Electric field induced motion of metallic droplets: application to submicron contactor

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Abstract: This letter reports the monitoring of reversible displacement of a gallium droplet on a tungsten submicron wire deposited by focused ion beam from tungsten hexacarbonyl precursor. We demonstrate that by applying a voltage to the wire terminals, the internal electric field created along the wire produces the motion of the droplet. Since the matter involved in this displacement is conductive, we show that it is possible to build a submicron electrical switch. Contact can be switched on and off between two electrodes separated by a submicron gap, by electrical monitoring the position of the conductive droplet.

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In nanoscience, connections of nano-objects are needed to demonstrate the peculiar electrical properties of such objects. A lot of studies have been devoted to direct writing of high quality metallic wires by local decomposition of gaseous molecules induced by tightly focused ion or electron beams¹⁻⁴. These techniques are particularly suitable to link the micron world (optical lithography) to the nano world. Cross beam systems coupling a Focused Ion Beam (FIB) and a Scanning Electron Microscope (SEM) equipped with a gas injection system (GIS) have been widely used in microelectronic industry for integrated circuit rewiring at the submicron scale. The deposited materials are often contaminated by C and O coming from the decomposition of the ligands of the precursor molecules, usually organometallic or metalorganic molecules. Furthermore extra contamination of the deposited wires occurs by implantation of gallium atoms coming from the Ga⁺ ion beam itself. Consequently the metal wire resistivity is usually as high as 100 times the bulk one.

Tungsten nanowires (200nm wide by 80nm thick) has been fabricated following a focussed ion beam induced deposition process⁵⁻⁶ (FIBID) from dissociation of WC0₆ molecules and studied in our cross beam system equipped with in situ electrical measurements. A current density above a threshold value⁵⁻⁶ of 1.4×10^7 A/cm² induces a wire annealing by Joule effect which improves their electrical conductivity. The high current density induces the exudation of gallium out of the wire, and its merging as several droplets (Fig. 1). The exudation of gallium is closely related to the light 3% decrease of the wire resistance between 1.1V and 4V (Fig 1). Droplet formation along the wire takes place when the voltage reaches about 2.5V. This value increases with the initial value of the resistance of the wire to get the current density threshold required. Increasing the voltage up to 4V drives all the droplets towards the lowest potential electrode, leading to the formation of a unique droplet of about 550nm in diameter (SEM image inserted on Fig 1). For voltage above 4V, the

resistance of the wire drops due to the non reversible crystallisation of the tungsten wire⁵⁻⁶.
The value of the resistance after the first ramping can be 10 times less than the value of the
resistance of the as deposited wire. The droplet contains about 90at% of gallium atoms and 10at% of oxygen atoms as observed by ex situ energy dispersive X-ray analysis. Most of the oxygen contained in the droplet is a consequence of the sample transfer through air from one SEM to the other.

A gallium droplet, which has been formed at one extremity of a wire, as explained previously⁵⁻⁶, migrates towards the opposite terminal if the voltage is reversed. The droplet is always driven towards the lowest potential electrode when the voltage applied ($\sim 2.5\text{V}$) induces a current density of about $1.4 \times 10^7 \text{ A/cm}^2$ through the tungsten wire. Droplet position change was followed in-situ by SEM. Fig. 2 displays 3 images grabbed from a SEM video, separated in time by 0.6s between 2a and 2b and 0.4s between 2b and 2c. We started with one droplet nearby the left electrode and slowly increased the voltage. One can see that the left droplet diameter decreases slightly while the right one increases. It demonstrated that the gallium transfer occurs from the left electrode to the right one by changing the polarity of the voltage between the two electrodes. During the transfer, the W wire remains unchanged between the initial and final state. It should be stressed that instead of moving at once from one point of the wire to the other, the droplet diameter decreases while simultaneously other droplets pop-out of the wire at different places close to the other extremity of the wire. Thus, it appears that gallium atoms transfer can be done following two paths the channels between W grain boundaries or on the wire surface. One can note that gallium atoms move in opposite direction of the electrons. The Ga atom motion is not due to a direct transfer of electron momentum. Finally, for a voltage over 4V, all the droplets end at the lowest potential electrode, merging again into a single droplet. Reversing the bias polarity leads to gallium transport towards the opposite electrode. The round trips (fig 3) of the gallium droplet were

