First-principles study of transport properties of Al wires: Comparison between crystalline and jellium electrodes

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(Received 16 May 2005; published 15 September 2005)

We investigate effects of electrode structures on transport properties of Al monatomic wires sandwiched between Al electrodes by using first-principles calculations. To exhibit the effects, we compare the conductance obtained by employing the jellium electrode with that by the crystalline one. Although the conductance spectrum of the jellium-electrode model is considerably different from that of the crystalline-electrode one, setting Al(001) atomic layers on the jellium electrode surface improves the conductance values of the jellium-electrode models at the Fermi energy.

DOI: 10.1103/PhysRevB.72.113407 PACS number(s): 73.21.Hb, 73.23.Ad, 73.63.Rt

I. INTRODUCTION

In recent years nanostructured materials such as atomic and/or molecular wires can be experimentally synthesized owing to the continuous developments in nanofabrication techniques,[1] and it has been getting important to investigate the electronic properties of nanostructures from the viewpoints of fundamental physics and relevant practical applications. In particular, electron transport through nanostructures has received a great deal of attention due to the appearance of unique phenomena such as the quantization of conductance[2] and the negative differential conductance,[3] and now theoretical investigations on transport properties are rapidly emerging as an active research field in nanophysics.

One of the standard approaches of theoretical investigation is to calculate the transmission probability through a nanostructure on the basis of a scattering theory. In these calculations, metallic electrodes sandwiching the nanostructure are often modeled by using jellium electrodes without any atomic structures instead of crystalline ones, because of the computational simplicity. There have been a large number of theoretical works reported so far within the jellium model.[4–15] The jellium electrode, however, might lead to some serious problems in evaluating the transmission probability, because Bloch states in the crystalline electrode are not described properly. Although the jellium model has been widely employed so far, to our knowledge, there have been few works that examine in detail the effects of the jellium electrode.[16,17]

In this paper, we present first-principles calculations for the transport properties of the Al wire sandwiched between two Al(001) electrodes. We focus on the following question: can the jellium model describe the transmission properties of the Al wire successfully? We first investigate the transmission properties of the Al wire sandwiched between two jellium electrodes, and then examine how the transmission properties change when some Al atomic layers are loaded on the jellium substrate. Finally, we compare the results of the jellium model with those of the crystalline model and exhibit the effects of the jellium electrode.

The organization of the paper is as follows. Section II describes the theoretical method to calculate the electron transport. Section III is devoted to the results and discussion. Finally, we summarize our results in Sec. IV.

II. METHOD OF CALCULATION

We now consider several systems consisting of a three-atom wire and two semi-infinite jellium electrodes, as shown in Fig. 1. In the system A, the Al wire is directly connected to the bare jellium electrodes. In the systems B–D, the four-atom base, the base and the one Al(001) monolayer, and the base and the two Al(001) monolayers are inserted between the edge of the wire and the jellium electrode at each side, respectively. We also consider the wire with two bases sandwiched between two semi-infinite crystalline Al(001) electrodes, as shown in Fig. 2. Three atoms of the wire in all systems are fixed with an equispacing of 5.45 a.u. The distance between the jellium electrode surface and the Al atoms bordered on the jellium one is taken to be one-half of the interlayer distance, i.e., $0.25a_0$, where a_0 (=7.65 a.u.) is the lattice constant of the Al crystal. The distance between the edge atom of the wire and the base is set to be $0.5a_0$, as well as that between the base and the Al(001) surface. The x and y directions are taken perpendicular to the wire and periodic boundary conditions are employed for these directions, while the z axis is set parallel to the wire and an open boundary condition is employed. We use a unit cell with a length of 15.3 a.u. in the x and y directions, which corresponds to the Al(001)-$c(4\times4)$ surface.

