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2007 J. Phys.: Condens. Matter 19 056010

(http://iopscience.iop.org/0953-8984/19/5/056010)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **19** (2007) 056010 (7pp) doi:10.1088/0953-8984/19/5/056010

Conductance oscillation and quantization in monatomic Al wires

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Received 31 August 2006, in final form 16 December 2006
Published 15 January 2007

Abstract
We present first-principles calculations for the transport properties of monatomic Al wires sandwiched between Al(100) electrodes. The conductance of the monatomic Al wires oscillates with the number of constituent atoms as a function of the wire length, either with a period of four atoms, for wires with the typical interatomic spacing, or with a period of six atoms, for wires with the interatomic spacing of bulk face centred cubic aluminium, indicating a dependence of the period of conductance oscillation on the interatomic spacing of the monatomic Al wires.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Atomic wires, which can be produced by means of either scanning tunnelling microscopy [1], mechanically controllable break junction techniques [2], or transmission electron microscopy [3], have attracted increasing attention both theoretically and experimentally [4]. For monovalent atomic wires, the even–odd oscillations of the wire conductance with the wire length have been analysed explicitly and demonstrated numerically [5], and perfect transmission is found for atom chains of an odd number of atoms and a smaller transmission in the even number case. The even–odd oscillations of conductance have been confirmed experimentally for atomic wires formed by noble-metal Au, Pt and Ir atoms [6]. An anomalous oscillation of conductance is found for atomic chains composed of alkali-metal atoms (such as Na, Cs) [7–9], with perfect transmission in the case of even number. However, for polyvalent atomic wires such as Pb and Al, multiple conductance channels contribute to the conductance, the situation becomes more complicated, and more interesting phenomena are expected.

The transport properties of monatomic wires formed by Al atoms have been studied with the first-principles method by several groups [10–12]. Lang has investigated the resistance of Al atomic wires connecting two semi-infinite metallic electrodes [10]. Kobayashi *et al* [11] have studied the conducting channels for monatomic Al wires by the recursion–transfer matrix method. They reveal that three open channels can contribute to the current through the Al atomic wire, and channel transmissions are sensitive to the geometry of the wire. Ono and Hirose [12] have studied the electronic conductance of a three-Al-atom form suspended between genuine semi-infinite aluminium crystalline electrodes. They have demonstrated that just one conducting channel is widely open at the Fermi level. However, we notice that, in the above calculations, the interatomic spacing between Al atoms is assumed to be the same as the bulk fcc one. The influence of the electrode model structure on the sandwiched wire has been clarified by Fujimoto *et al* [13]. Conductance oscillations with a period of four atoms have been shown by Thygesen and Jacobsen with the help of a plane-wave based pseudopotential code for the Al wire between two Al(111) electrodes [14]. The interatomic spacing of the wire is set to be 2.39 Å, the typical interatomic distance of the Al wires.

In the present paper, we perform first-principles calculations on the conductance of the monatomic Al wires attached to a pair of Al(100) bulk electrodes so as to clarify the effect of the interatomic spacing of the wire. Our results are consistent with those obtained in [14], when a smaller interatomic atomic spacing (the typical interatomic distance of Al wires) is used, while if we choose the interatomic distance of the bulk fcc aluminium, the six-atom period of the conductance oscillation is observed, which is different from the former case. Our results suggest that the period of conductance oscillation depends on the interatomic spacing of the wire.

2. Methodology

The calculations were performed by using a recently developed first-principles package TranSIESTA-C [15–17]. The package is based on the combination of density functional theory (DFT) implemented in the well tested SIESTA method with the nonequilibrium Green function technique. TranSIESTA-C is capable of modelling self-consistently the electrical properties of nanoscale devices that consist of an atomic scale system coupled to two semi-infinite electrodes. The system considered can be divided into three parts: the left electrode, the scattering region and the right electrode. The central region also consists of two layers of surface atoms on the left and three layers of surface atoms on the right in order to include the interaction between the electrode and the atomic wire. The end of the Al atomic wire is fixed at the hollow sites of the Al(100). The distance between the end atom of the wire and the electrode edge is set to be 1 Å. In our calculations two values of interatomic spacing of the wire are used: one is 2.86 Å, the same distance as in the bulk fcc aluminium; the other is 2.39 Å, a typical Al–Al atom spacing. The z-direction is set to be parallel to the wire axis. Figure 1 shows the system that we considered: a monatomic Al wire is sandwiched between a pair of Al(100) electrodes.

