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Relationship between the geometric structure and conductance oscillation in nanowires

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Abstract

A theoretical analysis of the electron transport properties of plain and bumpy jellium nanowires suspended between semi-infinite jellium electrodes is carried out, and the possibility of the experimental observation of the conductance oscillation with a period longer than the two-atom length is discussed. In both the nanowires, the transmission trace as a function of the nanowire length exhibits oscillatory behaviour. The period of the oscillation of the plain nanowire corresponds to π divided by the Bloch wavenumber of the electrons in the nanowire region. However, the period of the oscillation of the bumpy nanowire results in the least common multiple of π divided by the Bloch wavenumber and the geometric period of the nanowire. Our result indicates that the conductance oscillation with a period longer than the two-atom length can be experimentally observed if nanowires without any defects are formed in experiments.

1. Introduction

As downsizing of electronic devices progresses, the electron transport properties through atomic-scale structures have attracted more interest. When a system's dimensions reach the order of the electron mean free path, the electron transport becomes ballistic and its conductance is quantized in units of $G_0(= 2e^2/h)$ contrary to diffusive electron transport in a macroscopic system, where e is the electron charge and h is Planck's constant. In experiments, metallic nanowires are formed using a scanning tunnelling microscope or mechanically controllable break junctions [1]. In both cases the conductance quantization is observed in the last stage of pulling atomic nanowires.

On the theoretical side, numerous attempts have been made to study the electron transport properties of nanowires between electrodes, and some of them have reported that the conductance quantization emerges in a single-row nanowire made of monovalent atoms [2–14].

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Ten years ago, Lang [2] found that the conductance of a nanowire consisting of sodium atoms suspended between jellium electrodes oscillates with a period of the length of two-atoms as the length of the nanowire is varied; this is called conductance oscillation. Ever since, many studies regarding the conductance oscillation with a period of two-atom length have been reported [2, 3, 5–14]. In 2003, the conductance oscillation was observed experimentally by Smit *et al* [15] in Au, Pt and Ir nanowires. In addition, Thygesen *et al* [16] found that the oscillation of a four-atom period emerges in an Al nanowire while oscillation with such a long period has not yet been observed in experiments.

Some of the theoretical studies [2, 7, 11] concluded that the conductance oscillation could be understood in terms of the occupation number of the highest occupied molecular orbital (HOMO) of the system: the transmission is almost unity (minimum) when the HOMO is fractionally (fully) occupied and the occupation number changes alternately as the number of atoms constituting the nanowire increases. However, this interpretation cannot explain the oscillation with a period longer than the two-atom length such as that reported by Thygesen *et al* [16]. Recently, we studied the conductance oscillation of nanowires using atomistic and jellium nanowires and reported that the conductance oscillation is observed in the jellium nanowire even though there is no atomic configuration [13]; the period of the conductance oscillation depends on the Bloch wavenumber of the electrons in the nanowire region. Although the origin of the conductance oscillation has been revealed, there remains an unsettled question: why has the oscillation with a long period of more than the two-atom length not been observed experimentally?

In this paper we give a theoretical analysis of the electron transport properties of a plain jellium nanowire and a bumpy jellium nanowire, which is composed of wide and narrow parts, and also explore the possibility of experimental observation of the conductance oscillation with a period longer than the two-atom length. Our results indicate that the period of the oscillation is closely related to the Bloch wavenumber of the electrons in the nanowire region in the case of the plain nanowire. On the other hand, in the case of the bumpy nanowire, the period of the oscillation is affected by the geometric period of the nanowire as well as the Bloch wavenumber. Although the period of the oscillation is the least common multiple of the geometric period of the nanowire and the value of π divided by the Bloch wavenumber, a conductance oscillation with a period longer than the two-atom length will be observed in experiments if an ideal nanowire, which is straight and with uniform interatomic distance, can be formed.

The rest of the paper is organized as follows. The computational method is described in section 2. Our results and discussion are presented in section 3. We summarize our findings in section 4.

