Finite-Size Scaling in Silver Nanowire Films: Design Considerations for Practical Devices

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Supplementary Information

Percolation Simulations

The simulations used in this work are based on the algorithm of Li and Zhang[1]. The simulation domain is sub-divided into a set of boxes which are occupied by a nanowire with a fixed probability. For each box, intersections with other nanowires are checked by indexing boxes within a kernel (related to the length of the nanowire in that box). If only a sub-region of the domain is to be considered when calculating the network resistance, then nodes in the graph of the network are discarded based on their intersection coordinates. Figure S1 shows an example of the system geometry for a low density network over a sub-region of the domain.

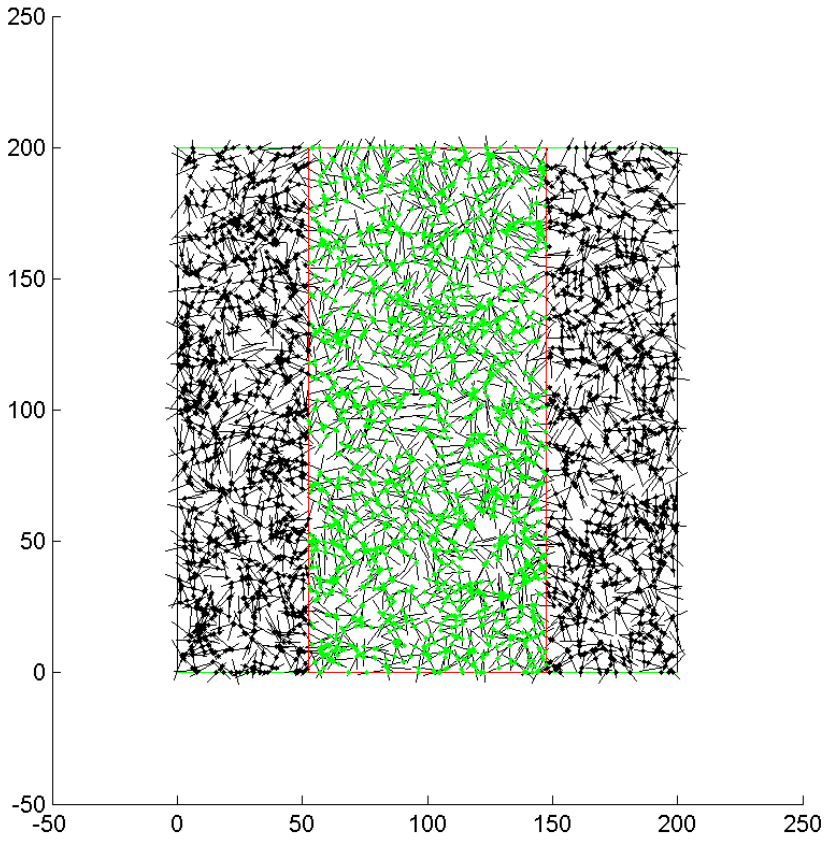

Figure S1: Example of the network structure in a simulation of a narrow track. The nanowire junctions included in the resistance calculation are shown in green, and those excluded are shown in black. The network resistance is calculated between the top and bottom edges of the domain. Units are in μm.

Once all of the nanowire intersections are determined, then an Incidence matrix representation U of the network is constructed, along with an associated weight matrix W. The Incidence matrix is an $n \times m$ matrix (where the network contains n nodes and m edges) which represents the (arbitrary) direction of each edge incident on a given node. The weight matrix is an $m \times m$ diagonal matrix with the weight (or conductivity) $w_{ij} = 1/r_{ij}$ of each edge (joining nodes i and j) along the main diagonal. This representation is related to the Laplacian matrix $\mathcal{L}$ by;

$$\mathcal{L} = D - A = UWU^T$$

where D is the degree matrix (a diagonal matrix of the sum of weights of edges incident on each node) and A is the adjacency matrix (where the entry $A(i,j) = w_{ij}$; the edge weight if an edge connects nodes i and j, or 0 otherwise).

The resistance of the system between two arbitrary nodes a and b can in principle be calculated two different ways. The standard method is by solving Kirchhoff's laws over the network by using the Moore-Penrose pseudoinverse of the Laplacian, $\mathcal{L}^+$, and calculating the total power dissipation P (which is related to the effective network resistance, R_{eff});

$$\mathcal{L}v = i$$

$$P = i_{ext}^2 R_{eff} = v^T i = v^T \mathcal{L} v = v^T U W U^T v$$

$$v = \mathcal{L}^+ i$$

where v is a vector of node potentials, and i is a vector containing the net currents flowing into each node (0 for all except for the source $i_a = i_{ext}$ and sink $i_b = -i_{ext}$). i_{ext} is the externally supplied current to the network (if taken to be unitary, then the case simplifies to $P = R$). Solving for the vector v is best done using an iterative algorithm, rather than directly evaluating the pseudoinverse $\mathcal{L}^+$ (since, even if $\mathcal{L}$ is sparse $\mathcal{L}^+$ is structurally dense).

