

Friedel oscillations in the presence of transport currents in a nanowire

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(Received 17 April 2007; revised manuscript received 28 June 2007; published 17 October 2007)

We investigate the Friedel oscillations in a nanowire coupled symmetrically to two macroscopic electrodes of different potentials. We show that the wavelength of the density oscillations monotonically increases with the bias voltage, whereas the amplitude and the spatial decay exponent of the oscillations remain intact. Using the nonequilibrium Keldysh Green functions, we derive an explicit formula that describes voltage dependence of the wavelength of the Friedel oscillations.

DOI: [10.1103/PhysRevB.76.165419](https://doi.org/10.1103/PhysRevB.76.165419)

PACS number(s): 73.63.Nm, 71.45.Lr

I. INTRODUCTION

Transport properties of nanosystems, e.g., nanowires or single molecules, have recently been receiving significant attention mainly due to their possible application in future electronic devices.¹⁻⁴ These properties strongly differ from those of macroscopic conductors. The most important obstacle in theoretical investigations of the transport phenomena originates from the coupling between nanosystem and macroscopic leads. Because of this coupling, analysis of the electron correlations is more difficult than in the equilibrium case.

In nanosystems the charge carriers are usually distributed inhomogeneously. There exist several reasons for such an inhomogeneity:

(i) First, it may originate from a spatial confinement.⁵ In analogy to the case of a quantum well, one may expect that due to a small size of nanosystems, electrons are inhomogeneously distributed. In particular, recent scanning tunneling spectroscopy has shown the presence of the electronic standing waves at the end of a single-wall carbon nanotube.⁶

(ii) Additionally, in the transport phenomena it may originate from the applied voltage.^{1,7,8} In this case the system properties are determined by the chemical potentials of the left and right electrodes. Different values of these potentials may lead to an inhomogeneous distribution as well.

(iii) Similarly to the macroscopic case, inhomogeneous charge distribution in nanosystems should occur in the presence of impurities.⁹

(iv) Nanowires or molecular wires represent quasi-one-dimensional conductors. Therefore, phenomena typical for low dimensional systems, e.g., charge density waves may occur as well.¹⁰⁻¹⁴ Recently it has been shown that the charge density waves are strongly modified by the bias voltage.¹⁵ Apart from the low-voltage regime, they are incommensurate and the corresponding wave vector decreases discontinuously with the increase of the bias voltage.

In this paper we focus on the impurity-induced inhomogeneities. It is known that an impurity in the electron gas produces local changes of the carrier concentration, known as the Friedel oscillations,⁹ that asymptotically decay with the distance from the impurity. The most of recent theoretical investigations of the Friedel oscillations concerned the influence of the electronic correlations, that is of crucial importance in one-dimensional systems.¹⁶⁻²² It has been shown

that correlations suppress the decay of the density oscillations.^{17,19} It is interesting that these oscillations give information about the impurities¹⁹ as well as the electron-electron interaction in Luttinger liquid systems.^{18,20} In macroscopic systems, the Friedel oscillations are characterized by the wave vector $Q=2k_F$, where k_F is the Fermi momentum. If the nanosystem is isolated (or more generally, is in equilibrium), k_F is a well defined quantity. However, in the transport experiments the nanosystem is coupled to two macroscopic leads with different Fermi levels and the difference between these Fermi energies increases with the bias voltage. Therefore, the meaning of k_F is ambiguous. Since the properties of a nanosystem are determined by the chemical potentials of the left and right electrodes, one may expect that the Friedel oscillations should depend on the voltage as well. In this paper we analyze this dependence using the formalism of the nonequilibrium Keldysh Green functions. In particular, we derive an explicit formula for the voltage dependence of the wavelength of the Friedel oscillations.

