Which nanowire couples better electrically to a metal contact: armchair or zigzag nanotube?

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Abstract

The fundamental question of how chirality affects the electronic coupling of a nanotube to metal contacts is important for the application of nanotubes as nanowires. We show that metallic-zigzag nanotubes are superior to armchair nanotubes as nanowires, by modeling the metal-nanotube interface. More specifically, we show that as a function of coupling strength, the total electron transmission of armchair nanotubes increases and tends to be pinned close to unity for a metal with Fermi wave vector close to that of gold. In contrast, the transmission probability of zigzag nanotubes increases to the maximum possible value of two. The origin of these effects lies in the details of the wave function, which is explained.

A nanotube's chirality is of prime importance in determining its electronic properties. Chirality determines whether a nanotube is metallic or semiconducting.[1] Ref. 2 showed that the bandgap change with tensile and torsional strain has a rather universal dependence on nanotube chirality. The electronic properties of zigzag and armchair nanotubes (two distinct chiralities) are also affected in very different manners upon bending.[3] From the view point of nanotubes in applications such as nanowires, it is critical to understand the physics of metal-nanotube coupling. We find that the overlap between nanotube and metal wave functions depend significantly on chirality. As a result, metallic-zigzag nanotubes [which are represented by (3 times integer,0)] are superiror to armchair nanotubes as nanowires.

We consider a single wall carbon nanotube coupled to a metal block in the side-contacted geometry [Fig. 1]. The metal contact is treated in the context of a free electron metal with a rectangular cross section in the (x,z) plane, and infinite extent in the y-direction as in most experiments. The surface Green's function of the metal contact is calculated using standard procedures. The nanotube is treated using the π orbital tight binding Hamiltonian. The coupling between the metal and the nanotube is modeled using a tunneling-type Hamiltonian, which is included to all orders (and not just Born approximation) in calculating the transmission probability. The details of modeling the metal-nanotube coupling can be found in reference 4. The total transmission (T) is the sum over the transmission probability of all modes at an energy. T at energy E is given by,[5] $T(E) = Tr\left[G^r(E)\Gamma_m(E)G^a(E)\Gamma_c(E)\right]$, where Γ_m and Γ_c are matrices that represent coupling between the metal and a semi-infinte nanotube region either to the left or right of the nanotube section shown in Fig. 1. G^r (G^a) is the full retarded (advanced) Green's function of the nanotube with coupling to metal and semi-infinite nanotube regions included.

The coupling strength of the metal contact to the nanotube is given by the diagonal component of Γ_m which is $|t_{mc}|^2\rho_m$, where ρ_m is the density of states of the metal surface and t_{mc} represents the hopping strength between nanotube atoms and metal in the Hamiltonian.[4] The electrical contact length (Fig. 1) between the metal and nanotube in this work is dictated by the available computational resources. The largest electrical contact length

considered is thirty nanotube unit cells (approximately 72 $\AA$ and 125 $\AA$ for armchair and zigzag nanotubes respectively). The dimensions of the metal contact are $L_x = 400 - 750\AA$ and $L_z = 750\AA$. The length of nanotube-metal electrical contact is kept constant at thirty nanotube unit cells, and the transmission is calculated as a function of coupling strength ($|t_{mc}|^2\rho_m$). Three values of metal Fermi wave vector (k_f) are considered, 1.75 $\AA^{-1}$ (Aluminum), 1.2 $\AA^{-1}$ (Gold/Silver) and 0.9 $\AA^{-1}$, where free electron metals with k_f close to the assumed values are indicated in the parentheses.

