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Effects of the grain size of CVD diamond films on the detector performance

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Although the unique properties of CVD diamond films have made it a candidate material for radiation detectors, the detector performance is strongly dependent on the film quality. In this paper, three CVD diamond films with different grain size were grown by using a hot-filament chemical vapor deposition (HFCVD) technique and the ratio of the grain size to the film thickness is high to 50%. 5.9 keV ^{55}Fe X-rays measured the photocurrents and the pulse height distributions (PHDs) of these CVD diamond detectors. The detector performance is improved with the grain size increasing. The dark-current of 16.0 nA and the photocurrent of 15.9 nA are obtained at an electrical field of 50 kV·cm^{-1} and the PHD peak is well separated from the noise pedestal.

1. Introduction

The many exceptional properties (electronic, optical, mechanical, etc.) of synthetic diamond films grown by CVD techniques give it excellent features in term of high radiation resistance [1], low leakage current, high temperature operating capability [2], high chemical inertness [3], and tissue-equivalence [4]. Thus, it has been expected that CVD diamond radiation detectors can be used in many applications, e.g. nuclear detectors [5]. The high resistivity of diamond permits a simple detector structure and the high band-gap results in an extremely low number of free carriers leading to very low noise and power dissipation, while the high carriers' mobilities and saturation field allow fast charge collection [6]. Even more important is the high radiation hardness and high temperature operating capability of CVD diamond detectors, since particle fluxes in the next generation of particle accelerators will be so high as to be beyond the operational limit of the conventional silicon based detectors [2].

The polycrystalline nature of films makes the performance of CVD diamond detectors strongly dependent on the film quality [7], but the effects of the grain size of CVD diamond films on the detector performance was scarcely studied [8]. In this work, the responses of CVD diamond detectors based on different grain size to 5.9 keV X-rays were investigated.

2. Experiment

CVD diamond films were deposited on (100) *p*-type silicon substrates with a resistivity of 4–7 Ω·cm by a HFCVD technique, using a hydrogen-acetone precursor mixture. Three CVD diamond films with different grain size were obtained by adjusting the deposition parameters [9] and characterized by SEM and Raman spectroscopy. After processed in the solution of strong H_2SO_4 + 50%H_2O_2 for 30 min to remove the non-diamond on the film surface and then annealed at 500°C in an Ar atmosphere for 1 h to improve the film quality, all films were sequentially thermally evaporated Cr and Au on the growth side. All detectors have a same sandwich structure (Au/Cr-diamond-Si). The frontside contact facing the X-ray irradiation was made from one 1 mm-diameter circle pad of Cr: Au double layers, 50 nm: 150 nm thick. In addition to mechanical support, the Si substrate was used as the backside electrode. To obtain ohmic contacts, all detectors were annealed at 450°C in an Ar atmosphere for 45 min [10].

Keithley 4200-SCS was used to measure the current characteristics of CVD diamond detectors with and without 5.9 keV X-ray irradiation [55]. Fe used as an X-ray source was placed above the detectors at a distance of 1 cm in atmosphere at room temperature. The frontside electrode used as the signal output was connected, through a charge sensitive preamplifier, a spectroscopy amplifier (Gain = 12 k; Shaping-time = 3 μs), to a multichannel pulse height analyzer [11].

3. Results and discussion

The surface morphology indicates a typical microcrystalline structure (as shown in Fig. 1a–c) with an average grain size increase from 0.5 to 10 μm. It also reveals that the film texture changes from a typical [110] (Fig. 1a) to a [100] dominant texture (Fig. 1b and c). A distinct columnar structure is observed at the cross-section of the film (c), while it is not evident in the films (a) and