Before presenting a detailed analysis of the nonequilibrium case, we present some equilibrium arguments, which support the above scenario. Let us consider a nanowire described by a tight-binding Hamiltonian, with the nearest-neighbor hopping. We assume for a moment an equilibrium case with the nanosystem connected only to the left lead with the chemical potential being the same as in the transport experiments, i.e., $\mu_L=eV/2$, where V is the bias voltage. For $eV=0$ one gets the average occupation number $n=1$, Fermi momentum $k_F=\pi/2$, and the wave vector of the Friedel oscillations is $Q=\pi$. Then, the increase of μ_L modifies the occupation number ($n=1+\Delta n=n_L$) and shifts Q ($Q=\pi+\Delta Q$). One may assume that the Friedel oscillations decay according to $\cos(Qx)/x^\delta=\cos(\pi x)\cos(\Delta Qx)/x^\delta$, for integer x . Now one can carry out a similar reasoning for the case when the system is connected only to the right lead with the chemical potential $\mu_R=-eV/2$. In this case $n=1-\Delta n=n_R$, $Q=\pi-\Delta Q$, and, because of the parity of the cosine function, the Friedel oscillations decay exactly in the same way. In this picture the wave vector Q vanishes when eV is equal to the bandwidth. Then, in a system connected to the left lead the occupation number is 2 and the Fermi momentum is π , whereas in the system connected to the right lead the number of electrons as well as the Fermi momentum vanish. The aim of this paper is to show that the Friedel oscillations behave in the same way also when the nanosys-

tem is connected symmetrically to both the leads. In this case, the occupation number becomes an average of n_L and n_R and remains independent of the bias voltage.

The paper is organized as follows. In Sec. II we discuss a microscopic model and details of calculations. Numerical results are presented in Sec. III. Approximate analytical formulas are derived in Sec. IV. The last section contains a discussion and concluding remarks.

II. MODEL AND THE CALCULATIONS SCHEME

We investigate a one-dimensional nanowire with its ends coupled to macroscopic leads. The system under consideration is described by the Hamiltonian

$$H = H_{\text{el}} + H_{\text{nano}} + H_{\text{nano-el}}, \quad (1)$$

where H_{el} , H_{nano} , and $H_{\text{nano-el}}$ describe leads, nanowire, and the coupling between the wire and leads, respectively. We assume that electrodes are described by the free electron gas with a wide energy band

$$H_{\text{el}} = \sum_{\mathbf{k}, \sigma, \alpha} (\varepsilon_{\mathbf{k}, \alpha} - \mu_{\alpha}) c_{\mathbf{k}\sigma\alpha}^{\dagger} c_{\mathbf{k}\sigma\alpha}, \quad (2)$$

where μ_{α} is the chemical potential and $\alpha \in \{L, R\}$ indicates the left or right electrode. $\mu_L - \mu_R = eV$, with V being the bias voltage. $c_{\mathbf{k}\sigma\alpha}^{\dagger}$ creates an electron with momentum \mathbf{k} and spin σ in the electrode α . The Hamiltonian of the nanosystem is given by

$$H_{\text{nano}} = -t \sum_{\langle ij \rangle \sigma} d_{i\sigma}^{\dagger} d_{j\sigma} + U \sum_{\sigma} n_{l\sigma}. \quad (3)$$

Here, $d_{i\sigma}^{\dagger}$ creates an electron with spin σ at site i of the nanosystem, $n_{i\sigma} = d_{i\sigma}^{\dagger} d_{i\sigma}$ and U is the impurity potential. We have assumed a single impurity localized at site l . This impurity is responsible for scattering of electrons between all possible states in the momentum representation. The coupling between the nanowire and the leads is given by

$$H_{\text{nano-el}} = \sum_{\mathbf{k}, i, \alpha, \sigma} (g_{\mathbf{k}, i, \alpha} c_{\mathbf{k}\sigma\alpha}^{\dagger} d_{i\sigma} + \text{H.c.}). \quad (4)$$

In the following, we assume that the matrix elements $g_{\mathbf{k}, i, \alpha}$ are nonzero only for the edge atoms of the nanowire.