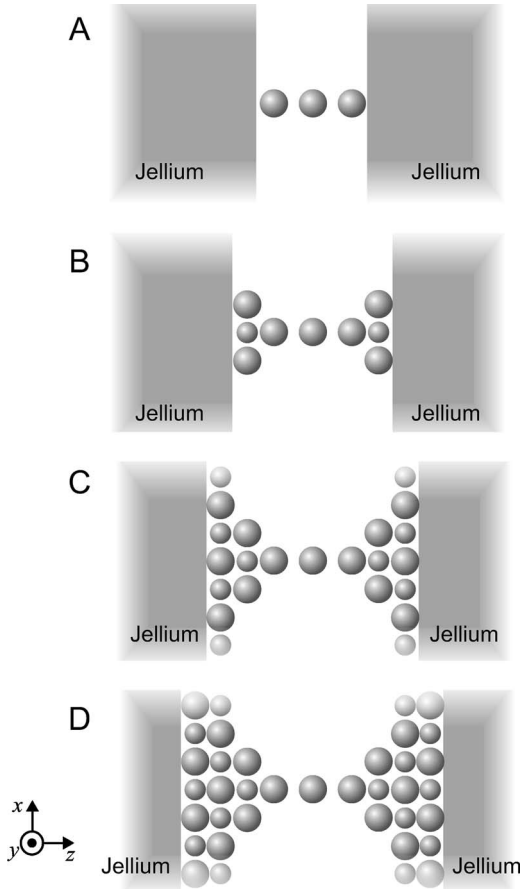

FIG. 1. Models of Al wires sandwiched between two jellium semi-infinite electrodes. In the system A, the wire is directly connected to the electrode. In the system B, the four-atom bases are inserted between the wire and the jellium electrode at each side. In the system C (D), one (two) Al(001) monolayer(s) is (are) inserted between the base and the jellium surface.

In the calculations of the jellium models shown in Fig. 1, we employ the Lippmann-Schwinger equation. The electronic structures and transport properties are calculated self-consistently. The scattering wave functions are expanded using two-dimensional plane wave basis sets with a cutoff energy of 9.0 Ry in the x-y direction and divided into the real-space grids with a size of 0.5 a.u. along the z direction. The electron density of the jellium electrodes is taken equal to the average valence-electron density of the Al bulk (r_s =2.0 a.u.). The details of the computational method has been described elsewhere.[4,7,13]

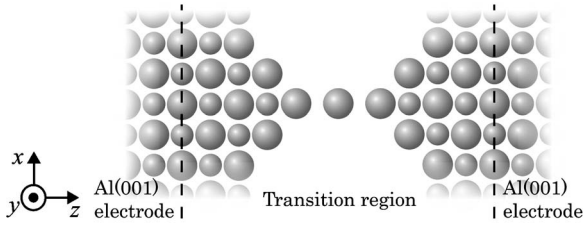

FIG. 2. Model of the Al wire with two bases sandwiched between two crystalline Al(001) semi-infinite electrodes. The transition region is chosen to be the area sandwiched between two dashed lines.

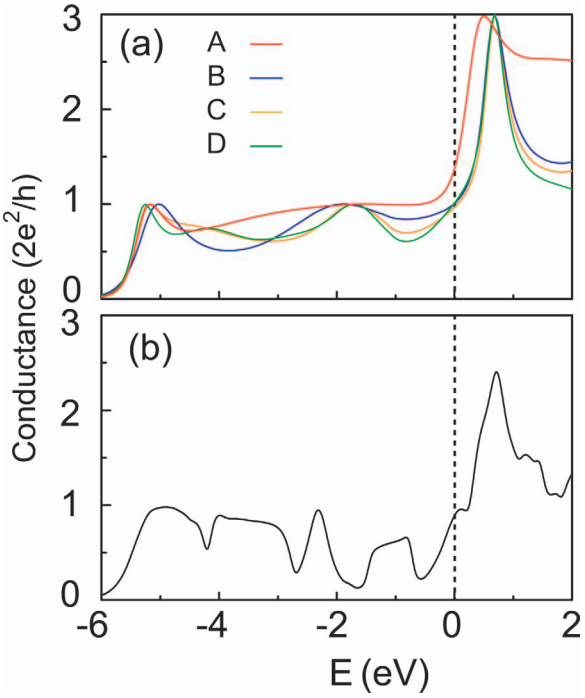

FIG. 3. (Color) Conductance spectra as a function of the electron incident energy for (a) the systems A–D and (b) the system shown in Fig. 2. The Fermi energy is zero.

