Metallic nanowires: multi-shelled or filled?

G. Bilalbegović

Department of Physics, University of Rijeka, Omladinska 14, 51 000 Rijeka, Croatia
(to be published in Computational Materials Science)

Abstract

The room temperature structure of aluminum, copper and gold infinite nanowires is studied. The molecular dynamics simulation method and the same type of the embedded atom potentials made by Voter and coworkers are used. It was found that multi-shelled and various filled metallic nanowires exist depending on the metal and the initial configuration. The results were compared with previous simulations for gold nanowires using different type of the potential.

Pacs: 61.46.+w,68.65.+g,81.05.Ys

Keywords: nanowires, metals, surfaces, molecular dynamics simulation, embedded atom potentials, aluminum, copper, gold

1. INTRODUCTION

The studies of nanostructures are important for advances in micro-electronics. Cylindrical nanostructures, i.e., nanowires were prepared for various materials [1–3]. The most studied are single and multi-walled carbon nanotubes [1,2]. Nanowires made of metals are also important [1,4,5,17]. Interest for metallic nanowires especially increased after the tip-surface contact experiments performed in Scanning Tunneling Microscope (STM) [3]. This was followed by a discovery of cylindrical metallic nanostructures in the table-top contact [3]. Experimental and theoretical results show that the quantized conductance exist for these metallic nanocontacts [4]. Molecular Dynamics (MD) simulation method was also used for studies of metallic nanowires and nanocontacts [8,17]. Structural, mechanical, and vibrational properties were investigated.

Carbon nanotubes often exist as multi-shelled structures [1,2]. Multi-walled nanowires were also found for several inorganic layered materials, such as WS$_2$, MoS$_2$, and NiCl$_2$ [3]. Electronic shells in metallic nanowires were obtained in the jellium model calculation using the local-density-functional based shells correction method [13]. This was recently confirmed by the conductance measurements for sodium nanowires formed in the contact between two bulk sodium electrodes [19]. MD simulations have shown an existence of finite [15] and infinite [16] multi-shelled gold nanowires. The radius of these gold nanowires was of the order of 1 nm, whereas the length of simulated finite nanowires was from one to several nm. Similar gold nanostructures were observed in the STM and transmission electron microscope experiments [20,21].
Copper, aluminum, and gold are technologically important metals. They are widely applied in macroscopic electric circuits, as well as in mesoscopic devices. For example, these metals are used for interconnections in the chips. Because of their importance special methods of producing metallic parts in integrated systems are developed, such as the electroless copper deposition [22]. Recently applied methods for making nanoscale devices are also the X-ray lithography and the technique of using an atomic force microscope cantilever for the stencil [23]. These methods are producing wires thinner than 0.1 µm. Computer simulations of metallic nanowires should help in developing new methods for their fabrication. The computational methods also provide access to microscopic information about physical properties of nanowires. In this paper the MD simulation study of internal structure of aluminum, copper, and gold nanowires has been carried out. The goal is to determine whether multi-shelled structures, already found in MD simulations for finite and infinite gold nanowires [15,16], exist for other metals.

II. COMPUTATIONAL METHOD

Embedded atom and effective medium potentials are widely used in simulation of metals [24,25]. They provide a good description of metallic bonding for bulk, surfaces, and clusters. In comparison with the pair potentials, in the embedded atom model there is an additional term for the energy to embed an atom in the electron density of its neighbours. Several versions of these potentials are available. They differ in the fitting procedure and in the selection of the properties which were taken for the fit. In this work the embedded atom potentials for copper, aluminum, and gold made by A. Voter and coworkers were used [26–28]. The preparation of these potentials included the fitting of the lattice constant, the bulk modulus, the cohesive energy, the three cubic elastic constants, the vacancy formation energy, as well as the bond length and bond energy of the diatomic molecule.

The classical MD simulation method was applied. The temperature control was realized through velocity rescaling. A time step of 2.64 × 10^{-15} s was used for aluminum, 4.06 × 10^{-15} s for copper, and 7.14 × 10^{-15} s for gold. The number of atoms in nanowires was between 1032 and 1152. The periodic boundary conditions were used along the wire axis. The simulation started from ideal fcc structures at T = 0 K. The cross-sections were (111) oriented. In previous simulation of infinite gold nanowires (using the so-called glue model potential) it was found that this initial orientation produces better multi-shelled structures than (110) and (100) [16]. The cylindrical MD boxes included all atoms whose distance from the axes was less than 1.2 nm. The same radius produced a pronounced five-shelled structure for an infinite (111) oriented gold nanowire in the previous simulation [16]. Following the annealing and quenching procedure, the nanowires were analyzed at T = 300 K, and after evolution of up to 10^6 time steps.