The electron distribution has been determined with the help of the nonequilibrium Keldysh Green functions. Here, we follow the procedure used by Kostyrko and Buřka in Ref. 7. In particular, the local carrier density is expressed by the lesser Green function

$$\langle d_{i\sigma}^{\dagger} d_{i\sigma} \rangle = \frac{1}{2\pi i} \int d\omega G_{i\sigma, i\sigma}^{<}(\omega). \quad (5)$$

This quantity, in turn, is determined by the retarded and advanced Green functions in the following way:

$$\hat{G}^{<}(\omega) = i \sum_{\alpha \in \{L, R\}} \hat{G}^r(\omega) \hat{\Gamma}_{\alpha}(\omega) \hat{G}^a(\omega) f_{\alpha}(\omega), \quad (6)$$

where

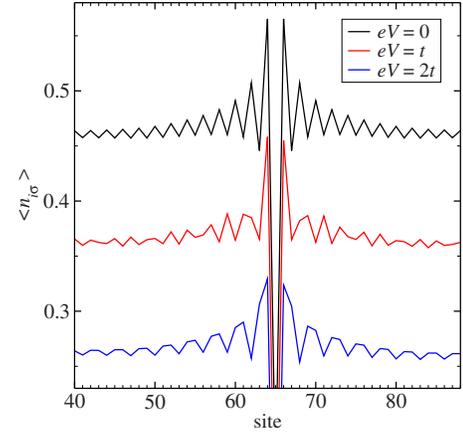


FIG. 1. (Color online) Occupation of sites in the vicinity of the impurity for a 129-site nanowire. The bias voltage is explicitly indicated in the legend. For the sake of clarity, the curves for $eV=t$ and $eV=2t$ have been shifted downward by 0.1 and 0.2, respectively. $k_B T = 0.01t$ and $U = 2t$ have been assumed.

$$[\hat{\Gamma}_{\alpha}(\omega)]_{ij} = 2\pi \sum_{\mathbf{k}} g_{\mathbf{k}, i, \alpha}^* g_{\mathbf{k}, j, \alpha} \delta(\omega - \varepsilon_{\mathbf{k}, \alpha}). \quad (7)$$

The Fermi distribution functions of the left and right electrodes read

$$f_L(\omega) = f\left(\omega - \frac{eV}{2}\right), \quad f_R(\omega) = f\left(\omega + \frac{eV}{2}\right), \quad (8)$$

with $f(x) = [\exp(x/k_B T) + 1]^{-1}$. The retarded Green function can be calculated from the following formula:

$$\hat{G}^r(\omega) = [\omega \hat{I} - \hat{H} - \hat{\Sigma}^r(\omega)]^{-1}, \quad (9)$$

where \hat{H} consists of the matrix elements of H_{nano} , i.e., $H_{mn} = -t\delta_{m, n\pm 1} + U\delta_{m, l}\delta_{n, l}$ and the retarded self-energy is determined by the coupling between the nanowire and the leads

$$\hat{\Sigma}^r(\omega) = \frac{1}{2} \sum_{\alpha \in \{L, R\}} \left[\frac{1}{\pi} P \int d\Omega \frac{\hat{\Gamma}_{\alpha}(\Omega)}{\omega - \Omega} - i\hat{\Gamma}_{\alpha}(\omega) \right]. \quad (10)$$

III. NUMERICAL RESULTS

We have solved numerically the system of Eqs. (5)–(10) for nanowires consisting of up to $N=129$ lattice sites, with a single impurity in the middle of the wire. The only nonvanishing elements of $\hat{\Gamma}$'s have been assumed to be frequency independent $[\hat{\Gamma}_L(\omega)]_{11} = [\hat{\Gamma}_R(\omega)]_{NN} = \Gamma_0$, where the sites in the chain are enumerated from 1 to N . We have taken the nearest neighbor hopping integral t as an energy unit and assumed the coupling between the nanosystem and the leads as $\Gamma_0 = 0.1t$. The temperature of both the leads is equal. Figure 1 shows the spatial distribution of electrons in the nanowire for $U=2t$ and various values of the bias voltage. One can see strong oscillations in the vicinity of the impurity. However, due to the coupling to the leads the electron distri-

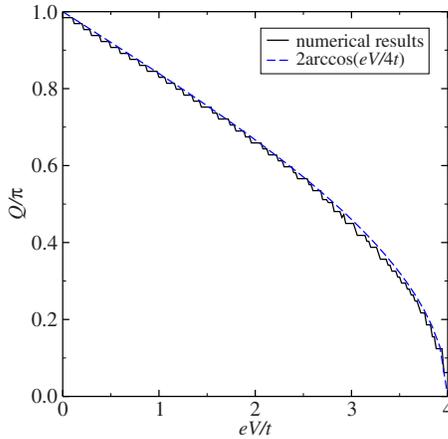


FIG. 2. (Color online) The wave-vector Q of the charge oscillations calculated by means of the fast Fourier transform of the numerical solution of Eqs. (5)–(10) (solid line). The dashed line shows the fit given by Eq. (12). The numerical results have been obtained for the same parameters as in Fig. 1. The discreteness of $Q(V)$ comes from the finite number of the lattice sites.

