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Effects of quantum recoil forces in resistive switching in memristors

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Memristors – electric devices whose resistance "remembers" the history of the previously passed current [1] – are appealing circuit elements for neuromorphic applications beyond the von Neumann architecture [2]. Current-controlled resistive switching in such devices typically occurs due to ion migration in a dielectric layer between the anode and the cathode resulting in the growth or rupture of conductive filaments [3]. The physics of this key process still remains quite obscure because of the stochasticity of the filament dynamics and a possible interplay of various scales and effects, especially when it comes to filaments several atoms thick exhibiting quantized conductance [4, 5]. Because of the attractiveness of such thin filaments for encoding discrete states of the memristor [6], in this letter, we analyze the role of electron-induced, "recoil" forces acting on the ions [7] in the dynamics of these filaments, in particular, in the context of conductance quantization. In fact, in our recent study [8], we identified that recoil forces should be large enough at voltages $V \sim 1$ V and conductances near the conductance quantum $G_0 = e_0^2/\pi\hbar \approx 77.5$ μS (e_0 is the elementary charge, $\hbar$ is the Planck's constant) to compete with the interatomic forces, potentially affecting the evolution of the filament; our experiments also favored the presence of such forces. However, the model in [8] lacked the dynamics and used a semi-quantitative continuous description of the filament in the form of a liquid drop with a certain profile. In contrast, in the present work we develop an atomistic quantum theory of electron recoil forces, estimate the latter ones within a toy model of a 1D atomic chain, and further incorporate quantum recoil into a molecular-dynamics (MD) framework with realistic interatomic potentials. Using the latter, we observe a quantum analogue of electromigration triggering a resistive switching event independently of the conventional electrostatically-driven ion migration mechanism. Our results thus indicate that quantum recoil should be taken into account when describing memristive nanodevices and could potentially be used in their rational design to achieve better retention characteristics of their quantized-conductance states.

We use a semiclassical, Born–Oppenheimer description of the nanofilament supporting a quantum current in terms of the tight-binding model:

$$\hat{H} = \hat{H}^{(\mathrm{el})} + \sum_{i=1}^{N} \frac{\mathbf{P}_i^2}{2M_i} + U(\mathbf{X}), \tag{1}$$

$$\hat{H}^{(\mathrm{el})} = \sum_{\sigma} \int \hat{\psi}_\sigma^\dagger(\mathbf{x}) \Big[-\frac{\hbar^2 \nabla^2}{2m} + u(\mathbf{x}; \mathbf{X}) \Big] \hat{\psi}_\sigma(\mathbf{x}) \mathrm{d}^3 x = \\ = \sum_{i,j,\sigma} t_{ij}(\mathbf{X}) \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma}, \tag{2}$$

where m is the electron mass, $\mathbf{X} = (\mathbf{X}_1, \ldots, \mathbf{X}_N)$ and $\mathbf{P}_i = M_i \dot{\mathbf{X}}_i$ are the (classical) coordinates and the momenta of the ions, respectively, $U(\mathbf{X})$ is the "bare" ion-ion potential, $u(\mathbf{x}; \mathbf{X})$ is the effective (Kohn–Sham) potential. We take the electron field operator $\hat{\psi}_\sigma(\mathbf{x})$ with the spin projection $\sigma = \pm 1/2$ expanded over a set of annihilation operators $\hat{c}_{i\sigma}$, one per each atom, yielding transfer integrals/on-site energies t_{ij}. The Ehrenfest equations for the atomic coordinates now give $\dot{\mathbf{P}}_i \approx -\partial U/\partial \mathbf{X}_i + \mathbf{F}_i^{(\mathrm{el})}(\mathbf{X})$, where the "recoil" force acting due to electrons can be written using nonequilibrium Green's functions (NEGFs) [9]:

$$\mathbf{F}_i^{(\mathrm{el})} = \sum_{j,k} \mathbf{f}_{jk}^i \sum_{S=\mathrm{L,R}} \int_{E_0}^{\mu_S} \frac{\mathrm{d}E}{\pi} \left[G_{\mathrm{F}} \Gamma_S G_{\mathrm{F}}^\dagger(E) \right]_{kj}. \tag{3}$$

In this formula, $\mu_{\mathrm{L,R}}$ are the chemical potentials of the two leads, $V = (\mu_{\mathrm{R}} - \mu_{\mathrm{L}})/e_0$ is the applied bias, E_0 is the conduction band bottom, while $G_{\mathrm{F}}(E)$ and $\Gamma_{\mathrm{L,R}}(E)$ are the filament's retarded Green's function and the broadening operators, respectively. The force coefficients $\mathbf{f}_{jk}^i$ are suppressed except for neighboring atoms and can be approximately expressed in terms of the t_{mn} matrix and the inverse atomic radius ζ.

