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Observation of large low-frequency resistance fluctuations in metallic nanowires: Implications on its stability

Aveek Bid,[1,*] Achyut Bora,[1] and A. K. Raychaudhuri[1,2,†]
[1]*Department of Physics, Indian Institute of Science, Bangalore 560 012, India*
[2]*Unit for Nanoscience and Technology, S.N. Bose National Centre for Basic Sciences, Salt Lake, Kolkata 700098, India*

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We have measured the low frequency ($1\text{ mHz} \leqslant f \leqslant 10\text{ Hz}$) resistance fluctuations in metallic nanowires (diameter 15 nm to 200 nm) in the temperature range 77 K to 400 K. The nanowires were grown electrochemically in polycarbonate membranes and the measurements were carried out in arrays of nanowires by retaining them in the membrane. A large fluctuation in excess of conventional $1/f$ noise which peaks beyond a certain temperature was found. The fluctuations with a significant low frequency component ($\simeq 1/f^{3/2}$) arise when the diameter of the wire $\simeq 15$ nm and vanish rapidly as the diameter is increased. We argue that Rayleigh-Plateau instability is the likely cause of this excess noise.

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The resistance fluctuation (noise) in a nanowire is an important issue both as a problem of basic physics and 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 thermal noise and the $1/f$ noise are the known sources of noise in a metallic system. At very low temperatures shot noise also makes a contribution. The equilibrium white thermal noise (the Nyquist noise) of a wire of resistance R at a temperature T can be estimated from the spectral power density $S_{th} \approx 4k_BTR$. However, in a current carrying nanowire a larger contribution (of $1/f$ type spectral power density) is expected to arise from the "conductance" or "resistance" noise. There is no *a priori* way to estimate the $1/f$ noise, it needs to be experimentally measured. In this report we measure low-frequency resistance fluctuations ($1\text{ mHz} < f < 10\text{ Hz}$) in arrays consisting of Ag and Cu nanowires with diameters down to 15 nm for $77\text{K} \leqslant T \leqslant 400$ K. In addition to the usual thermal and $1/f$ noise there are additional sources of resistance noise in metallic nanowires when the diameter is reduced down to 15 nm. These are not present in wires of larger diameter grown by the same method. We find that the source of this extra noise can be traced to Rayleigh instability[1–3] which is expected to occur in systems with large aspect ratio. This extra noise has important implications on stability of the wire. This instability, if large, can ultimately lead to break down of the wire.[4] To our knowledge, this is the first time that this particular issue has been investigated by measurement of electrical noise in nanowires of this dimension.

The Rayleigh-Plateau instability sets in when the force due to surface tension exceeds the limit that can lead to plastic flow. This occurs at a diameter $< d_m \approx 2\sigma_S/\sigma_Y$, where σ_S is the surface tension and σ_Y is the yield force.[3] For Ag and Cu (using bulk values for the two quantities) we estimate that $d_m \approx 15$ nm. We find that low-frequency resistance fluctuation (noise) measurements, being a very sensitive probe, can observe the instabilities, even though they are small and may not be seen through other measurements.

The Ag and Cu nanowires of approximate length of 6 μm and diameters 15 nm, 20 nm, and 200 nm were grown within polycarbonate templates (etched track membranes) from $AgNO_3$ and $CuSO_4$ (Refs. 5 and 6), respectively, using electrochemical deposition. During the growth one of the electrodes was attached to one side of the membrane and the other electrode was a microtip (radius of curvature $\simeq 100$ μm) carried by a micropositioner. This can be placed at a specific area on the membrane and thus the growth can be localized. The wires grow by filling the pores from end to end and as soon as one or more wires complete the path from one electrode to the other the growth stops. The wires can be removed from the membranes by dissolving the polymer in dichloromethane. The wires were characterized by x-Ray, SEM, and TEM. A typical image of a 100 nm wire is shown in Fig. 1 along with the XRD data. The XRD data matches well with the data taken on a bulk silver sample. We find that typical samples (arrays) on which the measurements were done had 2 to 50 wires. It is very difficult to estimate the exact number of wires that actually grow from end to end. One can use the methods outlined in Ref. 7. We find that the estimate made by these methods can be uncertain by a factor of 2. As a result we do not use the exact number of wires in any of our calculation.

The noise measurement was carried out using a digital

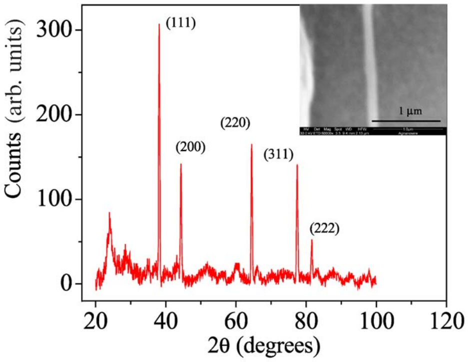

FIG. 1. (Color online) XRD of the 15 nm Ag nanowires. The inset shows the SEM image of a 100 nm wire.

