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Calculation of the charge spreading along a carbon nanotube seen in scanning tunnelling microscopy (STM)

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Abstract

Investigating the distribution of the scanning tunnelling microscopy (STM) current through a nanostructured material is a subject of great current interest. For this work, we calculated the transmission of an electron wave packet through a supported carbon nanotube by numerically solving the three-dimensional (3D) time-dependent Schrödinger equation. The spreading of the wave packet along the nanotube during the tunnelling event is determined from the wave function. It is shown that this spread influences the length segment of the nanotube 'sampled' by the tunnelling current. The results are compared to experiments. The prime novelty of the paper is the 3D time-dependent study of tunnelling through a supported nanotube. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Scanning tunnelling microscopy (STM); Modeling; Computer simulation; sp2 Bonding

1. Introduction

Scanning tunnelling microscopy (STM) provides information both about the topographic and the electronic structure of carbon nanotubes (CNTs) on a nanometre scale [1]. However, the interpretation of the images is delicate because of the complex geometry of the system. Due to this, computer simulation of the imaging process [2,3] is important in understanding STM and STS results. In this paper, the full three-dimensional (3D) transmission of electron wave packets (WPs) was calculated through a jellium potential model of a carbon nanotube (CNT) under the tip of a scanning tunnelling microscope (STM). This work is an extension of our former two-dimensional (2D) calculations [3,4] where the tunnelling process was examined only in the plane perpendicular to the CNT.

2. Calculation method and results

A jellium potential barrier was used as a model of the STM tip–nanotube–support system. The model system geometry used in our calculation is shown in the upper left panel of Fig. 1. The CNT is modelled by a cylinder of 0.5 nm radius floating above the support plane at a distance of 0.335 nm. The STM tip is taken as a hyperboloid of 0.5 nm apex radius. The chosen value of 0.4 nm for the tip-CNT tunnel gap [3] is consistent with that estimated from the apparent geometric distortion of the CNT atomic lattice [2] seen in STM experiments.

By numerically solving the time-dependent 3D Schrödinger equation, the evolution of a Gaussian WP approaching the tunnel junction from inside of the tip bulk was calculated. The $\rho(x, y, z, t) = |\varphi(x, y, z, t)|^2$ time-dependent probability density function is visualised by snapshots of a constant density surface in Fig. 1.

In the panel $t = 0.0$ fs of Fig. 1, the initial WP is shown. The sphere surface is clipped at the upper boundary of the presentation box. At $t = 1.4$ fs, the WP has already penetrated into the tip apex region. The part reflected back into the tip bulk forms interference patterns with the incoming wave. A fraction of the WP just begins to enter into the tip–CNT interface. At $t = 2.1$ fs, the WP flows around the tube and simultaneously

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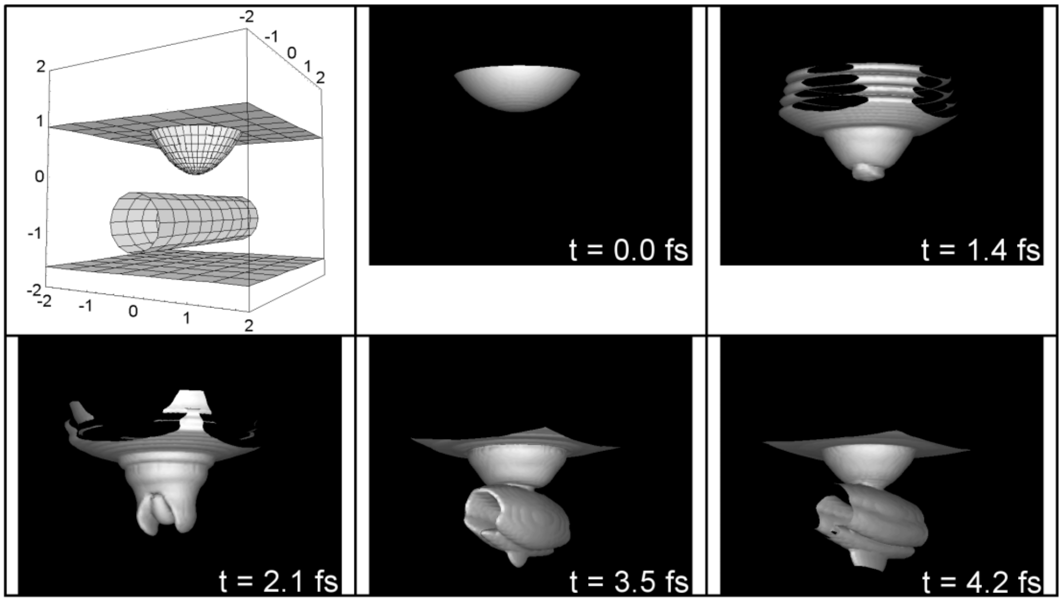

Fig. 1. Time evolution of the probability density of the wave packet approaching the STM junction from the tip bulk and tunnelling through the nanotube into the support. The upper left image is the model system used in the calculation. The labelled box is the presentation box. All dimensions are in nanometres. The subsequent images show snapshots of a constant probability density surface. This surface is clipped at the presentation box boundaries. See the text for details.

tunnels through it. The incoming and outgoing waves form interference patterns in the tip apex region. When the two WP parts (one moving on the left side and another on the right side of the tube) meet at the lowest point, standing wave patterns begin to form along the tube circumference. Subsequently, the WP tunnels through the CNT-support junction and enters into the support surface (at $t=3.5$ fs). In the meantime, the probability density is gradually spreading along the tube axis. At $t=4.2$ fs the CNT-support tunnel channel begins to open along the tube axis.

