

Comment on “Density Functional Simulation of a Breaking Nanowire”

In a recent Letter [1], Nakamura *et al.* described first principles calculations for a breaking Na nanocontact. Their system consists of a periodic one-dimensional array of supercells, each of which contains 39 Na atoms, originally forming a straight, crystalline wire with a length of 6 atoms. The system is elongated by increasing the length of the unit cell. Aside from a discontinuity of the force occurring at the transition from a crystalline to an amorphous configuration during the early stages of elongation, they were unable to identify any simple correlations between the force and the number of electronic modes transmitted through the contact. An important question is whether their model is realistic, i.e., whether it can be compared to experimental results [2] obtained for a single nanocontact between two macroscopic pieces of metal. In this Comment, we demonstrate that, with such a small unit cell, the interference effects between neighboring contacts are of the same size as the force oscillations in a single nanocontact.

In order to understand how the close proximity of the nanocontacts in the model of Ref. [1] may alter the energetics of the system, we consider a system of two identical nanocontacts in series, connecting two macroscopic wires. We model the metallic nanocontacts as constrictions in a free electron gas, with hard-wall boundary conditions, and obtain the energetics of the system from the electronic scattering matrix [3]. The scattering matrix of the compound system may be obtained as a geometric series in the scattering matrices of the individual contacts (which are taken to be symmetric under inversion, for simplicity), while the scattering matrix of a single contact may be evaluated using the adiabatic and WKB approximations [3], which are quite accurate for contacts of smooth shape [4]. The total grand canonical potential of the system is found to be the sum of the contributions of the individual contacts, plus an interference term

$$\Delta\Omega = -\frac{2}{\pi} \int_0^{E_F} dE \times \sum_{\nu} \tan^{-1} \frac{R_{\nu}(E) \sin[2\theta_{\nu}(E)]}{1 + R_{\nu}(E) \cos[2\theta_{\nu}(E)]},$$

where $R_{\nu}(E)$ and $\theta_{\nu}(E)$ are the reflection probability and scattering phase shift, respectively, of the ν th electronic mode for a single nanocontact.

The correction to the cohesive force in the supercell arrangement of Ref. [1] arising from interference effects between neighboring supercells is $\Delta F = -\partial[\Delta\Omega]/\partial L_{\text{cell}}$, where L_{cell} is the unit cell length. Interference between more widely separated supercells would lead to an additional correction. Figure 1(b) shows that, for the unit cell size considered in Ref. [1] ($L_{\text{cell}} = 17\text{--}31 \text{ \AA} = (2.5\text{--}4.5)\lambda_F$), the interference correction to the cohesive force is comparable to the force oscillations of an indi-

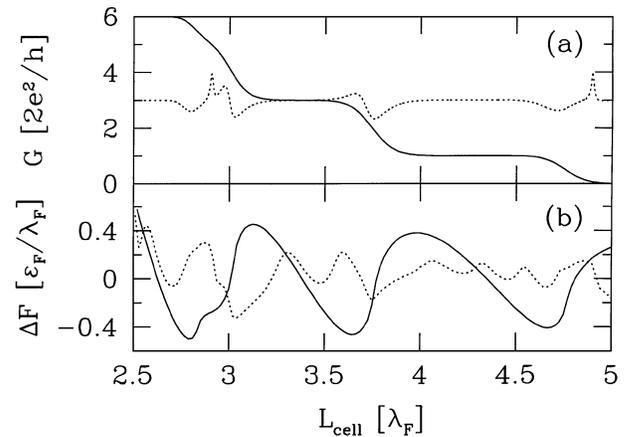


FIG. 1. Conductance and force for a metallic nanocontact with dimensions comparable to that of Ref. [1]: initial radius is λ_F and initial length is $2.5\lambda_F$. (a) Conductance for a single contact (solid curve), and interference term (dotted curve, offset by 3). (b) Force oscillations for a single nanocontact [3] (solid curve), and interference term $\Delta F = -\partial[\Delta\Omega]/\partial L_{\text{cell}}$ (dotted curve).

vidual nanocontact. For comparison, the conductance of a single nanocontact and the interference correction thereof are shown in Fig. 1(a). For a single contact, there is a clear correlation between the conductance steps and the force oscillations. However, the large interference correction would strongly suppress any correlations between the force calculated in the supercell arrangement of Ref. [1] and the conductance of a single contact.

In order to explain the correlations between cohesion and conductance observed experimentally in metallic nanocontacts [2], it is essential to treat the energetics and transport of the system on an equal footing. This has been achieved in our free-electron model [3]. The interference term scales as $\Delta F \sim \mathcal{O}(L_{\text{cell}}^{-1})$ [since $\theta_{\nu}(E) \propto L_{\text{cell}}$], so it would be worthwhile to perform larger-scale “first principles” simulations to address this question.

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