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Investigation of the effect of helium ion (He^+) irradiation on the fluorescence properties of microdiamonds grown by chemical vapour deposition

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In this study, we explore the effects of bulk defects introduced using high energy helium ion (He^+) irradiation on the fluorescence properties of microdiamonds grown using the chemical vapour deposition (CVD) method. 2 MeV He^+ ions at varying fluences were employed to create lattice defects at several micrometre depth with respect to the crystal surface. Lattice damage was estimated using the Stopping and Range of Ions in Matter (SRIM) software package. When combined with confocal fluorescence microscopy, the results show that the fluorescence primarily arises from the silicon-vacancy $(Si\text{-}V)^-$ colourcentres as well as the nitrogen-vacancy (N-V) Frenkel pairs. After irradiation with He^+ ions, the fluorescence output increased significantly (by around 8 times), suggesting that the proposed irradiation treatment increases the number density of fluorescent defects in diamonds.

Small diamond crystals containing deliberately induced point defects located far away from the crystal surface are of interest for a variety of applications, including biomedical imaging, quantum computing, and magnetic sensing [1]. The formation of these optically active point defects after growth is a two-step process. Vacancies are generated by high-energy electron or ion irradiation within the diamond bulk, and then mobilised ('activated') via high temperature annealing causing them to migrate within and interact with the crystal lattice. The location of a defect or impurity within a crystal can be tailored via careful selection of the species and incident energy of ions [2]. In addition, some residual strain may already be present within the as-grown crystals which might affect their optical properties [3]. In the case of wide band gap materials such as diamond, the electronic excitations may also produce additional point defects in the irradiated crystal, which can either increase (radiative decay) or decrease (non-radiative) its photoluminescence.

Here we report on the characterisation of microdiamonds irradiated with high energy He^+ ions. The effect of irradiation fluence on the formation of radiative defect centres as well as the fluorescence output of diamond crystals was investigated using both widefield and confocal fluorescence microscopy. The nature of the fluorescent defects within the microdiamond crystals was determined using a spectrometer attached to the confocal microscope. The microdiamonds were grown on a silicon (Si) substrate by the microwave plasma-enhanced CVD method; the details of seeding and growth can be found in reference [4]. A sample of as-grown microdiamonds was used as a control (CVD-U). Two samples were irradiated with 2MeV He^+ ions: one (CVD-9) at a lower dose of 10^9 ions/cm^2 (1.6×10^{-10} C/cm^2); another (CVD-12) at a higher dose of 10^{12}ions/cm^2 (1.6×10^{-7} C/cm^2). Surface sputtering and monolayer collision statistics were collected for a total of 10^4 simulated irradiated ions.
SRIM predicts that all the irradiated He^+ ions having an initial energy of 2 MeV, will pass through a 1.2 µm sized diamond crystal, producing on average 2.4 vacancies in the target crystal. The predicted damage densities (10^{13} and 10^{16} vacancies/cm^3 for low-dose (CVD-9) and high-dose (CVD-12) samples) are well below the graphitisation threshold (10^{22} vacancies/cm^3) for diamond [5]. The widefield fluorescence imaging results showed a small increase (around 2 times) in the number fluorescent defect centres observed in low-dose (1×10^9 ions/cm^2) sample. However, a significant increase (around 28 times) in the number density of fluorescent centres was observed for the high-dose (1×10^{12} ions/cm^2) sample. Confocal fluorescence microscopy results also showed an increase (by 8 times) in the fluorescence intensity of ion-irradiated microdiamonds. However, no additional peaks could be detected in the ion-irradiated samples.

Hence, our results suggest that even with the creation of only a small number of vacancies, high energy He^+ ion irradiation is very effective in producing more highly fluorescent microdiamonds.

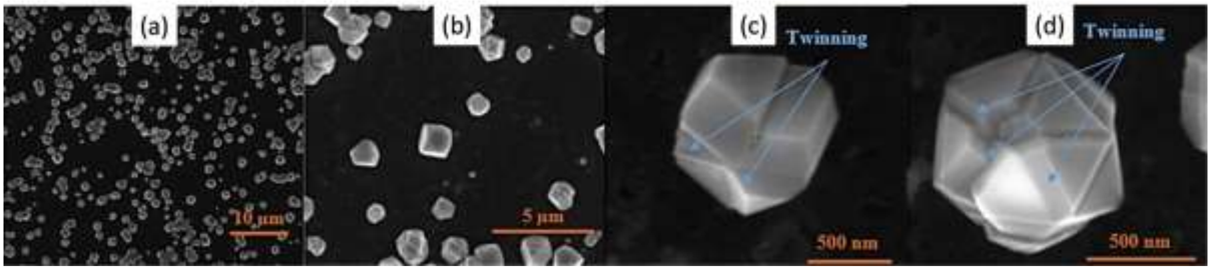

Figure 1. Figure 1 SEM images of CVD grown microdiamonds (a) Overview showing the number density of crystals, (b) zoomed in region of the sample shown in (a), confirming variable sizes and morphologies of the crystals, (c) and (d) Individual microdiamonds having crystallographic twinning in the diamonds possibly due to higher nucleation density.

