Driving-voltage-induced mechanical force oscillations in metal quantum-point contacts

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We predict that the mesoscopic tensile force fluctuations in metal quantum-point contacts (nanowires) arise as a result of finite electric voltage on the contact. They are due to reconfiguration of the electronic subsystem and are correlated with the nonlinearities of the current-voltage characteristics of the contact. The observation of the effect would directly confirm the recently suggested ''free-electron'' mechanism of mesoscopic force fluctuations observed in nanowires under deformation. The related magnetic susceptibility fluctuations and role of topology of the wire cross section are discussed as well. [S0163-1829(98)00147-7]

The quantization of electrical conductance of three-dimensional (3D) quantum-point contacts[1] (QPC) has been observed in a variety of metal mesoscopic contacts.[2] The simplest model of the 3D QPC is a ''nanowire'' of length L and diameter $d \sim \lambda_F$, connecting two bulk conducting reservoirs, where λ_F is the Fermi wavelength in the system (see Fig. 1), and the underlying atomic structure is the positively charge ''jellium.'' It is the quantized character of transverse motion of electrons that is revealed in conductance quantization as a function of d. Since the applied driving voltage changes the population of occupied subbands, nonlinear current-voltage dependence quantum-point contacts, was predicted.[3–6] The theory is in a good agreement with experiment on 2D and 3D systems.[7] On the other hand, the nonlinear conductivity of 3D bismuth QPC (Ref. 8) shows only qualitative agreement with the theory, while measurements on 3D gold QPCs (Ref. 9) did not show the predicted type of nonlinearities altogether.[10]

The experiments on metal QPCs under deformation showed that the mechanical stress in the wire fluctuates as a function of its elongation, and the fluctuations are correlated with the conductance jumps.[11] This seemed to require a more complex mechanism, and the often-used explanation of these phenomena invokes the atomic rearrangement processes, and is supported by molecular-dynamics calculations (e.g., Ref. 12).

Recently, an elegant alternative explanation of the force fluctuations was suggested, based on the reaction of the free electrons to the mechanical deformation of the contact region.[13] The role of the atomic structure of the wire was again reduced to providing a ''jellium'' background. In this ''free-electron'' model, the longitudinal force, being the coordinate derivative of the thermodynamic potential, is sensitive to the positions and occupancy of electronic subbands in the wire. The latter depends on the shape of the cross section and on the elongation of the wire (assumed to take place at a constant volume). The positions of cusps in F as a function of the elongation would naturally coincide with those of the conductance steps.

In this paper we show that the ''free-electron'' mechanism of tensile force fluctuations will lead to related effects: mechanical force and magnetic susceptibility fluctuations in a nanowire as a function of applied driving voltage at the same values of eU, as the features of the differential conductance $G_{\mathrm{diff}}(eU)$. Investigation of these effects can be done using existing experimental techniques and would provide an independent test of the mechanism suggested in Ref. 13 and confirm the role of an electronic subsystem in determining both transport and mechanic properties of metal quantum contacts.

We start from the expression for the grand potential of the electronic subsystem at zero temperature and voltage for a wire of uniform cross section (Blom *et al.*[13]):

$$\Omega(E_F) = -\frac{4}{3} L \sqrt{\frac{2m^*}{\pi^2 \hbar^2}} \sum_n \left[E_F - E_n(L,V)\right]^{3/2} \times \theta\left[E_F - E_n(L,V)\right]. \tag{1}$$

Here m^* is the effective mass of an electron, $\theta(x)$ is the Heaviside step function, and E_n is the energy of nth electronic transverse mode in the wire, which depends on the wire length L and volume V. We assume that the length of the wire is much larger than its diameter d, which allows us to set the electrical potential to zero in the wire. (Due to

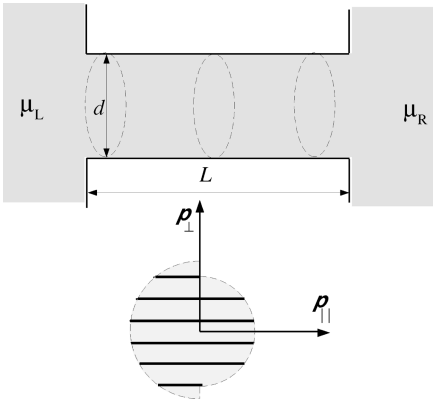

FIG. 1. Schematic view of a metal quantum-point contact (nanowire) and distribution function of right- and left-moving electrons in the wire at finite driving voltage. Solid lines correspond to quantized values of transverse momentum.

screening in metal the effects of finite bias will be felt only at distances $\approx d$ from the ends; see Ref. 4 and references therein.)