obtained at least hundred times on the same sample without any change of the volume of the gallium droplet. The transfer of gallium from one electrode to the other needs a slow ramping of the voltage from zero to 4V (~15s) to prevent melting of the wire which would occurs when pulse voltage are applied.

Each time the voltage is switched and the threshold of current density is reached, gallium ends agglomerated into one droplet at the vicinity of the lowest potential electrode. Such spherical pure gallium droplets have been already observed on structures prepared with gallium⁺ FIB⁷. We have checked that the volume of the droplet corresponds to the amount of gallium⁺ ions implanted in the tungsten wire⁵⁻⁶ during the deposition process. The electrical treatment alters the initial mixture of tungsten and gallium, and leads to the separation of the two components according to the phase diagram⁸. The application of a voltage of few volts induces a high density current flow that leads to a wire temperature increase. The power dissipated in such a wire can reach 15mW without damaging the circuit. At this power, the temperature in the middle of the wire can be estimated to be around 1000°C⁹. Considering that the temperature of the two electrodes remains close to room temperature, it exists two strong temperature gradients in opposite directions between the middle of the wire and both electrodes. Thus, the driving force for the gallium motion in only one direction cannot be related to a temperature gradient but to the strong electric field applied to the wire.

In order to better understand the driving force which induces gallium transfer, we have designed a test chip integrating a Z-shaped wire to check the possibility of driving gallium droplets along different circuits. The “Z” wire, shown in figure 4, is made of three wires prepared as the single straight wires. By applying the same value of the current density as for a straight wire (same diameter), identical states are observed: at the low voltages (<4V), gallium droplets appear along the wire, and when the voltage is increased gallium tends to accumulate in one droplet at the lowest potential electrode (Figure 4a). The presence of a

droplet nearby the lowest potential in such a structure demonstrates that the driving force is the internal electric field along the wire and not the electric field created between the two aluminium electrodes separated by ten microns apart. Despite the high electric field generated between the electrodes, which can reach 25kV/cm, the gallium contained in the Z structure can be moved along it. The inversion of the bias polarity induces the reverse motion of gallium, leading to the formation of the gallium droplets towards the opposite electrode (Fig. 4b).

Since the droplet is made of at least 90at% of gallium⁵⁻⁶, it is conductive and may be used to establish a contact between two electrodes separated by a distance of the order of the size of the droplet. Such a liquid contactor may be switched on and off by applying an electric field to position the droplet inside or outside the gap between the two electrodes.

To demonstrate the switching ability, we have drawn by FIBID an original circuit presented in Fig 5a. First, a cross consisting of two tungsten wires was deposited, and connected to four electrodes. The conduction through both branches (AD and BC) of the cross was checked. The resistance of each branch was almost the same for AD and BC wires (950Ω). Then, two grooves were milled by FIB across the BC wire, along the edges of the AD wire, close to the intersection between both wires (see Fig. 5a). The insulations between A-B and A-C were checked by measuring on time the resistance during the milling process. Milling was stopped as soon as the resistance reached a value which insured a low conduction (450kΩ).