3. Results and discussion

We first calculate the conductance of the monatomic Al wire with the typical interatomic spacing 2.39 Å, as a function of the number of constituent aluminium atoms. The results are shown in figure 2(a). The figure clearly shows an oscillation with a period of four atoms which

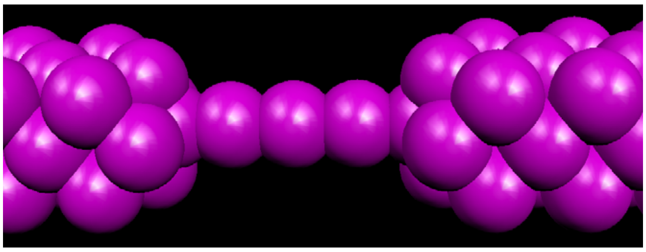
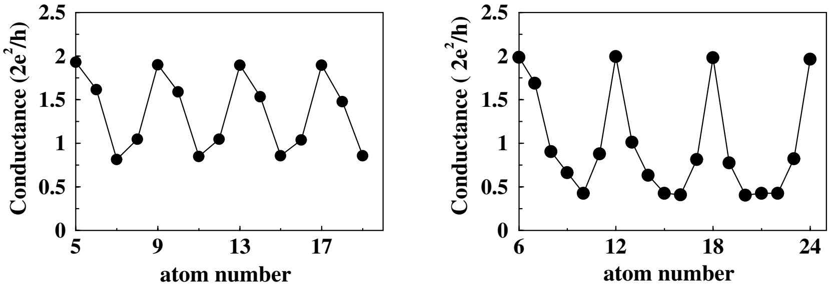


Figure 1. A schematic representation of an aluminium atom chain with five Al atoms sandwiched between a pair of Al(100) electrodes.



(a)

(b)

Figure 2. Conductance of monatomic Al wires connecting to a pair of Al(100) electrodes as a function of the number of Al atoms. (a) The Al interatomic spacing is 2.39 Å. (b) The interatomic spacing is 2.86 Å which is the same as that of the bulk fcc aluminium.

is similar to the one found in [14]. However, it is worth pointing out that in our calculations, the equilibrium conductance of the system varies between 0.8 and 1.9 G_0 ($G_0 = 2e^2/h$), and the maximal conductance occurs as the number of wire atoms takes the values 5, 9, 13, . . ., while in [14], the maximal values of the conductance correspond to the atom numbers 3, 7, 11, . . ., and the conductance range is 0.5–1.7 G_0. The different phases and amplitudes of the conductance oscillations may originate from the geometrical structure difference of the electrodes. We use a pair of semi-infinite Al(100) electrodes while two Al(111) electrodes are adopted in [14].

One may ask whether, if the monatomic Al chain is elongated, namely, the spacing between Al atoms is increased, can one expect the same four-atom period for the conductance as a function of the number of Al atoms? To answer this question, we calculate the conductance of Al atom wires coupled to the same pair of Al(100) electrodes as the number of Al atoms forming the chains, as the spacing of the chains is chosen the same as the interatomic spacing of the bulk fcc aluminium, 2.86 Å. The results are shown in figure 2(b). To our surprise, the conductance exhibits oscillations with a period of six atoms. Such an observation suggests that the period of the conductance oscillations of monatomic Al wires is related to the interatomic spacing of the wires.

To confirm our observation of the conductance oscillation with a period of four atoms or six atoms for the monatomic Al wire with different interatomic spacings sandwiched between a pair of Al(100) electrodes, we also calculate the transmission spectra of the system and show the results in figure 3. One can see a clear level splitting as the number of resonant

