Theory and first principles calculations of current-induced atomic dynamics

Mads Brandbyge[1], Jing-Tao Lü[1], Tue Gunst[1], and Per Hedegård[2]
[1]Dept. of Micro and Nanotechnology,
Technical University of Denmark, DTU building 345 east,DK-2800 Kgs. Lyngby, Denmark
[2]Niels Bohr Institute, Copenhagen University, Universitetsparken 5, 2100 København Ø

e-mail: mads.brandbyge@nanotech.dtu.dk

INTRODUCTION

The influence of an electronic current on atomic dynamics is an important and intriguing problem in nanoelectronics. Interaction of electrons with phonons has undergone intense study. For the inelastic phonon signals in the current has been addressed theoretically with some success for molecular contacts between metals, e.g. using parameters from DFT [7], for voltages around excitation threshold. For higher voltage bias ($\sim$1V), a high electronic current may strongly influence the motion of atoms, even for weak electron-phonon(e-p) coupling. This is not only due to Joule heating. Recently, new effects has been pointed out which may be important for the atomic dynamics and stability. These include run-away behavior due to excitation of "water-wheel" modes [3]–[5], [8], and a laser-like amplification of the phonons for contacts with a n-p-junction character [6].

METHOD

We derive and employ a semi-classical Langevin equation(SCLE) obtained to describe the ionic dynamics of a molecular-scale junction in the presence of electrical current. The electronic environment is treated as a non-equilibrium bath [1], [2], [4] which we assume to be: (i) unperturbed by the ionic motion and in a steady-state, (ii) coupled linearly to the atomic displacements via the electron-phonon coupling ($M_{\rm ep}$). Its effect on the ions is derived via the Feynman-Vernon theory. We obtain forces due to the current on the ions, valid to 2nd order in $M_{\rm ep}$. Denoting the (mass-scaled) displacements by of the phonon by x with frequency ω_0, we get the SCLE,

$$\ddot{x} = -\omega_0^2 x - \int_{t_0}^{t} \Pi^r(t,t')x(t')dt' + f(t)\,. \qquad (1)$$

Here the electrons are influencing the motion via both deterministic and random forces. The retarded time-kernel (e-p self-energy), Π^r, yield the back-action at time t after propagation in the electronic "bath" due to the motion of x at previous times. The force f is random and describes the unknown state of the bath, include quantum fluctuations, and is also given by e-p self-energies.

RESULTS

The random force, f, is in equilibrium related to Π^r by the fluctuation-dissipation theorem, while in nonequilibrium it contains a term describing the Joule heating (see Fig. 1) after a threshold ($V > \hbar\omega_0$). This is not the only excitation mechanism due to the current, and using NEGF-DFT calculations we have examined other effects for simple, yet realistic, systems (Fig. 2). The deterministic forces (Π^r) contain the energy non-conservative(NC) wind force, an well as an Lorentz-like force due to the Berry phase of the non-equilibrium electrons. These can couple the phonon-modes into polarized modes with circular motion. In effect, the atoms can pick up energy from the flowing electrons due to the NC force, akin to a waterwheel, see Fig. 2. The deterministic forces also include the electronic "friction", $-\gamma_{eh}\dot{x}$, which in the presence of current can change sign and turn into phonon "amplification" for n-p (donor-acceptor) type systems, illustrated in Fig. 3. Both effects will in the harmonic approximation lead to a runaway instability beyond a critical

voltage. Molecular dynamics simulations including anharmonic forces in the presence of current is a possible extension of the SCLE approach [5].

ACKNOWLEDGMENTS

We thank Profs. T. N. Todorov, D. Dundas and A.-P. Jauho for helpful discussions. We are grateful to grants from the DCSC for computational resources and the Lundbeck foundation.

REFERENCES

[1] N. Bode, S. V. Kusminskiy, R. Egger, and F. von Oppen. Current-induced forces in mesoscopic systems: A scattering-matrix approach. *Beilstein Journal of Nanotechnology*, 3:144–162, 2012.

[2] M. Brandbyge and P. Hedegård. Theory of the Eigler switch. *Phys. Rev. Lett.*, 72(18):2919–2922, 1994.

[3] D. Dundas, E. J. McEniry, and T. N. Todorov. Current-driven atomic waterwheels. *Nature Nanotech.*, 4(2):99–102, FEB 2009.

[4] J. T. Lü, M. Brandbyge, and P. Hedegård. Blowing the fuse: Berrys phase and runaway vibrations in molecular conductors. *Nano Lett.*, 10(5):1657–1663, May 2010.

