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FRIEDEL OSCILLATIONS IN NANOWIRES AT FINITE BIAS VOLTAGE

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Abstract. We investigate the charge density oscillations in a nanowire coupled asymmetrically to two leads. Depending on this asymmetry, the Friedel oscillations can either be characterized by a single wave-vector or become a superposition of oscillations with different wave-vectors. Using the formalism of nonequilibrium Keldysh Green functions, we derive a simple equation that determines bias voltage dependence of the wave-length of the oscillations. Finally, we discuss limitations of the commonly used formula that describes the spatial character of the Friedel oscillations.

Keywords: nanowire, Friedel oscillations, nonequilibrium Green functions

4.1 Introduction

The conductance of nanosystems has recently been the subject of intensive experimental and theoretical investigations, because of their possible application in the electronic devices. This ongoing research concerns the transport properties of various systems, e.g., nanowires, single molecules and quantum dots [2,3,4,8,18]. The properties of nanosystems differ significantly from the transport properties of macroscopic materials, what shows up in nonlinear or even irregular current–voltage characteristics.

In many cases, the charge carriers are distributed inhomogeneously in the nanosystem. On one hand, such inhomogeneity may be caused by the mechanisms, which are well known from the analysis of macroscopic materials. In particular, it may originate from impurities [7] or/and correlations which lead to charge density waves [12, 15, 17, 19, 23]. On the other hand, inhomogeneous distribution of the charge carriers may originate from mechanisms, which are typical for nanosystems. In analogy to the case of a quantum well,

one may expect that electrons are inhomogeneously distributed just due to the spatial confinement at the nanoscale [13]. The standing waves have recently been observed at the end of a carbon nanotube [14]. Inhomogeneous charge distribution may originate also from the applied bias voltage [2, 6, 11].

Recent theoretical analysis of the Friedel oscillations in nanosystems has been carried out for zero or vanishingly small bias voltage V . The most of these investigations concerned the electron correlations [1, 5, 10, 21, 22] which, as has been shown, suppress the decay of oscillations [3, 20]. In this paper we neglect the electron–electron interaction. Instead, we focus on the nonequilibrium case that occurs for finite V . We investigate a nanowire that is connected to two macroscopic particle reservoirs with different Fermi energies. Since this difference increases with V , the meaning of the Fermi momentum, k_F , becomes ambiguous. Therefore, the well known $2k_F x$ spatial dependence of the density oscillations (that holds also in the presence of correlations) may be modified for $V \neq 0$. We have recently considered a nanowire that is symmetrically coupled to the leads. We have shown that the wave-vector of the oscillations decreases monotonically with bias voltage and vanishes when V is equal to the band width. These results hold both for the charge density waves [16] and the Friedel oscillations [9]. Here, we extend our previous analysis of the Friedel oscillations and discuss the case of asymmetric coupling. We account also for a linear potential drop in the nanowire. We demonstrate that these effects seriously affect the spatial dependence of the density oscillations.

4.2 Model

We investigate a nanowire coupled to two macroscopic leads. Therefore, we consider a Hamiltonian, that consists of three terms H_{el} , H_{nano} , and $H_{\text{nano-el}}$, which describe the electrodes, nanowire and the coupling between the electrodes and the nanowire, respectively

$$H = H_{\text{el}} + H_{\text{nano}} + H_{\text{nano-el}}. \quad (4.1)$$

The electrodes are modeled by the electron gas:

$$H_{\text{el}} = \sum_{\mathbf{k}, \sigma, \alpha} (\varepsilon_{\mathbf{k}, \alpha} - \mu_{\alpha}) c_{\mathbf{k}\sigma\alpha}^{\dagger} c_{\mathbf{k}\sigma\alpha}, \quad (4.2)$$

where μ_{α} is the chemical potential, $\alpha \in \{\text{L}, \text{R}\}$ indicates the left or right electrode. $c_{\mathbf{k}\sigma\alpha}^{\dagger}$ creates an electron with momentum \mathbf{k} and spin σ in the electrode α . We assume that the nanowire can be described by the tight-binding Hamiltonian:

$$\mathcal{H}_{\text{nano}} = -t \sum_{\langle ij \rangle \sigma} d_{i\sigma}^{\dagger} d_{j\sigma} + \sum_{i\sigma} \varepsilon_i n_{i\sigma} + U \sum_{\sigma} n_{l\sigma}, \quad (4.3)$$

where $d_{i\sigma}^\dagger$ is the creation operator of an electron with spin σ at site i of the nanosystem, $n_{i\sigma}$ – occupation number operator, ε_i – atomic energy level. In order to investigate the Friedel oscillations we have introduced a single impurity with the potential U . This impurity is located in the site l . The coupling between the nanowire and the leads is given by:

$$H_{\text{nano-el}} = \sum_{\mathbf{k}, i, \alpha, \sigma} \left(g_{\mathbf{k}, i, \alpha} c_{\mathbf{k}\sigma\alpha}^\dagger d_{i\sigma} + \text{H.c.} \right), \quad (4.4)$$

where the matrix elements $g_{\mathbf{k}, i, \alpha}$ are nonzero only for the edge atoms of the nanowire.

4.3 Results and discussion

The electron density fluctuations have been determined by means of the nonequilibrium Keldysh Green functions. First, we have calculated the local carrier density, that is expressed by the lesser Green function,

$$\langle d_{i\sigma}^\dagger d_{i\sigma} \rangle = \frac{1}{2\pi i} \int d\omega G_{i\sigma, i\sigma}^<(\omega). \quad (4.5)$$

$G_{i\sigma, i\sigma}^<$ is, in turn, determined by the retarded and advanced Green functions:

$$\hat{G}^<(\omega) = i \sum_{\alpha \in \{L, R\}} \hat{G}^r(\omega) \hat{\Gamma}_\alpha(\omega) \hat{G}^a(\omega) f_\alpha(\omega), \quad (4.6)$$

where

$$\left[\hat{\Gamma}_\alpha(\omega) \right]_{ij} = 2\pi \sum_{\mathbf{k}} g_{\mathbf{k}, i, \alpha}^* g_{\mathbf{k}, j, \alpha} \delta(\omega - \varepsilon_{\mathbf{k}, \alpha}), \quad (4.7)$$

and $f_\alpha(\omega)$ stands for the Fermi distribution function of the electrode α . We have assumed that the energy bands of the leads are wide enough, and one can neglect the frequency dependence of $\hat{\Gamma}_\alpha$. Finally, the retarded self-energy is determined by the coupling between the nanowire and the leads

$$\hat{\Sigma}^r(\omega) = \frac{1}{2} \sum_{\alpha \in \{L, R\}} \left[\frac{1}{\pi} P \int d\Omega \frac{\hat{\Gamma}_\alpha(\Omega)}{\omega - \Omega} - i \hat{\Gamma}_\alpha(\omega) \right]. \quad (4.8)$$

Numerical analysis of the above equations allows one to investigate spatial distribution of charge carriers and the properties of the Friedel oscillations. In the equilibrium case, these oscillations asymptotically decay with the distance x from the impurity as $\cos(Qx + \phi_0)x^{-\delta}$, where $Q = 2k_F$ and η parametrizes the interaction. As we have pointed out in the introduction, the wave-vector of the oscillations, Q , may nontrivially depend on the bias voltage. This dependence has been obtained from the fast Fourier transform of the the spatial

electron distribution. On one hand, this approach enables accurate investigations of the Friedel oscillations for arbitrary model parameters. On the other hand, such numerical analysis does not explain the underlying physical mechanisms.

The bias voltage dependence of the wave-vector Q obtained from the above numerical analysis is exactly the same as previously reported for the charge density waves (CDW) [16]. Therefore, one may expect that there is a common physical mechanism that determines $Q(V)$ in both cases. In the following, we investigate a correlation function, that in the equilibrium case is related to the CDW instability. On the one hand, it allows us to explain $Q(V)$ obtained for the charge density waves. On the other hand, it provides a simple (even if only phenomenological) description of $Q(V)$ in the case of Friedel oscillation. The discussed above numerical analysis allowed us to confirm the applicability of this description.