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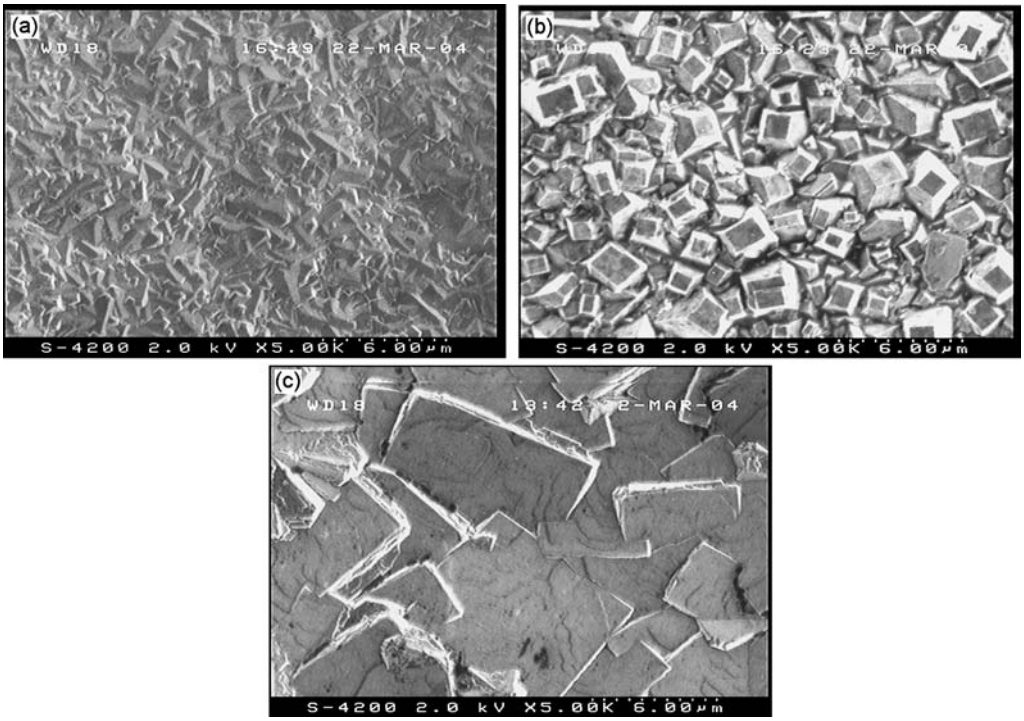

Figure 1 SEM photographs of three CVD diamond films.

(b). The film thicknesses are all of 20 μm (not shown here). The ratio of the grain size to the film thickness is about 50% in the film (c), much larger than the reported value of the order of 10–20% [12, 13], which is important to grow 'detector grade' or single crystalline CVD diamond.

All Raman spectra (as shown in Fig. 2) present a well-defined diamond peak at around 1332 cm^{-1} (as seen in Table I) together with a featureless and broad band at 1400–1600 cm^{-1} and a luminescence background. The graphite with characteristic peak at 1580 cm^{-1} is absent in all spectra. Although the quality of these CVD diamond films is different, they all have high diamond quality.

TABLE I Diamond peak and FWHM of three CVD diamond films obtained by Raman spectra

Sample	(a)	(b)	(c)
Diamond peak (cm^{-1})	1334.4	1335.0	1331.7
FWHM (cm^{-1})	11.26	11.10	10.84

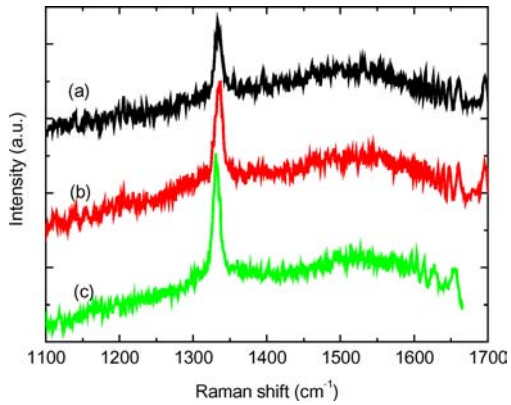

Figure 2 Raman spectra of three CVD diamond films.

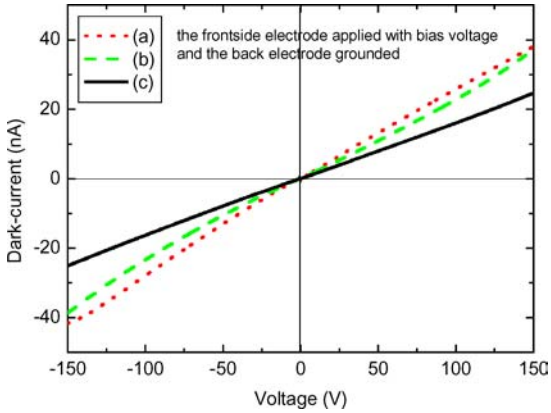

Figure 3 Dark-current vs. the detector bias voltage.