[5] J.-T. Lü, T. Gunst, P. Hedegård, and M. Brandbyge. Current-induced dynamics in carbon atomic contacts. *Beilstein Journal of Nanotechnology*, 2:814–823, 2011.

[6] J. T. Lü, P. Hedegård, and M. Brandbyge. Laserlike vibrational instability in rectifying molecular conductors. *Phys. Rev. Lett.*, 107(4):046801, Jul 2011.

[7] M. Paulsson, T. Frederiksen, and M. Brandbyge. Modeling inelastic phonon scattering in atomic- and molecular-wire junctions. *Phys. Rev. B*, 72(20):201101, 2005.

[8] T. N. Todorov, D. Dundas, A. T. Paxton, and A. P. Horsfield. Nonconservative current-induced forces: A physical interpretation. *Beilstein Journal of Nanotechnology*, 2:727–733, 2011.

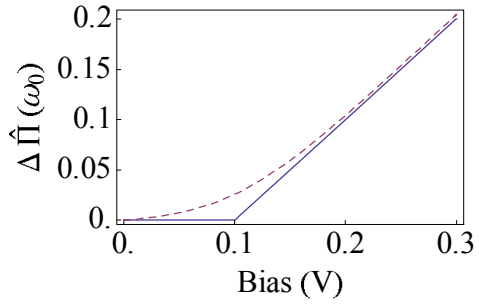

Fig. 1. Example of the nonequilibrium noise spectrum at T=0K (solid) and 300K (dashed) for a given phonon mode with frequency $\hbar\omega_0 = 0.1$ eV. We plot the nonequilibrium part of $\langle f(t)f(t')\rangle = \hbar(\Pi^{>}(t-t') + \Pi^{<}(t-t'))/2 \equiv \hbar\hat{\Pi}(t-t')$.

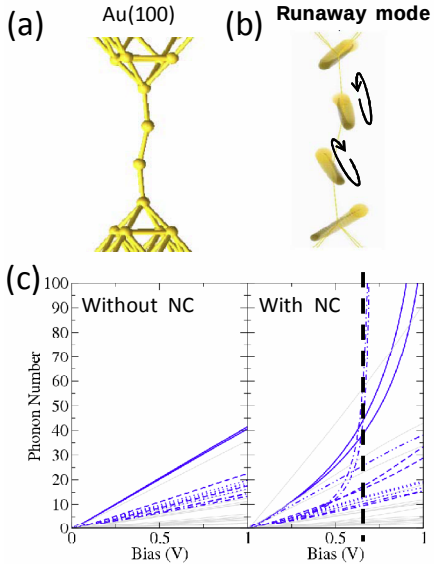

Fig. 2. (a) A model atomic conductor consisting of a short gold chain between electrodes. (b) Calculated phonon-occupations as a function of bias for phonons inside the chain region without(left) and with(right) the energy non-conserving force. The line indicates the voltage where a runaway mode is appearing. (c) The motion of atoms in the runaway mode. The arrows indicate a circular motion.

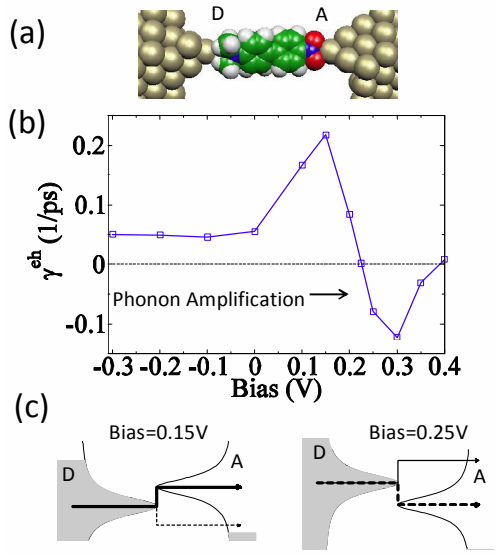

Fig. 3. (a) A donor-acceptor (D-A) molecule (4-dimethylamino-4'-nitrobiphenyl) bridging Au(111) electrodes. (b) Calculated phonon-friction of a phonon in the D-A molecule in the presence of electronic current as a function of bias. At a certain voltage the friction turns to amplification. (c) Schematic showing the DOS on the D-A groups for different voltage. The amplification is due to dominance of phonon-emission processes, which follows a region of extra damping where phonon-absorption dominates (cooling).