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Nano tracks in fullerene film by dense electronic excitations

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ABSTRACT

In the present work, we investigate the formation of nano tracks by cluster and mono-atomic ion beams in the fullerene (C_{60}) thin films by High Resolution Transmission Electron Microscopy (HRTEM). The fullerene films on carbon coated grids were irradiated by 30 MeV C_{60} cluster beam and 120 MeV Au mono-atomic beams at normal and grazing angle to the incident ion beams. The studies show that the cluster beam creates latent tracks of an average diameter of around 20 nm. The formation of large size nano tracks by cluster beam is attributed to the deposition of large electronic energy density as compared to mono-atomic beams.

1. Introduction

Fullerenes are important materials for the fabrication of nano-devices such as field effect transistors, flat panel display based on field emission [1,2]. The controlled conducting nano-wire patterns formation in films by ion beam has been demonstrated [3,4]. Phase transformation of fullerene by ion beam has been a topic of fundamental research interests. Ion irradiation induced phase transformation is one of the most important phenomena in the field of ion beam modification of fullerenes [5,6]. It is known that energetic heavy ions with velocities comparable or higher than the Bohr velocity provide a unique tool to create the extremely localized conditions of temperature and pressure in nanometric volumes for an ultra-short time, typically of the order of picoseconds. Each ion induces a continuous trail of damage having a few nanometer diameter and typically several tens of micrometers length. There have been several studies in the past, including a few from our group of irradiation effects of high energy heavy ions in fullerene films [7–10]. Special feature of high energy heavy ion is that the energy lost by the ion (that brings the changes in the film under ion irradiation) is dominantly via inelastic collisions, in contrast to low energy ions where the energy loss is dominantly via elastic collisions. The existence of an electronic stopping power threshold (dE/dx) for the phase transformation is well established for different types of materials (dielectric, semiconductors and metals) [11–13]. Vast experimental data are now available concerning the dependence of induced disorder on (dE/dx) but the damage mechanism is still an open question. Various mechanisms have been proposed to explain the transfer of energy from the energy deposited by ion to the atomic motion in the material, two main models are the thermal spike [14] and the Coulomb explosion [15].

Now-a-days, the availability of cluster ion beams in the MeV range extends the possibilities of increasing the density of energy deposition [16,17]. As the cluster ions pass through the solids, they break up within the first few atomic layers and the constituent atoms deposit their energy simultaneously. The trajectories of atoms (of fragmented cluster) remain strongly correlated over a distance that depends on the initial energy and number of constituents [18]. After a certain distance, strong correlation of trajectories is lost and hence the density of electronic energy deposition reduces drastically. Recently, the formation of latent tracks in semiconductors (Si, Ge, GaAs) by cluster beams has been reported at room temperature [19,20], whereas tracks are not created by mono-atomic beams in these materials. In the present study we investigate dense electronic energy deposition effects in fullerene

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films, which have semiconducting properties with optical band gap ~2 eV. In this material, there have been a few studies of the damage induced by swift heavy ions, using FTIR, Raman and other techniques [7,10,21,22]. The formation of tracks in fullerene films was first explained by Dufour et al. [23], in the electronic energy loss region from 3 keV/nm to 10 keV/nm by thermal spike calculations. They reported that tracks should be formed above a threshold $(dE/dx)_e$ value of about 3–4 keV/nm. Although there have been a number of studies on damage of fullerene by ion beam [3,4,7–10] indicating the possible formation of ion tracks, no direct evidence of tracks in fullerene by microscopy techniques has been reported. The damage process under the dense electronic excitations in such a complicated system needs to be investigated in more detail. In the present work, we study the formation of tracks in fullerene films by high resolution transmission electron microscopy. For this purpose, we irradiated fullerene films on carbon coated grids by 30 MeV C_{60} and 120 MeV Au ion beams.

2. Experimental details

Fullerene thin films were deposited on carbon coated transmission electron microscopy (TEM) grids in a vacuum of 1×10^{-6} Torr by resistive heating using commercially available 98% pure C_{60} in a tantalum boat. The thickness of the film as measured by quartz crystal thickness monitor was about 30 nm. The samples were irradiated with 30 MeV C_{60} cluster and 120 MeV Au mono-atomic ion beams from Tandem accelerator at IPN Orsay, France and 15 UD Pelletron at IUAC New Delhi, India, respectively. Samples were irradiated with C_{60} cluster ions at normal and grazing incidence up to fluence of 10^{10} ions/cm^2. This range of fluences was chosen in order to avoid any spatial overlap of tracks. In the case of mono-atomic ions, TEM samples were irradiated with fluence up to 2×10^{11} ions/cm^2. All irradiations and TEM measurements were performed at room temperature. The electronic energy loss and nuclear energy loss were estimated by a computer code Stopping and Range of Ion in Matter (SRIM-2013) [24]. The energy loss per incident cluster ion is the sum of the energy loss of individual carbon atoms as experimentally checked by energy loss measurements for various cluster ions [25]. The electronic energy loss by 30 MeV C_{60} and 120 MeV Au ion beam were ~40.78 keV/nm and ~13.7 keV/nm, respectively. Similarly, nuclear energy loss by 30 MeV C_{60} and 120 MeV Au ion beam were ~0.752 keV/nm and ~0.162 keV/nm, respectively. The samples irradiated at grazing incidence (about 80° with respect to normal from surface) have the advantage that the evolution of the track shape during the slowing-down of the projectile can be studied by TEM observations. The samples were analyzed using a JEOL 2010 UHR transmission electron microscope (TEM) operated at 200 kV, at Institute of physics, Bhubaneswar, India. The used high resolution transmission electron microscopy has 0.19 nm point to point resolution. The electron irradiation during TEM measurement was observed to be very destructive for fullerene [26,27]. To avoid any modification in the fullerene matrix by electron beam during TEM measurement, the electron flux was kept as low as possible and the electron beam spot was shifted periodically during acquisition.

