Nonlinear current-induced forces in Si atomic wires

Zhongqin Yang and Massimiliano Di Ventra

Department of Physics, Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061

Abstract

We report first-principles calculations of current-induced forces in Si atomic wires as a function of bias and wire length. We find that these forces are strongly nonlinear as a function of bias due to the competition between the force originating from the scattering states and the force due to bound states. We also find that the average force in the wire is larger the shorter the wire, suggesting that atomic wires are more difficult to break under current flow with increasing length. The last finding is in agreement with recent experimental data.

PACS numbers: 73.40.Jn, 73.40.Cg, 73.40.Gk, 85.65.+h

Current-induced atomic motion (electromigration) has always been of concern in microelectronics since it was found to be a major failor mechanism in aluminum conductors. It is a well-known fact that the smaller the material dimensions as it is the case, for instance, for metallization lines in conventional circuits. It does not come as a surprise then that this effect has recently attracted considerable interest in nanoscience. In particular, current-induced forces in atomic and molecular wires and in the case of metallic gold (jellium model). The interior electron density of the electrodes is taken equal to that for ideal metals (jellium model). The electron wave functions are computed by solving the Lippman-Schwinger equation iteratively to self-consistency in steady state. Exchange and correlation are included in the density-functional formalism within the local-density approximation. The current is computed from the wave functions of the electrode-molecule system. The force \( \mathbf{F} \) acting on a given atom at position \( \mathbf{R} \) due to the external bias is given by the Hellmann-Feynman-type of theorem developed in Ref. \( \mathbb{5} \) 7

\[
\mathbf{F} = - \sum_i \langle \psi_i | \frac{\partial H}{\partial \mathbf{R}} | \psi_i \rangle - \lim_{\Delta \to 0} \int d\mathbf{E} \langle \psi_\Delta | \frac{\partial H}{\partial \mathbf{R}} | \psi_\Delta \rangle. \tag{1}
\]

The sum and integral in Eq. (1) include spin variables also. The first term on the RHS of Eq. (1) is the usual Hellmann-Feynman contribution to the force due to localized electronic states \( |\psi_i\rangle \). The second term is the contribution to the force due to the continuum of states. It is calculated by constructing, for each energy in the continuum, square-integrable wavefunctions \( |\psi_\Delta\rangle \) in an energy region \( \Delta \)

\[
|\psi_\Delta\rangle = A \int_\Delta d\mathbf{E} \psi, \tag{2}
\]

where \( A \) is a normalization constant and the \( \psi \)'s are single-particle wavefunctions in the continuum, solutions of the Lippmann-Schwinger equation. The continuum integration \( \sigma \) covers the part of the spectrum occupied by the electrons at a given bias. Finally, the total force on the atom includes a trivial ion-ion interaction.

We start the calculations by first relaxing the atomic positions at zero bias. For all different wire lengths the relaxed Si-jellium surface bond length is about 2 a.u., and the relaxed Si-Si bond distance is about 4.2 a.u. In Fig. (a),(b),(c) we plot the total force as a function of bias on each Si atom for wires composed of two, three
and four Si atoms, respectively. The atomic positions have been fixed at the equilibrium position at zero bias, and the atoms are labeled with increasing number starting from the closer to the left electrode (see Fig. 1). In Fig. 1(d) we plot the average force (sum of the forces on each atom divided by the number of atoms in the wire) as a function of bias and the force for a wire composed of a single Si atom. Positive force pushes the atom to the right, i.e., opposite to electron flow (the left electrode is positively biased). It is clear from Fig. 1(a), (b), (c) that the force on atoms is a nonlinear function of the bias (except for the single-atom wire, see below and Ref. [18]) while the average force Fig. 1(d) saturates with the number of atoms in the wire. We first discuss the nonlinear behavior of the forces and then discuss the average force.

