Instabilities in Focused Ion Beam-patterned Au nanowires

J.P. Naik [a,*], P.D. Prewett [a], K. Das [b], A.K. Raychaudhuri [b]

[a] *School of Mechanical Engineering, University of Birmingham, Birmingham B15 2TT, UK*
[b] *S.N. Bose National Centre for Basic Sciences, JD Block, Salt Lake-Sector 3, Kolkata 700098, India*

A B S T R A C T

Focused Ion Beam (FIB) technology has become an indispensable enabling tool for micro nano fabrications. One important application is to use FIB for patterning conducting nanowires of metals down to a few tens of nanometre for applications such as interconnects, heaters and temperature nanosensors. A series of experiments on Au nanowires fabricated by FIB on Si_xN_y membrane show that nanowires with width $\leqslant$50 nm have structural instabilities. These are liquid like and first show-up as undulations in nanowire width with clearly defined wave lengths. For smaller widths ($\sim$20 nm) the instabilities grow and the wires eventually break-up into spherical balls. Further experiments show that the nanowires can be made stable to smaller widths by the use of a Cr underlayer to enhance surface wetting. The observed behaviour is due to the Rayleigh-Plateau instability which occurs for systems in which surface energy dominates.

1. Introduction

The growth and fabrication of metallic nanowires are of considerable interest for its application in nanoscale devices. In order to use the metallic nanowires in nano-device applications such as interconnects they must be stable and of controlled geometry. It has previously been observed that nanowires with small lateral dimensions may show instabilities which can eventually lead to break up into small spheres [1,2]. Even nanowires formed so as to be embedded in templates show oscillations/fluctuations in diameters leading to corresponding fluctuations in resistance [3]. Such structural or morphological instabilities have been attributed to the Rayleigh-Plateau instability [4–6] and are a major potential concern to the nanotechnology community.

In a fluid column with length > diameter, the Rayleigh-Plateau instability occurs when the radius falls below the value $R_C \frac{T}{\sigma_Y}$, where T Nm^{-1} is the surface tension and σ_Y Nm^{-2} is the liquid yield stress. Below this critical radius ($R < R_C$), the surface stress is larger than the yield stress and the instability can grow and lead ultimately to fragmentation. In solids the yield force is many orders larger than that of liquids and as a result the phenomena so ubiquitous in liquids cannot be seen in metals until the lateral dimension falls below a few tens of nanometre. For Au, the material investigated in this paper, $\sigma_Y \approx 100$ MPa and $T \approx 1.3$ Nm^{-1}, so that $R_C \approx 13$ nm. For Cu, with higher yield force $\sigma_Y \approx 201$ MPa, R_C has the significantly lower value of 7 nm [7]. It is thus expected that for metal nanowires with lateral dimensions below a few tens of nanometre such instabilities may occur. It is also clear that a number of factors like the degree of wetting of the substrate surface by the nanowires, impurities, strain and temperature may either enhance or inhibit the instabilities.

Past studies of liquid like instabilities in metal nanowires have been mostly carried out in cylindrical geometry [1–3] and the wires were not supported. These wires were typically made by chemical routes such as electrochemical deposition and were mostly single crystalline. The motivation of the present paper is to investigate liquid like instabilities in nanowires with non-cylindrical cross section made by "top-down" fabrication such as patterning by Focused Ion Beam (FIB). Such nanowires are more likely to be actually used in nano-device applications. In our case, the FIB patterned Au nanowires were formed on Si_xN_y membranes which are widely used to make nanosensors of various kinds.

2. Fabrication

The typical substrate used in our experiments is a 7.5×7.5 mm^2 silicon chip with an etched square window of 200 nm thick silicon nitride membrane, measuring 200 μm on its side, as shown in Fig. 1. The nitride window was first coated with a layer of sputtered gold to a thickness of 50 nm with an Agar Auto Sputter Coater with vacuum pressure of 0.07 mbar. The nanowires were fabricated by patterning with FIB using a FEI Strata^{TM} Dual Beam 235 machine with built-in Scanning Electron Microscope (SEM). The Focused Ion Beam of 30 keV gallium (Ga+) ions with spot size of 10–15 nm, was used to sputter the gold film from two sides to form a 5 μm long nanowire supported on the

* Corresponding author.
E-mail addresses: jpn779@bham.ac.uk, jpnaik_99@yahoo.com (J.P. Naik), p.d.prewett@bham.ac.uk (P.D. Prewett).

