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Quantum theory of metallic nanocohesion

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Abstract

The conducting and mechanical properties of ultrasmall metallic structures are calculated using the electronic scattering matrix, evaluated in the free electron approximation. Force oscillations of the order ε_F/λ_F are predicted when a metallic quantum wire is stretched to the breaking point, which are synchronized with quantized jumps in the conductance. Coherent backscattering from impurities is shown to lead to fine structure (a "quantum fingerprint") in the force oscillations.

Keywords: Nanocohesion: Conductance quantization

In a pioneering experiment, Rubio, Agraït, and Vieira recently measured simultaneously the force and conductance during the formation and rupture of an atomic-scale Au contact [1]. They observed oscillations in the tensile force of the order 1 nN under deformation, which were synchronized with quantized jumps in the conductance. In a previous paper [2], we showed that this intriguing behavior can be understood quantitatively using a simple jellium model for the conductance and cohesion of a ballistic metallic system, accounting explicitly for quantum-size effects via the electronic scattering matrix. Here, we extend the formalism of Ref. [2] to include the effects of multiple backscattering, which will occur due to the presence of impurities or irregularities in the shape of the contact. We find that multiple backscattering leads to fine structure in the force oscillations, a mechanical analogue of the so-called universal conductance fluctuations (UCF) [3].

The success of the jellium approximation in explaining the energetics of ultrasmall metal clusters [4, 5] motivates its application to mesoscopic (open) metallic systems, which are the subject of interest here. We investigate the conducting and mechanical properties of a nanoscopic constriction connecting two macroscopic metallic reservoirs. This is a quantum mechanical scattering problem. The formulation of electrical transport in terms of the scattering matrix was developed by Landauer [6, 7] and Büttiker [8], while the formulation of the free energy of open systems in terms of the scattering matrix was first developed by Dashen et al. [9], and was recently revived in the context of the persistent current problem by Akkermans et al. [10]. Neglecting electron–electron interactions, the electrical conductance of the system may be expressed

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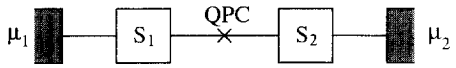

Fig. 1. inset

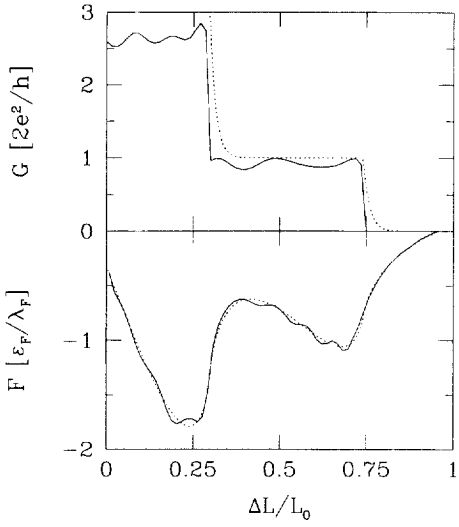

Fig. 1. Electrical conductance G and tensile force F of a metallic nanoconstriction versus the elongation $\Delta L/L_0$. Impurities with backscattering probabilities $\alpha_1 = 0.06$ and $\alpha_2 = 0.03$ were placed at opposite ends of the constriction, and the initial length and radius of the constriction were $k_F L_0 = 40$ and $k_F R = 5$, respectively. The dashed line indicates the results without impurity scattering. Inset: Effective circuit for the system.

as [6]

$$G = \frac{2e^2}{h}\int dE \frac{-df}{dE}\sum_v T_v(E), \tag{1}$$

and the grand canonical potential of the system may be written [2]

$$\Theta_v(E) = \begin{cases} \theta_v(E) + \tan^{-1}\left[\dfrac{\sqrt{\alpha_1\alpha_2}\sin 2\theta_v(E)}{1-\sqrt{\alpha_1\alpha_2}\cos 2\theta_v(E)}\right], & E > \varepsilon_v(0), \\ \theta_v(E) + \tan^{-1}\left[\dfrac{\sqrt{\alpha_1}\cos\theta_v(E)}{1-\sqrt{\alpha_1}\sin\theta_v(E)}\right] + \tan^{-1}\left[\dfrac{\sqrt{\alpha_2}\cos\theta_v(E)}{1-\sqrt{\alpha_2}\sin\theta_v(E)}\right], & E > \varepsilon_v(0). \end{cases} \tag{6}$$

