Channeling of protons in radially compressed carbon nanotubes

A Karabarbounis¹, S Sarros¹ and Ch Trikalinos²

¹ Department of Physics, Section of Nuclear and Particle Physics, University of Athens, Panepistimioupolis, Iliissa, 15771 Athens, Greece
² Department of Philosophy and History of Science, University of Athens, Panepistimioupolis, Iliissa, 15771 Athens, Greece

E-mail: stsarros@phys.uoa.gr

Abstract. Channeling of 10 MeV protons in radially compressed chiral carbon nanotubes is considered. Monte Carlo simulation program is used for the calculation of the trajectories, energy losses and angular distributions of protons in nanotubes of various lengths, where the potential in Doyle-Turner approximation is used to describe the interaction between a proton and a nanotube. Calculations were carried out for different incident angles between proton beam and the nanotube axis. The results show that a decreased angular distribution of the beam is observed, compared with propagation through a straight nanotube, in case when it enters from the compressed end of the nanotube. The energy and spatial distribution of channeled protons in compressed nanotubes is examined.

1. Introduction

Carbon nanotubes were discovered more than twenty years ago, multi-wall carbon nanotubes (MWCNT or MWNT) in 1991 [1] and single-wall carbon nanotubes (SWCNT or SWNT) in 1993 [2,3]. Since then, an advanced progress on the investigation of their properties and production methods has been achieved. Due to their unique physical properties, carbon nanotubes (CNTs) have attracted growing interest on various fields [4,5]. Carbon nanotubes are produced with various radii (from 0.2 nm up to several nm) and in a wide range of lengths (up to several μm and even mm). Although CNTs exhibit extremely high mechanical strength, recent studies have shown that they can undergo axial and radial compression [6]. This way it is feasible to obtain carbon nanotubes that are radially compressed at one end, that will resemble truncated nanocones.

Guidance of charged particle beams using CNTs has been investigated theoretically in recent years [7-11]. In those studies the possibility itself of channeling in single straight CNTs and nanotube bundles, as well the spatial and angular distribution of channeled particles have been examined. Also, in a recent study [11] the energy losses of channeled protons in straight chiral CNTs, presenting unusual results for their energy distribution, were examined.

Until now there have not been studies of propagation and channeling of charged particles in compressed CNTs. In particular, radially compressed carbon nanotubes at one end provide a suitable geometrical structure for particle focusing. Therefore, it is interesting to investigate the propagation and channeling of charged particles in such radially compressed CNTs and compare the results with the corresponding ones in uncompressed single CNTs.
2. Theory

Each nanotube can be described by two indices as \((n,m)\) and it may be considered as a collection of atomic rows parallel to the axis of the nanotube and arranged in a specific way along the perimeter of the cylindrical surface. In this way, from simple geometrical consideration, each pair of these indices define the nanotube radius \(R\) and helicity or chiral angle \(\theta\) (in practice, the angle under which the most closely packed rows of carbon atoms are wound on the cylindrical surface of the tube):

\[
R = \left( l \sqrt{3} / 2\pi \right) \sqrt{n^2 + nm + m^2} \quad (1)
\]

\[
\theta = \arctan \left( \sqrt{3m/(m+2n)} \right) \quad (2)
\]

where \(l = 0.142\) nm is the length of the bond between the carbon atoms [4, 5]. SWNTs with \(m = 0\) (\(\theta = 0^\circ\)) are called zigzag, those with \(m = n\) (\(\theta = 30^\circ\)) are called armchair, and all the others (\(0^\circ < \theta < 30^\circ\)) are called chiral. In the present study we examine only the case of chiral nanotubes.