The I-V curves (shown in Fig. 3) of three CVD diamond detectors show a linear and symmetric relationship from negative to positive voltage, which indicates that an ohmic contact, important for detectors, for bias voltage up to 150 V is formed between CVD diamond films and the electrodes. This contact may be consequent on the carbide formed between chromium and diamond during the annealing processing. The dark-currents of 25.8, 22.8 and 16.0 nA are found for the corresponding films (a), (b) and (c) at an electric field of 50 kV·cm^{-1}. The electric properties reveal that the detector (c) has a lower dark-current than the detectors (a) and (b). Larger the grain size is, lower the dark-current is and therefore lower the detector noise. The polycrystalline nature of CVD diamond films results in a great abundance of grain boundaries where a higher concentration of impurities and/or defects is found than in-grains [14]. These impurities and/or defects strongly

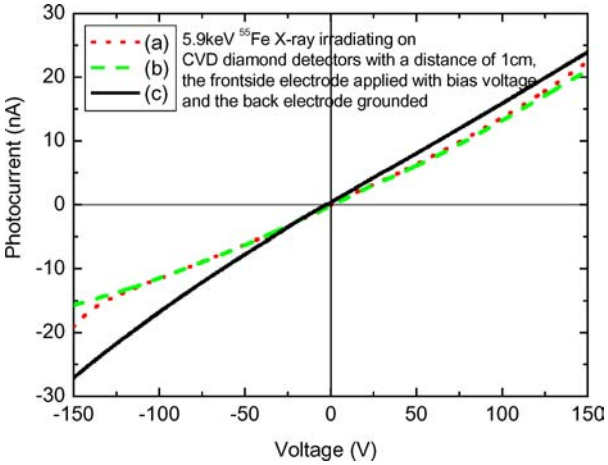

Figure 4 Photocurrent vs. the detector bias voltage under 5.9 keV X-ray irradiation, where the photocurrent denotes the net photocurrent which has been subtracted the dark-current from the total current.

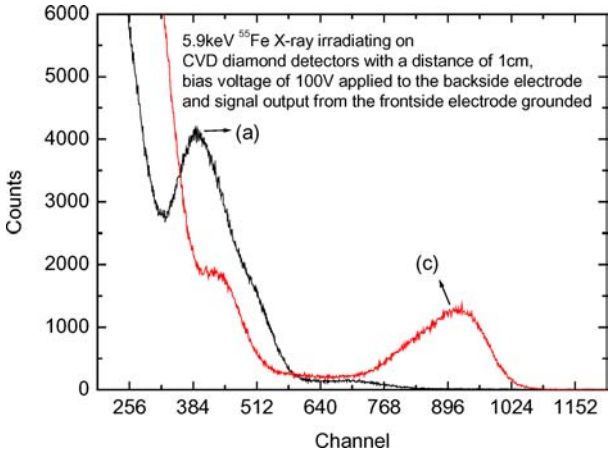

Figure 5 The pulse height distributions of CVD diamond detectors (a) and (c) obtained by 5.9 keV X-rays.

affect the electric performance of CVD diamond films. The detector (c) has larger grain size, which means less grain boundaries and therefore lower dark-current.

Fig. 4 illustrates the photocurrent responses of three CVD diamond detectors to 5.9 keV X-rays, where the photocurrent I_{ph} denotes the net photocurrent which has been subtracted the dark-current from the total current. At low electric field, the photocurrent proportionally increases with bias voltage, due to a linear relationship between the collected carriers and external electric field. In fact, according to a simple photo-generation and collection model [15], the photocurrent can be written as:

$$I_{\mathrm{ph}} = q F_0 \eta_{\mathrm{abs}} \mu\tau E/d \qquad (1)$$

where q is the electron charge, F_0 the number of incident photons per unit time, η_{abs} the optical absorption efficiency, $\mu\tau$ the mobility-lifetime product of photo-generated carriers, d the interelectrode spacing (i.e. the film thickness here) and E is the applied electric field.