An alternative method due to Bapat[2] is used in this work. This method relies on the calculation of the determinants of two sub-matrices of the Laplacian. Due to the very sparse nature of the matrix representation of this system, this allows for significant optimisation over the previously described algorithm. The notation $\mathcal{L}(i|j)$ is used to refer to the sub-matrix of $\mathcal{L}$ with the *i*th row and *j*th column removed. Bapat shows that the effective network resistance between nodes a and b can then be described as;

$$R_{eff} = \frac{\det \mathcal{L}(a, b|a, b)}{\det \mathcal{L}(a|a)}$$

Outlier Removal from Small Statistical Samples

The removal of statistical outliers from the data sets presented is challenging due to the small number of observations. This makes traditional methods such as pruning values with deviations from the mean of more than several times the standard deviation of the whole data set difficult, since the outliers significantly increase the standard deviation. We have developed and applied a simple method based on the use of a Chi-squared test statistic.

The set of n samples x are ordered according to their square deviation from the mean $\bar{x}$;

$$\bar{x} = \frac{1}{n}\sum_{i=1}^{n} x_i$$

Next we calculate the statistic

$$\chi^2 = \sum_{i=1}^{n-1} \left(\frac{x_i - \bar{x}}{\bar{x}}\right)^2$$

Note the exclusion of the nth value; i.e. the sample with the largest square deviation. If this statistic is greater than one, then the nth value is discarded from the set and the procedure repeated. This test is based on querying the likelihood that the nth sample is drawn from the same distribution as the other $n - 1$ samples. Figure S2 shows an example of this processing applied to two data sets; one with a larger number of outliers, and another with no outliers (to illustrate that the algorithm doesn't prune data points unnecessarily).

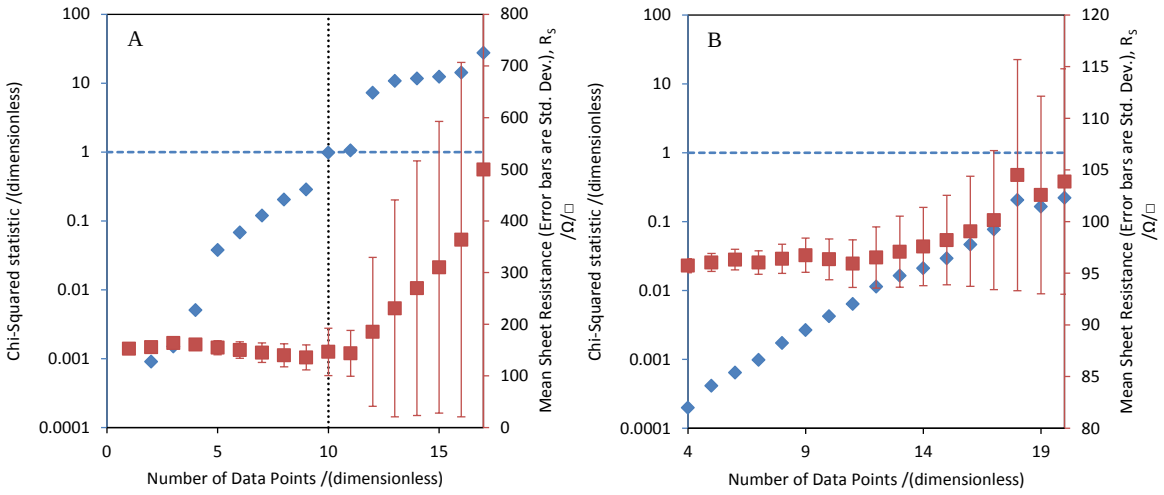

Figure S2: Examples of application of the data pruning algorithm to two data sets. (A) has a large number of outlier points; the points with the highest squared deviation from the mean are successively pruned (from right to left). (B) Sample with no outliers (since the initial value of $\chi^2 < 1$).

Application of Finite-Size Scaling to Silver Nanowire Films

Equation (1) is framed in terms of lattice percolation. However we know from the study of Li and Zhang[1] that there is good evidence that the finite-size scaling function X is universal to both lattice and continuum percolation models. As such, a change of variables should allow similar analyses on both simulation data and experimental measurements, within the FSS theory framework.

