Localization lengths of ultrathin disordered gold and silver nanowires

A. T. Costa Jr.
Departamento de Ciências Exatas, Universidade Federal de Lavras, 37200-000 Lavras, MG, Brazil

R. B. Muniz
Instituto de Física, Universidade Federal Fluminense, 24210-340 Niterói, RJ, Brazil

Jisang Hong and R. Q. Wu
Department of Physics and Astronomy, University of California, Irvine, California, 92697 U.S.A.

The localization lengths of ultrathin disordered Au and Ag nanowires are estimated by calculating the wire conductances as functions of wire lengths. We study Ag and Au monoatomic linear chains, and thicker Ag wires with very small cross sections. For the monoatomic chains we consider two types of disorder: bounded random fluctuations of the interatomic distances, and the presence of random substitutional impurities. The effect of impurity atoms on the nanowire conductance is much stronger. Our results show that electrical transport in ultrathin disordered wires may occur in the strong localization regime, and with relatively small amounts of disorder the localization lengths may be relatively small. The localization length dependence on wire thickness is investigated for Ag nanowires with different impurity concentrations.

Ultrathin metallic nanowires with decreasing diameters have been fabricated by ingenious methods. Wires with average cross sections containing just a few atoms have been reported recently. One extreme case is the remarkable manipulation of single add-atoms on surfaces using a scanning tunneling microscope, which allows wires to be built with atomic control of its composition. These ultrathin nanowires are very important for nanoelectronics. Not only they provide metallic contacts of nanoscopic dimensions, but the electronic confinement in one dimension substantially modifies the electrical characteristics, and offers exciting perspectives for both basic and applied physics.

Most of the currently available techniques for manufacturing ultrathin metallic nanowires produce structures with considerable amount of inhomogeneities. Disorder effects in such systems strongly influences the electrical transport, and may lead to localization. Under certain circumstances, they can reduce the average conductance $\bar{g}$ very effectively, making it fall off exponentially with the wire length $\ell$, for sufficiently long wires. The localization length $\Lambda$ is determined by the asymptotic decaying rate of $\bar{g}$ with $\ell$; more precisely, $\Lambda^{-1} = -\bar{g}^{-1}\partial \bar{g}/\partial \ell$ for large values of $\ell$. There are at least two additional characteristic lengths, relevant for discussing electrical transport in ultrathin nanowires, the electronic mean free path $\lambda$, and the average wire thickness or cross section diameter $\bar{d}$. For relatively short nanowires with $\ell \ll \lambda$, the transport is ballistic, whereas for sufficiently long wires with $\ell \gg \Lambda$, it becomes strongly localized. Clearly, both $\lambda$ and $\Lambda$ depend upon the amount and type of inhomogeneities present in the system, and they also vary with $\bar{d}$.

Electrical transport through ordinary metallic structures are usually far from the localization regime. On the other hand, non-interacting disordered low-dimensional systems (with dimensionality $\leq 2$) in the thermodynamic limit have localized states only. They are, strictly speaking, insulators at zero temperature. Localization effects were observed in very thin and short gold nanowires, and a remarkably small localization length ($\Lambda \leq 40 \text{ A}$) was estimated in such experiments. Electrical conductance through nanocontacts between various metallic systems, however, exhibits ballistic behavior.

Although some degree of disorder is clearly present in these highly constricted junctions, it is mainly effective on a relatively short length scale (of the order of a few nanometers). Even when disorder is sufficiently strong for localization to take place, the localization length needs to be compared with the dimensions of the disordered system, especially when the latter are relatively small. The observation of ballistic transport across those nanocontacts indicates that either the disorder is rather weak, or the localization length is much larger than the junction sizes.

The ability to fabricate thinner and longer disordered nanowires naturally brings up the issue of localization. Here we address the following questions: what is the order of magnitude of the localization length in disordered Ag and Au nanowires? How does the localization length vary with the type and degree of disorder that occur or may be introduced in these systems, and how does it depend upon the wire average diameter? Such estimates are particularly important when one considers the possibility of using these ultrathin metallic wires in devices, and exploiting the quantum character of electronic transport in such systems. In fact, disorder may be purposefully introduced to produce a desired effect. It has been recently shown, for example, that electrical current flowing through nanowires made of ferromagnetic disordered alloys can become highly spin-polarized.