III. RESULTS AND DISCUSSION

Figure 1 shows that this aluminum nanowire consists of the five coaxial cylindrical walls. The core of the nanowire is empty. The cylindrical walls are disordered. As presented in Fig. 2, the fcc(111) structure of the cross sections is preserved in the copper nanowire. The
side view in Fig. 2(b) shows that the perfect layered structure was formed. The faceted and filled gold nanowire is presented in Fig. 3. The parallel rows of atoms in the cross section are apparent. This should be compared to the initial fcc(111) cross section with the hexagonal symmetry. It was found that these copper and gold nanowires preserve their filled structures at higher temperatures. Therefore, the results show that cylindrical shells appear in the aluminum nanowire. However, it is not obvious why shells were not found for the copper and gold nanowires prepared in the same way. Moreover, previous simulations [15,16] for gold nanowires modeled by another type of the potential [29], have shown that multi-shelled structures of lasting stability appear. There was a possibility that simulations for copper and gold nanowires which employ the potentials from Refs. [26, 28] require more delicate simulation technique. To check this point a new set of simulations for aluminum, copper, and gold described by the potentials from Refs. [26–28] has been carried out. The same radius of 1.2 nm was selected. In these new simulations the starting geometry was the ordered configuration of the five-shelled nanowire which was obtained in simulations using the glue gold potential [16]. The aluminum nanowire shown here in Fig.1 is also five-shelled, but it is less ordered. In new simulations the same procedure of annealing and quenching was applied. The results are presented in Figs. 4, 5, and 6. The aluminum nanowire in Fig. 4 consists of the fcc(111) layers. Copper and gold nanowires in Figs. 5 and 6 also consist of the layers, but there in the cross sections the atoms close to the cylindrical walls follow circular paths. Therefore, various filled and multi-shelled metallic nanowires are stable within a long simulation time and a sensitive dependence of the initial conditions was found. Fluctuations in physical properties of metallic nanowires are studied theoretically within the jellium model [30]. The origin of these phenomena is the onset of chaos in classical and quantum billiard structures. It should be mentioned that we did not find such strong dependence on the initial configuration in gold nanowires modeled by the glue potential [15, 16]. In these simulations gold shells form almost immediately after the simulation starts and regardless of an initial configuration. The glue potential for gold, in comparison with the potential of Voter and coworkers, gives a better description of reconstruction on gold surfaces [31, 28]. The subtle details of inter-atomic interactions, that are responsible for surface reconstruction, also give rise to internal reconstructing of nanowires. Therefore, it is possible that the multi-shelled structure (found for the glue gold potential and here for the aluminum in Fig.1) is the most frequent in fcc metallic nanowires.

In conclusion, it was shown that multi-shelled and filled metallic nanowires exist for several fcc metals. The method of molecular dynamics simulation and the same kind of the embedded atom potentials for aluminum, copper, and gold were used. The structures obtained strongly depend on the initial configuration. In these simulations, no apparent correlation of the structure of nanowires with the properties of metals was found.

ACKNOWLEDGMENTS

This work has been carried under the CRO-MZT project 119206 - “Dynamical Properties of Surfaces” and the EC Research Action COST P3 - “Simulation of Physical Phenomena in Technological Applications”.
REFERENCES