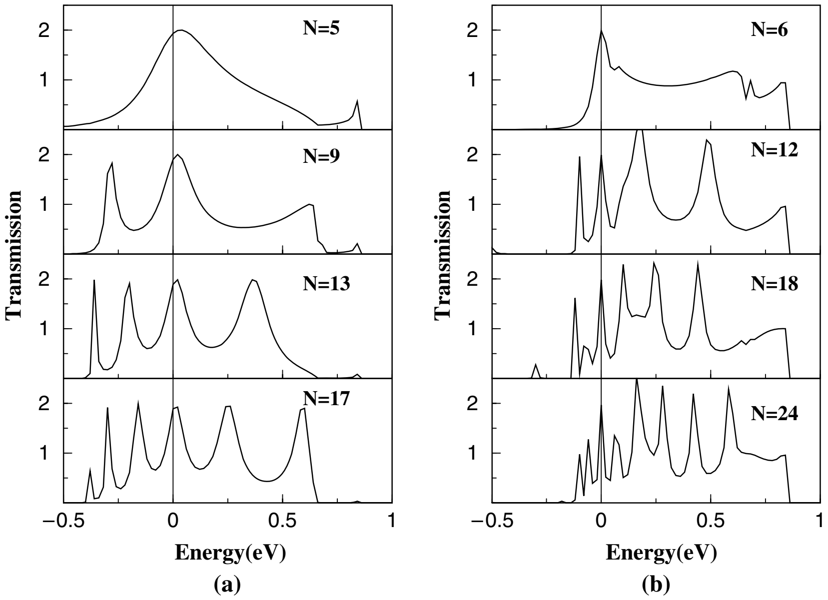


Figure 3. Transmission spectra for monatomic Al wires for different numbers of constituent Al atoms. (a) With the typical interatomic spacing of Al 2.39 Å. (b) With the interatomic spacing of the bulk fcc aluminium 2.86 Å.

peaks increases with increase of the number of constituent Al atoms. This can be easily tested from an analysis of the molecular projected self-consistent Hamiltonian (MPSH). The eigenstates of the MPSH are in fact the molecular orbitals renormalized by the molecule–electrode couplings. As an example, one would expect 12 eigenlevels for the wire containing 12 monovalent atoms. According the Landauer–Büttiker theory, the equilibrium conductance is proportional to the transmission probability at the Fermi level. Therefore the conductance oscillations of monatomic Al wires are equivalent to the coincidence of transmission peaks at the Fermi level for some specific numbers of Al atoms forming the wire. From figures 3(a) and (b), one clearly sees such a coincidence of transmission peaks, as the numbers of Al atoms are 5, 9, 13, 17 for the wire with the interatomic distance of the bulk fcc aluminium, and 6, 12, 18, 24 for the wire with the typical interatomic distance of Al.

The eigenchannel decomposition of transmission spectra provides some useful information about transport properties of two-probe systems. Therefore, we look at the transmission spectra of a three-Al-atom wire with the interatomic spacing of the bulk fcc aluminium 2.86 Å. The eigenchannel transmission spectrum is shown in figure 4. We note that the transmission spectra in this work are different from the previous results for the three-Al-atom wire based on various types of jellium electrode model. This confirms that the conductance of Al atomic wires is dependent on the detailed geometric structure of the electrodes [13].

It is well known that the valence electron configuration of the Al atom is $3s^23p^1$. The s and p_z orbitals constitute σ character channels and the p_x, p_y orbitals constitute degenerate π character channels, as clearly shown in figure 4. The σ channel contributes nothing to the current and thus the equilibrium conductance, because figure 4 tells us that the transmission of the σ channel at the Fermi level is zero. Two degenerate π channels contribute a small amount to the equilibrium conductance of the three-Al-atom wire, since the transmission probabilities of these two degenerate π channels are the same, with a value of about 0.07. We also calculate

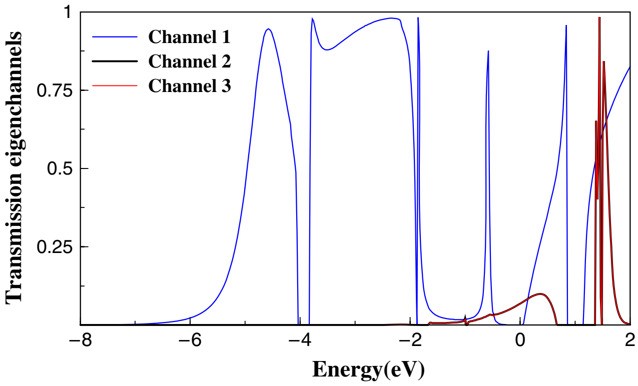


Figure 4. Eigenchannel transmission spectra of the σ channel (channel 1) and the two degenerate π transmission eigenchannels (channels 2 and 3) for the three-Al-atom wire with the interatomic spacing of the bulk fcc aluminium attached to a pair of Al(100) electrodes.

the eigenchannel transmission spectra for the monatomic Al wire with different numbers of constituent atoms. We find that for the Al wires containing 6, 12, 18 Al atoms, two degenerate π channels contribute mainly to the equilibrium conductance, each contribution to the conductance being about one conductance quantum: one G_0; and the contribution from the σ channel is still zero. Such a conductance quantization is suggested to be as a result of the charge neutrality and resonant character of the sharp tip structure of the wire–electrode contacts [18].