2. Computational method and model

We employ the computational method based on the real-space finite-difference approach [18–21], which is more useful than the existing plane wave approach, which is unable, for example, to describe strictly nonperiodic systems such as clusters and solid surfaces. The derivative arising from the kinetic energy operator in the Kohn–Sham equation is approximated by using finite-difference formulae. We discuss the transport properties of the plain jellium nanowire and also the bumpy jellium nanowire in order to explore the effect of the variation of the potential arising from the existence of atoms. In jellium models, the ions of the metal are replaced with a homogeneous positive background charge. Figure 1 shows an example of the calculation models, where the nanowires are suspended between semi-infinite jellium electrodes. In figure 1(b), the widths of wide and narrow parts of the nanowires, W_a and W_b, are 4.0 and 5.0 au, respectively, and both parts have the same lengths (L_a, $L_b = 2.0$ au). The potential in the nanowire and electrode regions is set at zero with hard-wall boundary conditions, while

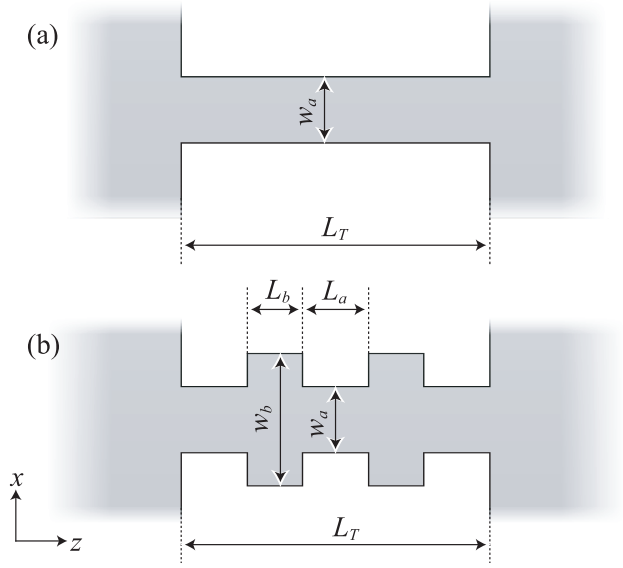

Figure 1. Computational models of (a) a two-dimensional plain jellium nanowire and (b) a two-dimensional bumpy jellium nanowire.

the other region is chosen to be sufficiently large to imitate the vacuum region ($125h^2/meW_b^2$), where m is the free electron mass. The grid spacing of 0.25 au is employed to describe the scattering wavefunctions and the three-point finite-difference formula is adopted for the kinetic energy operator. The scattering wavefunctions of the incident electrons from the left electrode are written as

$$\Psi_i^L = \begin{cases} \Phi_i^{\text{in}} + \sum_j r_{ij}^L \Phi_j^{\text{ref}} & \text{(in the left electrode)} \\ \phi_i^L & \text{(in the nanowire region)} \\ \sum_j t_{ij}^L \Phi_j^{\text{tra}} & \text{(in the right electrode),} \end{cases} \tag{1}$$

where ϕ^L is the wavefunction in the nanowire region, Φs are the bulk wavefunctions inside the electrodes, r^Ls are the reflection coefficients and t^Ls are the transmission coefficients. The reflection and transmission coefficients are computed by the overbridging boundary-matching method [9, 22], in which a wavefunction infinitely extending over the whole system can be determined by carrying out wavefunction matching based on a boundary-value problem in terms of the Green's function of a nanowire region. The Green's function determined by the inverse matrix of $(E - H)^{-1}$ which is calculated by the incomplete Cholesky decomposition conjugate gradient method [23]. Here, E is a Kohn–Sham energy and H is the Kohn–Sham Hamiltonian of the truncated nanowire region. When we investigate the ballistic electron transport through nanowires, the conductance G at the zero-bias limit is calculated by using the Landauer–Büttiker formula [24],

$$G = \frac{2e^2}{h} \sum_{i,j} |t_{ij}^L|^2 \frac{v_i}{v_j}, \tag{2}$$

where v_i (v_j) is the z component of the expectation value of the velocity for the ith transmitted (the jth incident) Bloch wave.