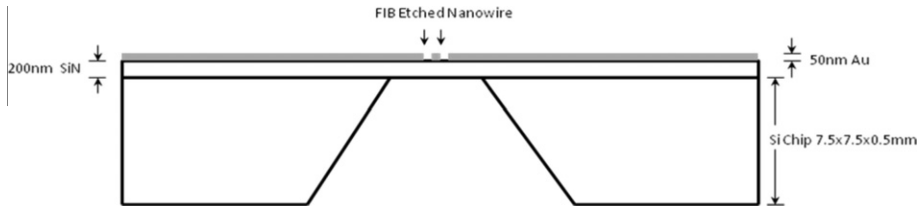

Fig. 1. Cross-section of the Si_xN_y membrane with FIB etched nanowire.

membrane. Nanowire widths ranged from more than 200 nm down to 20 nm with rectangular cross section. The dual beam capability of the FEI Strata^{TM} Dual Beam 235 FIB/SEM allows in situ imaging of the nanowires by SEM immediately after fabrication, without removing them from the specimen work chamber.

The patterning by FIB also leads to implantation of Ga^+ into the body of the film. The penetration depth of Ga^+ at this energy in Au is in the range of ~10 nm so that the ions stop inside the body of the film. However, the contamination by Ga^+ as detected by Energy Dispersive X-ray Analysis (EDAX) attached to the SEM is small, viz. less than 2%.

3. Results

Fig. 2 shows the SEM image of a gold nanowire of width about 200 nm. The image clearly shows that the wire is unperturbed and retains its as-fabricated rectangular cross-section along its length. However, as nanowire width is reduced to about 60 nm, undulations start to appear at some locations along the length of the wire, as shown Fig. 3. With further width reduction to below 50 nm the morphological instability becomes prominent. In Fig. 4, a nanowire of width below 50 nm shows clear spatial oscillation of width along the whole length of the wire, with well-defined wavelength along both edges (identified in Fig. 5.). The average wavelength (λ) calculated from the SEM image is found to be about 165 nm. The undulation instability grows progressively as the width is further reduced (diameter $\leqslant$ 20 nm) and this leads ultimately to fragmentation of the wire (see Figs. 5 and 6). The developing morphological instability of the nanowire breaks it like a liquid and the fragmented parts make near-spherical balls.

The main results from our experiments are that, for FIB fabricated nanowires of thickness 50 nm and width below ~50 nm, morphological instabilities become apparent in the nanowires even though they are supported by the silicon nitride substrate. The observed instabilities in the nanowires of rectangular cross-section are very similar to the liquid like instabilities observed in free standing nanowires of cylindrical cross-section of similar lateral dimension (diameter ~ width). The instabilities appear to have been initiated in absence of any explicit thermal treatment and as discussed below may be triggered by the ion beam process itself.

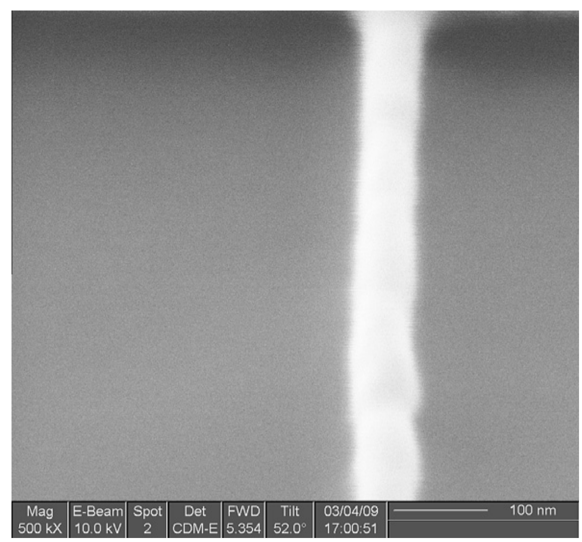

Fig. 3. SEM image showing 60 nm wire with undulating instability.