3. Discussion and conclusion

The 3D tunnelling calculation makes it, for the first time, possible to study the phenomenon of WP spreading along the CNT during the tunnelling event. This spreading can explain the features of the STM image shown in Fig. 2.

A short and thick multi-wall carbon nanotube (MWCNT) produced by the arc method was found during constant current STM imaging being adsorbed on a step edge in such a way that part of the tube is protruding from the step edge.

The tube part above the lower terrace is suspended above the HOPG surface at a distance of 1.3 nm. As shown by molecular mechanics calculations [5], such multi-wall structures behave like rigid objects. As a consequence, the short tube does not bend to make contact with the substrate on the lower terrace of the step. If bending to contact would occur, one should measure the same height value on the lower terrace as

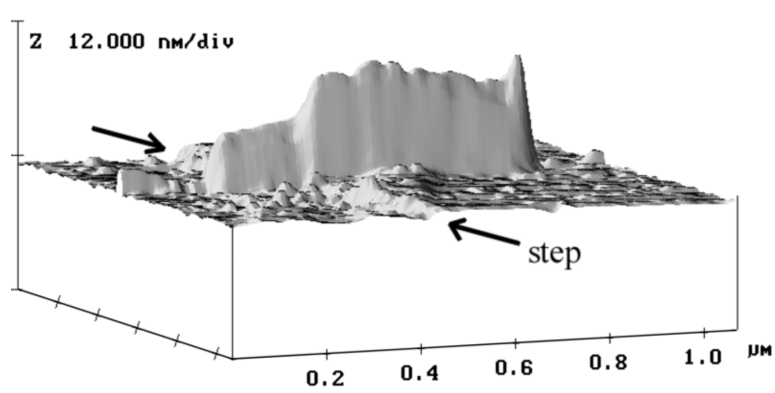

Fig. 2. Constant current 3D STM image of a multi-wall carbon nanotube crossing a step on the HOPG support surface. Note the transition region at the step edge and the gradual decrease of the apparent height of the tube section above the lower terrace.

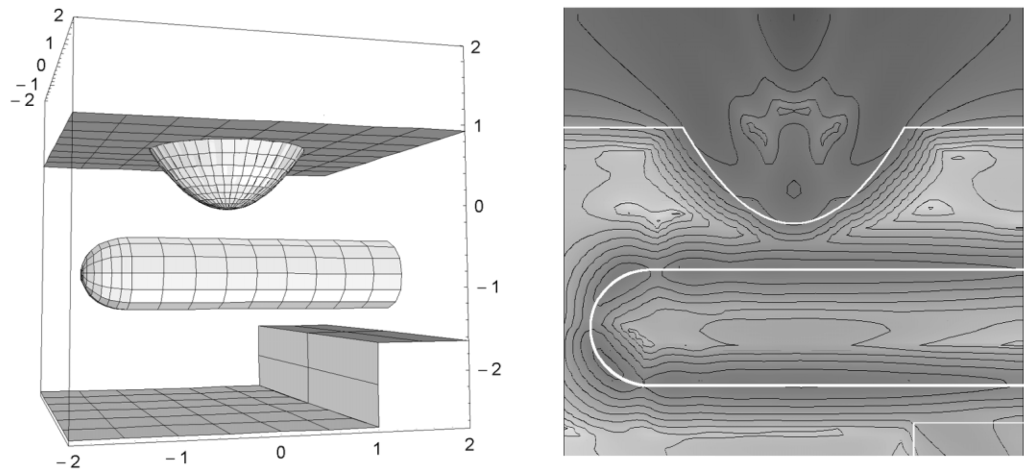

Fig. 3. (a) Model system of a nanotube crossing a HOPG step. (b) Probability density in the plane perpendicular to the support surface and including the tube axis. Contour lines are drawn on logarithmic scale. The thick white lines show the geometrical surface of the tip, nanotube and support.

on the upper one which was contrary to the experimental observation (see Fig. 2). The apparent height of the tube part above the higher terrace is constant (some noise is present) because of the translational symmetry along the tube. Right at the step edge there is a transition region in the apparent height; the width of this region corresponds to the length over which the charge spreading takes place along the tube.

We have performed WP tunnelling calculations for this geometry (see Fig. 3a). The snapshot of the probability density (see Fig. 3b) shows that no tunnelling can occur from the tube to the support. When the distance of the STM tip as measured from the step edge is larger than the lateral WP spreading length, the electrons can reach the support of the tube only after transport has occurred along the tube. This explains the ohmic-like decrease of the apparent height along the tube [6]. As a consequence, a more pronounced drop is found in the apparent height of the tube than the geometric height of the step in that part where the tube is not supported.

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