After the preparation of the circuit, the first step was to get a droplet nearby one of the electrodes A or D. This was obtained by applying the voltage threshold V_{AD} . For $V_{AD} < 0$ (resp $V_{AD} > 0$), the droplet is located nearby the A (resp the D) electrode. In order to create a contact between the AD wire and the counter electrode B, the droplet must be attracted into the gap towards this electrode. This was obtained by applying a constant voltage V_B on the B

electrode so that its potential remains lower than both V_A and V_D during the ramping of the V_{AD} voltage. Note that as long as the gap is open the electrical field is very high in this zone and can attract Ga atoms. The droplet motion can be followed in real-time by SEM imaging. As soon as a droplet closes the gap between the counter-electrode B and the AD wire, all the electrodes were grounded (Fig 5b). The resistance of the closed circuit formed between the electrode B and the wire was found to be 950Ω , that corresponds to the initial resistance which was measured before the milling step. Thus the electrical contacts between the gallium droplet and the tungsten wire do not produce any extra contact resistance. This confirms that the gallium droplet establishes a perfect electrical contact. Under vacuum during few hours we did not notice any change in the value of the contact resistance. To remove the contact between electrode B and the AD wire, a negative ramp voltage V_{AD} was applied to the wire, while a positive ramp voltage V_{BA} was applied between the electrodes B and A. The diameter of the droplet trapped in the gap was followed in situ by SEM imaging. When the electrical contact is removed, the gap is open and the value of the resistance between the wire and the electrode turns back to the value of $450k\Omega$. This experiment can be repeated many times under vacuum without noticeable change of the resistance of the open and closed circuit. This latter experience shows again that the driving force for gallium droplet motion is undoubtedly due to the electric field since we are able to position the droplet in the middle of the AD wire where the temperature is higher than other locations.

This study of submicron structures allows to evidence phenomena never observed before and opens a new field of research to get a better understanding of the influence of strong electric field on the displacement of atoms and liquid matter in nanostructures. Submicron system offers peculiar physical conditions: strong temperature gradient, strong electric field and high current densities. Electrical switching by transfer of a conductive material between two isolated electrodes is demonstrated. Such phenomenon could be used

for building memories if the commutation time for such circuits can be decreased. The increase of the commutation speed could be obtained by blocking a droplet of size smaller than the gap between the electrodes. In this case, the strong electric field applied between the droplet and the electrode could lead to a change of the droplet shape until the contact is established. Such a system could also offer a way to study the different steps of a metallic contact formation taking advantage of reversible tuneable quantum contacts. Low temperature experiments are required to address this point. It could be a way to establish a control quantum resistance between two metallic sinks. Finally, it is the first example of a control of the reversible transport of tiny quantities of metallic matter which opens the field of studies on metal migration and demixion induced by high current density.

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Figure captions

FIG 1 : Evolution of the resistance of the wire during the in situ electrical treatment. Insert on the graphic: SEM image of a tungsten nanowire after electrical treatment. One can see the accumulation of gallium in a droplet of 750nm in diameter nearby the negative electrode.

FIG 2 : Series (a,b,c) of SEM images extracted from a video during the transfer of a gallium droplet from left to right.

FIG 3 : A sequence (a,b,c,d) of SEM images of the same tungsten nanowire at different times for positive or negative applied voltage. The right electrode is the reference.

FIG 4 : SEM images of a Z structure for two different polarisations: a) Negative electrode on the right. b) Negative electrode on the left.

FIG 5 : Submicron switch elaborated by FIB at different states: a) the as built cross structure. b) on state, the gallium droplet create the contact with upper right electrode, and b) off state, after removing the gallium droplet from the gap

FIG. 1. H. Dallaporta,; M. Prestigiacomo, F. Bedu, D. Tonneau, D.Chatain, and P. Sudraud

FIG. 2. H. Dallaporta, M. Prestigiacomo, F. Bedu, D. Tonneau, D.Chatain, and P. Sudraud

FIG. 3. H. Dallaporta,; M. Prestigiacomo, F. Bedu, D. Tonneau, D.Chatain, and P. Sudraud

FIG. 4 : H. Dallaporta, M. Prestigiacomo, F. Bedu, D. Tonneau, D.Chatain, and P. Sudraud

FIG.5 : H. Dallaporta, M. Prestigiacomo, F. Bedu, D. Tonneau, D.Chatain, and P. Sudraud.