For the sake of simplicity, we will neglect both elastic and inelastic scattering in the nanowire. Their contributions are of order $(L,d)/l_s$, where l_s is the corresdponding scattering length.[14] The condition $(L,d)/l_s \ll 1$ is satisfied both for the elastic and electron-phonon scattering. In the latter case we use the estimate $l_{e\text{-}ph} \sim \hbar v_F/\lambda\omega_D$, where ω_D is Debye frequency, and $\lambda < 1$ is the electron-phonon coupling constant.

The electron-electron scattering length can be estimated as $l_{e\text{-}e} \sim \hbar v_F/\epsilon_F(\epsilon_F/eV)^2$, the bias eV playing the role of effective temperature. For the effects of electron-electron scattering to be small we need $d/l_{e-e} \ll 1$ (because the longitudinal momentum conservation and transverse quantization in the electronic subsystem suppress the electron-electron scattering inside the nanowire). The corresponding restriction on the applied voltage is $eV < \epsilon_F/\sqrt{N_\perp}$, where $N_\perp \sim d/\lambda_F$ is the number of transverse channels in the quantum contact. This condition is compatible with bias being of the same order as the interlevel spacing in the contact, $\Delta\epsilon \sim \epsilon_F/N_\perp$, which is necessary for the observation of nonlinear effects discussed in this paper.

Under the above assumptions, we can consider right-moving and left-moving electrons as two independent subsystems, with chemical potentials μ_L and μ_R, respectively[14,15] (Fig. 1):

$$\mu_L = E_F - (1-\beta)eU; \quad \mu_R = E_F + \beta eU. \tag{2}$$

The grand potential thus becomes

$$\Omega_{eU} = \frac{1}{2}\{\Omega(E_F+\beta eU) + \Omega[E_F-(1-\beta)eU]\}. \tag{3}$$

Parameter β determines the asymmetry of the voltage drop on the contact. [Usually symmetric voltage drop is assumed ($\beta = 1/2$), in which case the differential conductance is always a multiple of $\frac{1}{2}G_Q$.[3] Generally β can deviate from 1/2 (Ref. 7) and be voltage dependent.] It should be in principle determined self-consistently by solving the electrostatical problem for the wire and its surroundings at given eU.[10] We consider here two limiting cases: (a) symmetric voltage drop, $\beta = 1/2$, and (b) perfect screening. In the latter case $\beta(eU)$ is determined from the condition of no charge accumulation in the wire,

$$N(E_F+\beta eU) + N[E_F-(1-\beta)eU] = 2N(0), \tag{4}$$

where

$$N(E) = 2L\sqrt{\frac{2m^*}{\pi^2\hbar^2}}\sum_n \theta(E-E_n)\sqrt{E-E_n}. \tag{5}$$

It is easy to see, that the differential conductance of the system is given by

$$\begin{aligned} G_{\text{diff}}(eU) &= \frac{dI}{dU} = \frac{d}{dU}\frac{2e}{h}\int_{E_F-(1-\beta)eU}^{E_F+\beta eU} dE \sum_n \theta(E-E_n) \\ &= G_Q \sum_n \Big(\beta\,\theta(E_F+\beta eU-E_n) \\ &\quad + (1-\beta)\,\theta[E_F-(1-\beta)eU-E_n] \\ &\quad + U\frac{d\beta}{dU}\{\theta(E_F+\beta eU-E_n) \\ &\quad - \theta[E_F-(1-\beta)eU-E_n]\}\Big), \end{aligned} \tag{6}$$

where $G_Q = 2e^2/h$ is the unit quantum conductance. Therefore $G_{\text{diff}}(eU)$ shows a structure at the voltages when consequent transverse energy levels enter the current-carrying interval $[E_F-(1-\beta)eU, E_F+\beta eU]$. In the limit of perfect screening $\beta(eU)$ was found numerically for both models we considered: the wire of square cross section $d\times d$, and the round wire of diameter d.