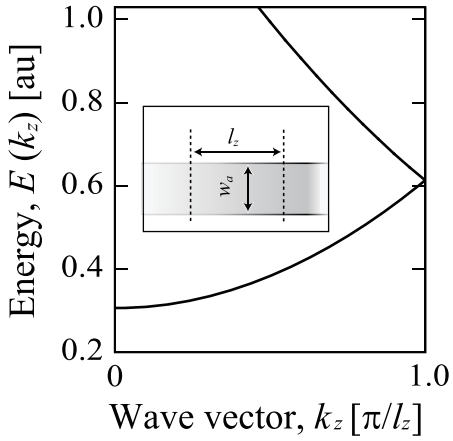

Figure 2. Electronic band structure of the infinite plain jellium wire. Inset: computational model for the infinite plain jellium wire.

3. Results and discussion

Figures 2 and 3 show the electronic band structure of the infinite plain jellium wire and the transmission properties of the plain jellium nanowire suspended between semi-infinite electrodes as a function of the length of the nanowire, respectively. The computational model of the infinite wire is inserted in figure 2, where l_z is $L_a + L_b (= 4.0$ au). We set the energy of the incident electrons to be $E(k_z)$ of figure 2, where k_zs are chosen to be $k_z = \pi/3l_z$, $k_z = \pi/2l_z$ and $k_z = 2\pi/3l_z$ for figures 3(a), (b) and (c), respectively. The transmission properties show oscillatory behaviour and the period of the oscillation is equal to π divided by the z component of the wavenumber, π/k_z. Although the oscillatory transmission behaviour was thought to arise from the occupation number of the HOMO [25], this result showing that the conductance oscillation with a period of π/k_z emerges in the plain jellium nanowires is strong evidence that the oscillation behaviour can be attributed to the z component of the Bloch wavenumber of electrons and that the occupation number of the HOMO plays the secondary role. Figures 4 and 5 illustrate the electron charge density distributions of the scattering waves Ψ^L. One can recognize that the length of the bunches is in agreement with π/k_z in all the distributions. In addition, all the bunches are elliptic when the transmission is unity (figure 4), whereas one of the bunches is deformed when the transmission is minimum (figure 5). These density distributions also support the idea that the quantum mechanical character of electrons is dominant in the transmission oscillation. In figure 3, the amplitude of the oscillations monotonically decreases as the z component of the Bloch wavenumber increases. This is because the length of the bunches of the scattering waves with low Bloch wavenumber is longer than that of the waves with high Bloch wavenumber and the waves with the long bunch hardly adjust their phase at the interface between the nanowire and electrodes. Moreover, according to the wavefunction matching formula [17], the amplitude of the transmission oscillation is inversely proportional to the z component of the Bloch wavenumber of the electrons in the nanowire region.

According to the above results, an oscillation with a period longer than the two-atom length will be observed experimentally. However, there remains another periodicity in atomistic nanowires, the spatial variation of the potential due to the existence of atoms. We next examine the transport properties of the bumpy jellium nanowire as shown in figure 1(b) in order to examine the relationship between the conductance oscillation and the geometric variation of