In the equilibrium case the wave-length of the oscillations can be analyzed with the help of an appropriate correlation function. Usually one assumes *periodic boundary conditions* and investigates

$$\chi(Q, \omega) = -\langle\langle \hat{\rho}(Q) | \hat{\rho}^\dagger(Q) \rangle\rangle, \quad (4.9)$$

where

$$\hat{\rho}(Q) = \sum_{i,\sigma} \exp(iQR_i) d_{i\sigma}^\dagger. \quad (4.10)$$

Since the nanowire is connected to macroscopic leads, application of periodic boundary conditions is unjustified and the choice of open boundary conditions seems to be much more appropriate. At this stage of our analysis we assume that $U = 0$ and $\varepsilon_i = \varepsilon_0 = \text{const}$. The second assumption implies that there is no potential drop inside the nanowire. Then, one can use the following unitary transformation to diagonalize H_{nano} (4.3):

$$d_{i\sigma}^\dagger = \sqrt{\frac{2}{N+1}} \sum_k \sin(kR_i) d_{k\sigma}^\dagger \quad (4.11)$$

where the values of k are given by: $k = \frac{\pi}{N+1}, \frac{2\pi}{N+1}, \dots, \frac{N\pi}{N+1}$ and N denotes the number of sites of the nanowire. It is straightforward to check that

$$\mathcal{H}_{\text{nano}} = \sum_{k\sigma} \varepsilon_k d_{k\sigma}^\dagger d_{k\sigma} \quad (4.12)$$

and the dispersion relation reads $\varepsilon_k = -2t \cos(k) + \varepsilon_0$.

In order to analyze $Q(V)$ we have transformed $H_{\text{nano-el}}$ according to Eq. (4.11). Then, we have calculated the correlation functions given by Eq. (4.9) with

$$\hat{\rho}(Q) = \sum_{i\sigma} \cos(QR_i) d_{i\sigma}^\dagger d_{i\sigma} = \sum_{kp\sigma} \mathcal{B}_Q(k, p) d_{k\sigma}^\dagger d_{p\sigma} \quad (4.13)$$

and

$$\mathcal{B}_Q(k, p) = \frac{1}{2} (\delta_{p, k-Q} - \delta_{p, Q-k} + \delta_{p, k+Q} - \delta_{p, 2\pi-(k+Q)}). \quad (4.14)$$

Using the equations of motion we have obtained the following form of the correlation function (4.9)

$$\chi(Q, \omega \rightarrow 0) = \sum_{kpq\sigma} \frac{\mathcal{B}_Q(k, p)}{\varepsilon_p - \varepsilon_k} \left(\mathcal{B}_Q^*(p, q) \langle d_{k\sigma}^\dagger d_{q\sigma} \rangle - \mathcal{B}_Q^*(q, k) \langle d_{q\sigma}^\dagger d_{p\sigma} \rangle \right) + \chi' \quad (4.15)$$

The second term of the above formula, χ' , is proportional to $\langle\langle d^\dagger c | d^\dagger d \rangle\rangle$ and it can be neglected for a weak coupling between the nanosystem and the leads. In order to proceed with the analytical discussion we have also neglected the off-diagonal elements of Γ_{kp}^α . Consequently, $\langle d_{k\sigma}^\dagger d_{q\sigma} \rangle \sim \delta_{kq}$ and the investigated correlation function takes on a simple form

$$\chi(Q, \omega \rightarrow 0) = \sum_{kp\sigma} |\mathcal{B}_Q(k, p)|^2 \frac{\langle n_{k\sigma} \rangle - \langle n_{p\sigma} \rangle}{\varepsilon_p - \varepsilon_k}. \quad (4.16)$$