Let us first consider the case of motion of fast positively charged particles (e.g. protons) in a straight isolated carbon nanotube. If such a particle enters a single crystal at small enough angle \(\theta_0\) with respect to an atomic row, then it can be assumed to be governed mainly by the continuum potential, i.e. the actual periodic potential of the rows averaged over the direction parallel to the rows [12]. Applying the concept of continuum potential to chiral nanotubes, it can be considered as the continuum potential of the (rolled-up) graphite plane, and thus we may average the actual potential of a nanotube over the circumference, i.e. the azimuthal angles. In the Doyle-Turner approximation to the atomic form-factor, the axially symmetrical continuum potential of a chiral nanotube (without taking into account the thermal vibrations of the atoms) can be described by the following expression [8]:

\[
U(r, \phi) = 3^{-3/2} 2\pi Ze^2 l^{-2} R \sum_{j=1}^{4} a_j b_j \exp \left[ -b_j (r^2 + R^2) \right] I_0 (2b_j R r) \quad (3)
\]

where \(Z = 6\) is the atomic number of the carbon atom, \(r\) is the distance from nanotube axis and \(a_j, b_j\) are dimensional parameters in the Doyle-Turner approximation:

\([a_j] = \{3.222, 5.270, 2.012, 0.5499\} \times 10^{-4} \text{ nm}^{-1}, [b_j] = \{10.330, 18.694, 37.456, 106.88\} \text{ nm}^{-1}\).

In case of a nanotube that is radially compressed at one end, its radius at each transverse plane at a point \(z\) of the axis is defined as:

\[
R = R_0 - z \cdot \tan \phi \quad (4)
\]

where \(R_0\) is the radius of the uncompressed nanotube and \(\phi\) is the angle of the slope of the walls of the nanotube after compression (see figure 1). This way, by introducing equation (4) into (3) instead of \(R\), we can obtain the potential in this compressed nanotube (see figure 2). The above approach is valid, since we consider small radial compression of the nanotube, so that the geometry of the nanotube doesn’t change significantly and therefore we can use the same expressions for the nanotube potential.
Because the de Broglie wavelength of a 10 MeV proton under consideration is very small compared to atomic dimensions, classical treatment of the problem can be adopted. We use the right-handed Cartesian coordinate system for the geometry of our problem, where $z$-axis coincides with the axis of the nanotube, and $x, y$ axes lie on the transverse plane.

In order to investigate this problem we use a Monte Carlo simulation program for individual proton trajectories. The individual proton trajectories were calculated by solving the classical equations of ion motion and taking into account energy loss and multiple scattering at each integration step.

We take into account energy losses calculated by phenomenological expression for the local stopping power given by Lindhard [13]:

$$
\frac{\Delta E}{\Delta z} = S(E) = \frac{4\pi Z_e^2 e^4 Z_{val}}{m v^2} \left[ (1 - \alpha) + a n_z(r) \right] \ln \left( \frac{2 m v^2}{I} \right)
$$

where $Z_e$ and $v$ are the ion charge and velocity, respectively (for proton $Z_e = 1$), $Z_{val}$ is the number of valence electrons per atom (for carbon atom $Z_{val} = 4$), $n_z(r)$ is the normalized local valence electron density at a point in the space, through which the particle moves, and $m$ is the electron mass. The coefficient $\alpha$ describes that part of close collisions which, according to Lindhard’s model, is proportional to the electron density (in our case, is 0.5, according to Bohr's equipartition rule). The quantity $I$ is the average excitation potential, $I = I_0 Z$, where $I_0 \cong 13.5$ eV [14] and $Z$ is the atomic number of target atoms. The normalized electron density $n_z(r)$ is calculated by using the Fourier component of the electron density in carbon atom [8]:

$$
f^{(e)}(k) = Z \sum_{j=1}^{5} a_j^{(e)} \exp \left[ -\frac{k^2}{4(2b_j^{(e)})^2} \right]
$$

with the five pairs of fitting parameters:

$$
\{a_j^{(e)}\} = \{0.3499, 0.3014, 0.2103, 0.0946, 0.0438\}
$$

$$
\{b_j^{(e)}\} = \{17.300, 11.400, 75.501, 155.24, 7.596\} \text{ nm}^{-1}
$$

Thus, by using equation (6) it is easy to show that for a chiral nanotube the normalized electron density averaged over the azimuthal angles is determined from the following expression:

