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On universality in metallic nanocohesion

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Abstract

A semiclassical trace formula for the mesoscopic oscillations of the cohesive force in a metallic nanowire of constant cross section is derived. It is shown that the force oscillations are *universal*, in the sense that their r.m.s. amplitude is independent of the area of the cross section, only for cross sections with a continuous one-dimensional symmetry. For a wire of rectangular cross section, the r.m.s. amplitude of the force oscillations is shown to be proportional to the aspect ratio of the cross section.

Keywords: Nanocohesion; Semiclassical techniques

In a seminal experiment [1,2], the cohesive force of atomic-scale gold contacts was found to exhibit oscillations of order one nanonewton as they were pulled apart, which were synchronized with steps of order $2e^2/\hbar$ in the contact conductance. A natural interpretation of these results is that the conductance channels act as delocalized bonds which determine the cohesion of the contact [3]. Refs. [3–6] modeled a metallic nanowire as a gas of independent electrons confined by hard walls, and found that the mesoscopic force oscillations have a characteristic amplitude of order E_F/λ_F, where λ_F is the de Broglie wavelength at the Fermi energy E_F. In this article, we examine under what conditions the amplitude of the force oscillations is *universal*, i.e., proportional to the material parameter E_F/λ_F and independent of the area of the contact.

The mesoscopic oscillations of the cohesive force may be expressed as [3]

$$\delta F = \frac{1}{\beta}\int_0^\infty \mathrm{d}E \frac{\partial[\delta g(E)]}{\partial L}\ln(1+\mathrm{e}^{-\beta(E-\mu)}), \tag{1}$$

where $\delta g = g(E) - g_{\mathrm{Weyl}}(E)$ is the oscillatory part of the electronic density of states (DOS), L is the applied strain, and $\beta = 1/k_B T$. For a system with an f-dimensional symmetry, δg may be expressed using a generalization of Gutzwiller's trace formula as [7]

$$\delta g(E) = \frac{2}{\pi\hbar}\frac{1}{(2\pi\hbar)^{f/2}}\sum_\Gamma \frac{T_\Gamma V_\Gamma}{|\det \tilde{M}_\Gamma - 1|^{1/2}} \times \cos\left(\frac{S_\Gamma}{\hbar} - \frac{\sigma_\Gamma \pi}{2} - \frac{f\pi}{4}\right), \tag{2}$$

where the sum is over f-dimensional families Γ of degenerate periodic orbits, T_Γ, S_Γ, σ_Γ, and $\tilde{M}_\Gamma$ are the period, action, Maslov index, and stability matrix, respectively, of a periodic orbit in Γ, and V_Γ is the f-dimensional volume spanned by the family Γ. Eq. (2) gives the leading-order term of an asymptotic expansion of δg in powers of $\hbar/S_\Gamma$.

The metal forming the nanowire may be taken to be incompressible to the first approximation [4], so that the volume of the nanowire remains constant as it deforms under the applied strain. If the nanowire has a z-independent cross section, the transverse eigenenergies will scale with the applied strain as $\partial\varepsilon_\nu/\partial L = \varepsilon_\nu/L$, and the transverse DOS must have the form

$$\delta g_\perp(E) = \sum_\Gamma \frac{C_\Gamma}{E}(k_E^2 A)^{(2+f)/4}\cos(c_\Gamma\sqrt{k_E^2 A} + \Phi_\Gamma),$$

where $k_E = (2mE/\hbar^2)^{1/2}$, and the constants C_Γ, c_Γ, and Φ_Γ do not depend on the energy or the cross-sectional

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area A. The full three-dimensional DOS δg may be computed from $\delta g_\perp$ by integration over the DOS of the longitudinal motion $g_{||}$; the asymptotic result for large E and A is

$$\delta g(E) = \int_0^E \mathrm{d}E' \delta g_\perp(E') g_{||}(E - E')$$

$$= \sum_\Gamma \frac{C_\Gamma k_E L (k_E^2 A)^{(f+1)/4}}{\sqrt{2\pi c_\Gamma E}} \cos\left(c_\Gamma \sqrt{k_E^2 A} + \Phi_\Gamma - \frac{\pi}{4} \right).$$

This formula could also be derived directly from Eq. (2) with an $f+1$-dimensional degeneracy, including translational invariance.

Inserting the result for $\delta g(E)$ into Eq. (1), we obtain in the limit $k_B T \to 0$

$$\delta F = \sqrt{8\pi} \frac{E_F}{\lambda_F} \sum_\Gamma \frac{C_\Gamma}{c_\Gamma^{3/2}} (k_F^2 A)^{(f-1)/4}$$

$$\times \cos\left(c_\Gamma \sqrt{k_F^2 A} + \Phi_\Gamma + \frac{\pi}{4} \right). \quad (3)$$

The amplitude of the force oscillations is independent of A if and only if the transverse orbits have a one-dimensional degeneracy ($f = 1$), which implies that the transverse motion is integrable, as for circular, elliptic, rectangular, etc. cross sections.

In particular, for a rectangular cross section with sides of length $\alpha^{1/2} D$ and $D/\alpha^{1/2}$, we have to insert $f = 1$, $c_\Gamma = 2\sqrt{m^2\alpha + n^2/\alpha}$, $\Phi_\Gamma = -\pi/4$, and $C_\Gamma = \delta_{mn}/\pi^{3/2}[m^2\alpha + n^2/\alpha]^{1/4}$ in Eq. (3), where δ_{mn} is zero if $m = n = 0$, one if only one of m and n is 0, and 2 otherwise [8]. The mean-squared force oscillations are found from Eq. (3) to be

$$\langle \delta F^2 \rangle = \left(\frac{E_F}{\lambda_F} \right)^2 \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \left(\frac{\delta_{mn}/\pi}{m^2\alpha + n^2/\alpha} \right)^2. \quad (4)$$

For large α, the shortest orbit ($m = 0$, $n = 1$) dominates sum (4), and the root-mean-square amplitude of the force oscillations is proportional to the aspect ratio,

$$\mathrm{rms}\, \delta F \simeq \frac{\alpha E_F}{\pi \lambda_F}, \quad \alpha \gg 1. \quad (5)$$

However, large aspect ratios are energetically unfavorable due to the increased surface energy [3], and will not occur spontaneously.

In conclusion, we find that the mesoscopic oscillations of the cohesive force of spontaneously forming metallic nanowires are indeed universal, i.e. independent of the area of contact, and $\sim E_F/\lambda_F$, provided the cross section possesses a one-dimensional symmetry. The fact that *axial symmetry* is favored by surface tension may explain the universality of the force oscillations observed in gold nanowires [1,2].

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