For a small external bias (0.01 V, i.e., linear-response regime), the current-induced forces in the 3-Si wire satisfy the zero-force sum rule while for the 2-Si and 4-Si wire all atoms are pushed opposite to the current flow with almost equal force as it can be inferred from symmetry considerations [1]. On the other hand, strong nonlinearities in the current-induced forces appear at voltages above 0.1 V. For instance, in the 2-Si wire the Si atom closer to the left electrode (Si_1 in the inset of Fig. 1) is pushed against the electron flow for biases less than 0.5 V and moves along the electron flow for larger biases. On the other hand, the second Si atom (Si_2 in the inset of Fig. 1) is pushed against the electron flow at all external voltages. Similar effects, involving different atoms in the wire, occur at even smaller voltages for the 4-Si wire. In the same vein, for the 3-Si wire the zero-force sum rule is not satisfied already at 0.1 V. It is interesting to note that for large biases the largest force occurs on the second Si atom from the left. The break-up of the wires is thus likely to nucleate from the bonds of this atom. In general, each atom experiences a force due to current flow that is nonlinear in the external bias. Such nonlinearities affect the atomic redistribution in the wire and eventually its resistance to current-induced rupture. In order to understand this nonlinear behavior, we study the different contributions to the forces as a function of bias: (a) the contribution from the electrodes without the atoms in between (direct force [1]), (b) the contribution from the continuum part of the spectrum, and (c) the part of the force originating from the discrete spectrum. The force originating from the ion-ion interaction does not depend on the bias and we thus do not discuss it here. For the sake of simplicity we discuss the 3-Si wire case. Similar considerations are valid for other wire lengths. We plot in Fig. 2 the three different contributions considered. As expected, the direct force is almost linear with the bias. Small deviations from linearity appear due to the small deviations from linear decay of the electrostatic potential close to the electrode surfaces. [17, 18] Furthermore, due to this deviation, the force on the central atom in the wire is larger than the force on the two atoms close to the electrode surfaces at any bias (see Fig. 2). The latter atoms also experience a force of similar magnitude with deviations occurring at large voltages. From Fig. 2 it is also evident that for biases larger than 0.5 V the major contribution to the force on each atom comes from the direct force. The nonlinearities in the total force then originate from the competition between the force due the states in the continuum (scattering states) and the force due to the bound states (in this case, those states below the band bottom of the left electrode). The force on the central atom of the wire (Si_3) due to the scattering states pushes the atom to the left, i.e., along with the electron flow. This force is almost linear with the bias. The force on the same atom due to the discrete spectrum, on the other hand is almost zero even at very large voltages. This behavior can be understood by looking at the extra charge localized on the atom at any given bias. This quantity can be estimated by integrating the charge around a given atom in a box centered on that atom with one face parallel to the electrode surfaces and whose size in the perpendicular direction is equal to the bond distance between the atoms. The difference between this charge at zero bias and the corresponding charge at any given bias is plotted in Fig. 3 for the three different Si atoms. Apart from small fluctuations as a function of bias, the extra charge on the central atom is zero. The charge on this atom is thus practically constant at any bias. Since, in this system, the number of bound states does not change with bias, their contribution to the force is constant [see Fig. 3(c)]. On the other hand, the force due to the continuum [second term on the RHS of Eq. (1)], increases linearly with bias due to the larger energy integration in Eq. (1) for larger voltages. [18]

The continuum and discrete contribution to the force is nonlinear for the atoms closer to the electrodes (Si_1 and Si_3 of Fig. 3). For the Si_1 atom, for instance, the continuum force is almost constant up to about 1 V, while the force from the discrete spectrum increases in magnitude with bias in the same voltage range (the sign of the force corresponds to the atom pushed along with the current flow). For biases above 1 V, the continuum force increases in magnitude and the force due to bound states is almost constant. An opposite trend is observed for the Si_3 atom (see Fig. 3). The opposite trend can be explained again by looking at the extra charge on these atoms (Fig. 3). This charge is of similar magnitude but of opposite sign indicating that there is a charge transfer from the right electrode to the left electrode when current flows with consequent creation of a local electric dipole. [18]

While it is difficult to extract general trends in the overall resistance of the wires to electromigration by looking at the forces on each atom, the average force follows quite a simple trend as a function of bias and wire length. In Fig. 2(d) we plot such a quantity for wires of two, three and four Si atoms. For comparison we also plot the force for a single Si atom. The average force is almost linear as a function of bias even for large biases (deviations occur above 2.5 V for the 2-Si wire). Furthermore, the force is larger the smaller the wire length and, in particular, it
is almost equal for the 3- and 4-Si wires. This trend indicates that short wires are easier to break than longer wires, and the average force reaches a “bulk” value at wire lengths of only three atoms, i.e., wire lengths of less than 10Å. Similar trends have been observed in experiments with Au atomic wires.

We conclude this paper by studying the effects of current-induced atomic relaxations on the current-voltage (I-V) characteristics of atomic wires. It was found by Di Ventra et al. that current-induced atomic relaxations in molecular wires do not substantially affect the absolute value of the current for large voltages and current densities. We find that this trend is also valid in the present case of atomic wires, suggesting it is a general trend valid for both atomic and molecular wires when current flow is mainly coherent. This is illustrated in Fig. 4 where we plot the I-V characteristics for the 3-Si wire with and without current-induced atomic relaxations. In the inset of the same figure we show the relaxed atomic positions for each atom at selected biases. It is clear from Fig. 4 that small changes in the absolute value of the current when the atoms are relaxed are observed even at large biases. Similar results are valid for the other wires as well.

In conclusion, we have reported first-principles calculations of current-induced forces in Si atomic wires. We find that these forces are generally strongly nonlinear as a function of bias. Since the direct force from the bare electrodes is almost linear as a function of bias, the nonlinearity originates from the competition between the scattering-state and discrete-spectrum force. We also find that the average force in the wire is larger the shorter the wire, suggesting that atomic wires are more difficult to break under current flow with increasing length. Finally, current-induced relaxations are found to change only slightly the absolute value of the current even at large voltages. A similar effect has been predicted for molecular wires, indicating it is a general trend for nanoscale wires when coherent scattering is the main transport mechanism.