Behind this theoretical proposition, however, lies the hypothesis that the spin diffusion length in such wires can
be much larger than $\Lambda$. Is this reasonable? In order to answer some of these questions we have calculated the conductances of disordered Au and Ag ultrathin wires, as functions of wire lengths. We start with monoatomic chains, and consider two types of disorder: bounded random fluctuations of the interatomic distances, and the presence of random substitutional impurities. The conductance is calculated within linear response theory, with the use of Kubo’s formula. A multi-orbital tight-binding Hamiltonian with nine bands (the five d-bands and the 4sp complex) is employed to describe the wire electronic structure. The tight-binding parameters were obtained by least-square fitting of first principles full potential linearized argument plane-wave (FLAPW) band structure calculations of pure Au and Ag one dimensional wires. They reproduce the lowest six FLAPW bands extremely well.

For an arbitrary wire it is useful to divide it in unit cells along the wire axial direction, and express the conductance $\Gamma$ in terms of the one-electron propagators connecting those cells. In units of the quantum of conductance $\Gamma_0 = \frac{2e^2}{h}$ we may write

$$\Gamma = \text{ReTr}(\tilde{G}_{00t01}\tilde{G}_{11t10} - t_{01}\tilde{G}_{10t01}\tilde{G}_{10}),$$

(1)

where $\tilde{G}_{ij}$ represents the difference between retarded and advanced one-electron propagator matrices, evaluated at the Fermi energy, connecting cells $i$ and $j$; $0$ and $1$ labels two adjacent cells (the choice is arbitrary due to current conservation), and $t_{01}$ is the tight-binding hopping matrix between such cells. ReTr stands for the real part of the trace over all orbitals and sites within an unit cell. For 1-atom thick chains, the unit cell obviously contains a single atom.

We consider infinite wires with disorder restricted to a sector of length $\ell$ along the wire axis. This is equivalent to assuming the current is injected into and collected from the disordered nanowire by semi-infinite perfect leads. The average wire conductance is calculated by taking into account 4,000 random configurations of the disordered sector. The required one-electron propagators are obtained as follows: first the tip Green functions of the semi-infinite perfect leads are generated by numerical methods that are, by now, well established and efficient. Then, the cells of the disordered wire sector are placed, one by one, atop the injector lead. This is carried out by using Dyson’s equation to switch on the hopping matrix between the added cell and the substrate. Finally, the whole structure is connected, by the same procedure, to the collector lead.

We shall start by examining disordered monoatomic chains in which the interatomic distances $d$ vary randomly, within pre-established bounds, around the bulk equilibrium value $d_0$. If the variations $\Delta d$ are not excessively large, they may be modeled by scaling the hopping integrals. Within certain limits, the two center integrals $\gamma$ approximately follow power laws of the type $\gamma(d) = (d/d_0)^n\gamma(d_0)$, where $n$ depends upon the pair of orbitals involved. We have adopted two different sets of exponents: one proposed by Harrison, and another by Papaconstantopoulos. More comprehensive scaling laws are presently available, but those two prescriptions suffice for the kind of estimate we wish to make. Figure depicts our results for the conductances of Au and Ag disordered wires, calculated as functions of wire lengths for different values of interatomic distance variation bounds $\alpha = d_m/d_0$; $d_m$ represents the maximum atomic separation. The exponential decaying behavior of $\Gamma$ with $\ell$, clearly shows that this type of disorder induces localization. The localization lengths $\Lambda$ deduced from the corresponding decaying rates are plotted against $\alpha$ in the inset. Our results show that $\Lambda$ is rather large for relatively low values of $\alpha$. For instance, we found $\Lambda \approx 100\text{nm}$ for $\alpha = 0.05$ with the use of Harrison’s scaling exponents. Qualitatively, both prescriptions give similar results: $\Lambda$ decreases with $\alpha$ approximately following a power law $\Lambda \approx \Lambda_0\alpha^{-2}$, with $\Lambda_0 \approx 0.62\text{nm}$ and $\approx 0.25\text{nm}$, for the Harrison and Papaconstantopoulos scaling rules, respectively. Much larger bond length variations, corresponding to $\alpha \approx 0.3$, have been observed in stretched Au nanowires. However, there are strong indications that some of those apparently very large values of $d$ are due to the presence of impurities.