The above equation resembles the Lindhard's form of the correlation function that is well known from the equilibrium physics. However, there exist two important differences. The first one originates from the open boundary conditions, what shows up in the presence of the factor $\mathcal{B}_Q(k, p)$. The second one accounts for the fact that the nanowire is not in equilibrium with a single thermostat. Therefore, the average $\langle n_{k\sigma} \rangle$ is determined by a lesser Green function and cannot be expressed by a single Fermi function. In order to make these differences more visible, we have assumed that $\Gamma_{kp}^R = a_\Gamma \Gamma_0 \delta_{kp}$ and $\Gamma_{kp}^L = \frac{1}{a_\Gamma} \Gamma_0 \delta_{kp}$. Here, we have introduced the parameter $a_\Gamma \in (1, \infty)$ that describes the degree of asymmetry of the coupling to the electrodes. For $\Gamma_0 \rightarrow 0$ (which is equivalent to the weak coupling regime) one can explicitly calculate the integral over frequencies in Eq. (4.5). The obtained value of $\langle n_{k\sigma} \rangle$ reads:

$$\langle n_{k\sigma} \rangle = \frac{a_\Gamma}{1 + a_\Gamma^2} \left(a_\Gamma f_R(\varepsilon_k) + \frac{1}{a_\Gamma} f_L(\varepsilon_k) \right) \quad (4.17)$$

and the correlation function becomes a sum of two functions, stemming from both the electrodes:

$$\chi(Q, \omega \rightarrow 0) = \frac{a_\Gamma^2}{1 + a_\Gamma^2} \chi^R(Q) + \frac{1}{1 + a_\Gamma^2} \chi^L(Q), \quad (4.18)$$

where

$$\chi^{R(L)}(Q) = \sum_{kp\sigma} |\mathcal{B}_Q(k, p)|^2 \frac{f_{R(L)}(\varepsilon_k) - f_{R(L)}(\varepsilon_p)}{\varepsilon_p - \varepsilon_k}. \quad (4.19)$$

The Fermi distribution functions of the left and right electrodes depend on the bias voltage. Namely, $f_{R(L)}(\varepsilon_k) = f(\varepsilon_k - \mu_{R(L)})$, with $f(x) = (\exp(x) + 1)^{-1}$

and $\mu_L - \mu_R = eV$. In the case of a symmetric coupling one usually puts $\mu_L = eV/2$ and $\mu_R = -eV/2$. However, for $a_\Gamma \neq 1$ it may be invalid. Therefore, we have introduced a second parameter, a_V , that determines the asymmetry of the potential drop that takes place at contacts between the nanowire and the electrodes. Namely, we assume that $\mu_L = eVa_V$ and $\mu_R = -eV(1 - a_V)$ and consider $a_V \in (0, 1)$.

A comment on the applied approximations ($\Gamma_{kp} \sim \delta_{kp}$ and $\Gamma_0 \rightarrow 0$) is necessary. These crude approximations may affect many properties of the nanowire. In particular, they are inapplicable in the investigations of the current–voltage characteristics. However, as will be demonstrated in the following discussion, the wave-vectors of the oscillations determined from the Fourier transform of Eq. (4.5) and from Eq. (4.18) are the same. It means, that the wave-length of the density oscillations is determined predominantly by the fact that the nanowire is coupled to two particle reservoirs with different chemical potentials.

First, we discuss the Friedel oscillations in the presence of the asymmetric coupling between the nanowire and the leads. This asymmetry occurs when the $a_\Gamma \neq 1$ or $a_V \neq 1/2$. Figure 4.1 shows the spatial distribution of charge carriers as well as correlation function χ for the case when the asymmetry originates only from a_Γ . We have found that a_Γ does influence the correlation function. It means that the wave-length of the density oscillations is independent of this quantity and is exactly the same as obtained for a symmetric coupling. This conclusion can also be drawn from the presented electron distribution. Although, $\langle n_{i\sigma} \rangle$ depends quantitatively on a_Γ , the periodicity of the density fluctuations remains unchanged. For a vanishing bias voltage, χ takes on the maximal value for $Q = \pi$. Since in the half-filled case $k_F = \pi/2$