Fig. 2 shows the transmission probability as a function of coupling strength for a (5,5) armchair nanotube. The results show the dramatic effect that T is pinned close to unity for $k_f = 1.2$ and $0.9\AA^{-1}$. Close to the Fermi energy (nanotube band center), two subbands carry current in both the positive and negative directions. The above result indicates that only one of the two subbands couples well to the metal. For $k_f = 1.75$, T is well above unity, implying that both subbands couple to the metal. The wave functions of the crossing bands of the two positive going states of a (N,N) armchair nanotube are:[6]

$$\phi_{ac1} = e^{\frac{im_a k_a a_0}{2}}(-1)^{m_a}\,[1\ 1] \text{ and } \phi_{ac2} = e^{\frac{im_a k_a a_0}{2}}(-1)^{m_a}\,[1\ -1]\ , \tag{1}$$

where k_a is the axial wave vector of the nanotube, m_a is an integer that denotes the cross section along the axial direction [inset of Fig. 2], and $[u_1\ u_2]$ is the wave function of a unit cell of the underlying graphene sheet. For an armchair nanotube, there is *no modulation* of $[u_1\ u_2]$ around the circumferential direction. The wave function of one of the two subbands (ϕ_{ac2}) is rapidly oscillating with the nodes separated by $a_0 = 1.4\AA$. In comparison, the nodes of a metal wave function (ϕ_m) with $k_f = 0.9$, 1.2 and 1.75 $\AA^{-1}$, are separated by 6.3, 3.4, and 2 $\AA$ respectively, taking into account that the axial wave vector has to be at least 0.75 $\AA^{-1}$.[4] As a result of this, the integral entering the Born approximation for scattering rate,

$$\int \phi^*_{ac2} H_{c-m} \phi_m\ , \tag{2}$$

(H_{c-m} is the nanotube-metal coupling Hamiltonian) is very small for $k_f = 0.9$ and $1.2\AA^{-1}$, and is larger for $k_f = 1.75\AA^{-1}$, in that order. Thus T is pinned close to unity for

$k_f = 0.9$ and 1.2Å^{-1}, and is larger for $k_f = 1.75\text{Å}^{-1}$. Recently, Ref. 7 discussed an alternate mechanism by which only one of the two crossing subbands of an armchair nanotube contributes to transport. The nanotube can be divided into regions where the nanotube atoms make and do not make contact to the metal atoms. A shift in the band structure between these two regions by about 1.5 eV causes a reflection of electrons incident from the metal into one of the two crossing subbands, at the interface between the two regions, as proposed in reference 7. Our work includes such a shift but in comparison to reference 7, we find that the conductance can be around unity (for $k_f = 0.9$ and 1.2Å^{-1}) even when this shift is smaller than 1.5 eV. Also, we propose that the crossing subband with the smaller angular momentum contributes more significantly to transport.

Fig. 3 shows the metal-nanotube total transmission (T) as a function of coupling strength for a (6,0) zigzag nanotube. In stark contrast to the armchair case, T does not saturate at unity. With increasing coupling strength, T approaches two, the maximum value possible. That is, both positive going subbands contribute to transmission from metal to nanotube. The wave function of the two crossing subbands of a zigzag nanotube are:

$$\phi_{zz1} = e^{\frac{-i\sqrt{3}m_a k_a a_0}{2}} e^{\frac{i2\pi m_a}{3}} e^{\frac{i4\pi m_c}{3}} [u_1\ u_2] \text{ and } \phi_{zz2} = e^{\frac{-i\sqrt{3}m_a k_a a_0}{2}} e^{\frac{i4\pi m_a}{3}} e^{\frac{i8\pi m_c}{3}} [u_1\ u_2]\ , \qquad (3)$$

where, m_a is an integer that denotes the cross section along the axial direction and m_c is an integer denoting the various unit cells along the circumferential direction as shown in Fig. 3. The wave function along the circumferential direction varies much more slowly than the armchair wavefunction:

$$\phi_{zz}(m_a, m_c) + \phi_{zz}(m_a, m_c + 1) + \phi_{zz}(m_a, m_c + 2) = 0\ , \qquad (4)$$

which corresponds to a distance of $3a_0$ (7.5 Å) over which the wave function adds up to zero. As a result of this feature [Eq. (4)], both crossing subbands of a zigzag nanotube couple with metals. In Figs. 2 and 3, it is noted that for small coupling strengths, T is larger for the armchair nanotube than the zigzag nanotube case. This is because as a result of the small circumferential wave vector of ϕ_{ac1}, ϕ_{ac1} couples more strongly to the metal