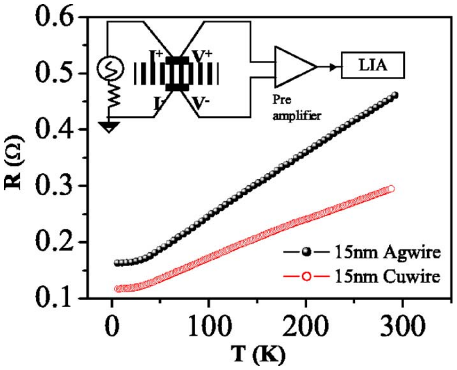

FIG. 2. (Color online) Resistance of the 15 nm Ag and 15 nm Cu samples as a function of temperature. The inset shows a cartoon of the measurement scheme.

signal processing (DSP) based ac technique (using a lock-in amplifier) which allows simultaneous measurement of the background noise as well as the bias dependent noise from the sample.[8,9] The apparatus was calibrated down to a spectral power density $S_V(f)=10^{-20}V^2/\mathrm{Hz}$ by measuring the $4k_BTR$ Nyquist noise at a known temperature T of a calibrated resistor. We have used a transformer preamplifier to couple the sample to the lock-in amplifier as shown in the inset of Fig. 2. 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 noise appears as the side bands of the carrier frequency. The output of the lock-in amplifier is sampled at a rate of 1024 points/s by a 16 bit A/D card and stored in the computer. This forms the time series (consisting of nearly 3 million points) from which the spectral power density $S_V(f)$ is obtained by using FFT. The relative variance of the resistance fluctuation $\langle(\Delta R)^2\rangle/R^2$ within the detection bandwidth ($f_{\min}\rightarrow f_{\max}$) was obtained by integrating the power spectrum $\equiv(1/V^2)\int_{f_{\min}}^{f_{\max}}S_V(f)df$. The lower frequency limit of 1 mHz is determined by the quality of the temperature control which is within ±40 ppm. The output low-pass filter of the lock-in amplifier had been set at 3 ms with a roll off of 24 dB/octave which has a flat response for $f\leqslant 10$ Hz. This determines the upper limit of our spectral range.

The resistivity and noise measurements were carried out by retaining the wires within the membrane and the contacts were made with silver epoxy. Though the measurements were made with the wires retained within the membrane, the individual wires are well separated by the insulating walls of the pores in which they were grown. Each wire thus acts as an independent source of noise.The temperature variation for the resistivity measurements were done down to 5 K in a bath-type liquid helium cryostat and the noise was measured down to liquid nitrogen temperature in a similar cryostat.

We note that contact can be a serious issue in noise measurements and we carried out a number of checks to rule out any predominant contribution of the contact to the measured noise. In addition to silver epoxy contacts we also used evaporated silver films and Pb-Sn solder to make contact. We find that they give similar results to within ±15%. In case of the Pb-Sn contact the change in R of the sample as we go below the superconducting transition temperature ($\sim$7 K) of the solder is negligibly small ($<2\%-3\%$) implying a small contribution of the contact to the total R measured. We have also measured the noise in different samples with varying numbers of wires in them. The resistance of the array, due to different numbers of wires in them vary. Also such samples will have significantly different contact areas and hence different contact noise, if any. Yet we find that the normalized noise $\langle(\Delta R)^2\rangle/R^2$ in different arrays of the same diameter wire lie within ±15%. All these tests rule out any predominant contribution from the contacts.

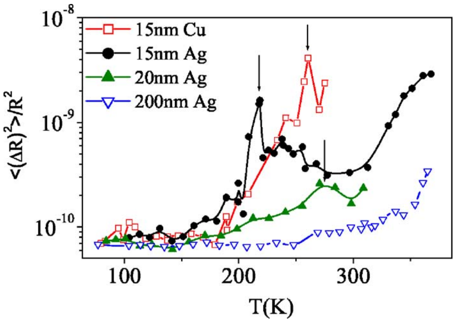

FIG. 3. (Color online) The relative variance of the resistance fluctuation $\langle(\Delta R)^2\rangle/R^2$ as a function of temperature for the 15 nm, 20 nm, 200 nm Ag and 15 nm Cu nanowires.

