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Channeling of heavy ions through multi-walled carbon nanotubes

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Abstract

Making use of molecular dynamics with analytical potentials we theoretically study channeling of heavy ions with keV energies through multi-walled carbon nanotubes. We show that under certain conditions on the tube alignment with respect to the ion beam and on ion energies, the ions can move through the empty cores of the MWNT with a very low probability of dechanneling. We further discuss how, by employing the experimental techniques routinely used nowadays for handling nanotubes on substrates, one can create a nanotube-based conduit for energetic ions, which should work as an aperture and allow one to manipulate the beam at the nanoscale.

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1. Introduction

Channeling of energetic ions through solids is a phenomenon which should be accounted for in the present-day semiconductor technology [1,2], as it gives rise to deeper implantation and less lattice disorder. The channeling effects are particularly important for materials with high crystallinity and anisotropic atomic structure. One can expect that the effective channeling of ions is possible in carbon nanotubes [3] (CNTs), as they have hollow cores, high aspect ratio and a low concentration of defects.

The motion of high-energy light ions (protons) through single-walled nanotubes (SWNTs) has received considerable attention [4,5]. The driving force for these studies was the possibility to use SWNT bundles for steering the beams of high-energy (GeV) protons, which would otherwise require cumbersome and expensive magnetic systems. However, despite an extensive theoretical analysis and first experimental results [6] it is not clear at all if CNTs can in practice be used for this

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purpose, as the sample will quickly be destroyed by the beam as experiments on proton irradiation of SWNTs indicate [7].

Contrary to light ions, channeling of heavy ions through CNTs has not yet been studied. At the same time, this issue is of fundamental interest, as the well-controlled atomic structure of CNTs makes it possible to check (e.g. by the transmission electron microscope) the fundamentals of the channeling theory in general, and specifically in graphitic systems [8]. Besides this, as CNTs can easily be bent and manipulated at the nanoscale, developing a CNT-based conduit for energetic ions could facilitate the further progress in many applications, such as the solid-state quantum computing [9,10].

In this paper, we theoretically study channeling of heavy ions with keV energies through multi-walled nanotubes (MWNTs). We show that under certain conditions on the tube alignment with respect to the ion beam and on ion energies, the ions can channel through the empty cores of the MWNT with a very low probability of dechanneling.

2. Simulation method

To describe collisions of energetic ions with the nanotube, we employed molecular dynamics [11] (MD) with analytical potentials. To model carbon–carbon interactions, we used the Brenner II interatomic potential [12]. We chose Ar as the typical heavy ion. The interaction between Ar ions and C was modelled with the Ziegler–Biersack–Littmark universal repulsive potential [13]. A very large cutoff range of 4 Å was used for the Ar–C interaction, as we found that shorter cut-off ranges effected the results. We did not account for the electronic stopping as the ion energies considered were low and the nuclear slowing down completely governed the collisional phase. Besides this, the electron density in the MWNT cores is very low. The simulations were carried out at zero temperature. Other details of the simulation methods can be found in our previous publications [14,15].

3. Results and discussions

Effects of ion irradiation on nanotubes have been studied both experimentally [7,16–18] and theoretically [19] (and references therein). When estimating the ion ranges, it has been assumed that the beam direction is perpendicular to the tube shells. Now, to address channeling through the tubes, we must consider the opposite case: the ion beam direction is nearly parallel to the tube axis, see Fig. 1.

To understand the nature of ion interactions with MWNTs, we considered first the collision of the ion with the inner shell of a MWNT, see Fig. 1. The neglect of all other MWNT shells is well motivated in this case as the shells are spatially separated by 3.2 Å and they are only weakly bounded to each other via van der Waals-type forces [3].

We started with the following question: What happens during the collision of an Ar ion with a

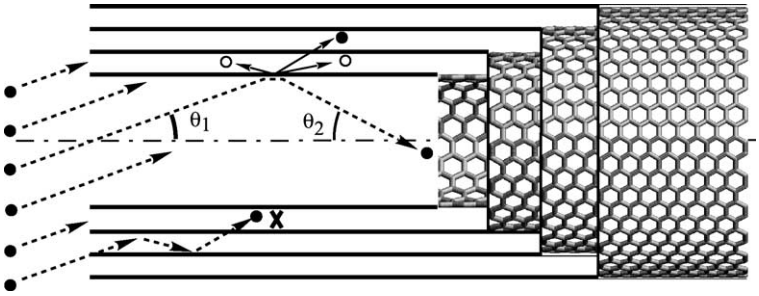

Fig. 1. Schematic representation of a beam of Ar ions colliding with a multi-walled carbon nanotube with an open end. Depending on ion energy, impact point, and angle Θ_1, the ion hitting the inner shell of the tube can either return to the core region (with or without creating the damage to the tube) or go through the shell.

single-walled nanotube (SWNT) and how does the ion trajectory depend on the ion energy E and the incidence angle Θ_1 (the angle between the tube axis and the original ion velocity vector, see Fig. 1)? We considered ions with energies of 0.1–20 keV and $\Theta_1 = 8$–$20°$. For each ion energy and angle we simulated 100 impacts and collected the statistics. The polar angle (the projection of the ion velocity vector onto the plane perpendicular to the tube axis) and the impact points were randomly chosen. In this work we considered (17,0) zigzag SWNTs with a diameter of about 1.3 nm.

