Experimental study of Rayleigh instability in metallic nanowires using resistance fluctuations measurements from 77K to 375K

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ABSTRACT

Nanowires with high aspect ratio can become unstable due to Rayleigh-Plateau instability. The instability sets in below a certain minimum diameter when the force due to surface tension exceeds the limit that can lead to plastic flow as determined by the yield stress of the material of the wire. This minimum diameter is given \( d_m \approx 2 \sigma_S/\sigma_Y \), where \( \sigma_S \) is the surface tension and \( \sigma_Y \) is the Yield force. For Ag and Cu we estimate that \( d_m \approx 15 \text{nm} \). The Rayleigh instability (a classical mechanism) is severely modified by electronic shell effect contributions. It has been predicted recently that quantum-size effects arising from the electron confinement within the cross section of the wire can become an important factor as the wire is scaled down to atomic dimensions, in fact the Rayleigh instability could be completely suppressed for certain values of \( k_F a_0 \). Even for the stable wires, there are pockets of temperature where the wires are unstable. Low-frequency resistance fluctuation (noise) measurement is a very sensitive probe of such instabilities, which often may not be seen through other measurements. We have studied the low-frequency resistance fluctuations in the temperature range 77K to 400K in Ag and Cu nanowires of average diameter \( \approx 15 \text{nm} \) to \( 200 \text{nm} \). We identify a threshold temperature \( T^* \approx 220 \text{K} \) for the nanowires, below which the power spectral density \( S_V(f) \sim 1/f \). As the temperature is raised beyond \( T^* \) there is onset of a new contribution to the power spectra. We link this observation to onset of Rayleigh instability expected in such long nanowires.

Keywords: resistance fluctuations, nanowire, Rayleigh instability, stability.

1. INTRODUCTION

The resistance fluctuation (noise) in a nanowire is an important issue both as a problem of basic physics and also as an important input for the feasibility of using them in nanoelectronic circuits as interconnects. It sets the limit to the best signal to noise ratio one can get in a practical device having such nanowires as components. The equilibrium white thermal noise (the Nyquist Noise) of a wire of resistance \( R \) kept at a temperature \( T \) can be estimated from the spectral power \( S_{th} \approx 4k_B T R \). However, in a current carrying nanowire a larger contribution is expected to arise from the “excess noise” or “resistance noise”. This can be appreciated if we do a rough calculation to estimate the noise in a nano system. Assume we have a sample which is a cube of side 10nm. The resistance fluctuation in this sample can be estimated using the empirical Hooge’s formula

\[
\frac{<\Delta R^2>}{R^2} = \int_{f_{min}}^{f_{max}} \frac{S_V(f) df}{V^2}
\]

where

\[
S_V(f) = \frac{\gamma_H V^2}{N_f}
\]

Taking the value of \( \gamma_H \) to be \( \approx 10^{-3} \) as is typical of a bulk metal and the bandwidth of operation to be from 1Hz to 1GHz, we get \( \sqrt{\frac{<\Delta R^2>}{R^2}} \approx 1.5\% \). For 1nm cube this value comes out to be \( \approx 5\% \) which sets the limit to best signal to noise ratio one can get in a practical device having such nanowires as components.

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Another important issue is the stability of these wires. A metallic nanowire acts essentially as an incompressible fluid of electrons, at least for simple monovalent metals, such as the alkali or noble metals. It is therefore expected that a sufficiently long cylindrical nanowire will break apart due to the Rayleigh instability. But long gold nanowires have experimentally been observed and appear to be very stable with lifetimes of the order of seconds. This stability has been theoretically shown to arise due to a competition of shell-effects (comparable to what happens in metal clusters) and surface tension producing a complex stability diagram, with regions of stability extending to very high temperatures for certain “magic” resistance values. On the stability boundary, the surface exhibits critical fluctuations, and the nanowire becomes inhomogeneous. Some stability fingers coalesce at higher temperatures, or exhibit overhangs, leading to reentrant behavior. The picture of the surface dynamics of metastable metal nanowires which emerges from theoretical analysis is that there is a separation of time scales: on short time scales, the surface oscillates rapidly about the cylindrical equilibrium shape (arising from the inertial dynamics of the ionic background and leading to acoustical phonons), while, on much longer time scales, surface atoms diffuse irreversibly (driven by a gradient in chemical potential produced by surface deformation).