The 15 nm Ag and Cu nanowire arrays have a fairly linear temperature dependence of resistance down to 75 K and R reaches a residual value below 50 K with residual resistivity ratio (RRR)($\rho_{300\ \mathrm{K}}/\rho_{4.2\ \mathrm{K}}$)$\sim 3$ for both the 15 nm wires (shown in Fig. 2). If the mean free path l_e is significantly small due to disorder, then $k_Fl_e\approx 1$ (k_F=Fermi wave vector) and the wires would show an upturn in resistivity at low temperatures due to electron localization. The absence of upturn at low temperature rules out significant disorder in the system which can give rise to such effects as localization. Another parameter that points to the quality of the wires grown is the temperature coefficient of resistivity $\beta=1/R(dR/dT)$ as the presence of a large degree of structural imperfections and impurity can reduce β significantly. For the wires grown β lies in the range $\sim 4\times 10^{-3}$ K and $\sim 2.5\times 10^{-3}$ K at 300 K. For high purity bulk Ag and Cu $\beta\simeq 3.8\times 10^{-3}$ K and 3.9×10^{-3} K, respectively,[10] at 300 K.

In Fig. 3 we show a plot of $\langle(\Delta R)^2\rangle/R^2$ as a function of T. The spectral power density $S_V(f)$ is $\propto V^2$. The figure clearly shows the enhanced fluctuations in the 15 nm wires occuring for $T>200$ K in comparison to wires having a larger diameter. Absence of a large noise in the 200 nm wires establish that the observed features are intrinsic to the 15 nm wire and that they do not arise due to the presence of the polycarbonate membrane. In both the 15 nm Ag and the Cu nanowires, $\langle(\Delta R)^2\rangle/R^2$ increases as T is raised and shows a prominent peak at $T=T^*$ (indicated by arrows in Fig. 3), where T^* is $\approx$220 K for the Ag nanowire and $\approx$260 K for the Cu wire. In the 20 nm wire the fluctuations, though distinctly smaller than that seen in 15 nm wire, show a shallow peak at T^* $\simeq$275 K. The observed behavior are reproducible and for different samples the variablity in the data is within experi-

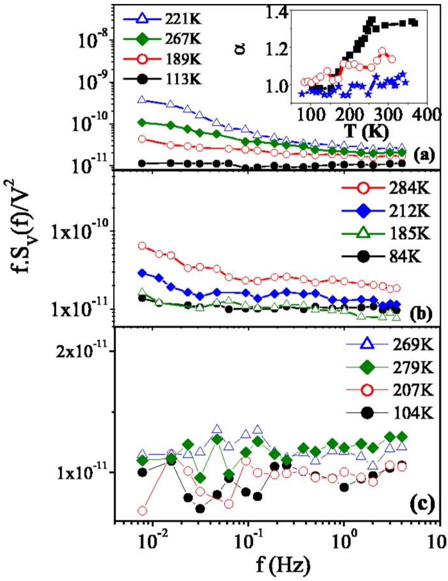

FIG. 4. (Color online) The spectral power density (after subtracting out the measured background noise and multiplying by the frequency) as a function of f at a few representative temperatures for the (a) 15 nm wire, (b) 20 nm wire, and (c) 200 nm wire. We have plotted the spectral power density as $fS_v(f)/V^2$ in order to accentuate the deviation from the $1/f$ dependence. The inset of the 4(a) shows the temperature dependence of the measured α for the wires (filled square, 15 nm; open circle, 20 nm; and filled star, 200 nm wire).

mental errors of about 15%. For $T > T^*$, $\langle(\Delta R)^2\rangle/R^2$ shows a shallow shoulder before beginning to rise again. The rise in the fluctuation beyond 300 K are qualitatively similar in all the samples. The rise is observed even in films of thickness $\simeq 100-200$ nm.[12]

In Fig. 4(a) we show $fS_V(f)/V^2$ as a function of f at a few representative temperatures for the 15 nm Ag wire. The $S_V(f)$ follows the relation $S_V(f) \propto 1/f^{\alpha}$. The data have been plotted as $fS_V(f)/V^2$ in order to accentuate the deviation of α from 1. The temperature variation of α for the sample is shown as an inset. Similar data were also obtained for the Cu wires. At $T < T^*$, $\alpha \approx 1$ while for $T \geqslant T^*$ we find that α deviates from unity. For the 15 nm wire α increases gradually to $\approx 1.35 - 1.4$ at T^* and stays at that level until the highest temperature measured. For the 20 nm Ag wire also $\alpha \approx 1$ for $T < 200$ K and increases to a value $\alpha \approx 1.2$ at T^*, while for the 200 nm wire $\alpha \approx 1$ for the complete temperature range covered. One thus sees that the spectral decomposition of the large noise present in the 15 nm wire is qualitatively different from that present in the 200 nm wire. The fluctuations in 15 nm wires are not only large but also have a large low frequency component.

The results indicate that the nanowires of diameter 15 nm have significantly different resistance fluctuations. There is a jump in $\langle(\Delta R)^2\rangle/R^2$ at a certain temperature (which we denote by T^*) and $S_V(f)$ deviates from the usual $1/f$ dependence for $T > T^*$. The nature of the $1/f$ noise in the 200 nm wire is qualitatively similar to that of a Ag film and this we think is the usual $1/f$ noise in the metallic conductors which can generally be explained by the Dutta-Horn model which gives $\alpha \approx 1$.[11] The extra fluctuation observed in the 15 nm wires at the temperature range around T^* and above is thus expected to arise from a different origin.