Disorder caused by impurities leads to much stronger localization effects. In order to illustrate and investigate the role played by impurities in the conductance characteristics of these nanowires, we consider substitutional Pt atoms randomly distributed along the system. Pt in a noble metal matrix represents a relatively weak scattering impurity potential. We simulate its presence simply by modifying the atomic energy at the impurity site so as to guarantee local charge neutrality. More elaborate ways of describing the impurity potential are certainly possible, but we recall that our main interest here is just to estimate the order of magnitude of the localization length induced by it. Hence, such an approximate treatment of the impurity suffices for our purposes.

In figure we show results for the conductances of monoatomic chains of $\text{Pt}_x\text{Au}_{1-x}$ and $\text{Pt}_x\text{Ag}_{1-x}$ disordered alloys, calculated as functions of $\ell$, for different Pt concentrations $x$. We note in the inset that $\Lambda$ decreases rapidly with $x$, as expected from the one-dimensional character of the disordered wire. We found $\Lambda \approx 16\text{nm}$ for just 1% of Pt impurities in Au-based nanowires. It is also noteworthy that $\Lambda$ becomes rather low for sufficiently large $x$, reaching values of the same order of magnitude as that reported in ref. [6].

One may rightfully argue that the effect of impurities is being overestimated in those monoatomic linear chains, and that such systems are not so easy to realize in practice. Moreover, it is rather difficult to observe localization effects in systems with higher dimensionalities. Therefore, it is important to investigate the dependence of the
localization length on the wire thickness. Some kind of crossover from a localized to a non-localized regime may occur as the wire cross section increases, for a fixed impurity type and concentration. A particularly useful information is the range of thicknesses in which a finite disordered wire is expected to be found in the regime of strong localization. To gain some insight about this matter, we have calculated the conductance of thicker $\text{Pt}_x\text{Ag}_{1-x}$ disordered wires as functions of $\ell$, for several values of $x$. We consider wires with cross sections containing four and eight atoms, as schematically shown in figures 3 and b, respectively. The tight-binding parameters for the 4-atoms-thick Ag wire were obtained by a crude fitting of the band structure calculation reported in reference 10, whereas for the 8-atoms-thick one we simply used bulk Ag parameters. It is worth to mention that our results should not be much sensitive to details of the electronic structure, since impurity-related effects are expected to be much stronger than small variations in the hopping integrals.

Our results depicted in figure 5 show that the localization lengths in these thicker (but still ultrathin) disordered wires may be rather short for moderately low impurity concentrations. In both cases, the localization lengths vary with the degree of disorder, apparently following a power law. Even more intense localization effects are expected for impurities which are associated with stronger scattering potentials as shown in figure 4. The most important information, however, is the fact that those thicker wires also display localization on a nanometric length scale for sufficiently large degree of disorder. These results support our previous hypothesis that the localization length in ultrathin wires made of ferromagnetic metallic alloys may be much smaller than the spin diffusion length. With the increasing ability to control the dimensions and composition of nanowires, we envisage the possibility of using localization as an useful parameter to tailor electrical characteristics in devices of nanoscopic dimensions. Disordered nanowires form a rich laboratory for testing some very nice physical ideas developed on extensive work following the classical article by Abrahams et al.

To summarize, we have calculated the conductance of disordered Ag and Au nanowires using realistic band structures. Our results show that electrical transport flowing through such systems may occur in a strong localization regime. The localization lengths in monoatomic linear chains can be remarkably small for moderately low impurity concentrations. Localization persists in thicker disordered wires, which also exhibit rather small localization lengths for modest amounts of disorder. We hope our findings will stimulate further investigation on these systems. If $\Lambda$ is comparable to the wire length, to the electronic phase breaking or to the spin coherence lengths, localization effects will be of primary importance in the determination of the electronic behavior of such systems.

Enlightening discussions with D.L. Mills are gratefully acknowledged. A.T.C. and R.B.M acknowledge financial support from CNPq and the Milenium Institute for Nanoscience (Brazil). A.T.C also acknowledges financial support from FAPEMIG (Brazil).