[1] C. Dekker, Phys. Today, V/22 (1999).
[2] S. Iijima, Nature 354, 56 (1991); MRS Bulletin 19, 43 (1994).
[3] R. Tenne, L. Margulis, M. Genut, and G. Hodes, Nature 360, 444 (1992); L. Margulis, G. Salitra, R. Tenne, and M. Tallenker, Nature 365, 113 (1993); Y. R. Hacohen, E. Grunbaum, R. Tenne, and M. Tallenker, Nature 395, 336 (1998).
[4] J. I. Pascual, J. Mendez, J. Gomez-Herrero, A. M. Baro, N. Garcia, U. Landman, W. D. Luedtke, E. N. Bogachek, and H. P. Cheng, Science 267, 1793 (1995).
[5] J. I. Pascual, J. Mendez, J. Gomez-Herrero, A. M. Baro, and N. Garcia, Phys. Rev. Lett. 71, 1852 (1993).
[6] J. L. Costa-Krämer, N. Garcia, P. Garcia Mochales, and P. A. Serena, Surf. Sci. 342, L1144 (1995).
[7] J. M. van Ruitenbeek, cond-mat/9910394, to appear in “Metal Clusters on Surfaces: Structure, Quantum Properties, Physical Chemistry”, K. H. Meiwes-Broer, ed. (Springer-Verlag, Berlin).
[8] M. R. Sorensen, K. W. Jacobsen, and P. Stoltze, Phys. Rev. B 53, 2101 (1996).
[9] A. M. Bratkovsky, A. P. Sutton, and T. N. Todorov, Phys. Rev. B 52, 5036 (1995).
[10] G. M. Finbow, R. M. Lynden-Bell, and I. R. McDonald, Mol. Phys. 92, 705 (1997).
[11] H. Mehrez and S. Ciraci, Phys. Rev. B 56, 12632 (1997).
[12] R. N. Barnett and U. Landman, Nature 387, 788 (1997).
[13] O. Gülerseren, F. Ercolessi, and E. Tosatti, Phys. Rev. B 51, 7377 (1995).
[14] O. Gülerseren, F. Ercolessi, and E. Tosatti, Phys. Rev. Lett. 80, 3775 (1998).
[15] G. Bilalbegović, Phys. Rev. B 58, 15412 (1998).
[16] G. Bilalbegović, Solid State Commun. (2000).
[17] H. Ikeda, Y. Qi, T. Cagin, K. Samwer, W. L. Johnson, and W. A. Goddard III, Phys. Rev. Lett. 82 2900 (1999).
[18] C. Yannouleas and U. Landman, J. Phys. Chem. B 101, 5780 (1997).
[19] A. I. Yanson, I. K. Yanson, and J. M. van Ruitenbeek, Nature 400, 144 (1999).
[20] H. Ohnishi, Y. Kondo, and K. Takayanagi, Nature 395, 788 (1998).
[21] A. I. Yanson, G. Rubio Bollinger, H. E. van der Brom, N. Agrait, and J. M. van Ruitenbeek, Nature 395, 783 (1998).
[22] J. Li, R. Blower, and J. W. Mayer, MRS Bulletin 18, 18 (1993).
[23] R. Lüthi, R.R. Schlittler, J. Brugger, P. Vettiger, M. E. Welland, and J. K. Gimzevski, Appl. Phys. Lett. 75, 1314 (1999).
[24] M. S. Daw, S. M. Foiles, and M. I. Baskes, Mater. Sci. Rep. 9, 251 (1993).
[25] K. W. Jacobsen, J. K. Nørskov, and M. J. Puska, Phys. Rev. B 35, 7423 (1987).
[26] C. L. Liu, J. M. Cohen, J. B. Adams, and A. F. Voter, Surf. Sci. 253, 334 (1991).
[27] A. F. Voter and S. P. Chen, Mater. Res. Soc. Symp. Proc. 82, 175 (1987).
[28] S. P. Chen and A. F. Voter, Surf. Sci. 244, L107 (1991).
[29] F. Ercolessi, M. Parrinello, and E. Tosatti, Philos. Mag. A 58, 213 (1988).
[30] C. A. Stafford, D. Baeriswyl, and J. Buerki, Phys. Rev. Lett. 83, 4836 (1999); C. Höppler and W. Zwerger, Phys. Rev. B 59, R7849 (1999).
[31] M. Bernasconi and E. Tosatti, Surf. Sci. Rep. 17, 363 (1993).
FIGURES

FIG. 1. Infinite aluminum nanowire with an average radius of 1.2 nm, shown at 300 K, after $10^6$ time steps, and starting from an ideal fcc(111) structure: (a) top view, (b) side view. The trajectory plots refer to a time span of $\sim 7$ ps and include all atoms in the slice of the thickness of 4 nm along the wire axis.

FIG. 2. Copper nanowire simulated starting from the fcc(111) orientation: (a) top view, (b) side view. Other details as in Fig. 1

FIG. 3. Gold nanowire simulated starting from the fcc(111) orientation: (a) top view, (b) side view. Details as in Fig. 1

FIG. 4. Aluminum nanowire simulated starting from the five-shelled structure: (a) top view, (b) side view. Details as in Fig. 1

FIG. 5. Copper nanowire simulated starting from the five-shelled structure: (a) top view, (b) side view. Details as in Fig. 1

FIG. 6. Gold nanowire simulated starting from the five-shelled structure: (a) top view, (b) side view. Details as in Fig. 1
This figure "Fig1.jpg" is available in "jpg" format from:

http://arxiv.org/ps/cond-mat/0005203v1
This figure "Fig2.jpg" is available in "jpg" format from:

http://arxiv.org/ps/cond-mat/0005203v1
This figure "Fig3.jpg" is available in "jpg" format from:

http://arxiv.org/ps/cond-mat/0005203v1
This figure "Fig4.jpg" is available in "jpg" format from:

http://arxiv.org/ps/cond-mat/0005203v1
This figure "Fig5.jpg" is available in "jpg" format from:

http://arxiv.org/ps/cond-mat/0005203v1
This figure "Fig6.jpg" is available in "jpg" format from:

http://arxiv.org/ps/cond-mat/0005203v1