We found that at low energies the ion is always bounced back by the wall and it remains inside the core region without creating any damage to the tube, but giving rise to pressure waves and local heating. At higher energies the ion can sputter 1–4 carbon atoms from the tube, but it still remains inside the tube. At a certain energy E_{dc} (the dechanneling energy – the maximum energy for channeling) which depends on Θ_1, the ion goes through the wall, as one can expect from the momentum and energy conservation laws. The probability of dechanneling (the number of dechanneled ions per incident ion) as a function of ion energy is shown in Fig. 2 for the zigzag SWNT. It is seen that E_{dc} is critically dependent on the angle, but even when the ion energy is higher than E_{dc}, the ion has a finite probability to stay inside the tube after the collision. This is related to different local geometries at the impact points and thus different ways of momentum transfer to the tube atoms.

Using the data given in Fig. 2, one can plot E_{dc} versus the angle, or the other way around, one can present the maximum, or critical, angle ψ_c as a function of ion energy, see Fig. 3. The simulations results shown in Fig. 3 can be described by a universal curve fit to the data. We found that the results can well be reproduced by an equation

$$\psi_c = \text{const}/\sqrt{E}, \tag{1}$$

see also Fig. 3. This is in line with a general equation [1]

$$\psi_c = \sqrt{U(r_c)/E}, \tag{2}$$

where $U(r_c)$ is the ion potential energy at the critical approach distance r_c. This equation has been derived within the framework of the continuum theory of channeling. Note that Eq. (2) is of limited validity (see e.g. [1] and references therein), as only one row or plane of atoms is taken into account to calculate $\psi_c(E)$. However, the open

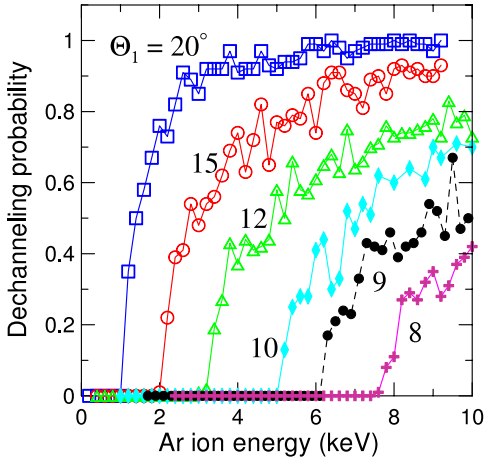

Fig. 2. Dechanneling probability as a function of Ar ion energy at different angles of incidence Θ_1 for a zigzag (17,0) SWNT.

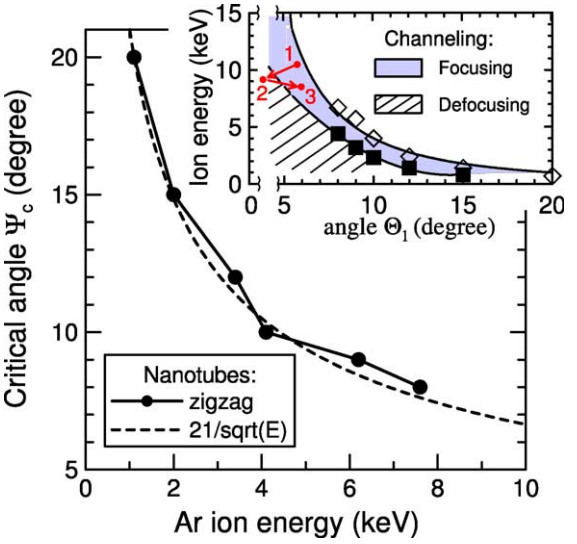

Fig. 3. Critical angle as a function of ion energy for zigzag nanotubes. The dashed line is a fit to the data. The inset: Parameter ranges for different regimes of ion channeling. Dots with numbers show relation of the parameters before and after the ion collision with the tube. Symbols show the simulation results, curves are approximations to the MD data.

structure of SWNTs motivates well this approximation, as our simulation data obtained with account for the interaction of the ion with all C atoms within the cutoff range indicate.

The next highly important issue to be addressed is the average scattering angle Θ_2 of the ion, see Fig. 1. One can expect that an ion propagating through the MWNT core will collide many times with the inner wall of the MWNT. Thus, if Θ_2 grows after each impact, this should inevitably result in ion dechanneling. On the contrary, if Θ_2 decreases, after several collisions the ion trajectory will be nearly parallel to the tube axis thus decreasing the probability of dechanneling.