The detectors (a) and (b) have a similar behavior: the photocurrent is proportional to bias voltage up to 150 V in the positive condition, and linearly increases at low voltage and then levels off over 50 V in the negative condition. Different to the detectors (a) and (b), although the detector (c) behaves the same way in the positive condition, in the negative, with an initially linear increase at lower voltage, larger photocurrent at higher voltage can be observed. At an electrical field of 50 kV·cm^{-1}, the photocurrents about 13.7, 13.2 and 15.9 nA are obtained by the detectors (a), (b) and (c), respectively. Compared with the dark-current, the detector (c) indicates the best detection performance.

When CVD diamond films are irradiated by X-rays, free carriers (electron-hole pairs) are generated which, under external electric field, move towards and then induce a transient signal on the device electrodes. Since the collected charges are mainly originated from electrons at positive voltage and holes at negative voltage, the difference between the detectors (a) or (b) and (c) may be due to the different grain size. The films (a) and (b) have smaller grain size, which means that there exist much more grain boundaries in the films (a) and (b) than in the film (c) with larger grain size. Because grain boundaries are abundant of trapping centers which would capture free carriers, the detector (c) exhibits a stronger photocurrent than the detectors (a) and (b). The reverse behavior between the detectors (a) or (b) and (c) in high negative voltage may be correlated with the grain size and the hole-conductivity nature of CVD diamond films. At high voltage, the dark-current and the photocurrent are all high. The dark-current of the detectors (a) and (b) increase more rapidly than the photocurrent, while that of the detector (c) is on the contrary, due to the grain size.

Fig. 5 shows the typical pulse height distributions (PHDs) generated by 5.9 keV X-rays from a ^{55}Fe source for the detectors (a) and (c) at an electrical field of 50 kV·cm^{-1}. The broad feature of the PHDs indicates that spectral resolutions of CVD diamond X-ray detectors are generally insufficient for spectrometric applications. Since the PHD peak lies well above the noise threshold, high counting efficiencies and low detection limits can be generally expected. The detector will work well as an X-ray radiation monitor, for which a precise particle energy is not of special interest, and usually the type of radiation source is known. Especially, for medical purposes, for example, X-ray and gamma dosimetry, diamond has a unique advantage of being a tissue-equivalent material, i.e. having an atomic number ($Z = 6$) close to that of the human body ($Z \approx 7.4$) [4]. Furthermore, the material is non-toxic, thus suitable for *in-vivo* usage, including implantation.

Because the detector (a) behaves similarly with the detector (b) in Figs 3 and 4, the pulse height distributions generated by X-rays were only compared by using the detectors (a) and (c). Although the signal peak is well separated from the noise for both detectors, i.e. the valley formed between the noise and the PHD peak, it is clear that the detector (c) has a higher PHD maximum and deeper valley between the noise and the PHD peak, which means a higher counting efficiency and thus a higher signal-noise ratio, consistent with the dark-current and photocurrent results. In general, the performance of a semiconductor detector is governed mainly by the quality of the semiconductor material

[16]. Thus, the difference between the performance of the detectors (a) and (c) can be directly explained by the film quality. The detectors (a) and (c) have exactly the same structure and closely diamond quality, therefore the observed difference in the PHDs originates from the difference in the grain size of CVD diamond films used for these detectors. Figs 3 and 4 also show that the detectors (a) and (b) with a different texture and a similarly low grain size behave similar current characteristics. Hence, the film texture may affect the radiation response, but the grain size of the films may be a more evident contribution to improve the detector performance, due to the trapping centers (impurities and/or defects) abundant on grain boundaries. Bergonzo *et al.* [17] had also confirmed no significant improvement of the detection properties in an oriented material.

4. Conclusions

The response to 5.9 keV X-rays reveals that the detector (c) based on the film with the largest grain size exhibits the best detection performance. The dark-current of 16.0 nA and the photocurrent of 15.9 nA are achieved at an electrical field of 50 $kV \cdot cm^{-1}$. The PHD peak is well separated from the noise, which means high counting efficiencies, high signal-noise ratio and low detection limits. Therefore, the detector performance would be drastically improved by increasing the grain size of CVD diamond films.

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