3. Results and discussion

The nano-track evolution in fullerene matrix by cluster ion (C_{60}) is induced in the fullerene films irradiated up at normal and grazing angle to a fluence of 1×10^{10} ions/cm^2. Fig. 1 shows a bright field (BF) image of latent tracks formation in fullerene irradiated by 30 MeV C_{60} ions at normal incidence at room temperature. The image permits to determine the density of the tracks as well as their diameter. The tracks are randomly distributed all over the surface of the sample and the track density is consistent with the fluence of projectiles. Fig. 2 depicts a high resolution TEM (HRTEM) images of the pristine (Fig. 2(a)) and irradiated fullerene films at normal incidence (Fig. 2(b)). The lattice fringes of pristine C_{60} film are seen in Fig. 2(a), with spacing $d \sim 0.82$ nm, which matches with reported values [22]. An impact of one cluster ion is observed in Fig. 2(b). From the HRTEM image, the impact region appears to be amorphous and non-impact region to be crystalline in the nature. Low magnification image (shown in Figs. 1 and 3) were taken under the conditions of BF imaging (low magnification BF images that are

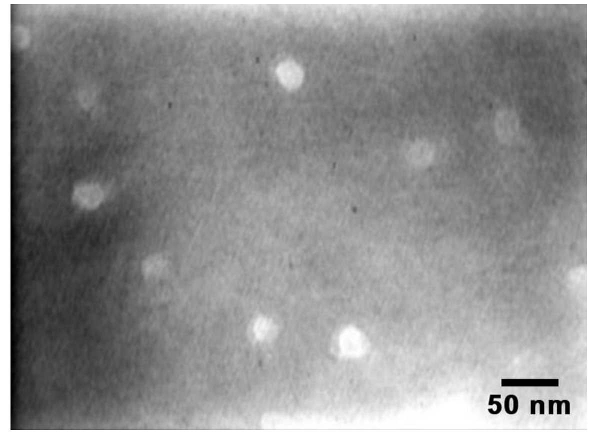

Fig. 1. Transmission electron micrograph of latent tracks formed in fullerene film under 30 MeV C_{60} cluster beam irradiation. The direction of beam was normal to the sample surface.

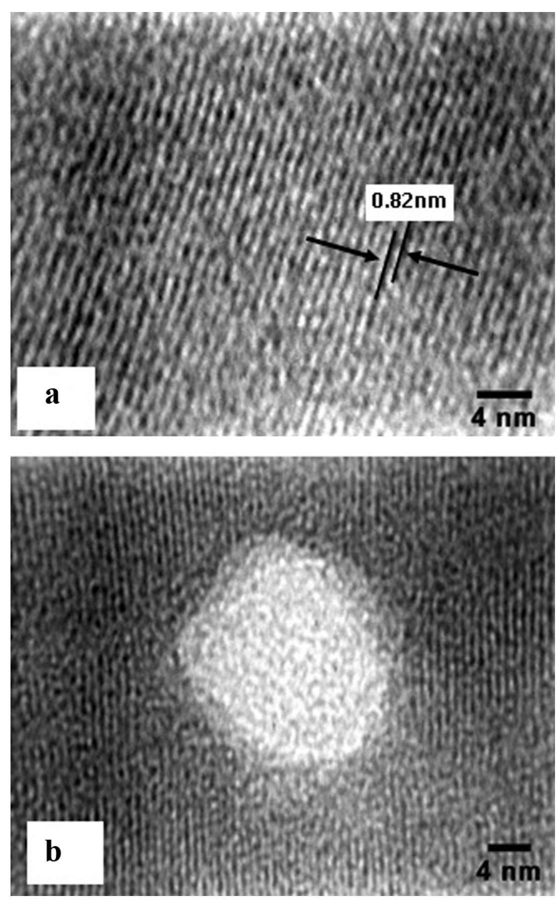

Fig. 2. HRTEM images of (a) pristine fullerene matrix, (b) latent track formation under 30 MeV C_{60} cluster beam irradiation.

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