and on the loading force, as well as on the elastic modules of the sample and probe coating materials. Roughly, one can estimate $D_p \sim R_p \approx 70$ nm for NT-MDT® NSG-11 DLC probes (according to the vendor's specifications) that is consistent with the sizes of the current channels in Figure 2b. *I–V* curve of the contact between the AFM probe and the SiO_2:NP-Au/n^+-Si(001) film measured inside a current channel is shown in Figure 3. The current channel, wherein the *I–V* curve was measured, is marked by a circle in Figure 2b. A well expressed NDR at the positive bias V_g applied to the n^+-Si substrate relative to the AFM probe (corresponding to the injection of the electrons from the AFM probe into the SiO_2:NP-Au/Si film) has been observed.

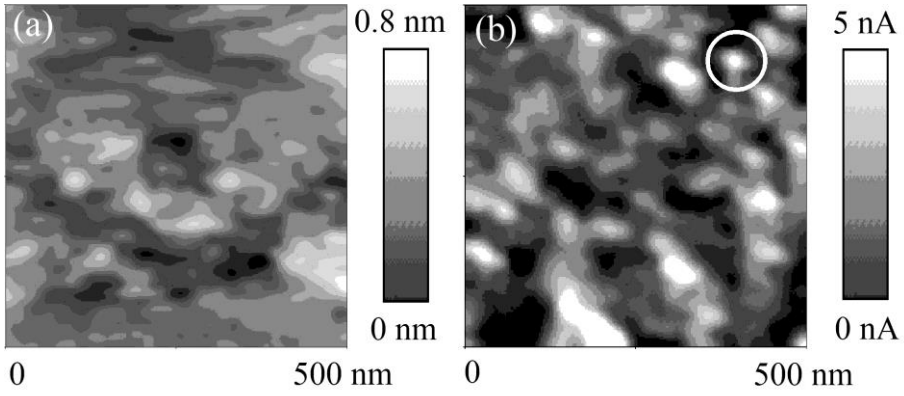

Figure 2. (a) AFM and (b) current images of the SiO_2:NP-Au/$n^+$$-Si(001) film. V_g = 4.5 V.

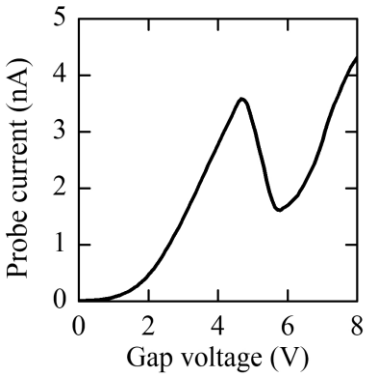

Figure 3. *I–V* curve of the probe-to-sample contact measured in the current channel marked by a circle in Figure 2b.

The NDR was attributed to the resonant electron tunneling from the AFM probe coating into the n^+-Si substrate via the quantum confined electron states in the Au NPs [2]. A one-dimensional band diagram of the contact between the DLC-coated AFM probe and the SiO_2:NP-Au/n^+-Si film across an Au NP of 1 nm in thickness embedded into the middle of a 3.5 nm thick SiO_2 film calculated for $V_g = 4.5$ V is presented in Figure 4a. The calculation procedure was described in details in Ref. [2]. One can see in Figure 4a that the SiO_2/Au/SiO_2/Si stack represents a double barrier structure (DBS) for the electrons similar to the one of the resonant tunnel diode [6].

Figure 4b presents the calculated tunnel transparency spectrum T(E) (300 K) of the DBS presented in Figure 4a for $V_g = 4.5$ V, which resonant peak in the *I–V* curve has been observed in Figure 3. The procedure of the calculation was described in Ref. [2] as well. The peaks in T(E) correspond to the virtual quantum confined energy states in the Au NP. For example, the calculated envelope wavefunction $\chi(z)$ for the resonant state marked by the arrow in Figure 4b is shown in Figure 4a. The energy spacing between the resonant states in the Au NPs can be estimated as ≈ 1.5 eV that is much greater than the thermal energy kT at 300 K (≈ 26 meV). So far, the conditions for the manifestation of the resonant tunneling effect were readily available in the investigated films.