bution visibly differs from the standard Friedel oscillations

$$n(x) = \bar{n} + A \cos(Qx + \eta)/x^\delta, \quad (11)$$

where the wave vector $Q=2k_F$ and the parameters η and δ depend on the interaction. In our case, the actual value of Q has been obtained from the fast Fourier transform of the electron distribution $n(x)$.

Figure 2 shows the voltage dependence of the vector Q . In the equilibrium case ($V=0$) $Q=\pi$ (with the lattice constant $a=1$), so the charge oscillations are commensurate with the lattice. Since in the half-filled case $k_F=\pi/2$, this wave vector remains in agreement with the standard relation $Q=2k_F$. However, when the bias voltage is switched on, the situation changes and the oscillations are in general no longer commensurate. Moreover, one can see a strong dependence of Q on the applied voltage. Q is a monotonically decreasing function of voltage and vanishes for a sufficiently large V . A similar situation occurs in the transport phenomena through one-dimensional charge density wave systems.¹⁵ Our numerical results indicate that the obtained $Q(V)$ dependence can be very accurately described by a formula

$$eV = 4t \cos(Q/2). \quad (12)$$

The same form of $Q(V)$ can be obtained on the basis of the equilibrium assumptions discussed in the Introduction. In the following section we present an approximate analytical approach that explains such a form of $Q(V)$ in the nonequilibrium case. It is applicable to an arbitrary tight-binding Hamiltonian of noninteracting electrons and holds true in a wide range of the coupling strength Γ_0 . The numerical results presented in Fig. 2 allow us to test the applied approximations.

Up to this point, we have assumed one particular value of the impurity potential ($U=2t$) and one particular temperature of the leads ($k_B T=0.01t$). However, with the help of the

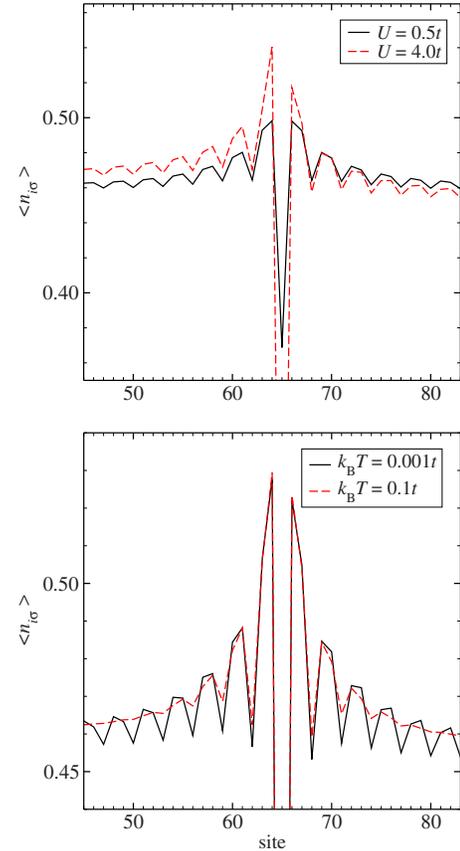


FIG. 3. (Color online) Temperature (lower panel) and impurity potential (upper panel) dependence of $\langle n_{i\sigma} \rangle$ obtained for a 129-site nanowire and for the bias voltage $eV=2t$. The lower panel shows results for $U=2t$, whereas the upper one for $k_B T=0.01t$.

Fourier transform, we have found that the voltage dependence of the wavelength of the oscillations, discussed above, is independent of these quantities. As it can be inferred from Fig. 3, the decrease of the impurity potential reduces the amplitude of the Friedel oscillations mainly in the vicinity of the impurity, whereas the increase of temperature causes faster decay of the oscillations away from the impurity. It can be seen in the upper panel in Fig. 3 that the occupation of sites is asymmetric with regard to the position of the impurity and this asymmetry increases with U and V . Strong impurity potential reduces the current and results in an enhancement (reduction) of the carrier concentration on the left (right) side of the impurity.