The wires nevertheless become unstable below a certain minimum diameter (when the force due to surface tension exceeds the limit that can lead to plastic flow) \(d_m \approx 2\sigma_S/\sigma_Y\), where \(\sigma_S\) is the surface tension and \(\sigma_Y\) is the Yield force. For Ag and Cu using bulk values for the two quantities we estimate that \(d_m \approx 15\text{nm}\). In this paper we use low-frequency resistance fluctuations \((3\text{mHz} < f < 2\text{Hz})\) to investigate this instability in 15nm Ag and Cu nanowires. Low-frequency resistance fluctuation (noise) measurement is a very sensitive probe of such instabilities which often may not be seen through other measurements. Measurements of the fluctuation were carried out over the temperature range 77K to 400K. We make the important observation that there exists an instability temperatures \(T^* (\approx 220\text{K})\) for the nanowire at which the fluctuation becomes rather large and for \(T > T^*\) the spectral power deviates from the usual \(1/f\) contribution. Our observations, as explained later on, are related to the onset of Rayleigh-Plateau instability in the nanowire and have implications on the issue of their stability at room temperature.

### 2. INSTABILITY OF A WIRE OF FINITE DIMENSION

#### 2.1. Instability of a wire of finite radius - Rayleigh instability

We here summarize the main results of the work of Plateau and Rayleigh on the instability of cylindrical fluid jets bound by surface tension. Consider an infinitely long incompressible cylindrical fluid column of radius \(r_0\), density \(\rho\) and surface tension \(\sigma_S\). The influence of gravity is neglected. We consider the evolution of infinitesimal varicose perturbations on the surface of the cylinder. Let the radius at a distance \(z\) along the axis of the perturbed column be

\[
r(z) = r_0(1 + b\cos(kz))e^{-wt}
\]

where the perturbation amplitude \(b \ll 1\), \(w\) is the growth rate of the instability and \(k\) is the wave number of the disturbance in the \(z\)-direction. The corresponding wavelength of the varicose perturbations is \(2\pi/k\).

The dispersion relation, that indicates the dependence of the growth rate \(w\) on the wavenumber \(k\) is given by:

\[
w^2 = \frac{\sigma}{\rho r_0^3}kr_0 I_1(kr_0)I_0(kr_0)(1 - k^2r_0^2)
\]

We note that the perturbing modes become unstable when

\[
kr_0 < 1
\]

which is the Rayleigh criterion for the stability.

The maxima of the plot of \(w^2\) as a function of \(kr_0\) will give the fastest growing unstable mode. This value of \(k\) we denote as \(k_{\text{fast}}\). For the kind of deformation considered here this will happen for \(kr_0 = 0.697\), i.e. when the wavelength of the disturbance is

\[
\lambda_{\text{fast}} \approx 9.02r_0, k_{\text{fast}} = 0.697/r_0
\]
2.2. Instability of a wire of finite length

In this section we study the stability of a wire of a finite length \( l \) against the type of axially-symmetric perturbations considered in the previous section. (Only axially-symmetric perturbations can lower the surface energy of a cylindrical object, and thus lead to an instability.) We again write the deformation as a Fourier series

\[
r(z) = r_0(1 + b \cos(kz))
\]

where \( \lambda = 2\pi /k \) is the wavelength of the perturbation. The value of \( b \) is determined by the condition that the volume of the wire be conserved under such deformations. Using this condition yields:

\[
b = -\frac{8 \sin(kl)}{2kl + \sin(2kl)}
\]

The resistance of an infinitesimal element of the wire at \( z \) is

\[
dR(z) = \frac{\rho dz}{\pi r^2}
\]

Integrating over the length of the wire yields:

\[
\frac{R}{R_0} = \frac{2}{k'(1-b)^2} \left[ \frac{1}{a^3} \tan^{-1} \left( \frac{1}{a} \frac{k'}{2} \right) - \left( \frac{b}{a^2} \right) \frac{\tan(k'/2)}{a^2 + \tan^2(k'/2)} \right]
\]

where \( a = \sqrt{\frac{1+b}{1-b}} \) and \( k' = kl \) and \( R_0 \) is the resistance of the unperturbed wire. This is plotted in figure 2. The value of \( k \) (or \( k' \)) for which the wire is stable is bound by several physical considerations:

- For the wire not to break up into globules, the value of \( b \) must lie between +1 and -1.

\[
1 < b = -\frac{8 \sin(kl)}{2kl + \sin(2kl)} < 1
\]

\[
\Rightarrow k'_{\text{min}} \approx 2.6
\]

For our wire \( (l = 6\mu m) \) this implies \( k_{\text{min}} = 4.5 \times 10^4 m^{-1} \)

- From the quantization condition on \( k \)

\[
k_n = \frac{2\pi n}{l}
\]
where \( n \) is an integer with a maximum possible value \( n_{\text{max}} \sim k_F l / \pi \). This gives

\[
k_{\text{max}} = \frac{2\pi k_F l}{\pi} = 2k_F
\]

(14)

For Ag \( k_F = 1.2 \times 10^{10} m^{-1} \). This gives \( k_{\text{max}} = 2.4 \times 10^{10} m^{-1} \)

- Rayleigh criterion states that the wire will be unstable for values of \( k \) such that \( kR_0 < 1 \). For the 15nm wires this will imply that \( k \) values below \( \sim 6.67 \times 10^7 m^{-1} \) will destabilize the wire. Thus the value of \( k_{\text{min}} \) obtained above is not the real minimum value, Rayleigh instability pushes the value higher to \( k_{\text{min}} = 6.67 \times 10^7 m^{-1} \)

Thus the values of \( k \) for which the wire is stable lie between \( 6.67 \times 10^7 m^{-1} < k < 2.4 \times 10^{10} m^{-1} \). As discussed in the previous section, the fastest growing unstable mode is given by \( k_{\text{fast}} r_0 = 0.697 \) (eqn. 6). At room temperature for a 15nm diameter wire this will give \( k_{\text{fast}} = 4.65 \times 10^7 m^{-1} \). So for a 15nm Ag wire at room temperature the dominant varicose perturbation that tends to destabilize it has a wavenumber \( k_{\text{fast}} = 4.65 \times 10^7 m^{-1} \). The deformation in the wire produced by this perturbation will produce a relative resistance change of the order \( < \frac{\Delta R}{R} > \simeq 10^{-4} \) (see figure 2).

3. EXPERIMENTAL TECHNIQUES

The Ag nanowires of approximate length of 6\( \mu m \) and diameters in the range of 6nm to 200nm were grown within polymeric templates (etched track membranes) using electrochemical deposition from AgNO\(_3\) salt.\(^3\) (In this paper we concentrate on the data obtained for wires of diameter 15nm as this corresponds to the stability limit for both Ag and Cu as discussed earlier). The resistivity and noise measurements were carried out by retaining the wires within the polymeric membrane. On each of the two sides of the membrane two electrical leads were attached using silver epoxy. Thus our measurement is a pseudo-4-probe measurement. Though the measurements were made with the wires retained within the membrane, the system is an array of parallel nanowires where the individual wires are well separated by the insulating membrane.

The noise measurement was done using a digital signal processing (DSP) based a.c technique which allows simultaneous measurement of the background noise as well as the bias dependent noise from the sample.\(^5,6\) The measured background noise (bias independent) was white and was contributed by the \( 4k_B T R \) Nyquist noise. The
apparatus can measure a spectral power down to $10^{-20} V^2 Hz^{-1}$. We have used a transformer preamplifier SR554 to couple the sample to the lock-in-amplifier. The carrier frequency was chosen to lie in the eye of the Noise Figure (NF) of the transformer preamplifier to minimize the contribution of the transformer noise to the background noise.