In Figure 5, a probe current waveform $I_t(t)$ recorded at $V_g \approx 5$ V is presented, i. e. V_g was within the NDR region of the *I–V* curve of the probe-to-sample contact (Fig. 4a). Nearly sinusoidal oscillations of I_t have been observed. The frequency of the oscillations matches the free oscillation frequency of the oscillating loop $f_0 \approx 24.3$ kHz.

Thus, the results of the present study demonstrate the possibility to build an oscillator based on the resonant tunneling effect in a single Au NP embedded into a tunnel transparent SiO_2/Si film.

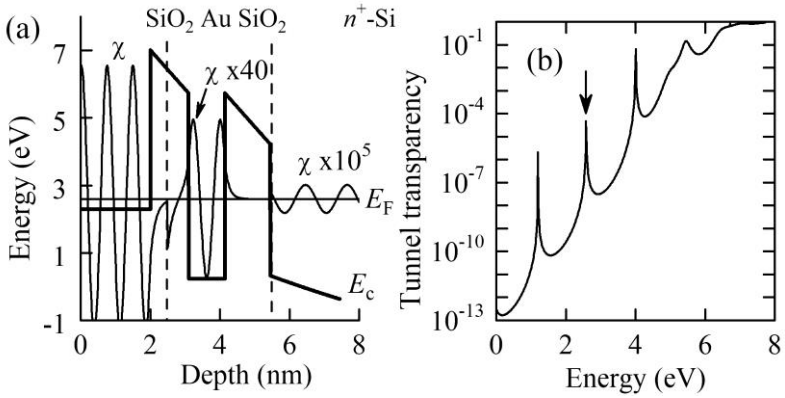

Figure 4. (a) Calculated band diagram and (b) tunnel transparency spectrum of a contact between the AFM probe and the SiO_2:NP-Au/n^+-Si film. $V_g = 4.5$ V.

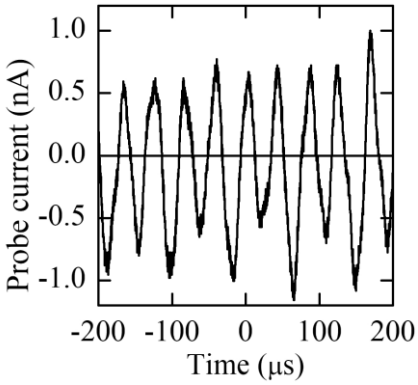

Figure 5. A waveform of the probe current oscillations.

Future prospects of the device application of these results are connected with the replacement of the AFM probe by a permanent point electrical contact to individual metal NPs. The present microelectronics is approaching the 10-nm critical dimension rapidly, so one can expect an electrical contact of ~ 10 nm in size with appropriate microstrip wiring to become possible in the nearest future.

It is worth noting that similar results obtained in Ref. [3] on an YSZ:NP-Au film and on the SiO_2:NP-Au ones in the present study point to the origin of the NDR in the *I–V* curves to be not the properties of the film materials, but the fundamental phenomenon of resonant tunneling via the ultrafine Au NPs.

It should be noted also that a low free oscillation frequency of the oscillation loop f_0 was selected in the present work for the reason of limited bandpass of the STM preamplifier only. Since the tunneling process itself is very fast, the operation frequency of the nanoelectronic oscillator device based on a single metal NP could be raised, in general, up to the THz band when an appropriate cavity is utilized.

4. Conclusion

In the present study, we have demonstrated experimentally the room-temperature operation of a prototype nanoelectronic oscillator device based on a single Au NP embedded into a tunnel-transparent SiO_2/Si film. The operation of the device is based on the resonant electron tunneling via the quantum confined energy states in an ultrafine Au NP. To provide a local tunnel electrical contact to a single NP inside the SiO_2 film, we applied a probe of an atomic force microscope. The results of the present study demonstrate wide prospects for future application of such nanoelectronic devices.

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