Finally, we briefly discuss the influence of the Coulomb correlations. Our analysis requires investigation of relatively large systems far from equilibrium. Therefore, the interaction cannot be taken into account exactly. Moreover, most of the approaches, which provide reliable results for low-dimensional systems, are inapplicable in this case. In order to get at least the qualitative picture we use the mean-field approximation. Similar investigations of the Friedel oscillations have recently been carried out for vanishing bias voltage.²³ For the sake of this analysis we extend H_{nano} by adding a term describing the electron-electron interaction at the mean-field level

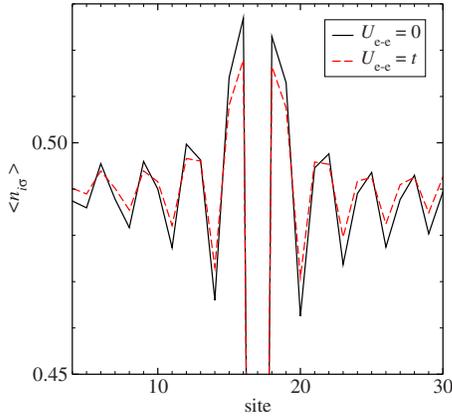


FIG. 4. (Color online) Influence of the Coulomb repulsion (U_{e-e}) on the occupation of sites. Results have been obtained for a 33-site nanowire and for the bias voltage $eV=2t$. $k_B T=0.01t$ and $U=2t$ have been assumed.

$$H_{\text{nano}} \rightarrow H_{\text{nano}} + U_{e-e} \sum_{i\sigma} \left(\langle n_{i-\sigma} \rangle - \frac{1}{2} \right) d_{i\sigma}^\dagger d_{i\sigma}, \quad (13)$$

where U_{e-e} denotes the potential of the on-site Coulomb repulsion and the average $\langle n_{i-\sigma} \rangle$ has to be calculated self-consistently from Eqs. (5)–(10). Being aware of the limitations of the mean-field approach, we have restricted our analysis only to small values of the Coulomb potential. In Fig. 4 we compare the Friedel oscillations calculated for $U_{e-e}=0$ and $U_{e-e}=t$. As expected, the local electron-electron repulsion reduces the amplitude of the Friedel oscillations and modifies the decay exponent. However, the wavelength of the oscillations is almost independent of U_{e-e} , at least for $U_{e-e} \leq t$. Similar to the uncorrelated case, the wavelength is determined predominantly by the bias voltage. Therefore, U_{e-e} will be neglected in the following discussion.

IV. ANALYTICAL DISCUSSION

Since the dependence $Q(V)$, described above, is the same as reported before for the charge density waves,¹⁵ a common explanation should be possible. In the equilibrium case, the eigenstates of an isolated system with periodic boundary conditions (PBCs) are built out of plane waves. The $2k_F x$ density oscillations are related to the maximum in the response function defined as a retarded Green function

$$\chi(Q, \omega) = - \langle \langle \hat{\rho}(Q) | \hat{\rho}^\dagger(Q) \rangle \rangle \quad (14)$$

calculated for $U=0$. Here,

$$\hat{\rho}(Q) = \sum_{i,\sigma} \exp(iQR_i) d_{i\sigma}^\dagger d_{i\sigma} = \sum_{k,\sigma} d_{k+Q\sigma}^\dagger d_{k\sigma}, \quad (15)$$

where the summation is carried out over all momenta k . In the following we demonstrate that this quantity helps one to explain the dependence $Q=Q(V)$ also in the nonequilibrium case of the Friedel oscillations.