The calculation of the conductance for the system shown in Fig. 2 is performed based on the real-space finite-difference approach.[19,20] The electronic structure of the transition region is determined self-consistently using the conventional supercell geometry under a three-dimensional periodic boundary condition with a mesh size of 0.76 a.u., which corresponds to a cutoff energy of 16.9 Ry. The effective potential obtained in this way is employed for evaluating the electron transmission. Then, the scattering wave functions are constructed from the overbridging boundary-matching (OBM) method, whose theoretical details are described in previous papers.[16–18]

In all cases of our calculations, the norm-conserving Troullier-Martins pseudopotentials are employed to describe the electron-ion interactions,[21,22] and exchange-correlation term is treated with the local-density approximation parametrized by Perdew and Zunger[23] within the density-functional theory. We calculate the conductance G associated with the transmission T by the Landauer-Büttiker formula $G=G_0T$, where $G_0=2e^2/h$ (e, the electron charge; h, Planck's constant).[24] We use only the Γ point in the two-dimensional Brillouin zone for the evaluation of the electron transmission.

III. RESULTS AND DISCUSSION

Figure 3 shows the conductance spectra of the Al wire systems as a function of an electron incident energy E, where the Fermi energy is set to be 0 eV. The spectra A–D in

Fig. 3(a) correspond to the systems A–D, respectively. For the system A, the conductance is negligible in an energy range of $E<-6$ eV. As the energy increases from −6 eV, the conductance grows rapidly and reaches a resonance peak with a value of $\sim 1G_0$ at $E=-5$ eV. After a plateau structure appears in a range of -4 eV$<E<0$ eV, the conductance rapidly increases again and is enhanced to $3G_0$ around $E=1$ eV. These conductance properties are roughly comprehensible from the electronic states of the infinite Al monatomic wire, as previously reported.[7,10] In the case of the infinite Al wire, the valence band bottom lies around −6 eV. From the bottom to the Fermi energy, there is only a σ state composed of s and p_z orbitals and around the Fermi energy π states consisting of p_x and p_y orbitals appear. Accordingly, the conductance of the system A has a negligible value below −6 eV. Only one channel (σ state) contributes to the conductance spectrum from −6 eV to ~ Fermi energy. Thus, the conductance takes $1G_0$ at most in this energy range. On the other hand, in an energy range of $E>E_F$, the contribution of two degenerate channels (π states) to the conductance appears. Therefore, the conductance in this energy range consists of three channels in total, yielding the maximum value of $3G_0$.

Concerning the contact between the electrode and the wire, the system A lacks the influence of the contact atomic configurations, such as the orientation of the electrode surfaces. We here investigate the effect of the four-atom base inserted between the wire and the jellium electrode (system B). The conductance spectrum of the system B is almost similar to that of the system A in an energy range below $E=-5$ eV, while it is quite different above −5 eV; three large dents are found around −4 eV, −1 eV, and 2 eV, especially, the decrease around 2 eV is remarkable. The peak of the system B above the Fermi energy moves towards the higher energy and its width becomes narrower than that of the system A. This implies that the interaction between the Al wire and the electrodes becomes weak.[25] In this way, the conductance spectrum changes largely due to the existence of the four-atom base.

We also investigate the effect of piling up Al(001) atomic layers on the jellium electrode surfaces (see the systems C and D). When one Al(001) monolayer is inserted between the base and the jellium electrodes (system C), the conductance spectrum considerably changes, in particular the changes around −4 eV and −1 eV are remarkable. A protrusion around −4 eV becomes noticeable in the conductance spectrum when one more Al(001) monolayer is added (system D). Thus, the pileup of the Al(001) monolayer induces the sizable change in the conductance spectrum. It is noted that the spectrum is almost unchangeable from $E=0$ to 1 eV.