than the sum of contributions from ϕ_{zz1} and ϕ_{zz2}. It is pointed out that at small coupling strengths, T is significantly larger in the case of the armchair nanotube. This is because both crossing subbands of the zigzag nanotube have enough angular momentum to make the overlap integral between the metal and nanotube wave functions small. With increasing coupling strengths, both crossing subbands of the zigzag nanotube however eventually couple well to the metal, unlike the armchair nanotube.

The calculations presented above consider the entire circumference of the nanotube to be coupled to the metal contact. Such a scenario is relevant to the experiment in Ref. 8, which resulted in a conductance of approximately $2e^2/h$. Other experiments involve the metal making contact to only part of the circumference of the nanotube.[9] We also perform calculations corresponding to this case. Sectors of varying lengths are considered, and the results do not change qualitatively from that presented below. The number of atoms around the nanotube circumference that couple to the metal contact is shown in the legend of Fig. 4. The main point is that the essential features of Figs. 2 and 3 are preserved when contact is made to a sector. The difference between a four and five atom sector is negligible in the zigzag case. The difference between the four and five atom sectors although small in the case of an armchair nanotubes, is larger than the difference for zigzag nanotubes. The reason for this, based on the discussion of scattering rate within the Born approximation above, is that the end odd atom [Fig. 2] corresponding to the wave function ϕ_{ac2} does not have a partner-atom to compensate (to make zero) its contribution to the scattering rate in Eq. (2).

Two practical issues, disorder/defects and length dependence, are discussed next. A ten percent random variation in coupling strength between the nanotube atoms and the metal does not cause a significant change in the results. From an experimental view point, a large random variation in coupling from atom to atom in a crystalline metal is unlikely. Defects in the nanotube such as the Stone-Wales defect will be more effective in destroying the discussed difference.

The transmission probability of an electron from the metal to the nanotube can be made

larger either by increasing the coupling strength or by increasing the area of electrical contact, between the nanotube and metal. From a technological perspective, the first alternative of small contact area (as assumed in this paper) along with strong coupling is more desirable. In typical experiments, the coupling between metal and nanotube is weak compared to the 0.2eV assumed for the largest coupling in Figs. 2 and 3, and the contact length is larger. The results of this paper are also qualitatively valid for a calculation where the coupling strength is constant and the electrical contact length is increased ('coupling strength' in the x-axis of Figs. 2 - 4 should be replaced by electrical contact length). Such a calculation however requires much larger computational resources because the computation scales as the cube of the number of atoms.

Many factors such as the role of curvature, torsion and tension of armchair and zigzag nanotubes play a role in determining the suitablility of nanotubes as nanowires. The small curvature induced band gap in large diameter metallic-zigzag nanotubes predicted by tight-binding theory is smaller than kT.[1] Further, reference 10 showed that a (6,0) nanotube is a perfect metal, contrary to the popular belief that all small diameter metallic-zigzag nanotubes have a small bandgap. This lends support to the use of metallic-zigzag nanotubes as nanowires. In this paper, we considered the role of the nanotube's electron wave function in determining the coupling strength to a metal contact, in the absence of significant defects. We find that zigzag nanotubes perform better than armchair nanotubes as nanowires. For Fermi wave vectors close to that of gold, the total transmission (T) of side-contacted armchair tubes is pinned close to unity. In contrast, the total transmission in case of zigzag tubes is close to the maximum possible value of two. This represents a two fold increase in the small bias current that can be driven through a zigzag nanoutube when compared to an armchair nanotube.

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Figure Captions:

Fig. 1: Nanotube lying on a metal contact. The metal contact is infinitely long in the y-direction (open boundaries), and thirty unit cells of the nanotube make electrical contact to the metal. Semi-infinite nanotube regions present to the left and right of the nanotube section are not shown. The total-transmission (T) is evaluated from the metal to either the semi-infinite nanotube region to the left or right.

