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Structural damage on single-crystal diamond by swift heavy ion irradiation

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ABSTRACT

Experimental evidence of synthetic single-crystal diamond amorphization under the effect of irradiation with swift heavy ions is shown. The type of sharp thresholding behavior that characterizes electronic damage is not observed within a range of electronic stopping power covering up to 14 keV/nm. Amorphization is assessed by Rutherford Back Scattering Channeling (RBS-c) measurements done with light ions, after swift heavy ion irradiation. Results are analyzed and discussed in order to confirm the hypothesis of a nuclear damage-induced process, whereby it is concluded that electronic effects are not relevant in the studied range of stopping powers. The evolution of the amorphized fraction as a function of the irradiated fluence and nuclear stopping power is measured and discussed as well.

1. Introduction

Experiments performed on many dielectric and semiconductor crystals have conclusively shown that structural damage can be induced in dielectric materials by swift ions in which the electronic stopping power is dominant. This type of damage has been shown to have peculiar features such as thresholding [1–4] and cumulative [3–7] character. Different phenomenological models are available in the literature to describe the different features of this type of process [1,2,8,9].

Ion-beam-induced damage on diamond crystals has been observed and studied in the past only either in the low ion energy range (i.e., <10 MeV) [10–13] or for light ion beams [14–18]. In both cases nuclear stopping power is far larger than its electronic counterpart. Diamond has some extreme properties, such as excellent heat conductivity, which make it very different from any of the other electrical insulators where electronic damage has been observed so far. According to the values of the key material physical parameters reported by the manufacturer (i.e., thermal conductivity, $\kappa = 2000$ W/m K at 20 °C; specific heat, 2000 W/m K at 20 °C, density, $\rho = 3.515$ g/cm^3 and melting point, $T_m = 3550$–3730 °C) and the model by Toulemonde et al. [2] one would expect a threshold for the formation of permanent structural damage (i.e., latent tracks) at a large value of the electronic stopping power (above 20 keV/nm), predicting that one may rely on a simple picture where only nuclear stopping has an effect; the material is not affected by electronic interactions at all. However, this hypothesis has not been proven experimentally in a wide range of electronic stopping power. This work provides such evidence, for electronic stopping values high enough for practically any irradiation conditions possible in mid-energy ion accelerators (i.e., up to 14 keV/nm). Hence, we confirm for the first time the validity of the simplified damage mechanism scheme as a guideline for materials processing in tailored conditions.

2. Experimental methods

Samples are optical-grade synthetic single-crystal diamond pieces obtained from Element Six [19] manufactured using chemical vapor deposition. Their dimensions are $3 \times 3 \times 0.3$ mm^3 and all samples were irradiated with different ion beams at CMAM [20,21]. Ion species used for that purpose were: silicon (to explore the low stopping power regime), gold (to study and disentangle the competing effects of nuclear and electronic stopping powers) and bromine (for a final systematic analysis covering a wide electronic stopping power range). Used beam energies ranged from a few MeV to a maximum of 40 MeV. The reference fluence used was 10^{14} cm^{-2} because for this value of fluence appreciable track radii (>1 nm) produce full surface amorphization. Lower values have been used for systematic fluence-dependent damage analysis.

Once irradiated, samples were analyzed at CMAM with Rutherford Backscattering in the Channeling configuration (RBS-c) using a 3 MeV hydrogen beam, in order to have a large enough depth range of analysis, thus ensuring the consistency of the results. Depth resolution was not critical, since only amorphization in the immediate vicinity of the surface has been analyzed quantitatively. For high-resolution damage depth profiling of a limited depth layer a He beam would be a more convenient choice. In all cases (both, for the irradiation and the analysis)

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the ion current was kept below 20 nA to avoid the sample heating. Sizes of the irradiation and RBS-c spots were 6 × 6 mm^2 and 2 × 2 mm^2, respectively.

RBS-c spectra were analyzed in order to understand and quantify the effect of nuclear and electronic stopping. Finally, the damaged fraction of the crystal in the upper 500 nm thick layer was quantified for each spectrum, by applying the usual formula: $f=(\chi_d-\chi_v)/(\chi_r-\chi_v)$, where χ stands for the integral of the RBS spectrum within the depth range chosen and the subscripts stand for damaged (d), random (r) and virgin (v) diamond [22]. Electronic stopping power was determined by using the SRIM 2013 package [23,24] and averaging over the corresponding depth layer.

3. Experimental results

3.1. Dependence of irradiation damage on stopping power

Fig. 1 shows the RBS-c spectra taken with a 3 MeV hydrogen beam corresponding to a series of samples irradiated under different conditions. As a reference the channeled spectrum of an unirradiated sample and the one corresponding to the unchanneled condition (equivalent to full damage) are also shown.

The damage peak produced by the ions is clearly seen in all spectra, very prominently in the case of Au and Br irradiations, and less markedly for Si. Its position ranges from ~1 μm down to ~3.8 μm for silicon irradiations, from ~2.5 μm down to ~4 μm for gold and from ~2.2 μm down to ~4.7 μm for bromine ones. The depth scale has been calculated taking as a reference the position of the surface peak and that of the nuclear stopping maximum, as given by SRIM. Silicon irradiated samples do not show any damage at the surface, whereas gold irradiated ones show a small level of damage (ca. 10% displaced fraction). Finally, the samples irradiated with Br show a transition behavior, wherein irradiations with the nuclear damage peak being closer to the surface do show some residual damage tailing all the way to the sample surface. The depth of the maximum nuclear stopping power, calculated using SRIM [23,24], is represented in the plots as vertical dashed lines. It can be seen that the correlation between this value and the peak of maximum damage is excellent. Hence, this is strong evidence suggesting that damage is coming exclusively from nuclear effects. In all cases the integration window corresponding to the upper 500 nm surface layer is indicated in the figures. This is the window used for further analysis.