In Fig. 3(b), we present the conductance spectrum of the crystalline-electrode model shown in Fig. 2. As mentioned above, the features of the conductance spectrum can be roughly understood in terms of the electronic states of the infinite Al wire. Below the Fermi energy, only one channel contributes to the conductance, showing a value less than $1G_0$, while above the Fermi energy the conductance has a large peak with a value of $\sim 2.5G_0$ due to the contribution of two degenerate channels. However, we find that the curve of the conductance spectrum for the crystalline model is considerably different from that for the jellium one, especially, the discrepancies between both models are remarkable in the range of -5 eV$<E<E_F$. We surmise that such discrepancies are mainly caused by the following two reasons: One is that unfavorable reflections of the incident electron waves might happen because of the existence of the artificial interface between the Al(001) atomic layer and the jellium electrode. The other is that the characters of the incident waves propagating through the crystalline electrode are much different from those through the jellium one. However, these questions remain unresolved. Further investigations should be addressed in future work.

TABLE I. Conductance values at the Fermi energy for the systems A–D and the crystalline electrode.

	System				
	Jellium electrode				
	A	B	C	D	Crystalline electrode
Conductance (G_0)	1.44	1.02	0.98	1.03	0.89

It is expected that the conductance spectrum of the jellium-electrode model gets close to that of the crystalline-electrode one with increasing the number of the Al(001) atomic layers inserted between the wire and the jellium electrode surface. Nevertheless, it seems difficult to reproduce satisfactorily the intricate form of the conductance spectrum of the crystalline model with a stack of a few Al(001) layers on the jellium electrode surface (see Fig. 3). In the limit of the zero-bias voltage, the conductance at the Fermi energy is of great importance. In Table I, we show the conductance values at the Fermi energy. The conductance values of the jellium models except the system A show almost the same value of $1.0G_0$, whereas the conductance of the crystalline model is approximately $0.9G_0$. These results show good agreement with the experimental results, $0.8G_0-0.9G_0$.[26,27] Eventually, it would be appropriate to investigate the transport properties of the Al wire near the Fermi energy by means of the jellium-electrode model, if some Al atomic layers are set on the jellium electrode surface.

One of the authors has reported the conductances of the gold monatomic wires sandwiched between two jellium Au electrodes and between two crystalline Au electrodes.[17] Their results have shown that the conductances of both models have almost the same values at the Fermi energy, nearly $1G_0$. The electronic state of Au near the Fermi energy is regarded as a simple metal because it is mainly determined by $6s$ valence electrons, while Al is also treated as a simple metal. Therefore, it might be available to replace a crystalline electrode of a simple metal by a jellium electrode when the transport properties near the Fermi energy are investigated.[28]

We finally mention effects of the electrode geometries on the transport properties. Palacios *et al.* reported the conductance spectra of Al wires sandwiched between two Al electrodes with a pyramidal shape.[29] The conductance spectrum in their study is quite different from that in our calculations, though the three-atom wire sandwiched between two four-

atom bases is treated in both cases. These discrepancies seem to be mainly attributed to the geometrical difference of the electrode, i.e., the pyramidal shape in their paper or the Al(001) face in our calculations. This indicates that the conductance spectrum is not determined by only the contact geometry between the Al wire and the electrode, and the electrode geometry plays an important role in evaluating the electron transport properties.

IV. SUMMARY

We have investigated the transport properties of the Al wires sandwiched between the metallic jellium electrodes as well as between the Al(001) crystalline electrodes. The whole conductance spectrum obtained by using the jellium electrodes is much different from that by the crystalline electrode. If some Al atomic layers are laid on the jellium electrode surface, however, the transmission properties in the jellium model near the Fermi energy exhibit similar behaviors in the crystalline model, especially, the conductances at the Fermi energy in both models are close to $1G_0$.

ACKNOWLEDGMENTS

The authors thank Dr. H. Kino and Dr. W. T. Geng for useful comments. This work was done in "Frontier Simulation Software for Industrial Science (FSIS)" project supported by IT program of the Ministry of Education, Culture, Sports, Science, and Technology of Japan (MEXT), and partially performed through Special Coordination Funds of MEXT. The numerical calculations were carried out at National Institute for Materials Science (NIMS) and at the Institute for Solid State Physics, University of Tokyo.

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