First we apply our model to a 1D chain of atoms with the filament comprised of one, central atom ($i = 0$) and all the other atoms assumed fixed (Fig. 1a); all nearest-neighbor transfer integrals $t_{i,i\pm 1}$ are equal to $t < 0$

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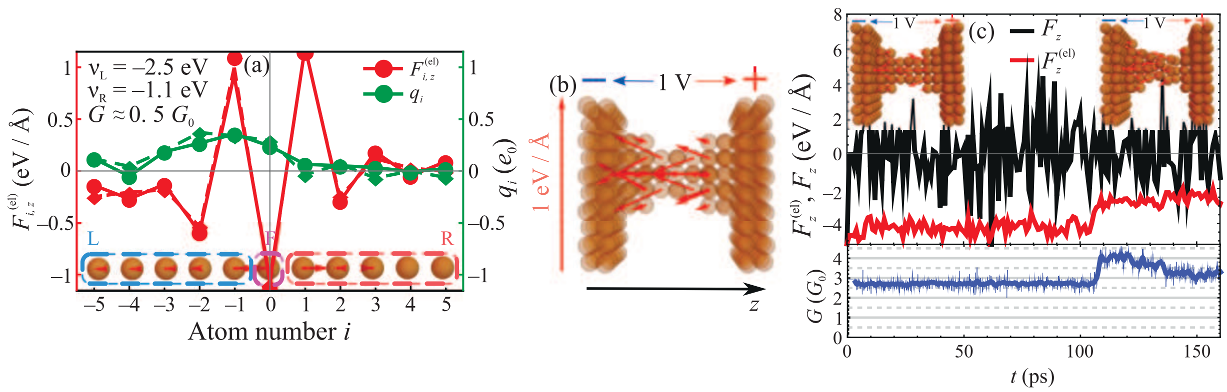

Fig. 1. (Color online) (a) – Forces acting on atoms of a 1D atomic chain and their partial charges at $V = 0$ and 1 V (solid and dashed lines, respectively). (b) – Renormalized recoil forces for a 3D copper filament. (c) – Recoil-driven resistive switching for the 3D filament (warmup and production MD at $T = 300$ K), with the plots showing the evolution of the total renormalized recoil force $\mathbf{F}^{(\mathrm{el})}$ acting on the whole filament, the total EAM + recoil force $\mathbf{F}$, and the conductance G

except $t_{0,\pm 1} = \upsilon_{\mathrm{R,L}}$. Figure 1a demonstrates that the recoil forces can be as large as 1 eV/Å; moreover, a nonzero bias can trigger a Peierls-like instability of the central atom's position [10] with the critical voltage

$$V_{\mathrm{crit}} = \frac{\pi|t|}{e_0|1/2-\beta|}\left[\frac{2q_{\mathrm{eff}}^2}{\zeta^2 a^3|t|} - 1 - 8/\pi\right], \qquad (4)$$

where q_{eff} is the partially screened charges of the ions, a is the interatomic distance, and $\beta = (\mu_{\mathrm{R}} - E_{\mathrm{F}})/e_0 V \in [0,1]$ is the percentage of the voltage drop between the cathode and the Fermi energy. For example, for $a = 1/\zeta = 2.5$ Å, $t = -2$ eV, $q_{\mathrm{eff}} = 0.8e_0$, and $\beta = 0$, the recoil-driven instability appears at $V_{\mathrm{crit}} \approx 1.7$ V, resulting in a voltage-controlled ion displacement.

Next, to simulate the effect of the recoil forces for a realistic copper filament at room temperature – beyond the above toy model at $T = 0$ – we developed a custom MD code implementing the embedded-atom model (EAM) of the interatomic Cu-Cu forces [11] together with the recoil forces (3). To avoid double counting, the latter ones were "renormalized" by subtracting their zero-bias values, which should already be included into the EAM force field. The recoil forces for the initial, fcc-like filament configuration are presented in Fig. 1b, and the MD results in Fig. 1c. During all the simulation time, the recoil force remains directed toward the cathode, facilitating ion migration in this direction, which results in a resistive switching around $t = 110$ ps. Note that the electrostatic field between the electrodes is absent in our simulations, and the ion transport occurs solely due to momentum transfer from conduction electrons. This is a novel mechanism of resistive switching we report here, which is important in the atomic-scale-filaments regime and could potentially be used for rational design of memristive devices.

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