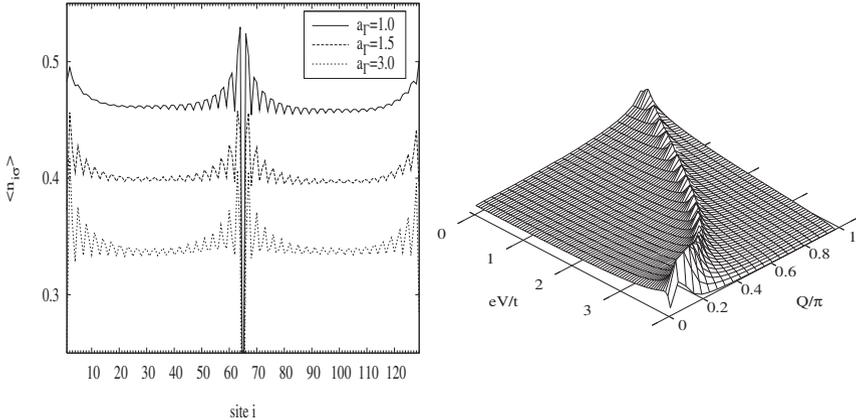


Fig. 4.1. Results obtained for $a_V = 0.5$, $\varepsilon_0 = 0$ and various a_Γ . Left panel shows occupation of sites of a 129-site nanowire for $eV = 2t$. Right panel shows the correlation function $\chi = \chi(Q, V)$, Eq. (4.18), calculated for $a_\Gamma = 3$.

this result remains in agreement with a standard relation, that holds in the equilibrium case $Q = 2k_F$. However, for a finite bias voltage the situation is very different. The correlation function reaches its maximum for a wave-vector Q that monotonically decreases with V and vanishes when the bias voltage exceeds the band width of an isolated nanowire. An explicit form of the voltage dependence of Q can be found from Eqs. (4.18) and (4.19) [9]. Namely, for $a_V = 1/2$ and $\varepsilon_0 = 0$ the maximum of the correlation function χ occurs for $Q = 2 \arccos(eV/4t)$. In the symmetric case, $a_\Gamma = 1$, the bias voltage changes the wave-length of the Friedel oscillations but the average occupation of sites is independent of V . On the other hand, for a fixed bias voltage, a_Γ modifies the occupation number but does not influence the wave-length of the oscillations. These results visibly differ from the equilibrium case, when the wave-length of the density fluctuation is uniquely determined by the occupation number.

A different form of the Friedel oscillations occurs for the asymmetric potential drop (see Fig. 4.2). Contrary to the previous case, the wave-length of the oscillations depends on a_V , provided the bias voltage is finite. One can see, that the correlation function achieves two local maxima, which occur for different values of Q . However, the results presented in Fig. 4.2 do not allow one to distinguish between the following possibilities: (i) the Friedel oscillations can be described by a single wave-vector, e.g., by Q that corresponds to larger value of the investigated correlations functions; (ii) the Friedel oscillations represent a superposition of two oscillations with different values of Q . Carrying out Fourier transform of $\langle n_{i\sigma} \rangle$ we have found that the second case