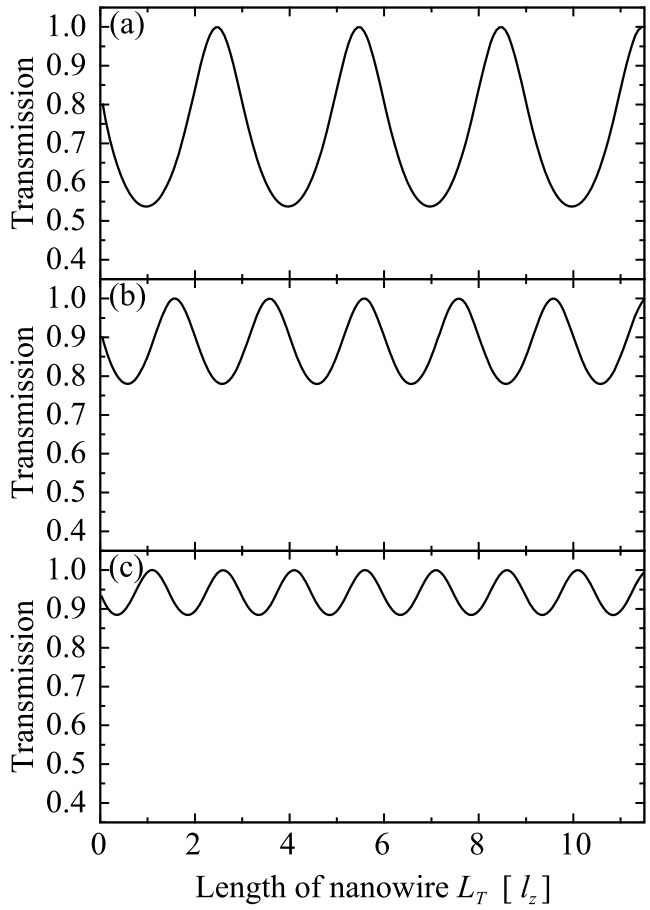

Figure 3. Transmission trace at the energy of $E(k_z)$ as a function of the length of the plain nanowire, where k_zs are chosen as (a) $\pi/3l_z$, (b) $\pi/2l_z$ and (c) $2\pi/3l_z$ in figure 2.

the potential. The computational model and the electronic band structure of the bumpy infinite wire are depicted in figure 6. Figure 7 shows the transmission properties as a function of the length of the bumpy nanowire. The energy of the incident electrons is set to be $E(k_z)$ of figure 6, where k_zs are set at $k_z = \pi/3l_z$, $k_z = \pi/2l_z$ and $k_z = 2\pi/3l_z$ for figures 7(a), (b) and (c), respectively, so that the Bloch wavenumbers correspond to those of the plain jellium nanowire. In the case of $k_z = \pi/2l_z$ ($k_z = \pi/3l_z$), the period remains $2l_z$ ($3l_z$). On the contrary, the period of the transmission oscillation is $3l_z$ in the case of $k_z = 2\pi/3l_z$ while that of the plain nanowire is $3l_z/2$. In order to study the origin of the change of the period in detail, we depict in figure 8 the electron charge density distributions of the scattering waves Ψ^L. The shapes of the distributions are almost inherited from those of the plain nanowire. The length of the bunches in figures 8(a) and (c) is equal to that in figures 4(a) and (c), while the bunches with the length of $3l_z/2$ disappear in figure 8(b) and the distribution has a period of $3l_z$ due to the disagreement between the spatial periodicity of the bumpy nanowire and the Bloch wavenumber. Taking into account the transmission properties of the bumpy nanowire, we can conclude that the period of oscillation results in the least common multiple of π divided by the z component of the Bloch wavenumber, π/k_z, and the geometric period of the nanowire, l_z.