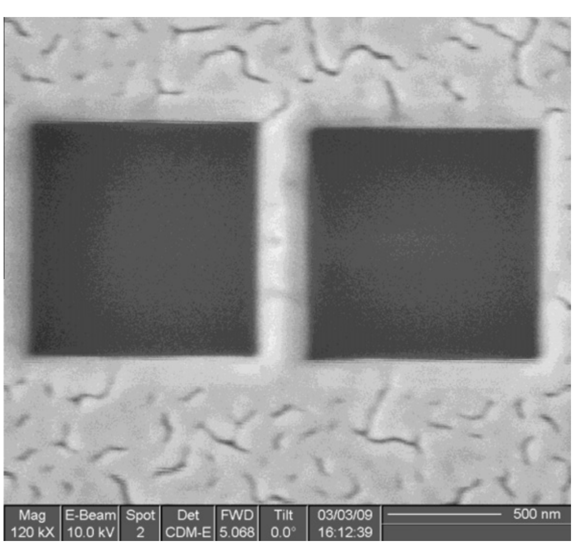

Fig. 2. SEM image of the 200 nm gold wire showing stable morphology.

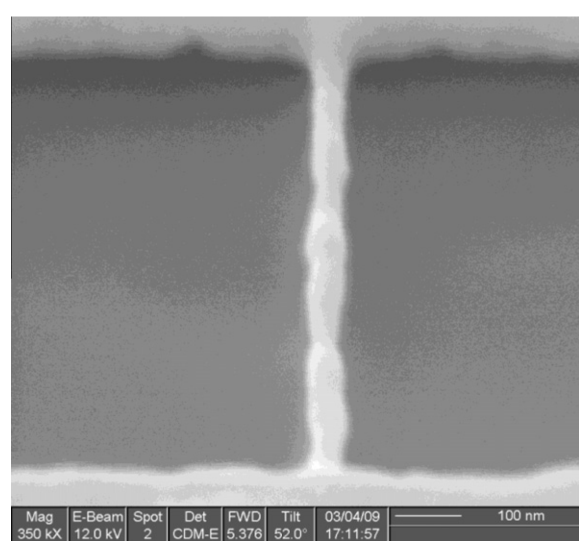

Fig. 4. SEM image of the nanowire below 50 nm in diameter with periodic undulation of the width.

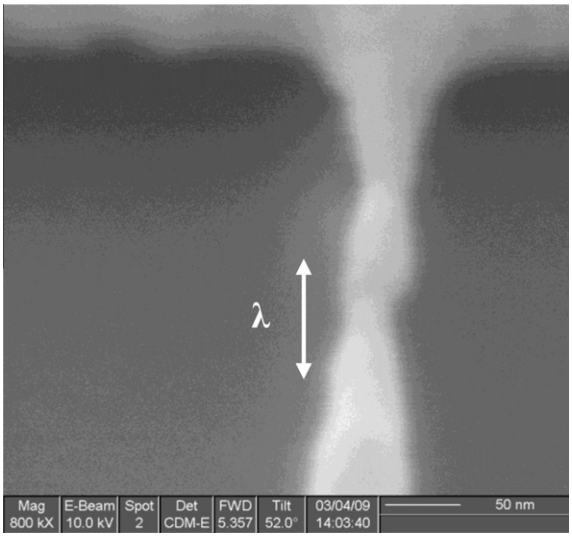

Fig. 5. SEM image showing onset of droplet formation in the unstable nanowire.

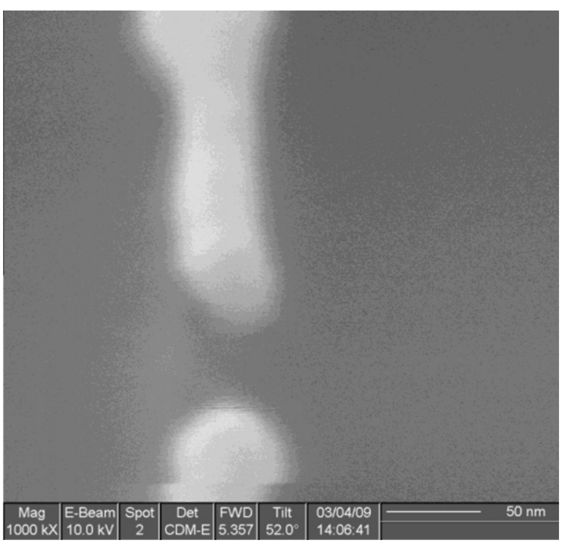

Fig. 6. SEM Image showing fragmented nanowire.