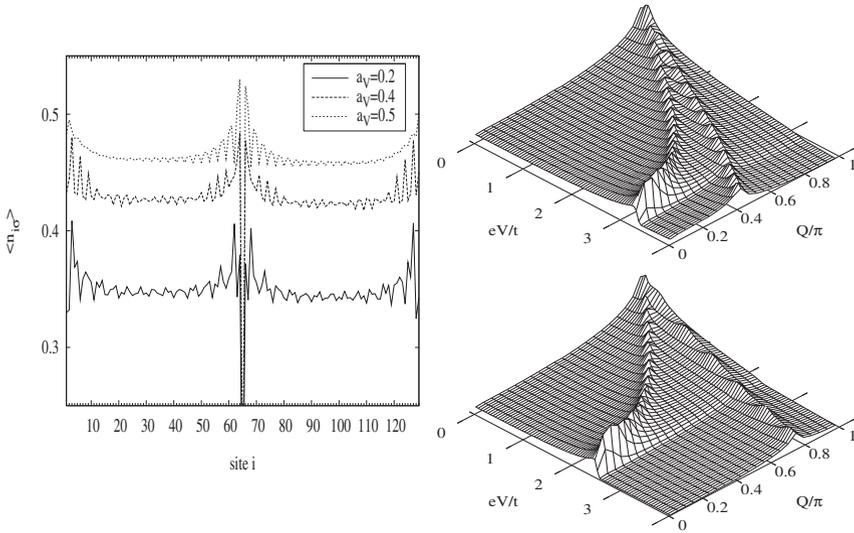


Fig. 4.2. The same as in Fig. 4.1 but for $a_\Gamma = 1$ and various a_V . The right upper and the right lower panels show $\chi = \chi(Q, V)$ obtained for $a_V = 0.4$ and 0.2 , respectively.

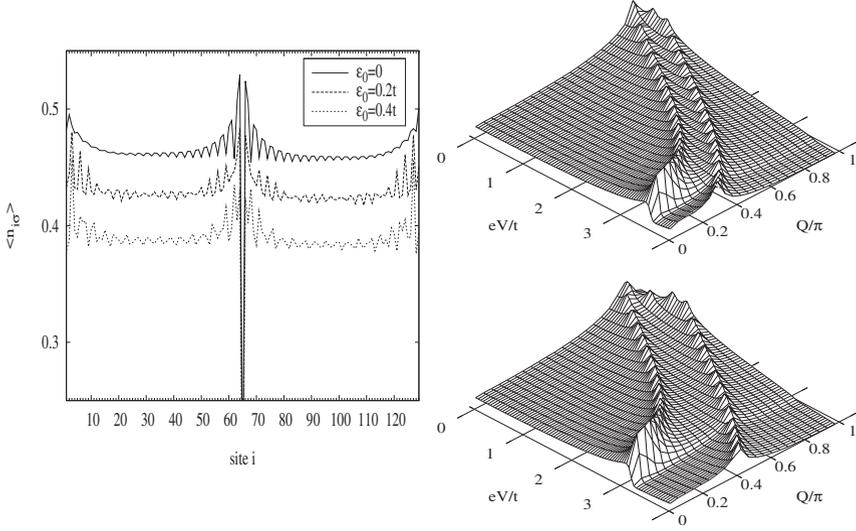


Fig. 4.3. The same as in Fig. 4.1 but for $a_\Gamma = 1$ and various ε_0 . The right upper and the right lower panels show $\chi = \chi(Q, V)$ obtained for $\varepsilon_0 = 0.2t$ and $0.4t$, respectively.

is actually realized in the system under investigations. Figure 4.3 shows that a similar situation occurs also for $\varepsilon_0 \neq 0$. The above discussion allows one to draw the following conclusion: The Friedel oscillations can be characterized by a single wave-vector Q provided $\mu_L + \mu_R = \varepsilon_0 = 0$. This result holds independently of the asymmetry of Γ . Otherwise, the spatial dependence of the electron concentration becomes a superposition of oscillations with two different wave-vectors Q .

In the above discussion, we have assumed that there is no potential drop inside the nanowire. In the following we relax this assumption, and consider a linear potential drop that modifies the on-site energy levels:

$$\varepsilon_i = \varepsilon_0 + \frac{eV}{2}\eta\left(1 - \frac{2i}{N+1}\right) \quad (4.20)$$

For $\eta = 0$ there is no potential drop, whereas for $\eta = 1$ the entire drop takes place inside the nanowire. The assumed linear dependence may be invalid in the presence of impurities. Therefore, we have put $U = 0$ and investigated the density oscillations that occur at the edges of the nanowire. For $\varepsilon_i \neq \text{const}$, i.e., for $\eta \neq 0$ H_{nano} cannot be diagonalized by the transformation given by Eq. (4.11). In order to proceed with the discussion, we have solved the arising eigenproblem numerically. Namely, we have introduced new fermionic operators $a_{a\sigma}$

$$d_{i\sigma} = \sum_m U_{im} a_{m\sigma}, \quad (4.21)$$

where the unitary matrix U leads to a diagonal form of H_{nano}

$$\mathcal{H}_{\text{nano}} = \sum_{n\sigma} \varepsilon_n a_{n\sigma}^\dagger a_{n\sigma}. \quad (4.22)$$