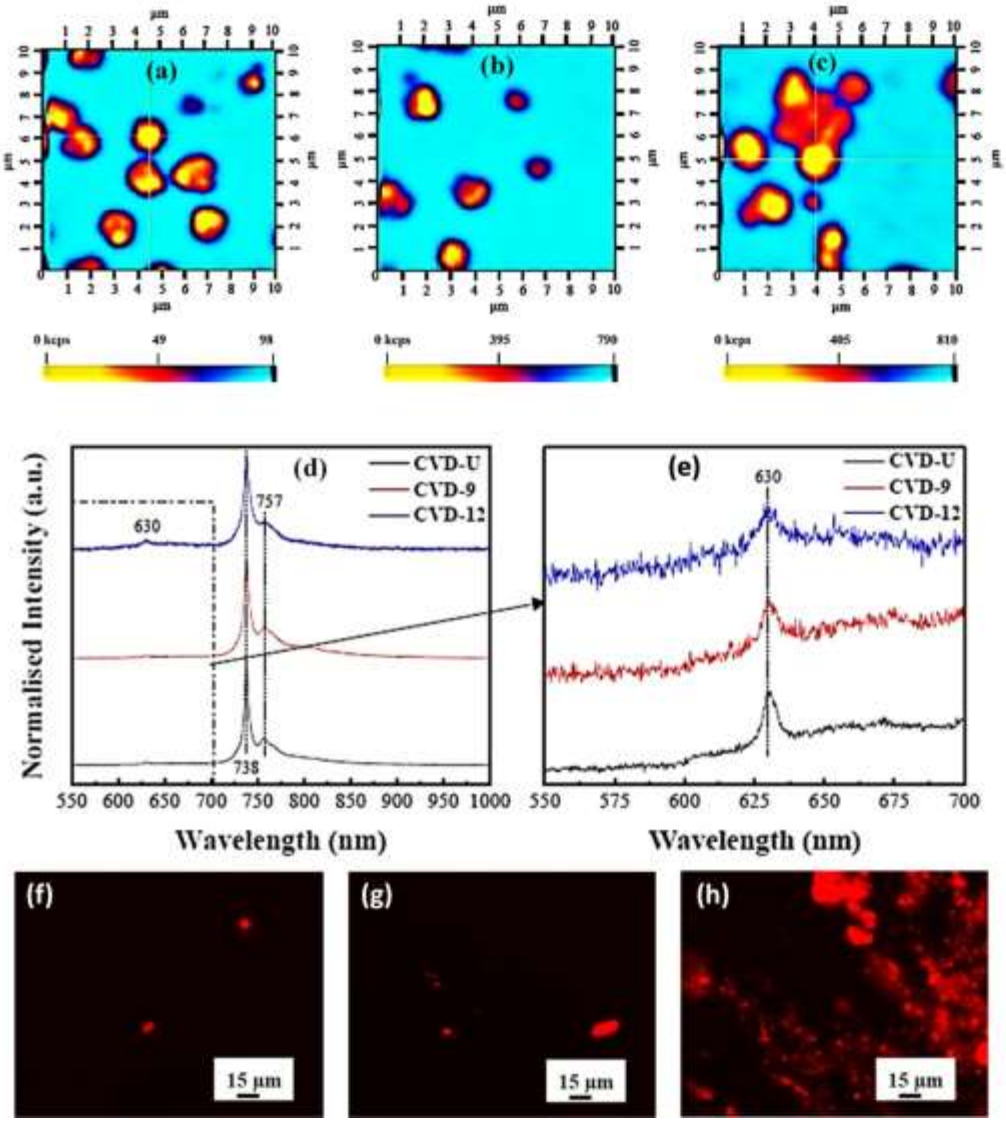

Figure 2. Figure 2 Confocal fluorescence images of (a) control (CVD-U), (b) low-dose (CVD-9) and (c) high-dose (CVD-12) samples, respectively. (d) Photoluminescence spectra for the control (CVD-U), low-

dose (CVD-9), and high-dose (CVD-12) samples. (e) Photoluminescence spectra zoomed in over the wavelength range of 550 – 700 nm. Widefield fluorescence images were obtained from (f) CVD-U, (g) CVD-9, and (h) CVD-12 samples.

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