We thank Y.-C. Chen for useful discussions. This work is supported in part by the National Science Foundation Grants No. DMR-01-02277 and DMR-01-33075, Carilion Biomedical Institute, and Oak Ridge Associated Universities. Acknowledgement is also made to the Donors of The Petroleum Research Fund, administered by the American Chemical Society, for partial support of this research. The calculations reported in this paper were performed on the beowulf cluster of the Laboratory for Advanced Scientific Computing and Applications at Virginia Tech.

---

* E-mail address: diventra@vt.edu.

1 See e.g., I. A. Blech and H. Sello, in *Physics of Failure in Electronics*, edited by T. S. Shilliday and J. Vaccaro (USAF, Rome Air Development Center, 1967), Vol. 5, p. 496.

2 M. Hauder, J. Gstottner, W. Hansch, and D. Schmitt-Landsiedel, Appl. Phys. Lett. 78, 838 (2001).

3 N. D. Lang, Phys. Rev. B 45, 13599 (1992); 49, 2067 (1994).

4 N. Agraït, C. Untiedt, G. Rubio-Bollinger, and S. Vieira, Phys. Rev. Lett. 88, 216803 (2002).

5 K. Itakura, K. Yuki, S. Kurokawa, H. Yasuda, and A. Sakai, Phys. Rev. B 60, 11163 (1999).

6 H. Yasuda and A. Sakai, Phys. Rev. B 56, 1069 (1997).

7 T. N. Todorov, J. Hoekstra, and A. P. Sutton, Philos. Mag. B 80, 421 (2000); T. N. Todorov, J. Hoekstra, and A. P. Sutton, Phys. Rev. Lett. 86, 3606 (2001).

8 B. Q., Wei, R. Vajtai, P.M. Ajayan, Appl. Phys. Lett. 79, 1172 (2001).

9 M. Di Ventra and S.T. Pantelides, Phys. Rev. B 61, 16207 (2000); M. Di Ventra, S. T. Pantelides, and N. D. Lang, Phys. Rev. Lett. 88, 046801 (2002).

10 See, e.g., *Molecular Electronics II*, A. Aviram, M. Ratner, and V. Mujica eds., (NY Academy of Sciences, NY, 2002).

11 M. Di Ventra, N.D. Lang, and S.T. Pantelides, Chemical Physics 281, 189 (2002).

12 A. I. Yanson, G. Rubio-Bollinger, H. E. van den Brom, N. Agraït, and J. M. van Ruitenbeek, Nature, 395, 783 (1998).

13 R. S. Sorbello, *Solid State Physics*, edited by H. Ehrenreich and F. Spaepen (Academic Press, New York, 1998), Vol 51, p. 159, and references therein; R. Landauer and J. W. F. Woo, Phys. Rev. B 10, 1266 (1974); A. K. Das and R. Peierls, J. Phys. C 8, 3348 (1975); L. J. Sham, Phys. Rev. B 12, 3142 (1975).

14 Note that this definition of direct force is not unique in literature (see e.g. Ref. 13).

15 N. D. Lang, Phys. Rev. B 52, 5335 (1995); 49, 2067 (1994); Z. Yang, A. Tackett, M. Di Ventra, Phys. Rev. B 66, 041405 (2002).

16 M. Di Ventra and N. D. Lang, Phys. Rev. B 65, 045402 (2002).

17 Note that a misprint appears in the expression of the force in Refs. 13, 14. A negative sign is missing in the definition of forces in these references. See M. Di Ventra, S.T. Pantelides, and N.D. Lang, Phys. Rev. Lett. 89, 139902 (2002).

18 Similar considerations explain the almost linear dependence of the force on the single-atom wire (see Fig. 1(d)). In this case the extra charge on the atom is almost zero at any bias with consequent zero contribution from the discrete spectrum, and linear contribution from the continuum spectrum as a function of bias.

19 It is interesting to note at this point that the sum of the continuum- and discrete-state contributions to the force is larger for the second atom from the right electrode at all biases. A similar result has been inferred by Brandbyge et al. [M. Brandbyge, N. Kobayashi, and M. Tsukada, Phys. Rev. B 60, 17064 (1999)] from the analysis of the voltage drop in gold wires, suggesting that this effect is material independent.

20 Using a tight-binding approach Todorov at al. have shown
that in linear response the largest current-induced force is almost constant for wire lengths of three atoms or more. We show here that this trend is also valid for the average force at any bias.

FIG. 1: Total force as a function of bias in atomic wires containing (a) two, (b) three, and (c) four Si atoms. The inset shows a schematic of one of the wires investigated. The atoms are labeled with increasing number from the left electrode (see inset). (d) The average force in the Si wires. The left electrode is positively biased.

FIG. 2: Different contributions to the total force for a 3-Si wire: (a) direct force (see text), (b) scattering-state contribution, and (c) contribution from the discrete spectrum.

FIG. 3: Extra charge (see text) on the three Si atoms in the 3-Si wire as a function of bias.
FIG. 4: I-V curve of the 3-Si wire with and without the effect of current-induced atomic relaxations. The inset shows the unrelaxed (open circles) and relaxed (full circles) atomic positions of the Si atoms between the two electrodes (vertical thin rectangles) at selected biases. The atoms move opposite to the electron flow.
(a) Direct

(b) Continuum

(c) Discrete