As shown in Equation (2), we first identify the percolation parameter of interest σ as $1/R_S$. For continuum percolation of rods, a typical density metric is that of the filling factor[3] η (defined as the total squared length of rods per unit area of the domain). From the work presented previously by the authors[4], we establish that the term $\eta/\eta_C - 1$ is equivalent to $\log T/\log T_C - 1$. Lastly, for the lattice case the domain dimension L is normalised to the lattice spacing; for the continuum

case considered by Li and Zhang[1], the (uniform) length of the rods was used to normalise L. For the present case we do not have rods of uniform length, and therefore must normalise the domain dimension to a statistic of the length distribution (e.g. the average length $\langle l \rangle$). We have found by a process of trial and error (considering the moments of the length distribution which appear in the expressions used to derive Equation (3)[4]) that a suitable substitution is;

$$L \to w \frac{\langle l \rangle}{\langle l^2 \rangle}$$

This leads to the form of Equation (2) presented. Figure S3 shows a plot of sample experimental data at two different mixing ratios of short (sonicated) nanowires with the rescaled parameter;

$$\ln\left[\frac{1}{R_S(T,w)}\left(w\frac{\langle l \rangle}{\langle l^2 \rangle}\right)^{\frac{\mu}{\nu}}\right]$$

Plotted against the parameter

$$\ln\left[\left(\frac{\log T}{\log T_C} - 1\right)\left(w\frac{\langle l \rangle}{\langle l^2 \rangle}\right)^{\frac{1}{\nu}}\right]$$

As is evident, the large-x trends for the two data sets are different. This is likely due to a variation in the average junction resistance between nanowires (R_j). It is established that the scaling exponent μ depends both on the ratio of the rod resistance (between junctions) and the junction resistance (between rods)[5], and on the breadth of the distribution of values of R_j [6]. As such, it is sensible to subtract this large-x trend from each data set, and aggregate the residuals.

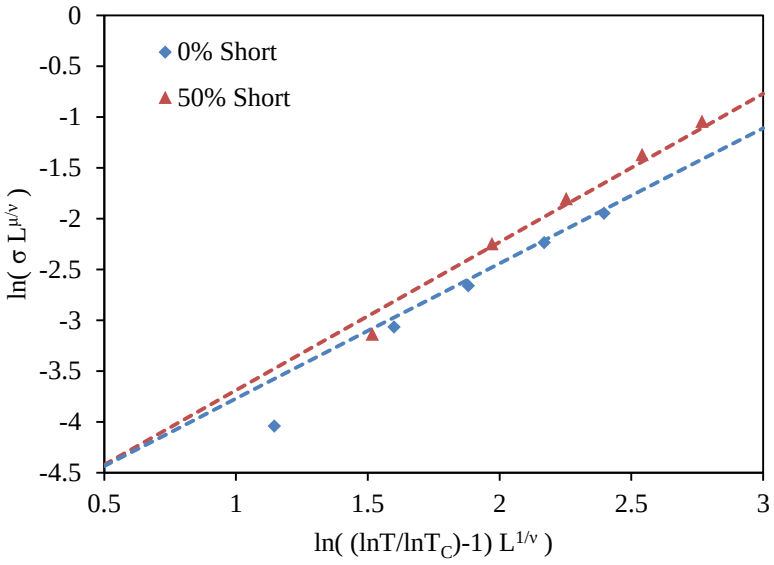

Figure S3: Experimental R-w data for two different length mixing ratios plotted according to the FSS Theory description. As can be seen, the two "infinite film" limits (dotted lines) are different. This is primarily due to variation of the percolative exponent μ.

The large-x trend represents the "infinite film" case of the percolative behaviour;

$$\frac{1}{R_S} = \frac{1}{M'}\,(\eta - \eta_C)^{\mu}$$

This expression can be rearranged into the form $y = mx + c$ (for the graph of Figure S3);

$$\ln\left[\frac{1}{R_S(T,\infty)}\left(w\frac{\langle l\rangle}{\langle l^2\rangle}\right)^{\frac{\mu}{\nu}}\right] = \mu \ln\left[\left(\frac{\log T}{\log T_C} - 1\right)\left(w\frac{\langle l\rangle}{\langle l^2\rangle}\right)^{\frac{1}{\nu}}\right] + \ln\left[\frac{\eta_C}{M'}\right]$$

As we can see, the gradient of the plot corresponds to the percolative exponent of the sheet resistance, and the intercept gives information regarding the material constant M'. Subtraction of this trend from our dataset for $R_S(T,w)$ on the log-log scale corresponds to division of the arguments of the logarithm terms;

$$\ln\left[\frac{1}{R_S(T,w)}\left(w\frac{\langle l\rangle}{\langle l^2\rangle}\right)^{\frac{\mu}{\nu}}\right]-\ln\left[\frac{1}{R_S(T,\infty)}\left(w\frac{\langle l\rangle}{\langle l^2\rangle}\right)^{\frac{\mu}{\nu}}\right]=\ln\left[\frac{R_S(T,\infty)}{R_S(T,w)}\right]$$

which is the form of the data plotted in Figure 4(A).

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