* Electronic address: antc@stout.ufla.br
† Electronic address: bechara@if.uff.br

[1] Y. Zhang and H. Dai, Appl. Phys. Lett. 77, 665 (2000); Y. Zhang, N.W. Franklin, R.J. Chen, and H. Dai. Chem. Phys. Lett. 331, 35 (2000); J.-L. Lin, D. Y. Petrovykh, A. Kirakosian, H. Raicher, F.J. Himpsel, and P.A. Dowben, Appl. Phys. Letters 78, 829 (2001).
[2] B.H. Hong, S.C. Bae, C-W. Lee, S. Jeong, K.S. Kim, Science 294, 348 (2001).
[3] N. Nilius, T.M. Wallis, and W. Ho, Science 297, 1853 (2002).
[4] D.J. Thouless, Phys. Rev. Lett. 39, 1167 (1977).
[5] P.A. Lee, Phys. Rev. Lett. 42, 1492 (1979).
[6] J.I. Pascual, J. Mendez, J. Gomezherrero, A.M. Baro, N. Garcia, U. Landman, W.D. Luedtke, E.N. Bogachek, H.P. Cheng, Science 267, 1793 (1995).
[7] H. Onishi, Y. Kondo, and K. Takayanagi, Nature 395, 780 (1998); A.I. Yanson, G.R. Bollinger, H.E. van den Brom, N. Agrait, and J.M. van Ruitenbeek, Nature 395, 783 (1998).
[8] G. Tamura et al., Phys. Rev. Lett. 83, 2030 (1999).
[9] N. Garcia et al., Phys. Rev. Lett. 82, 1699 (1999).
[10] A.T. Costa, R.B. Muniz, Phys. Rev. B 66, 113402 (2002).
[11] A. Umerys, Phys. Rev. B 55, 5266 (1997).
[12] M.P.L. Sancho, J.M.L. Sancho and J. Rubio, J. Phys. F: Metal Phys. 15, 851 (1985).
[13] W. A. Harrison, Electronic Structure and the Properties of Solids (Freeman, San Francisco, 1980).
[14] J.D. Shore and D.A. Papaconstantopoulos, Phys. Rev. B 35, 1122 (1987).
[15] C.E. Lekka, N.I. Papanicolau, G.A. Evangelakis, and D.A. Papaconstantopoulos, J. Phys. Chem. Solids. 62, 753 (2001).
[16] F.D. Novaes, A.J.R. da Silva, E.Z. da Silva, and A. Fazzio, Phys. Rev. Lett. 90, 036101 (2003).
[17] L.G.C. Rego, A.R. Rocha, V. Rodrigues, and D. Ugarte, Phys. Rev. B 67, 045412 (2003); S.B. Legoas, D.S. Galvão, V. Rodrigues, D. Ugarte, Phys. Rev. Lett. 88, 076105 (2002).
[18] E. Abrahams, P.W. Anderson, D.C. Licciardello and T.V. Ramakrishnan, Phys. Rev. Lett., 42, 673 (1979); P.A. Lee and T.V. Ramakrishnan, Rev. of Mod. Phys., 57, 287 (1985).
FIG. 1: Average logarithm of $\Gamma$ as a function of wire length for different values of $\alpha$, obtained using Harrison prescription. The inset shows the localization length $\Lambda$ as a function of bond length variation bounds $\alpha$, for Papaconstantopoulos (squares) and Harrison (circles) prescriptions.

FIG. 2: Conductance as a function of wire length for a Au (a) and Ag (b) atomic chains with Pt impurities, for various impurity concentrations $x$. In (c) we show how $\Lambda$ varies with $x$ for the Au (squares) and Ag (circles) wires.
FIG. 3: (Color online) Schematic representation of the 4 (a) and 8 (b) atoms cross-section wires. The atoms marked in red represent our choice of unit cell.

FIG. 4: Localization length as a function of the impurity potential for a 4 atoms thick Ag wire, with 2% of randomly distributed impurities (circles). The red curve is a fitting of the data to a power-law with exponent $\approx 1$. 
FIG. 5: Conductance of a four atoms cross section Ag fcc(110) nanowire as a function of wire length for various impurity concentrations. The inset shows the localization length as a function of impurity concentration for the four atoms (circles) and eight atoms cross section wires. The lines are power-law fittings to the data (see text).