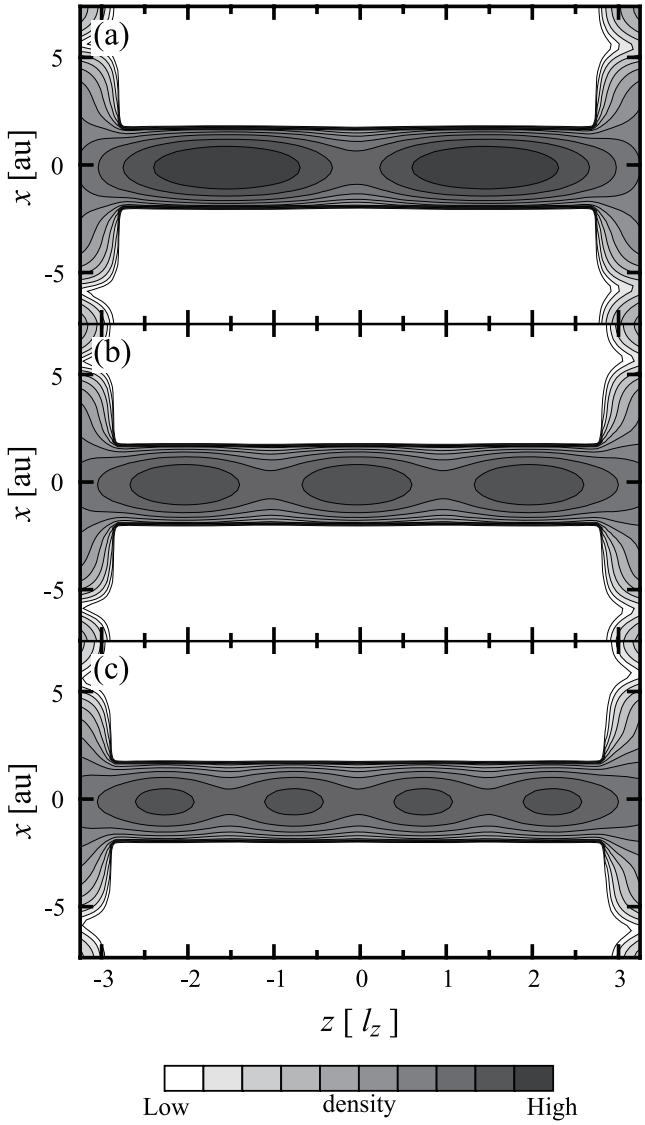

Figure 4. Electron charge density distributions of scattering wavefunctions Ψ^L with the length of L_T = (a) 5.4375 l_z, (b) 5.5625 l_z, (c) 5.6250 l_z at the energy of $E(k_z)$ where k_zs are chosen as (a) $\pi/3l_z$, (b) $\pi/2l_z$ and (c) $2\pi/3l_z$ in figure 2. Each contour represents twice or half the density of adjacent contour lines.

In the case of the Na and Au nanowires, since the band comprising s electrons crosses the Fermi level at $k_z = \pi/2d$, where d is the interatomic distance, the oscillation emerges. Smit *et al* [15] reported that the even–odd oscillation is also observed in Pt and Ir nanowires although Pt and Ir nanowires have 5d electrons at the Fermi level. Yet the 6s band of the Pt and Ir nanowires crosses the Fermi level at $k_z = \pi/2d$ and the contribution of the channel composed of the 6s electrons is observed in the experiment. Generally, the nanowire consisting of atoms with many valence electrons has plural energy bands crossing the Fermi level and a period of the conductance oscillation expected to be the least common multiples of the channels