We found that for slow ions the ratio Θ_2/Θ_1 is higher than unity, which corresponds to ion defocusing. However, at a certain crossover energy E_{foc} the ratio $\Theta_2/\Theta_1 < 1$, which means that, the other way around, every collision should decrease the angle. Energy-related changes in the ratio Θ_2/Θ_1 can be understood within the binary collision approximation: the impinging particle scattering angle increases at low velocities of the particle [13] due to a higher momentum transfer. Note also that contrary to the continuum theory, neither the transverse ion kinetic energy $E_{\text{tr}} = E\sin^2(\Theta_1)$ nor the energy projected to the tube axis $E_{\|} = E\cos^2(\Theta_1)$ is conserved in the MD approach. Depending on Θ_1 and E, every collision results in a decrease of the total energy by 0.05–0.3 keV. Thus, $\Theta_2/\Theta_1 < 1$ means larger losses in E_{tr} than in $E_{\|}$. This happens when the ion starts displacing carbon atoms from the tube, and E_{foc} correlates very well with ion energies at which carbon recoils appear. As our simulations indicate, energy losses at $E \sim E_{\text{foc}}$ are 0.05–0.1 keV per single collision.

The existence of the "focusing" region means that at relatively large angles considered and high ion energies the ion effectively transfers its kinetic energy to several atoms only, but not to the whole shell (row of atoms) – the binary collision approximation should work here better than the continuum theory. In the former approximation the scattering angle can be arbitrary small. In reality it cannot be smaller than a certain value due to the discrete nature of the atoms in the rows. Note also that graphite has highly anisotropic structure, so that the atoms can be displaced more easily in the direction perpendicular to the shell rather than within the shell.

Using the data presented in Fig. 3 and the results on the angles, one can plot a Θ_1 versus E diagram showing two regions of parameters corresponding to the two different regimes of channeling described above, see the inset in Fig. 3. The upper curve corresponds to the dechanneling energy, the lower to E_{foc}. The data points are the simulation results, the lines the approximations to the data. The presence of the "focusing" region indicates that the SWNT can be bent as an increase in the angle of incidence Θ_1 will be compensated by the corresponding decrease in Θ_2.

In practice, losses in kinetic energy and related changes in the angle of incidence for the next impact will result to a new set of the parameter points on the diagram after each impact. An example is also shown in the inset. The first impact decreases the angle (the regime of focusing, point 1) while the second impact will increase back the angle, point 2 in the inset, the energy losses are exaggerated. In practice, this means that the averaged angle will fluctuate around the value corresponding to E_{foc} in the diagram.

The number of collisions governed by the total length of the tube will set the lower limit on the ion energy for the tube of a given length. Knowing the dependence of Θ_2 on Θ_1 and E one can estimate the averaged number of collisions and the final energy of the ion. For example, if $\Theta_{\text{ini}} = 5$ and $E_{\text{ini}} = 12$ keV, the ion will go through a μm-long SWNT with an inner diameter of $d = 1.5$ nm and have at least 6 keV at the output from the other end of the tube. Detailed calculations of the output ion energy will be presented elsewhere [20].

Having analyzed propagation of energetic ions through SWNTs, we moved on considering effects of the high-dose irradiation on the atomic structure of a MWNT composed of (10, 10), (15, 15) and (20, 20) SWNTs. Details of these simulations will be published elsewhere [20]. We found that although the open end of the tube quickly becomes completely amorphous, the inner core remained open up to a dose of $\Phi = 4 \times 10^{15}\,\text{cm}^{-2}$. This means that MWNTs, especially the MWNTs with larger inner cores can survive "shooting" about hundred ions before the end of the tube is com-

pletely destroyed and closed. Note that a substantial part of defects in the MWNT should anneal due to the migration of carbon interstitials [21] and by saturating vacancy dangling bonds [15].

The efficient channeling of ions through MWNTs can be employed to make a MWNT-based apertures to steer the ion beam. Such an aperture can be produced by using a combination of techniques routinely used nowadays for handling supported CNTs. A MWNT deposited on a substrate can be straightened [22] and cut [23] to open its ends by the tip of the atomic force microscope, then by applying a combination of etching and electron-beam lithography techniques [24] the metal shield can be produced. The beam can be steered by moving the whole unit or, ideally by bending the MWNT. Thus, the target can be irradiated in predetermined positions by a beam just several nm across. Such a device can be used for implanting ions or even single ions, which is highly important for the further progress in the solid-state quantum computing [9,10].

4. Conclusions

To conclude, we theoretically studied channeling of Ar ions through MWNTs. We found that the ions can channel through the empty cores of the MWNT with a very low probability of dechanneling. We showed that the dependence of the critical angle on ion energy obeys a simple universal equation and the continuum theory of channeling should work well in nanotubes due to their open structure. As interactions of energetic ions with the target can well be described by the universal repulsive potential [13] such a behavior should be general not only for Ar but also for other heavy ions. We finally suggested a nanotube-based conduit for energetic ions, which should work as an aperture and allow one to manipulate the beam at the nanoscale.

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