The quantized levels in the wire are given by

$$E_{mn} = \begin{cases} E_0(m^2+n^2) & \text{(square)} \\ \dfrac{4E_0}{\pi^2}\gamma_{mn}^2 & \text{(round)}. \end{cases} \tag{7}$$

Here $E_0 = \pi^2\hbar^2/(2m^*d^2)$, and γ_{mn} denotes the nth positive zero of the Bessel function $J_m(z)$; m is the magnetic quantum number. The mechanical force is given by

$$\begin{aligned} F(u) &= -\left(\frac{\partial\Omega}{\partial L}\right)_V = F_0\sum_{mn}\{f[\epsilon_F+\beta(u)u;\epsilon_{mn}] \\ &\quad + f[\epsilon_F-(1-\beta(u))u;\epsilon_{mn}]\}, \end{aligned} \tag{8}$$

where $\epsilon_F = E_F/E_0$; $u = eU/E_0$; $\epsilon_{mn} = E_{mn}/E_0$; $F_0 = \pi^2\hbar^2/(2m^*d^3)$, and $f(x;y) = [(2/3)(x-y)^{3/2} - (x-y)^{1/2}y]\theta(x-y)$. The nonlinear dependence $F(eU)$ and $G_{\text{diff}}(eU)$ in both limiting cases is shown in Fig. 2. The nonlinear conductance is strongly dependent on the character of screening in the wire. On the other hand, the qualitative character of the force fluctuations is the same, and mechanical force and differential conductance still show singularities at the same applied voltages in both limits.[16] The absolute magnitude of force fluctuations for $d \sim 1$ nm is of order 1 nN, in agreement with earlier results.[11,13]

Another way of changing positions of quantized levels, and thus the properties of the contact, is by applying longitudinal magnetic field. The characteristic field sweep scale, corresponding to interlevel spacing, is though $\sim\Phi_0/d^2$, where $\Phi_0 = hc/e$ is the magnetic flux quantum.[1,6,17] For a metal QPC with $d \sim 1$ nm this yields unrealistic fields of order 10^3 T. This means that in metal contacts, appreciable dependence of the contact's properties on the magnetic field can take place only when the Fermi level is already very close to one of the quantized energy levels. This can be achieved, e.g., by mechanical deformation of the contact, or by applying finite driving voltage. We will concentrate on the latter possibility, which is reversible and promises better opportunities for the necessary fine tuning.

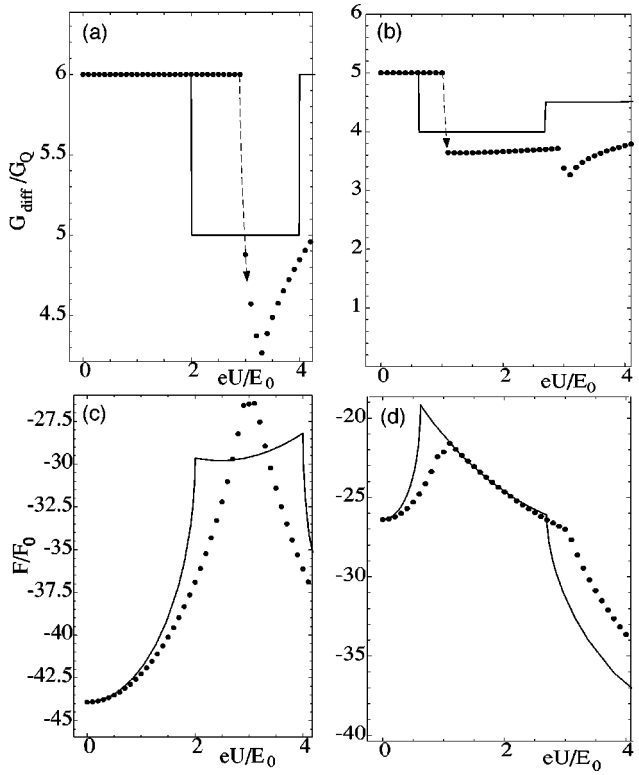

FIG. 2. Differential conductance and force fluctuations vs applied voltage in a nanowire of square cross section $d\times d$ [(a) and (c)] or circular cross section of diameter d [(b) and (d)]. The force and bias are measured in units of $F_0=\pi^2\hbar^2/(2m^*d^3)$ and $E_0=\pi^2\hbar^2/(2m^*d^2)$, respectively. The Fermi energy is $E_F=11E_0$. Solid line: symmetric voltage drop ($\beta=0.5$). Dots: perfect screening.