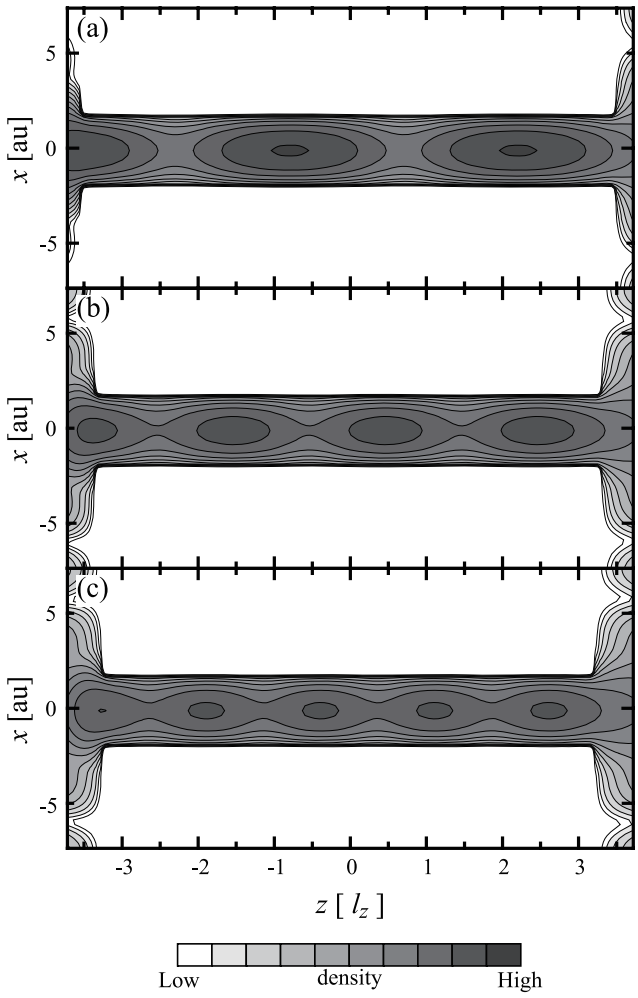

Figure 5. Electron charge density distributions of scattering wavefunctions Ψ^L with the length of L_T = (a) 6.9375 l_z, (b) 6.5625 l_z, (c) 6.3750 l_z at the energy of $E(k_z)$ where k_zs are chosen as (a) $\pi/3l_z$, (b) $\pi/2l_z$ and (c) $2\pi/3l_z$ in figure 2. Each contour represents twice or half the density of adjacent contour lines.

consisting of these bands as well as the geometric period of the nanowires. Indeed, in the cases of the Pt and Ir nanowires, the oscillation is obscure and the conductance decreases with increasing nanowire length [15]; there should be the certain contribution from other channels composed of the 5d electrons. Since the long distance periodicities easily break due to defects and thermal vibration of atoms, the conductance oscillation with a long period is expected to be hardly observed. However, the conductance oscillation with a period longer than the two-atom length can be observed if nanowires without any defects are formed in experiments.

4. Conclusion

We have studied the conductance oscillation of the plain and bumpy jellium nanowires suspended between semi-infinite jellium electrodes and discussed whether the conductance

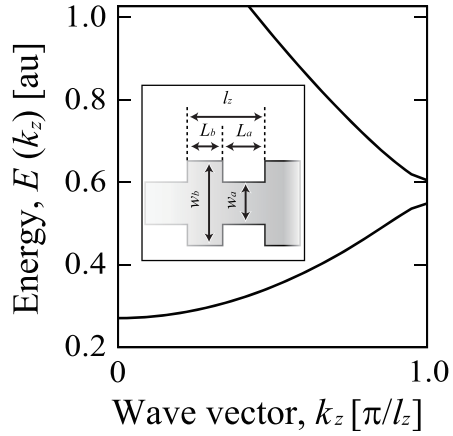

Figure 6. Electronic band structure of an infinite bumpy jellium wire. Inset: computational model for an infinite bumpy jellium wire.

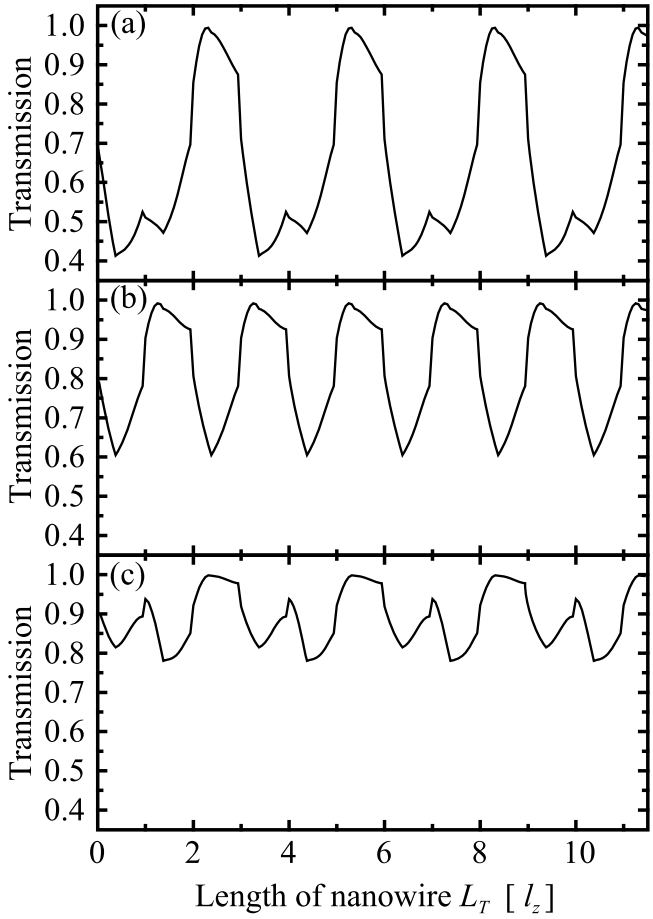

Figure 7. Transmission trace at the energy of $E(k_z)$ as a function of the length of a bumpy nanowire, where k_zs are chosen as (a) $\pi/3l_z$, (b) $\pi/2l_z$ and (c) $2\pi/3l_z$ in figure 6.

