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Conducting ion tracks generated by charge-selected swift heavy ions

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ABSTRACT

Conducting ion tracks in tetrahedral amorphous carbon (ta-C) thin films were generated by irradiation with swift heavy ions of well-defined charge state. The conductivity of tracks and the surface topography of the films, showing characteristic hillocks at each track position, were investigated using conductive atomic force microscopy measurements. The dependence of track conductivity and hillock size on the charge state of the ions was studied using 4.6 MeV/u Pb ions of charge state 53+, 56+ and 60+ provided by GANIL, as well as 4.8 MeV/u Bi and Au ions of charge state from 50+ to 61+ and 4.2 MeV/u ^{238}U ions in equilibrium charge state provided by UNILAC of GSI. For the charge state selection at GSI, an additional stripper-foil system was installed at the M-branch that now allows routine irradiations with ions of selected charge states. The conductivity of tracks in ta-C increases significantly when the charge state increases from 51+ to 60+. However, the conductivity of individual tracks on the same sample still shows large variations, indicating that tracks formed in ta-C are either inhomogeneous or the conductivity is limited by the interface between ion track and Si substrate.

1. Introduction

Tetrahedral amorphous carbon (ta-C) with sp^3 bond fraction around 80% exhibits diamond-like properties, including high hardness, high atomic density, and high electrical resistivity. So far, ta-C is the only material that undergoes a local transition from an insulating to a conducting phase along the trajectory of swift heavy ions (SHI), leading to highly conductive ion tracks with diameter of about 10 nm [1]. The amorphous highly sp^3 bonded low conducting phase of ta-C is converted into a sp^2-rich amorphous phase of lower density and highly increased conductivity. At the impact site of each ion, a small hillock is formed at the sample surface, which can be used to identify the position of an ion track. Previous works on ta-C irradiated with SHI showed that the conductivity of the tracks depends very sensitively on the electronic energy loss, i.e. on the energy, the mass and the charge state of the ions [2]. The hillock height was found to depend linearly on the energy loss [3]. At equilibrium charge state 1-GeV U ions provide largest possible electronic energy loss for monoatomic projectiles in ta-C of about 40 keV/nm. However, the conductivities of U tracks in a given sample vary up to an order of magnitude most likely due to still inhomogeneous tracks [4–6]. Several approaches, such as doping of ta-C by different elements like B, N, Fe and Cu or irradiation with swift C_{60} cluster ions, have been tried to optimize the track conductivities [7–9]. As the charge state of the ion determines the electronic energy loss, it will also influence the track conductivity. Through realization of charge state selected swift heavy beams with very high charge states, we expect to improve the conductivity of ion tracks.

In the past, the dependence of the electronic energy loss on the charge state as well as the evolution of the charge state of an ion towards its equilibrium value while passing through a target have been examined both theoretically and experimentally. A theoretical description of fluctuations of the charge state of MeV ions was given two decades ago [10]. Sigmund describes the statistics

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of the charge states and correlated phenomena during the passage of ions through matter [11]. Osmani and Sigmund [12], Sigmund and Schinner [13] as well as Lifshitz and Arista [14] provide a theoretical description for the charge state of high energy heavy ions in matter. Furthermore, the effective charge of molecular ions was studied theoretically [15]. The relationship between electronic energy loss and charge state when ions pass through thin foils was explored by Blazevic et al. [16,17], providing experimental evidence of a superlinear increase of the energy loss with increasing charge state. Experimental data on the charge state equilibrium after ion passing through thin films were carried out for different ion species and energies [18–21]. In several experiments it was shown that the ion charge state plays an important role on energy loss related effects in the near-surface area of an irradiated sample, such as electron or photon emission from surfaces or the formation of swift heavy ion induced craters in polymers [22–24].

Looking at the dimension of ion tracks (8 nm in diameter and several hundreds of nm in length), conducting tracks in ta-C can be viewed as quasi-one-dimensional conductors. So far, electronic transport in one-dimensional structures was studied mainly in the following systems: contacted carbon nanotubes [25,26], lithographic generated 1-D semiconductor hetero structures [27], 1-D metallic wires on surfaces, 1-D wires between the tip of a scanning tunneling microscope and surfaces [28], as well as metallic nanowires electro-deposited into polymer pores [29]. Here, the term "1-D" includes wire diameters between 1 nm up to several 100 nm. The objective of the mentioned studies is the realization of an ideal 1-D-electron gas, 1-D semiconductor diode-like junctions and charge transport, which is dominated by Coulomb blockade. Other activities in this field deal with electronic components based on ion track technology [30,31]. Compared to many other approaches, SHI tracks offer the advantages to synthesized nanostructures in a rather simple and reproducible manner yielding cylindrical structures with diameters below 10 nm and lengths up to several tens of micrometers. Ion tracks in ta-C could therefore represent model systems for one-dimensional electrical transport and may serve as a starting point for novel quantum dot structures.