Then, we have repeated the previous analysis and applied the same approximation scheme. In particular we have calculated $\chi(Q, \omega \rightarrow 0)$. The correlation function in this case takes on the form

$$\chi(Q, \omega \rightarrow 0) = \sum_{ij\sigma} \cos(QR_i) \cos(QR_j) \sum_{mn} U_{im} U_{in}^* U_{jm} U_{jn}^* \frac{\langle n_{n\sigma} \rangle - \langle n_{m\sigma} \rangle}{\varepsilon_m - \varepsilon_n}. \quad (4.23)$$

Figure 4.4 shows the numerical results. One see that the character of the Friedel oscillations strongly depends on the potential drop, that takes place inside the nanowire. The maximum of the correlation function occurs for Q that strongly depends on the bias voltage, similarly to the previously discussed cases. However, for $\eta \neq 0$ the value of this maximum decreases when V increases. For sufficiently high voltage, Q -dependence of the correlation functions smears out. In order to understand this result, we have divided the nanowire into small sections. Then, we have carried out the Fourier transform of $\langle n_{i\sigma} \rangle$ independently for each section. Surprisingly, we have found that Q depends on the position of the section of the nanowire. Therefore, for $\eta \neq 0$ the standard formula describing the Friedel oscillations $\cos(Qx + \phi_0)/x^\delta$ holds only locally, i.e., it holds at the lengthscale much smaller than the size of the

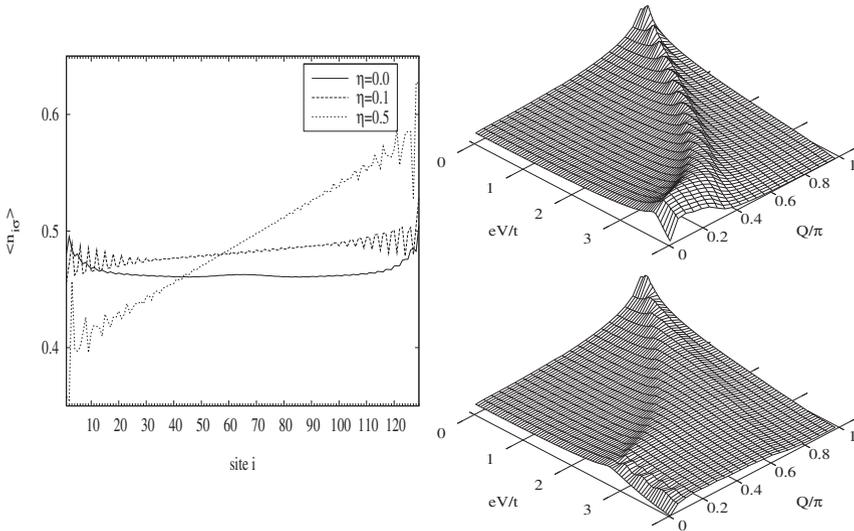


Fig. 4.4. Results obtained for $a_T = 1$, $a_V = 0.5$ and $\varepsilon_0 = 0$. The left panel shows the occupation of sites of a 129-site nanowire for $eV = 2t$. The right panels show $\chi = \chi(Q, V)$, Eq. (4.23). The upper and lower ones have been obtained for $\eta = 0.1$ and 0.5, respectively.

nanowire. Otherwise, one should consider Q as a slowly varying function of the position.

To conclude, we have analyzed the Friedel oscillations in the nanowire. Using the formalism of the Keldysh Green functions, we have focus on the nonequilibrium case that occurs for finite bias voltage. Generally, the wavelength of the oscillations decreases when the bias voltage increases. The spatial character of the Friedel oscillations strongly depends on the asymmetry of the coupling between nanowire and leads as well as on the potential drop inside the nanowire. As we have shown, experimental investigations of the Friedel oscillations may provide important information concerning the coupling between the nanowire and the leads.

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