The magnetic field can be taken into account in the perturbation theory,[18] valid in the limit of the weak field (large cyclotron radius, $r_c\gg d$) (Ref. 19) one finds for the quantized transverse levels,

$$\tilde{E}^s_{mn}(\eta)=\frac{4E_0}{\pi^2}\left(\gamma^2_{mn}+2m\eta+4\frac{m^*}{m_0}g\eta s\right)+O(\eta^2). \quad (9)$$

Here η is the magnetic field measured in units of $H_0=(hc/e)/(\pi d^2/4)$. Since $\eta\ll 1$, we keep in Eq. (9) only linear in η terms, including the spin splitting (the last term in the parentheses). Here $s=\pm 1$ is the projection of spin, g is the g factor, and m_0 is the free electron mass.

The differential conductance and force fluctuations are thus given by

$$G_{\rm diff}(\eta,u)=\frac{1}{2}G_Q\sum_{s=\pm 1}\sum_{n=1}^{\infty}\sum_{m=-\infty}^{\infty}\Big(\theta[\epsilon_F+\beta u-\tilde{\epsilon}^s_{mn}(\eta)]+\theta[\epsilon_F-(1-\beta)u-\tilde{\epsilon}^s_{mn}(\eta)]+u\frac{d\beta}{du}\{\theta[\epsilon_F+\beta u-\tilde{\epsilon}^s_{mn}(\eta)]-\theta[\epsilon_F-(1-\beta)u-\tilde{\epsilon}^s_{mn}(\eta)]\}\Big), \quad (10)$$

$$F(\eta,u)=\frac{1}{2}F_0\sum_{s=\pm 1}\sum_{n=1}^{\infty}\sum_{m=-\infty}^{\infty}\{f[\epsilon_F+\beta u;\tilde{\epsilon}^s_{mn}(\eta)]+f[\epsilon_F-(1-\beta)u;\tilde{\epsilon}^s_{mn}(\eta)]\}, \quad (11)$$

where $\tilde{\epsilon}_{mn}\equiv\tilde{E}_{mn}/E_0$. The factors of one-half before G_Q, F_0 reflect the spin splitting in the magnetic field of previously degenerate energy levels.

The magnetization of the wire is

$$\mathcal{M}(\eta,u)=-\frac{1}{V}\left(\frac{\partial\Omega}{\partial H}\right)_{V,T}=\frac{m_0}{m^*}\mu_B\sum_{s,m,n}\left(-\frac{\partial\tilde{\epsilon}^s_{mn}(\eta)}{\partial\eta}\right)\times\{[\epsilon_F+u/2-\tilde{\epsilon}^s_{mn}(\eta)]^{1/2}\theta[\epsilon_F+u/2-\tilde{\epsilon}^s_{mn}(\eta)]+[\epsilon_F-u/2-\tilde{\epsilon}^s_{mn}(\eta)]^{1/2}\theta[\epsilon_F-u/2-\tilde{\epsilon}^s_{mn}(\eta)]\}, \quad (12)$$

where $\mu_B=e\hbar/(2m_0c)$ is the Bohr magneton.

The effects of the applied weak magnetic field are described by the magnetoconductance coefficient $\sigma(u)\equiv(\partial G_{\rm diff}/\partial H)_{V,T;H=0}$, magnetotension coefficient $\Upsilon(u)\equiv(\partial F/\partial H)_{V,T;H=0}$, and the magnetic susceptibility $\chi(u)=(\partial\mathcal{M}/\partial H)_{V,T;H=0}$.

Keeping only the singular terms, we find the following expressions:

$$\sigma(eU)=-\frac{1}{2}G_Q\sum_{s,n,m}\left(\frac{\partial\tilde{\epsilon}^s_{mn}(\eta)}{\partial\eta}\right)_{\eta=0}\Big(\delta[\epsilon_F+\beta u-\tilde{\epsilon}^s_{mn}(0)]+\delta[\epsilon_F-(1-\beta)u-\tilde{\epsilon}^s_{mn}(0)]+u\frac{d\beta}{du}\{\delta[\epsilon_F+\beta u-\tilde{\epsilon}^s_{mn}(0)]-\delta[\epsilon_F-(1-\beta)u-\tilde{\epsilon}^s_{mn}(0)]\}\Big), \quad (13)$$

$$\Upsilon(eU)\approx\frac{F_0}{2H_0}\sum_{s,n,m}\tilde{\epsilon}^s_{mn}(0)\times\left(\frac{\partial\tilde{\epsilon}^s_{mn}(\eta)}{\partial\eta}\right)_{\eta=0}\left\{\frac{\theta[\epsilon_F+\beta u-\tilde{\epsilon}^s_{mn}(0)]}{[\epsilon_F+\beta u-\tilde{\epsilon}^s_{mn}(0)]^{1/2}}+\frac{\theta[\epsilon_F-(1-\beta)u-\tilde{\epsilon}^s_{mn}(0)]}{[\epsilon_F-(1-\beta)u-\tilde{\epsilon}^s_{mn}(0)]^{1/2}}\right\}, \quad (14)$$