Transport processes in sp^2 and sp^3 amorphous carbon volume material have been extensively studied. Our group has specifically investigated the charge transport in conductive ion tracks in ta-C [2,4,6,8,9,32–35]. However, the details of the transport mechanism (tunneling effects, variable range hopping, Poole–Frenkel conduction) are not yet fully understood. The investigations include ensemble conductivity measurements at low temperatures, determination of the conductivity of individual tracks and averaged conductivities of track ensembles, as well as measurements of field emission currents and the energy distribution of field-emitted electrons from irradiated ta-C layers [33,35]. Molecular dynamic simulations were performed to reveal the bond structure of the ion tracks [26].

For ion tracks formed on a given ta-C sample we usually observe a broad distribution of track conductivities [2]. So far, two approaches successfully achieved a more narrow conductivity distribution: (i) ta-C which was doped with a small concentration of about 1 at.% of Cu [27,28] and (ii) the electronic energy loss in ta-C was increased by using 30 MeV C_{60}^{2+} cluster ions (energy loss ~72 keV/nm). Given by the high electronic energy loss C_{60} clusters seems to produce continuous conducting filaments throughout the ta-C film [7,9]. However, this high energy loss is only effective on a coherence length of about 50–100 nm where the clusters ions act as an ensemble before disintegrating into smaller fragments. Thus, a C_{60} beam only allows track formation in rather thin films <100 nm.

Previous experiments have also shown that the electrical conduction mechanism will change from Frenkel–Poole conduction towards Ohmic conduction as the track conductivity increases. Such conducting ion tracks would be ideal for the realization of simple electronic components on the basis of Coulomb-blockade effects. Irradiation with ions of a well-defined high charge state appears as an additional promising approach to realize ion track ensembles with high and uniform track conductivities. To achieve this, it is necessary to magnetically select the ions of specific charge states after passing through a stripper foil prior to impact onto the sample. In this work, we will demonstrate that SHI irradiation with charge state selected ions significantly improves the track conductivities in ta-C thin films.

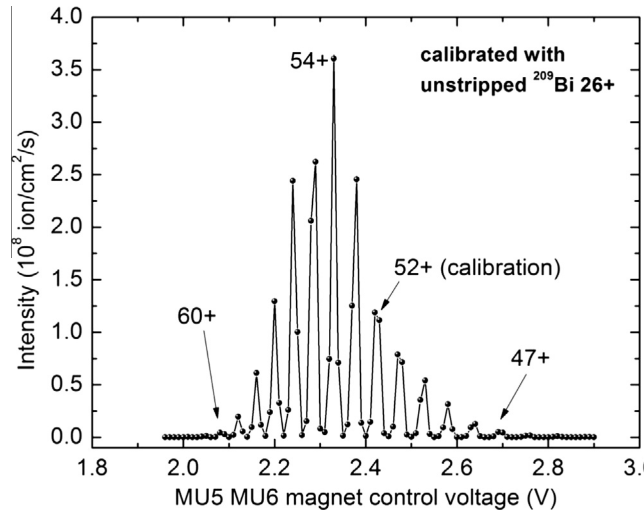

Fig. 1. Charge state distribution of ^{209}Bi ions after passing through a thin stripper foil as a function of the voltage of the MU5–MU6 magnets at the M-branch of the GSI UNILAC accelerator. The calibration of charge-state 52+ was done using the unstripped $^{209}Bi^{26+}$ ion beam.

2. Material and methods

The effect of the charge state on the conductivity of ion tracks in ta-C was examined by irradiation experiments at two different ion beam facilities. First tests were performed at GANIL accelerator with 946 MeV (4.57 MeV/u) $^{208}Pb^{23+}$ ions of charge states between 53+ and 60+. These high charge states were obtained by passing the accelerated $^{208}Pb^{23+}$ beam through a thin carbon stripper foil and selecting beams with charge state 53+ and 56+. Even higher charge states were generated after the passage of a second stripper yielding sufficiently high intensities for charge state 60+. Experiments using Au, Bi and U ions were also carried out at the GSI linear accelerator UNILAC. 4.2 MeV/u ^{238}U irradiations were performed with equilibrium charge state distribution by using a stripper foil in front of the ta-C targets. In contrast, ^{209}Bi and ^{197}Au ion irradiations at 4.8 MeV/u were done with selected charge states between 50+ and 61+. The initial beams of $^{209}Bi^{26+}$ and $^{197}Au^{26+}$ ions were stripped using a 0.62 μm (170 μg/cm^2) thin Al foil inserted behind the kicker magnet that guides the beam into the M-branch. The following deflection magnets MU5 and MU6 of the M-branch were then used for charge state selection. The charge state distribution of ^{209}Bi ions after passing through a thin stripper foil was Gaussian with a maximum at q_{max} = 54+ and an envelope with a width of $\Delta q \approx 2$–3 charge states (Fig. 1). The separation of individual charge states is excellent, with beam intensities between neighboring charge states dropping to almost zero. The small energy straggling induced by the stripper foils did not affect the quality of charge state selection. The energy loss of 4.8 MeV/u ^{209}Bi ions in the Al stripper foil as a function of charge state was calculated using the CasP code [36,37]. Within the