$$
n_z(r) = \frac{2 N Z_{val}}{\pi d_R^2} \sum_{j=1}^{5} a_j^{(e)} b_j^{(e)2} \exp \left[ -b_j^{(e)2} (R^2 + r^2) \right] I_0 (2b_j^{(e)} R r)
$$

where $N$ is the number of atomic rows in each sequence, and $d_R$ is the distance between neighboring atoms within each row [8].

We also take into account the electronic multiple scattering after each integration step, by calculating a normal distribution of the scattering angle with standard deviation:

$$
\sigma_{\text{ms}}^2 = \frac{m \Delta E}{2 m E}
$$

where $E$ and $\Delta E$ are the energy and the energy loss at each integration step, respectively.

We assume that protons are dechanneled if during their propagation inside a nanotube they approach the nanotube wall less than the critical distance $r_c = \sqrt{2} u_\perp$, where $u_\perp$ is the thermal vibration amplitude in the transverse plane [15]. In our case, we can use the value $8.5 \cdot 10^{-3}$ nm, which corresponds to that of graphite (at room temperature).

Our calculations were carried out for different angles between incident proton beam and the nanotube axis, and for different nanotube length. Beam protons are assumed to enter nanotube’s cross-sectional area homogeneously. In all simulations we take into account energy losses and multiple scattering.
3. Results and discussion

We examined 10 MeV monoenergetic proton beam channeled in radially compressed at one end (6,4) and (11,9) SWCNTs of various length. The radii of these nanotubes at the uncompressed end are 0.341 and 0.679 nm, respectively, calculated with the use of more accurate value of length of the bond between the carbon atoms mentioned above. We investigated two cases of incident angle, zero and half of the critical angle. The cases of nanotube types examined are the following: straight, radially compressed with the angle of slope of the walls of 0.005 degrees with entrance at both wide and narrow ends. For proton energies of 10 MeV the critical angles $\psi_{cr.}$, which were calculated as $\psi_{cr.} = (U(r_c)/E)^{1/2}$, for (6,4) and (11,9) nanotubes are 2.181 and 2.169 mrad, respectively. The initial proton beam was assumed to be well collimated ($\Delta \theta = 0$).

Figures 3 and 4 show the angular distributions for three types of (6,4) and (11,9) nanotubes of 1000 nm length, respectively, and for different incident angles (0 and 0.5$\psi_{cr.}$). We observe that in the case of the narrow end entrance, the beam exits more collimated than in the two other cases. This effect can be explained by examining the trajectory of the particle inside this nanotube in the x-z plane, where it moves in an increasing channel so that its motion resembles damping oscillation (see figure 5).
Also, it is clear that a ring-shaped pattern of angular distribution is being formed when the incidence angle is $0.5\psi_c$, (doughnut scattering effect) with radius close to the corresponding incident angle of the beam. Another interesting feature, which was analyzed in our previous work [11], is the formation of “zones” in the angular distribution at zero incident angle of the beam, where each zone corresponds to different initial incident region. In particular, channeled protons incident in a region closer to nanotube’s wall exit nanotube with larger angle. It is noteworthy, that these “zones” appear in the case of both types of radially compressed nanotubes as well.