The output low-pass filter of the Lock-in amplifier had been set at 3 msec with a roll off of 24 dB/octave. For a 3 msec time constant the output filter of the Lock-in amplifier with 24 dB/octave is flat to $f \leq 10$ Hz. This determines the upper limit of our spectral range. The output of the Lock-in amplifier is sampled by a 16 BIT A/D card and stored in the computer. At each temperature the data are taken by stabilizing the temperature with $\Delta T/T \approx 4 \times 10^{-3}$%. A single set of data are acquired typically for a time period of about fifty minutes or more at a sampling rate of 1024 points/sec. The complete data set of a time series at each temperature consisting of nearly 3 million points was decimated to about 0.1 million points before the spectral power density $S_V(f)$ is determined numerically. The frequency range probed by us ranges from 1 mHz to 10 Hz which allows us to probe time scales of the order of 15 msec to nearly 160 seconds. The frequency range is determined mostly by practical considerations. The lower frequency limit is determined by the quality of the temperature control. The sample resistance and the bridge output may also show a long time drift (in general such a long time drift is subtracted out by a least square fit to the data). Taking these factors into consideration the lower spectral limit in our experiment has been kept at 1 mHz.

4. RESULTS

The Ag nanowire array has a fairly linear resistivity ($\beta = \frac{1}{R} \frac{dR}{dT} \sim 4 \times 10^{-3}/K$) down to 75K and reaches a residual value below 50K with $\frac{\rho_{300K}}{\rho_{4K}} \sim 3$ (shown in figure 3a). $\beta$ is very close to the value seen for bulk Ag.

In figure 4(a) we show a plot the relative variance of the resistance fluctuation $\frac{\langle \Delta R^2 \rangle}{R^2} (\equiv \frac{1}{V^2} \int_{f_{min}}^{f_{max}} S_V(f) df)$ as a function of temperature for the 15nm Ag and Cu nanowires. For the Ag sample $\frac{\langle \Delta R^2 \rangle}{R^2}$ shows a prominent peak at $T = 220K$, we identify this as the instability temperatures $T^*$ as explained later in the text. (For the 15nm Cu nanowires $T^*$ is about 260K). For $T > T^*$ $\frac{\langle \Delta R^2 \rangle}{R^2}$ shows a shallow shoulder before beginning to rise again as the temperature is raised. To probe whether the features seen in the plot of $\frac{\langle \Delta R^2 \rangle}{R^2}$ are intrinsic to the sample or arise due to the confining effect of the polycarbonate membrane on the nanowires we have measured the noise in Ag wires of diameter 200nm grown inside polycarbonate templates in the same way. The $\frac{\langle \Delta R^2 \rangle}{R^2}$ obtained thus is plotted in figure 4(b). It can be seen that the effects that we are observing in the 15nm wires are completely absent in the wires of higher diameter. This shows that these effects arise purely due to size reduction of the wires and are not artifacts due the presence of the membrane. Figure 4(b) shows the $\frac{\langle \Delta R^2 \rangle}{R^2}$.
Figure 4. (a) Plot the relative variance of the resistance fluctuation $\frac{\langle \Delta R^2 \rangle}{R^2}$ as a function of temperature for the 15nm Ag and Cu nanowires. (b) Noise in Ag wires of diameter 200nm grown in polycarbonate templates and that measured on a Ag film (100nm thick).

The figure clearly shows the enhanced fluctuation in the 15nm wire occurring for $T \geq 220K$ in comparison to wires having a higher diameter or thin films.

Figure 5(a) shows a typical spectral power density of the resistance fluctuations for the 15nm Ag sample obtained at room temperature (the data in the Cu 15nm wire is similar to that obtained in the Ag wires). In figure 5(b) we show the spectral power as a function of $f$ at a few representative temperatures for the 15nm Ag wire. We find the appearance of new contributions to the power spectra of the 15 nm wire that makes it deviate rather appreciably from $1/f$ behavior for $T > T^\ast$. The 200nm wire and the film both have the spectral power density $S_V(f) \propto 1/f^\alpha$ with $\alpha \approx 1$ for all T. One can express the spectral power as $1/f^\alpha$ and the deviation from $1/f$ behavior can be expressed as a deviation of $\alpha$ from unity. A value of $\alpha$ close to unity generally indicates that the resistance fluctuation is arising due to the fluctuation of local defects and that there are no diffusion components present. The temperature variation of $\alpha$ for the 15nm Ag wire is shown in the inset of figure 5(b). $\alpha$ is close to 1 at low temperatures and increases gradually to $\approx 1.35 - 1.4$ at $T^\ast$ and stays at that level till the highest temperature measured.