When the nanosystem is connected to macroscopic leads, the PBCs become inappropriate since they do not reflect the

geometry of the experimental setup. Then, the choice of open boundary conditions (OBCs) seems to be more appropriate. For $U=0$, the Hamiltonian (3) with OBCs can be diagonalized with the help of the unitary transformation²⁴

$$d_{i\sigma}^\dagger = \sqrt{\frac{2}{N+1}} \sum_k \sin(kR_i) d_{k\sigma}^\dagger, \quad (16)$$

where the wave vectors k take on the following values:

$$k = \frac{\pi}{N+1}, \frac{2\pi}{N+1}, \dots, \frac{N\pi}{N+1}. \quad (17)$$

The specific form of this transformation accounts for vanishing of the one-electron wave functions at the edges of the nanosystem. In this representation one gets

$$H_{\text{nano}} = \sum_{k\sigma} \epsilon_k d_{k\sigma}^\dagger d_{k\sigma}, \quad (18)$$

where

$$\epsilon_k = -2t \cos(k). \quad (19)$$

Although the dispersion relation is exactly the same as for PBC, the values of k belong to $(0, \pi)$ instead of the first Brillouin zone $(-\pi, \pi)$. One can apply the above transformation also to the remaining terms in the Hamiltonian (1) and repeat calculations presented in Sec. II. The resulting equations have the same structure as Eqs. (5)–(10) with the real space variables i replaced by the wave vectors k . In the new representation, the Hamiltonian matrix \hat{H} is diagonal, however, the matrices $\hat{\Gamma}_\alpha$ take on much more complicated form

$$[\hat{\Gamma}_\alpha(\omega)]_{kp} = \frac{2}{N+1} \sum_{ij} [\hat{\Gamma}_\alpha(\omega)]_{ij} \sin(kR_i) \sin(pR_j). \quad (20)$$

In order to analyze the $Q(V)$ dependence, we investigate the correlation function given by Eq. (14) with

$$\hat{\rho}(Q) = \sum_{i\sigma} \cos(QR_i) d_{i\sigma}^\dagger d_{i\sigma} = \sum_{kp\sigma} \mathcal{B}_Q(k,p) d_{k\sigma}^\dagger d_{p\sigma}, \quad (21)$$

where

$$\mathcal{B}_Q(k,p) = \frac{1}{2} (\delta_{p,k-Q} - \delta_{p,Q-k} + \delta_{p,k+Q} - \delta_{p,2\pi-(k+Q)}). \quad (22)$$

The equations of motion allow one to calculate the correlation function that in the static limit takes on the form

$$\chi(Q, \omega \rightarrow 0) = \sum_{k,p,q,\sigma} \frac{\mathcal{B}_Q(k,p)}{\epsilon_p - \epsilon_k} (\mathcal{B}_Q^*(p,q) \langle d_{k\sigma}^\dagger d_{q\sigma} \rangle - \mathcal{B}_Q^*(q,k) \langle d_{q\sigma}^\dagger d_{p\sigma} \rangle) + \chi'. \quad (23)$$

The second term in the above equation χ' is proportional to $g \langle \langle d^\dagger c | d^\dagger d \rangle \rangle$ and will be neglected in the following analysis. Such simplification is justified only for a weak coupling between the nanosystem and the leads. In order to demonstrate the validity of this approximation we have calculated numerically the resulting correlation function for nanowires consisting of 20 and 40 sites (see Fig. 5). The discreteness of the system is clearly visible for short nanowires, whereas for