The energy distributions of channeled protons for three types of (6,4) nanotubes are presented in figure 6. The incident angle of the beam considered was zero. In these figures the length evolution of energy distribution of protons is demonstrated. The appearance of several small peaks is observed in all three nanotube types, the number of which increases with larger nanotube length. This effect was observed for the first time in straight nanotubes [11]. An explanation of these results can be achieved by examining the particle trajectory inside a nanotube in x-z plane (see figure 5 in case of (6,4) CNT with particles entering the compressed end). As indicated in [11], the trajectory period strongly depends on the distance of incidence point from the nanotube wall. The closer this point is to the wall, the less is the period of proton motion between nanotube walls. This, in turn, forces the proton to spend more time in regions close to the wall, where the energy losses are significant, compared to the region around nanotube axis, where they are close to zero. Thus, protons that where incident in a zone close to the nanotube walls, correspond to the small peak with the minimum energy (maximum energy loss), while other protons incident in zones farther from the wall correspond to the rest small peaks. In the same figure it becomes clear that the exit angle of the protons is larger, when it is incident closer to the wall, which in turn corresponds to the zone with larger exit angles in the angular distribution (see figure 3). Therefore, we can correlate the zones in angular distributions (figure 3) with the corresponding energy peaks (figure 6). Regarding the main peak of maximum energy, it comes from the protons incident in the region around nanotube axis, where energy losses are negligible.
Figure 7 shows the evolution of spatial distribution of protons propagating in a (6,4) radially compressed CNT, in case when particles enter the compressed end. Depending on the length of the compressed nanotube, focusing of particles in smaller area is different. This feature can be useful, in order to choose the proper length of CNTs to obtain the desirable focusing.

4. Conclusions
We studied proton channeling in radially compressed carbon nanotubes. The presented results of the angular distribution of channeled protons in compressed nanotubes are similar to the corresponding ones for straight nanotubes. The main difference is observed in the case when particles enter the compressed end, where the angular distribution is more focused than in other cases.

Moreover, we have studied energy losses of channeled protons in compressed nanotubes. The energy distribution of channeled protons incident with zero angle inside a compressed nanotube, in both cases of beam entering compressed and uncompressed end, appears to have many small peaks, depending on the length of the nanotube. Similar pattern was observed in straight nanotubes, and there is an indication that these peaks are correlated to the zones in the corresponding angular distribution plots.

Finally, we presented results of the evolution of spatial distribution of channeled protons incident at the compressed end of a (6,4) radially compressed CNT. The observed fluctuations in the focusing of the beam require farther investigation.

References
[1] Iijima S 1991 Nature 354 56-8
[2] Iijima S and Ichihashi T 1993 Nature 363 603-5
[3] Bethune D S, Kiang C H, de Vries M S, Gorman G, Savoy R, Vazquez J and Beyers R 1993 Nature 363 605-7
[4] Jorio A, Dresselhaus G and Dresselhaus M S 2008 Carbon nanotubes: Advanced Topics in the Synthesis, Structure, Properties and Applications (Berlin: Springer)
[5] Wong H-S P and Akinwande D 2011 Carbon Nanotube and Graphene Device Physics, (Cambridge: Cambridge University Press)
[6] Barboza A P M, Chacham H and Neves B R A 2009 Phys. Rev. Lett. 102 025501
[7] Zhevago N K and Glebov V I 1998 Phys. Lett. A 250 360-68
[8] Artru X, Fomin S P, Shul'ga N F, Ispirian K A and Zhevago N K 2005 Phys. Rep. 412 89-189
[9] Matyukhin S I 2008 Kinetics of Ion Channeling in Carbon Nanotubes (Orel: OrelGTU)
[10] Borka D, Lukic V, Timko V J and Borka Jovanovic V 2012 Nucl. Instrum. Meth. Phys. Res. B 279 198-201
[11] Karababounis A, Sarros S and Trikalinos Ch 2013 Nucl. Instr. Meth. Phys. Res. B 316 160-170
[12] Gemmell D S 1974 Rev. Mod. Phys. 46 129-227
[13] Lindhard J 1965 Kong. Danske Vid. Selsk., Mat.-Fys. Medd. 34 14
[14] Sigmund P 2006 Particle Penetration and Radiation Effects (Berlin: Springer)
[15] Lenkeit K, Trikalinos Ch, Balashova L L, Kabachnik N M and Shulga V I 1990 Phys. Status Solidi B 161 513-524