5. DISCUSSIONS

Thus our main experimental observations for the 15nm nanowires (both Ag and Cu) can be summarized as:

- There is a jump in the relative resistance fluctuation $\frac{\langle \Delta R^2 \rangle}{R^2}$ at a temperature, (which we denote by $T^\ast$).
- The spectral power density $S_V(f)$ deviates from $1/f$ dependence in the vicinity of $T^\ast$.

These seem to point to the fact that the nanowire has significant change in resistance fluctuation at temperatures around $T = T^\ast$. The instability that sets in at $T^\ast$ leads to an rms fluctuation (in the bandwidth of our measurement) of $\approx 30$ppm. While this is not large enough to cause a large change in the resistance, it can nevertheless be detected by very precision resistance measurements. On thermal cycling from low T upto 220K the observed noise as well as the resistance data are reproducible. However a thermal cycling beyond $T^\ast$ leads to small systematic changes in the noise as well as resistivity ($\approx 1 - 2\%$) implying that beyond $T^\ast$ instability does set in.

We propose that the observed resistance fluctuation is a manifestation of the Rayleigh-Plateau instability. As pointed out before, the critical diameter at which instability sets in for both Cu and Ag is about 15nm. This instability of the wire can lead to long wavelength fluctuations in the radius of the wire. Depending on various factors, the instability may either lead to break down of the wire into smaller droplets or it can lead to long lived...
fluctuations without droplet formation. We believe that the second case holds here because the wire diameter is same as the critical diameter 15nm. As calculated previously in this paper, for a wire of diameter 15nm the contribution of the fastest growing fluctuation mode (this will also be the most dominant one as the instability grows exponentially with time) to the resistance fluctuation is

\[ \langle (\Delta R R)^2 \rangle \simeq 10^{-8} \] (15)

Our experimentally obtained number

\[ \langle (\Delta R R)^2 \rangle \simeq 3 \times 10^{-9} \] (16)

is very close to this value.

As mentioned earlier in this paper, it has been proposed that the dynamics of the instability has two components -

1. occurring in short time scales due to surface phonons where the surface of the wire oscillates rapidly along an equilibrium shape

2. occurring at much longer time scale which has a diffusive component arising from surface diffusion of atoms driven by a gradient in chemical potential produced by surface deformation.\(^2\) This long wavelength fluctuation in wire radius can lead to a low frequency fluctuation in the resistance of the wire.

**Figure 5.** (a) Typical spectra obtained at room temperature for the 15nm and 200nm Ag samples. (b) The spectral power density (after subtracting out the measured background noise and multiplying by the frequency) as a function of \( f \) at a few representative temperatures for the 15nm wire. We have plotted the spectral power density as \( f S_V(f)/V^2 \) in order to accentuate the deviation from the \( 1/f \) dependence.
The slow (low-frequency) fluctuation that our experiment investigated can arise from this diffusive part. We also find that there is a large deviation from the $1/f$ spectral dependence at low-frequencies with the measured value of $\alpha \to 3/2$ near $T = T^*$. This is expected for diffusive motion which gives rise to a component $S_V(f) \simeq 1/f^{3/2}$.

The existence of the threshold $T^*$ is a consequence of this diffusive process, because this is likely to be an activated process and only at a threshold temperature can there be sufficient diffusion that can sustain this process and make a dominant and observable contribution to the fluctuation.

To summarize, the Ag nanowire of diameter 15 nm shows large resistance fluctuation in a certain temperature region. The fluctuation has a large low-frequency component that is not the usual $1/f$ noise. The measured resistance fluctuation can be interpreted as arising due to as shape fluctuation of the wire as a consequence of Rayleigh-Plateau instability and is, as expected, absent in wires of larger diameter. The magnitude of the noise matches to within 2-3 times the noise calculated for a Ag wire of the same diameter undergoing shape fluctuations. Our observation has impact in the context of stability of nanowires at or above 300K. It raises a fundamental question on use of nanowires as interconnects in nanoelectronics.

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