Results of the calculated surface damage (for the 500 nm thick surface layer) are summarized in Fig. 2 for the different samples, as a function of either the electronic or nuclear stopping power. The plot of the surface damage as a function of the electronic stopping power (Fig. 2a) looks rather chaotic, indicating that no relevant dependence exists in this case. On the other hand, the surface damage steadily increases as a function of the nuclear stopping power (Fig. 2b). Hence, it

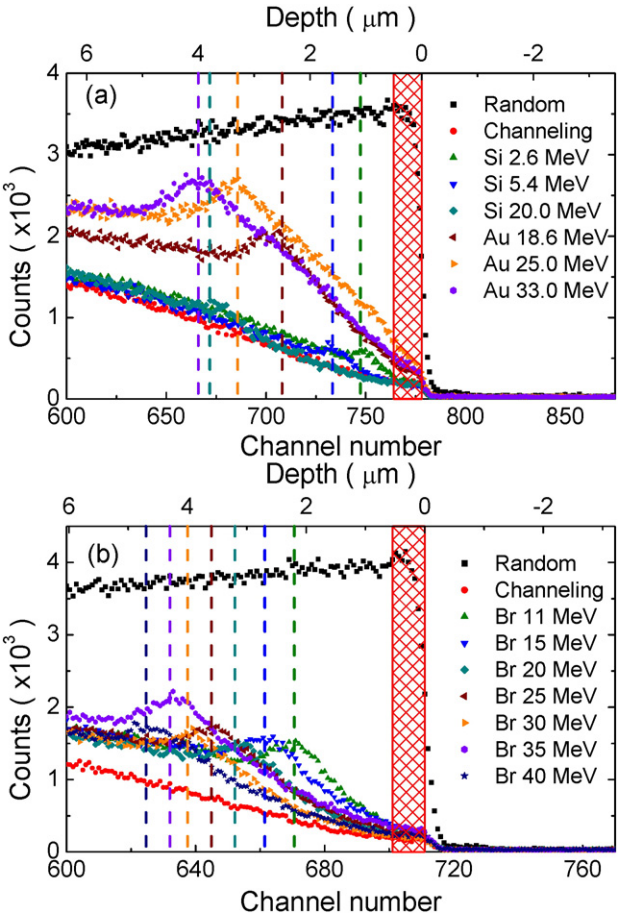

Fig. 1. RBS-c spectra taken with a 3 MeV H ion beam, on samples previously irradiated with 10^{14} cm^{-2} of (a) Si, Au and (b) Br ions. Undamaged sample spectra, for channeling and random configuration, are shown as a reference. Integration window for the subsequent analysis (upper 500 nm layer) is indicated with red area. Vertical dashed lines indicate the penetration depth of the ion beams, as calculated from SRIM.

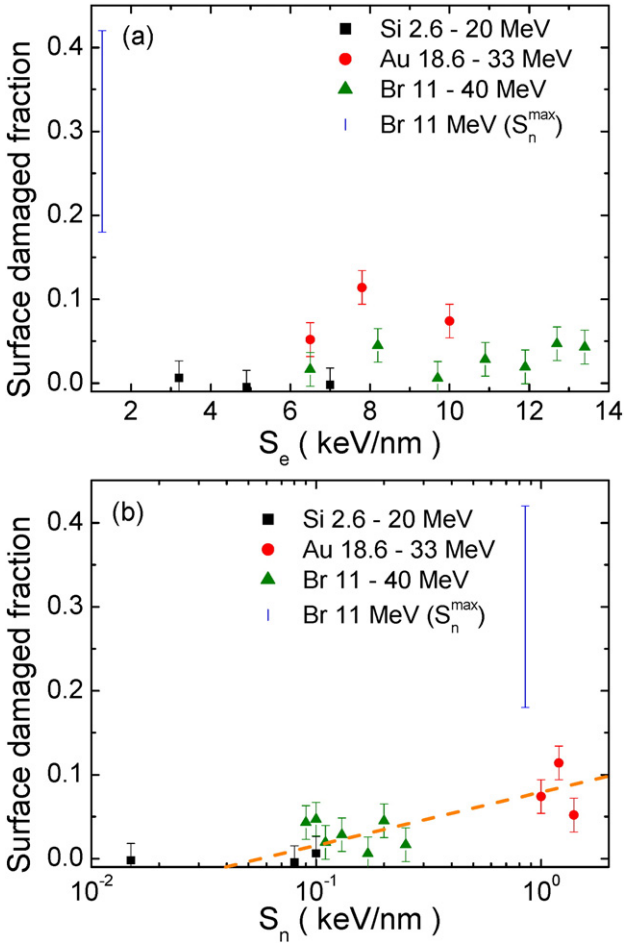

Fig. 2. RBS-c assessment of the damage produced by the previous ion beam as a function of either the (a) electronic or (b) nuclear stopping power. In all cases the fluence of the previous ion beam is 10^{14} cm^{-2}. Orange dashed line is just a guide to the eye. Symbols with the same color represent the different energies used for the same ion. Blue error bars represent an estimation of damage at the depth with the maximum value of nuclear stopping power (around 2.2 μm), for the sample irradiated with Br at 11 MeV.