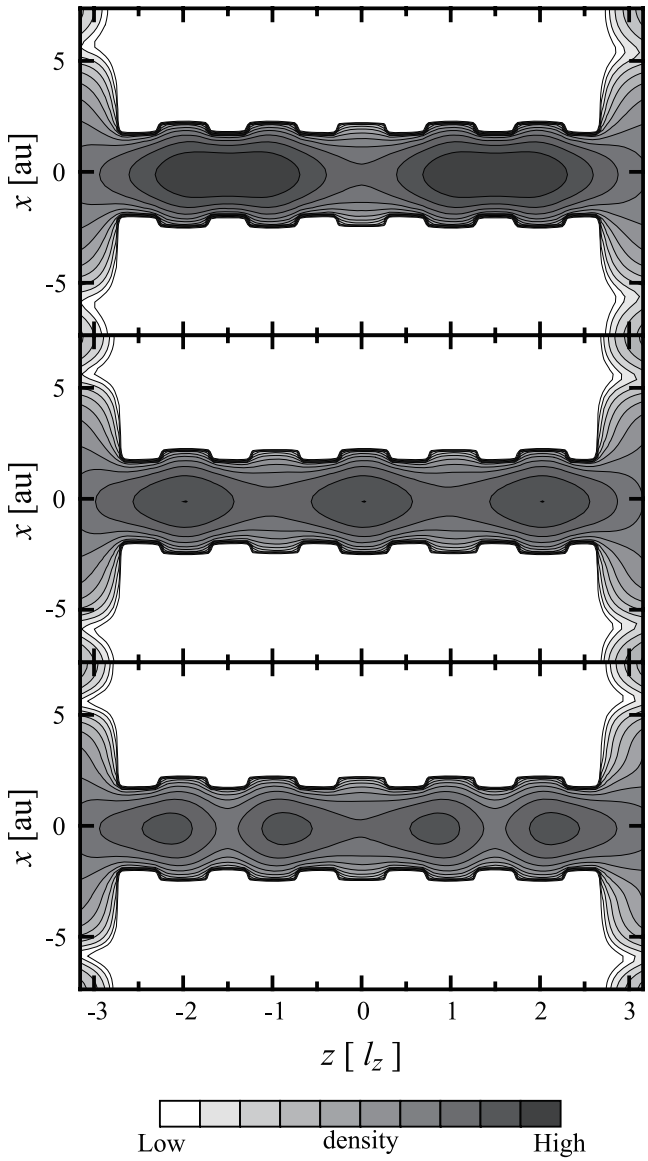

Figure 8. Electron charge density distributions of scattering wavefunctions Ψ^L with the length of L_T = (a) 5.3125 l_z, (b) 5.2500 l_z, (c) 5.3125 l_z at an energy of $E(k_z)$, where k_zs are chosen as (a) $\pi/3l_z$, (b) $\pi/2l_z$ and (c) $2\pi/3l_z$ in figure 6. Each contour represents twice or half the density of adjacent contour lines.

oscillation with a period longer than the two-atom length can be observed. The oscillation is driven by the quantum-mechanical wave character of electrons rather than the occupation of the states around the Fermi level. In addition, the period of the transmission oscillation corresponds to π divided by the z component of the Bloch wavenumber of the electrons in the nanowire region. In the case of a bumpy nanowire, the period of oscillation coincides with the least common multiple of π divided by the z component of the Bloch wavenumber and the geometric period of the nanowire. Although the disorders of atomic strings are hardly

eliminated in experiments, our result indicates that oscillation with at least a three- or four-atom period is conceivable.

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