Fig. 2: Plot of T versus coupling strength between metal and armchair nanotube. While for $k_f = 0.9$ and $1.2\mathring{A}^{-1}$, T is pinned close to unity, for $k_f = 1.75\mathring{A}$, T is larger.

Fig. 3: Plot of T versus coupling strength between metal and zigzag nanotube. In contrast to the armchair case, T increases to the maximum allowed value of two with coupling strength.

Fig. 4: Plot of T versus coupling strength between metal and nanotube for the case of a sector of the nanotube circumference making contact to the metal. The legend shows the number of contiguos atoms (see inset of Fig. 2) in a unit cell making contact. The essential features of Figs. 2 and 3 are retained. The metal Fermi wave vector was chosen to be close to that of gold ($1.2\ \mathring{A}^{-1}$).

Fig. 1 / Anantram

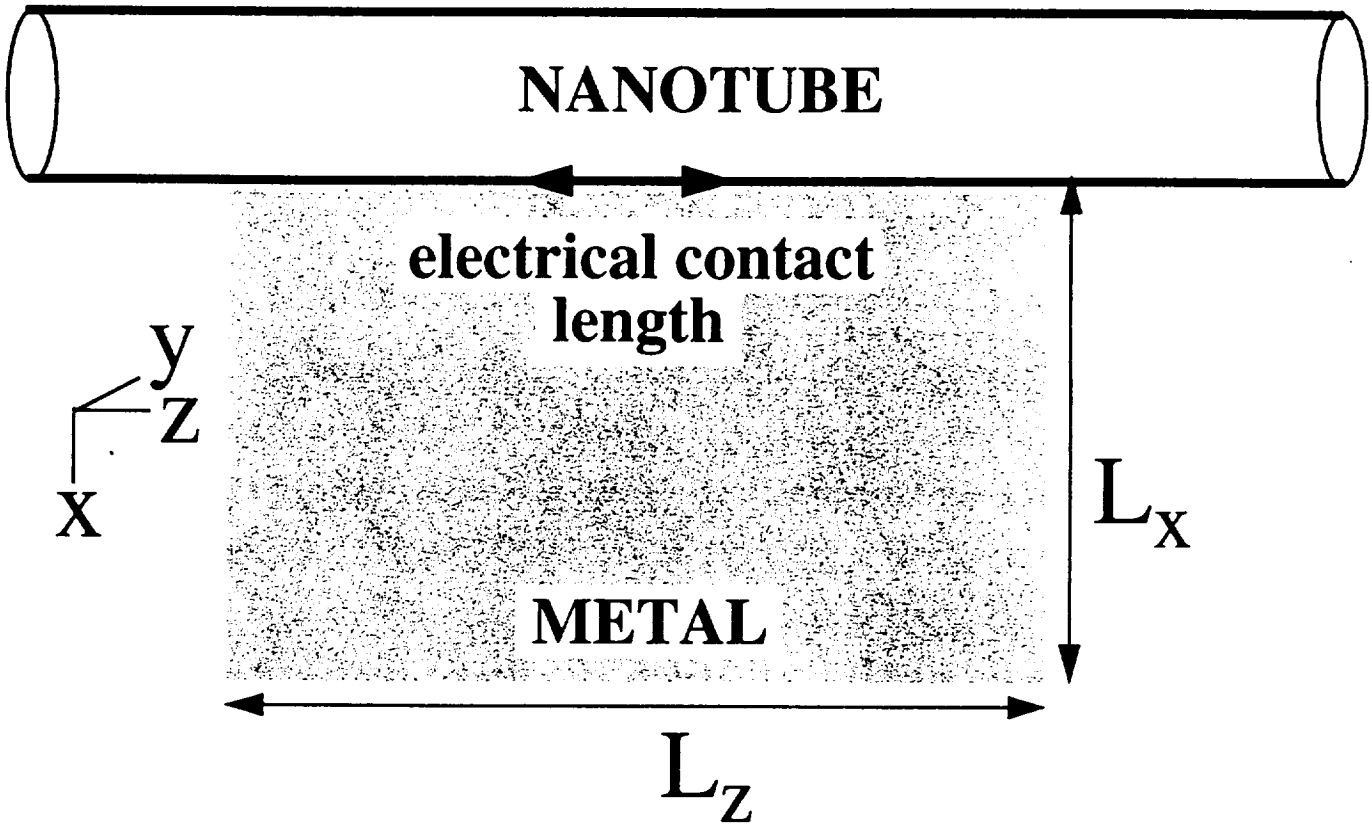

Fig. 2 / Anantram

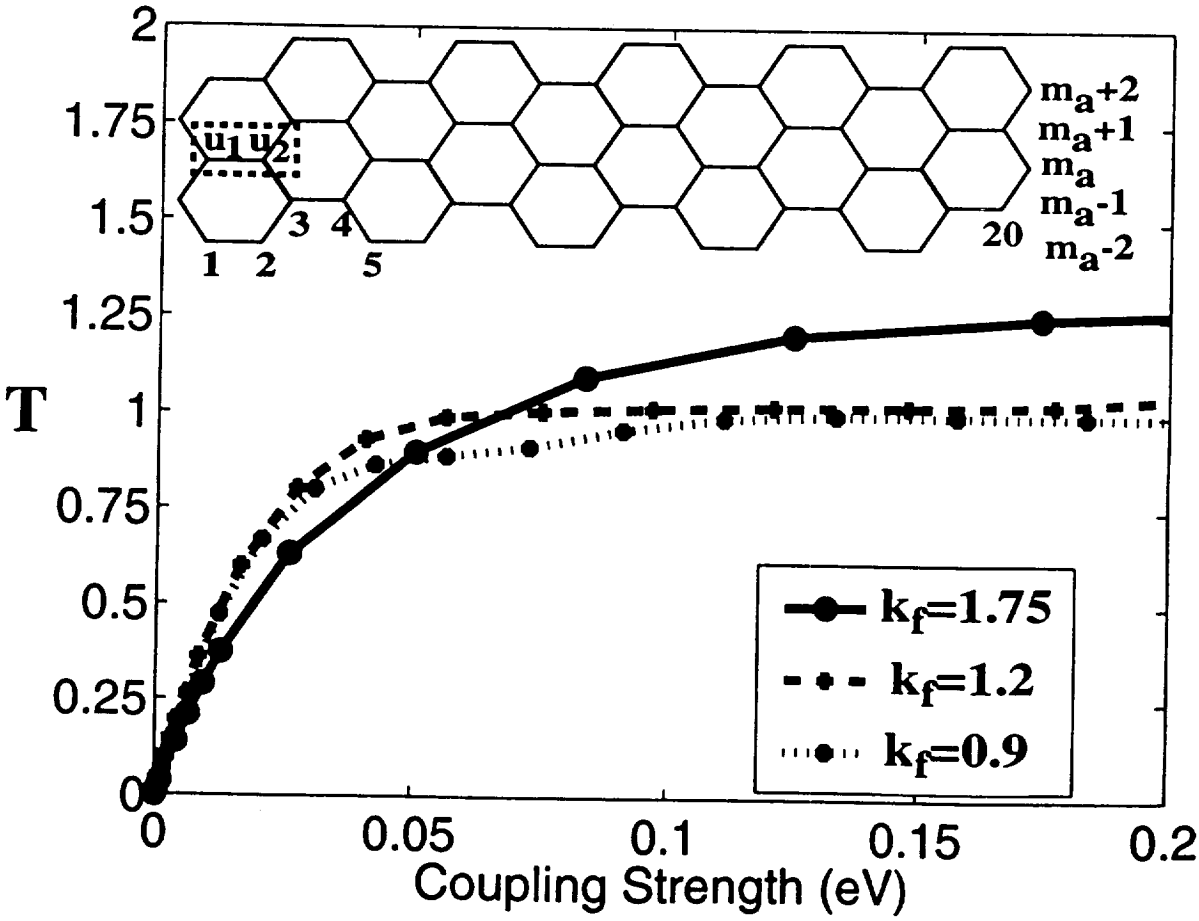

Fig. 3 / Anantram

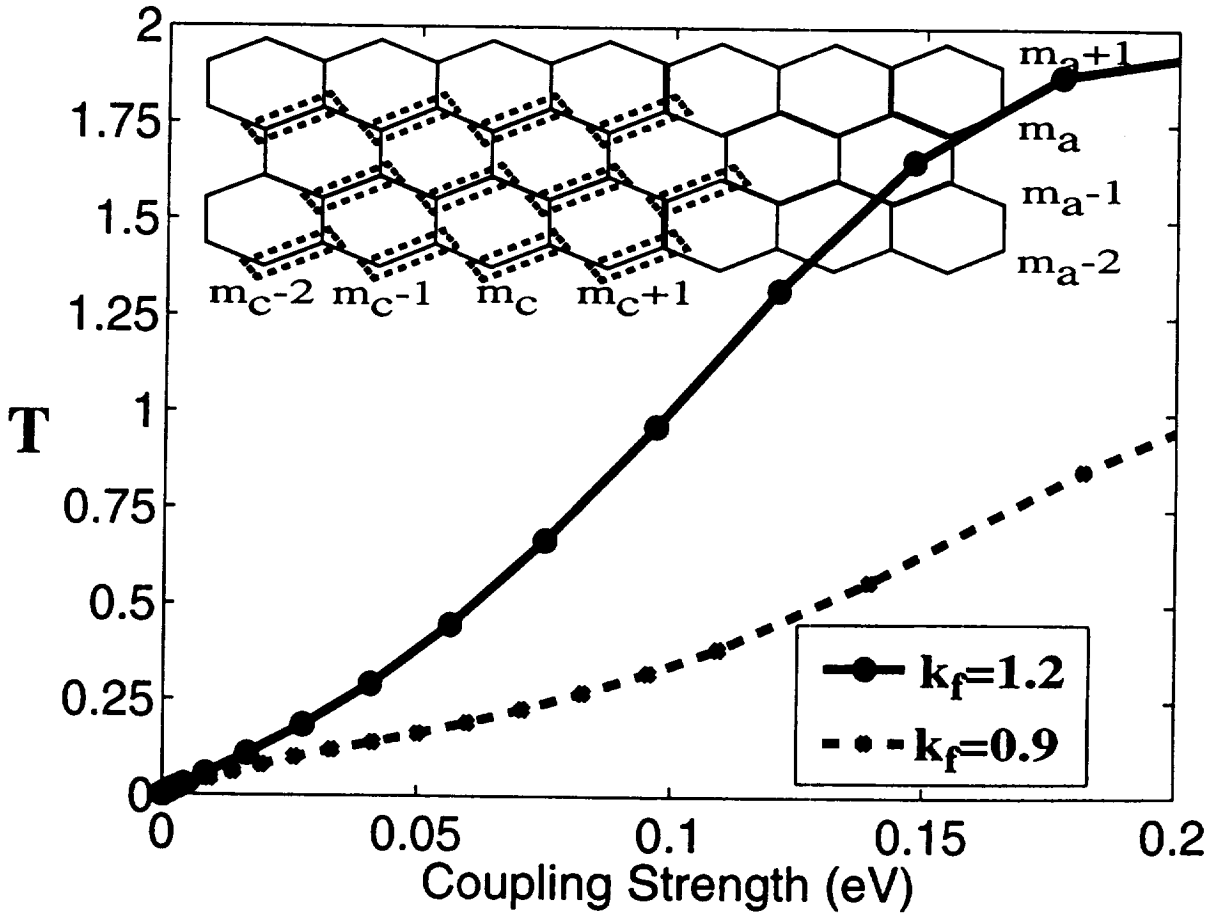

Fig. 4 / Anantram