$$\Omega = -\frac{2}{\pi}\int dE\, f(E)\sum_v \Theta_v(E), \tag{2}$$

where $f(E) = \{\exp[\beta(E-\mu)]+1\}^{-1}$ is the Fermi distribution function, and $T_v(E)$ and $\Theta_v(E)$ are, respectively, the transmission probability and scattering phase shift in scattering channel v. Eqs. (1) and (2) allow one to treat the conducting and mechanical properties of a confined electron gas on an equal footing, and provide the starting point for our calculation.

To be specific, let us consider an axially symmetric adiabatic constriction of length L in an infinitely long quantum wire, with electrons confined along the z-axis by a hard-wall potential at $r = r(z)$. The transverse motion is quantized, with mode energies $\varepsilon_v(z) = \hbar^2\gamma_v^2/2mr(z)^2$, where $\{\gamma_v\}$ are the zeros of the Bessel functions. As the constriction is elongated, $r(z)$ decreases to preserve the volume of the system (ideal plastic deformation). Backscattering is introduced by two scattering centers, to the right and left of the constriction (see inset of Fig. 1), with scattering matrices

$$S_j = \begin{pmatrix} \alpha_j^{1/2} & \mathrm{i}(1-\alpha_j)^{1/2} \\ \mathrm{i}(1-\alpha_j)^{1/2} & \alpha_j^{1/2} \end{pmatrix}\delta_{vv'}, \tag{3}$$

where α_j is the backscattering probability from impurity j (intermode impurity scattering is neglected). The scattering matrix for the combined system may be obtained straightforwardly, and Eq. (1) yields, in the limit $T = 0$,

$$G = \frac{2e^2}{h}\sum_{v<v_F}\frac{(1-\alpha_1)(1-\alpha_2)}{1+\alpha_1\alpha_2 - 2\sqrt{\alpha_1\alpha_2}\cos 2\theta_v(\varepsilon_F)}, \tag{4}$$

where

$$\theta_v(E) = (2m/\hbar^2)^{1/2}\int_0^L dz[E-\varepsilon_v(z)]^{1/2} \tag{5}$$

is the phase shift in the absence of impurity scattering. The total scattering phase shift in channel v is found to be

The tensile force under elongation is given by $F = -\partial\Omega/\partial L$, where Ω is calculated from Eqs. (2), (5), and (6).

Fig. 1 shows the conductance and force of a metallic nanoconstriction with impurity scattering as a function of the elongation, calculated from Eqs. (2)–(6). The results without impurity scattering are shown for comparison as a dashed line. The correlations between the force and the conductance are striking: $|F|$ increases along the conductance plateaus, and decreases sharply when the conductance drops. This behavior reflects the fact that cohesion, like conduction, is mediated by the quantized transverse modes in the constriction [2]. One sees that the conductance is suppressed below integer values due to backscattering, while the gross features of the force fluctuations are not shifted. Both the conductance and force exhibit fine structure, a "quantum fingerprint" of the microscopic disorder. Fig. 1 is remarkably similar to the experimental results of Ref. [1], both qualitatively and quantitatively. Inserting the value $\varepsilon_F/\lambda_F \cong 1.7$ nN for Au, we see that the calculated force is in quantitative agreement with the last two force oscillations shown in Fig. 1 of Ref. [1]. Fine structure in the tensile force similar to that shown here was also observed in Ref. [1], raising the intriguing possibility that the mechanical analogue of UCF may already have been observed. Further experimental work will be necessary to show that this fine structure is reproducible, and may thus constitute a quantum fingerprint of the disorder. Perhaps the most promising possibility is to measure the Aharanov–Bohm effect for two metallic point contacts in parallel, which can be shown to lead to a similar, flux-periodic, fine structure in the force oscillations, with an amplitude up to ε_F/λ_F.

In conclusion, we have investigated quantum interference effects on the mechanical properties of metallic nanostructures using the electronic scattering matrix, calculated in the noninteracting jellium approximation. This model is able to explain quantitatively recent experiments on the conducting and mechanical properties of ultrasmall metallic contacts [1], and should be applicable to a wide variety of problems in the rapidly evolving field of nanomechanics.

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