Note that the magnetoconductance and magnetotension coefficients contain the first power of $\partial\tilde{\epsilon}^s_{mn}(\eta)/\partial\eta$. Therefore they are *exactly* zero, due to cancellation of terms with opposite m,s:

$$\sigma=0; \quad \Upsilon=0. \quad (15)$$

This means, that the magnetoconductance and magnetotension in a metal quantum contact are the effects of second order in $\eta\ll 1$. They would thus appear as numerically small, extremely narrow peaks in voltage dependence of the corresponding functions, and cannot be in a satisfactory way investigated in our simple model, neglecting the effects of finite temperature and scattering.

On the contrary, the magnetic susceptibility contains the second power of $\partial\tilde{\epsilon}^s_{mn}(\eta)/\partial\eta$, and is thus nonzero:

$$\chi(eU)\approx\frac{m_e\mu_B}{2m^*H_0}\sum_{s,m,n}\left(\frac{\partial\tilde{\epsilon}^s_{mn}(\eta)}{\partial\eta}\right)^2_{\eta=0}\times\left\{\frac{\theta[\epsilon_F+\beta u-\tilde{\epsilon}^s_{mn}(0)]}{[\epsilon_F+\beta u-\tilde{\epsilon}^s_{mn}(0)]^{1/2}}+\frac{\theta[\epsilon_F-(1-\beta)u-\tilde{\epsilon}^s_{mn}(0)]}{[\epsilon_F-(1-\beta)u-\tilde{\epsilon}^s_{mn}(0)]^{1/2}}\right\}. \quad (16)$$

It demonstrates a series of inverse square-root singularities at the same values of driving voltage as the features of differential conductance and force fluctuations (see Fig. 3). The features of $\chi(eU)$ are better pronounced than those of the former coefficients, which could outweigh the small magnitude of the effect and make measurements of magnetic susceptibility of a metal point contact a more sensitive tool for investigation of electronic density and potential redistribution in metal point contacts.

In conclusion, using a simple model, we showed that finite driving voltage can lead to mechanical force fluctuations and singularities of magnetic susceptibility in metal quantum contacts. The mechanism of these effects is voltage-induced nonequilibrium redistribution of electrons over quasi-1D subbands in the contact. On the other hand, magnetoconductance and magnetotension coefficients are shown to be exactly zero, and the corresponding effects to be at least of order $(H/H_0)^2$, where $H_0\approx10^3$ T in a nanometer-size contact.

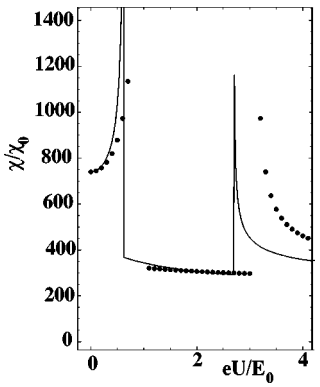

FIG. 3. Magnetic susceptibility vs applied voltage in a nanowire of round cross section. The unit $\chi_0=m_e\mu_B/(m^*H_0)$, where $H_0=8\hbar c/(d^2e)$. We chose $g=2$, $m^*=m_e$.

Experimental investigation of the predicted effects would clarify the role played by electronic subsystem in behavior of metal quantum contacts.

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[15] This picture remains qualitatively valid even in case of strong elastic scattering, $(L,d)/l_{\rm elastic}\gg1$, as long as $(L,d)\ll\sqrt{l_{\rm elastic}l_{\rm inelastic}}$. There will be still two distinct groups of electrons in every point of the contact, with maximum energies differing by eU, even though the elastic scattering will smear the sharp edges of the momentum distribution function of Fig. 1

(see I. O. Kulik, R. I. Shekhter, and A. G. Shkorbatov, Zh. Éksp. Teor. Fiz. **81**, 2126 (1981) [Sov. Phys. JETP **54**, 1130 (1981)]). The singularities that we are discussing here will be smeared as well in this limit (which is the case when there are numerous defects in the contact area).

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[19] Condition $r_c \gg d$ is equivalent to $\eta \ll k_F d \approx G_{\rm diff}/G_Q$. Since in our system the latter is at least of order unity, this ''weak-field'' condition is satisfied by any realistic field.