4. Discussion

The fact that the instabilities occur below a width of 50 nm and the nature of the undulation suggest that these are caused by the Rayleigh-Plateau instabilities. Due to the variation of etch rate with crystal orientation, some edge roughness due to the nanocrystalline nature of the deposit is to be expected [12]. However, the observed variations of nanowire width are on a significantly larger scale and are distributed along the wire with an identifiable wavelength which suggests a hydrodynamic origin. For a pristine Au nanowire with cylindrical geometry, such instability should start when the diameter falls below 25 nm [3–7]. The observed undulations in the width which we take as the onset of the instability, start at a somewhat higher value of the width ($\sim$50 nm), but the break-up and fragmentation occurs for a width of $\approx$20 nm which is very close to what one would expect for a cylindrical wire, even though our case the wire is deposited onto a substrate. This is consistent with relatively poor adhesion to the substrate, which means that surface tension effects in the nanowire overcome adhesion, allowing the observed hydrodynamic effects to occur.

In a Rayleigh-Plateau instability the wavelength for undulation in the wire diameter is linked to mass diffusion which sustains the instability. The hydrodynamic instability gives rise to a spectrum of modes with different wavelengths. There is a wavelength which grows fastest at the instability [6–8] which for a cylindrical wire (with isotropic surface energy) with radius R occurs for $\lambda_m = \sqrt{2}(2\pi R)$. The value of λ_m is dependent on a number of factors, including the mode of diffusion and isotropy/anisotropy of the surface energy. The observed undulation is thus expected to have a wavelength close to, λ_m although an exact match may not occur. For our case for instability onset occurs at a width of $\approx$50 nm. This corresponds to an effective radius $R_{eff} = \frac{circumference}{2\pi}$, so that for a wire of width of 50 nm and thickness = 50 nm, $R_{eff} = 32$ nm and the expected $\lambda_m = 282$ nm. The final fragmentation occurring at a width of $\approx$20 nm, $R_{eff} = 22$ nm, and we would expect $\lambda_m = 198$ nm. The observed average wavelength of $\sim$165 nm, though smaller, is close to the expected range of values. It is therefore proposed that the observed instabilities can are due to the Rayleigh-Plateau instability.

Since it involves mass transport, the Rayleigh-Plateau instability is often assisted by thermal processes which enhance the rate of mass diffusion [9,10]. These may include defects which can lower the activation energy and strains which can enhance mass migration and thus trigger the instability. Often instabilities in somewhat wider metal lines made by e-beam evaporation can be triggered by exposing them to ion beams leading to dewetting [11]. In our case, however, it is not clear what leads to the initiation of the instability; although ion-bombardment induced dewetting at the edges of the nanowire is a possible cause which should be investigated further.

To check the role of dewetting, a preliminary experiment was conducted using a 4 nm Cr adhesion layer on Si_xN_y membrane surface before the Au film was deposited. The film was then exposed to the same schedule of patterning using FIB. In this case, the instability was inhibited and the film began to fragment only when its width was below 15 nm. It is argued that the enhanced adhesion of the film due to the Cr layer inhibited the ion induced dewetting and delayed the onset of the Rayleigh-Plateau instability. The use of an adhesion layer to produce FIB-patterned nanowires with significantly reduced width is encouraging and will allow fabrication of stable ultra-narrow nanowires for a range of applications. These experiments are continuing and the results will be presented at length in a future publication.

5. Conclusions

An experimental investigation of the morphological stability of Au nanowires patterned using FIB on a silicon nitride membrane substrate has been completed. Below a certain width ($\sim$30–40 nm) the nanowires develop liquid like instabilities. The observed behaviour has been explained as manifestation of Rayleigh-Plateau instability which occurs below a critical width when the surface energy dominates. The use of an adhesion layer of Cr should inhibit the onset of the hydrodynamic instability, thereby allowing stable ultra-narrow FIB-etched nanowires to be fabricated for nanosensors and other applications; preliminary results support this assumption. Further work will investigate *inter alia* the effects of different substrates and doping elements on nanowire stability.

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