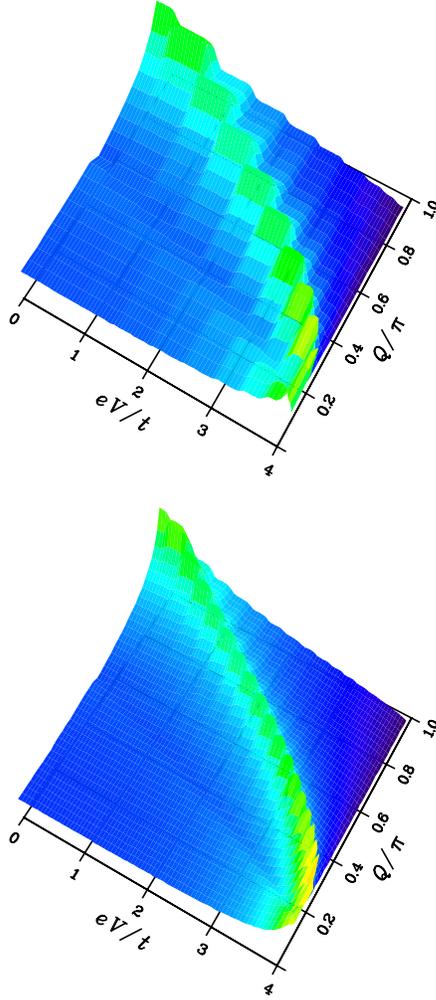


FIG. 5. (Color online) Correlation function $\chi = \chi(Q, V)$ [Eq. (23)] determined numerically for 20-site (upper panel) and 40-site (lower panel) chains. $k_B T = 0.01t$ has been assumed.

large systems the correlation function becomes smoother. In the latter case one can see that $\chi(Q, \omega \rightarrow 0)$ reaches its maximum value for Q given by Eq. (12), what justifies the applied approximation $\chi' \approx 0$.

The correlation function given by Eq. (23) is still too complicated for analytical discussion. The averages in Eq. (23) are determined by the lesser Keldysh Green functions that, in turn, depend on the retarded ones. Because of the coupling between the nanosystem and the leads, off-diagonal elements of the retarded Green function are nonzero [see Eqs. (10) and (20)]. In order to proceed with the discussion of the Friedel oscillations we apply an additional approximation

$$(\hat{\Gamma}_\alpha)_{kp} \approx \delta_{kp} \Gamma'. \quad (24)$$

Then, all matrices in Eq. (6) become diagonal. In the next step we assume $\Gamma' \rightarrow 0$, what allows one to calculate the integral over frequencies in Eq. (5). The resulting correlation function can be expressed as a sum of two Lindhard functions

$$\chi(Q, \omega \rightarrow 0) = \chi^L(Q) + \chi^R(Q), \quad (25)$$

where

$$\chi^{L(R)}(Q) = \sum_{k,p} |\mathcal{B}_Q(k,p)|^2 \frac{f_{L(R)}(\epsilon_k) - f_{L(R)}(\epsilon_p)}{\epsilon_p - \epsilon_k}. \quad (26)$$

The maximum of the response function occurs for such Q , that arguments of both the Fermi functions in the numerator in Eq. (26) vanish simultaneously. According to Eq. (8), it is easy to check that for both $\chi^L(Q)$ and $\chi^R(Q)$ this requirement is equivalent to Eq. (12).

At this stage a comment on the approximation given by Eq. (24) is necessary. It is a crude and generally inappropriate approximation that significantly affects most of the properties of the system. In particular, it would strongly modify the current-voltage characteristics. However, the correlation functions calculated from Eqs. (23) and (25) are almost indistinguishable, which *a posteriori* justifies the use of this approximation for the discussion of charge inhomogeneities. This surprising result gives some insight into the physical mechanism of the charge distribution in nanosystems in the presence of transport currents. This distribution seems to be independent of the details of the coupling between the nanosystem and the leads. However, it is determined by the fact that nanosystem is connected to two macroscopic particle reservoirs with different chemical potentials.

V. CONCLUDING REMARKS

Using the nonequilibrium Keldysh Green functions we have investigated the Friedel oscillations in a nanowire coupled symmetrically to two macroscopic electrodes. We have derived a simple formula for the correlation function that determines the wave vector Q of the oscillations. The approximate analytical expression fits the numerical results obtained from the Fourier transform of the electron distribution very accurately. Our analysis concerns nanosystems described by the tight-binding Hamiltonian with the nearest neighbor hopping. However, it can be straightforwardly extended to account for arbitrary hopping matrix elements.

The above discussion of the Friedel oscillations focuses on the voltage dependence of the wave vector Q . We have found that the envelope of the charge density oscillations is almost bias-voltage independent. This means that the remaining parameters characterizing the Friedel oscillations, i.e., the amplitude A and the spatial decay exponent δ , are determined predominantly by the internal properties of the nanowire, whereas the wavelength of the oscillations depends on the bias voltage. We believe that investigations of the Friedel oscillations in the transport phenomena should allow one to get insight into many important parameters of the experimental setup.

ACKNOWLEDGMENT

This work has been supported by the Polish Ministry of Education and Science under Grant No. 1 P03B 071 30.

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