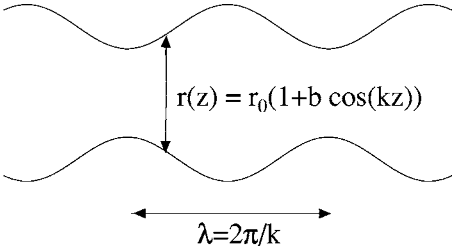

FIG. 5. Schematic of the model used to estimate the resistance fluctuation arising due to a volume conserving fluctuation in the radius of a wire of finite length.

We propose that the extra fluctuation is a manifestation of the Rayleigh-Plateau instability. The critical diameter d_m at which instability sets in for both Cu and Ag is about 15 nm. To quantitatively estimate the extent of the fluctuation, we make a model shown in Fig. 5, where the wire has a volume conserving fluctuation of wave vector k in the z direction. The radius of the wire at a distance z along its axis will be given by

$$r(z) = r_0[1 + b\cos(kz)] \tag{1}$$

where the perturbation amplitude $b \ll 1$ and r_0 is the radius of the unperturbed wire. For a volume preserving fluctuation k and b will be related by the relation

$$b = -\frac{8\sin(kl)}{2kl + \sin(2kl)}, \tag{2}$$

where l is the length of the wire. The fluctuation in radius will lead to a fluctuation in the cross section of the wire which in turn will lead to a fluctuation in the resistance which the measurement picks up. We can calculate the resistance R of a wire of length l whose radius $r(z)$ follows relations (1) and (2) and this is given by

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

where $a = \sqrt{(1+b)/(1-b)}$ and $k' = kl$. R_0 is the resistance of the unperturbed wire of radius r_0. We note that following the Rayleigh criterion[1] for the stability, the perturbing modes become unstable when $kr_0 < 1$, and the fastest growing unstable mode occurs for $kr_0 = 0.697$. Using these numbers we can estimate the resistance fluctuation from Eq. (3). We get an estimate of $\langle(\Delta R)^2\rangle/R^2 \simeq 10^{-8}$ for the 15 nm wire Ag wire at 300 K and this compares rather well with 3×10^{-9} observed experimentally within the bandwidth of our experiment. Our simple estimate thus suggests that the extra noise could be linked to the Rayleigh-Plateau instability. The fact that the extra noise appears when the diameter $\approx d_m$ and rapidly goes down even for the 20 nm wire where the diameter $\approx 1.3 d_m$, strongly points to this mechanism for the extra noise.

The instability of the wire which leads to long wavelength fluctuations in the radius of the wire may either lead to a break down of the wire into smaller droplets or can lead to long-lived fluctuations without droplet formation. We believe that the second case holds here because the wire diameter (15 nm) is the same as the critical diameter d_m. It appears that the electronic contribution to the surface energy can lead to a reduction of the fluctuation arising from the Rayleigh instability.[2] Theoretically it has been found that due to the electronic contribution the wire (depending on the factor $k_F r_0$) can have temperature ranges where it is stable and yet can have islands of instabilities.[3]

The power spectrum of the extra fluctuation deviates significantly from the conventional $1/f$ noise due to low frequency components. This low frequency component can extend over a range of frequency. However, what we measure in the experiment is limited by the bandwidth of our experiment. The Rayleigh instability is sustained by surface diffusion which is driven by a gradient in chemical potential produced by surface deformation. Diffusion generally contributes a spectral power with $S_V(f) \simeq 1/f^{3/2}$.[12,13] The observation that for $T \geqslant T^* \ \alpha \rightarrow 3/2$ would suggest existence of such modes. It is thus likely that the low frequency dynamics seen in the fluctuation arises from this diffusion process.

To summarize, we find that in metallic Ag and Cu nanowires of diameter 15 nm there are large low frequency resistance fluctuations beyond a certain temperature. The fluctuations differ in characteristics from those usually found in wires of larger diameters. The fluctuation is not the usual $1/f$ noise. It is proposed that the extra fluctuation originates from Rayleigh instability. The measured resistance fluctuation was found to have a magnitude similar to that estimated from a simple model of a wire exhibiting volume preserving fluctuation. The extra noise has a strong dependence on the wire diameter and it makes a sharp appearance when the diameter of the wire approaches the critical value. Our observation has impact in the context of stability of nanowires at or above 300 K. It raises a fundamental issue on the use of nanowires as interconnects in nanoelectronics.

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*Electronic address: avik@physics.iisc.ernet.in
†Electronic address: arup@bose.res.in

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