To explain the six-atom period of the conductance oscillation of monatomic Al wires with the interatomic spacing of the bulk fcc aluminium, we perform band structure calculations for an infinite Al wire with the typical interatomic distance 2.39 Å and the interatomic distance of the bulk fcc aluminium 2.86 Å attached to a pair of Al(100) electrodes. The results are shown in figure 5. The period of conductance oscillations is determined by the filling factor of the conduction bands with consideration of the local charge neutrality [14]: the filling factor is the inverse of the period of the conductance oscillation. Comparing the band structures of the infinite Al wire with the typical interatomic spacing and with the interatomic spacing of the bulk fcc aluminium, we find that the valence band—the bonding σ band, formed by the σ orbitals of Al atoms—is fully filled in both cases. For the infinite Al wires with the typical interatomic spacing, the conduction band is formed by the degenerate π orbitals, while the conduction band includes the contributions of both the anti-bonding σ band and the degenerate π band for the monatomic Al wires with the interatomic distance of bulk fcc aluminium. Such a dependence of the energy band structure of an infinite Al wire on the interatomic distance may be associated with the Pierce distortion effect, and is similar to the findings by Okano *et al* [19]. It is known that each Al atom provides three electrons with two occupying the bonding σ valence band and another occupying the conduction band. Therefore one finds, for infinite monatomic Al wires, a filling factor $1/4$ with the typical interatomic distance and a filling factor $1/6$ with the interatomic distance of the bulk fcc aluminium. It is such a filling factor imposed by local charge neutrality that causes the conductance oscillation with a period of either four atoms or six atoms, depending on the interatomic distance of Al atoms forming the wire. However, it is difficult to determine the phase of the conductance oscillations of the monatomic Al wires, due to the limitations of the software package TranSIESTA-C that we have used.

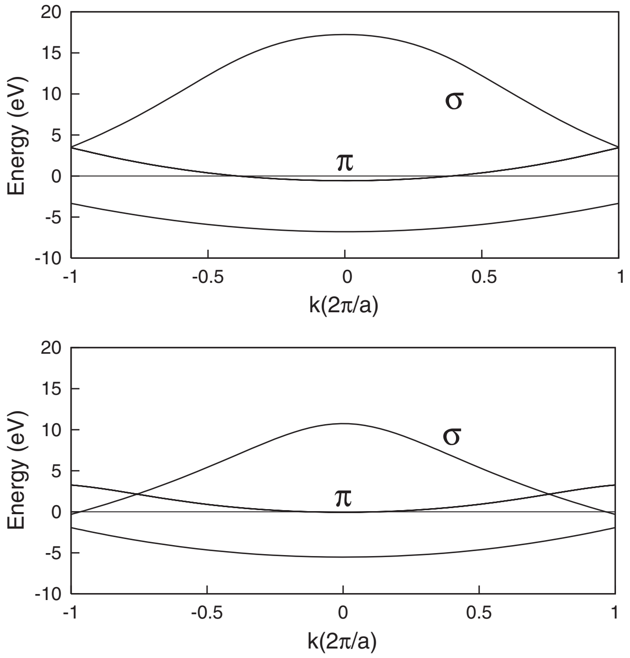


Figure 5. The energy band structure of infinite Al wires with the typical interatomic distance 2.39 Å (upper panel) and the interatomic distance of bulk fcc aluminium 2.86 Å (lower panel).

4. Conclusion

In conclusion, we have investigated conductance oscillations of monatomic Al wires with different interatomic spacings sandwiched between a pair of Al(100) electrodes with a well developed software package TranSIESTA-C. Conductance oscillations with periods of four atoms and six atoms are observed for the monatomic wire with the typical interatomic distance and the interatomic distance of bulk fcc aluminium. The period of the conductance oscillations is determined by the filling factor of the conduction band of the Al wires with specific interatomic spacing.

Acknowledgments

This work was supported by the National Natural Science Foundation of China under Grant No 10404010, the project sponsored by SRF for ROCS, SEM, the Foundation of Jiangxi Educational Committee under Grant No 112[2006], and the Talent Fund of Jiangxi Normal University under Grants Nos 1186 and 1187. BL was supported in part by an FRG grant of NUS.

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