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## Introductory review of diamond detectors

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#### **Abstract**

Diamonds offer good properties as nuclear particle detectors. In these review the main properties of diamonds as particle detectors are shown. In the first part the detection mechanism is explained. Later it is described how diamonds can be created in detector grades and their main problems are mentioned. A section is dedicated to the analysis of the main parameters to describe the detection properties. Some diamonds properties under neutron or protons irradiation are presented as these detectors are to be used in harsh radiation environment.

### 1 Introduction

Since the beginning of nuclear era diamonds were investigated as nuclear particle detectors. Their high chemical inertness, high thermal conductivity, high breakdown field make them suitable to work in harsh environments.

# 2 Physics of detection

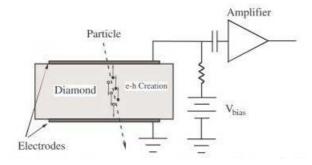


Figure 1. Basic semiconductor detector scheme

Carbon (C), whereof diamond is an allotrope, stand in the IVA group of the periodic table, with Silicon (Si) and Germanium (Ge), the most famous semiconductors. These kind of materials have a crystalline lattice that establishes allowed energy bands for electron that exist within that solid. A simplified model allow the electron to be in a lower energy band, called the valence band, or in a upper band, the conduction band, where electrons are free to move. The two bands are separated by a band gap. Electrons need energy to jump from the valence band, where they are bound in the outer-shell of the atoms, to the conduction band to let the charge moving. The band gap defines a material to be a conductor, a semiconductor or an insulator.

Diamonds have a 5.47 eV band gap, while Silicon 1.12 eV and Germanium only 0.66 eV at room temperature. Thermal energy of the crystal can promote an electron to the conduction band, creating an electron-hole pair. For each electron in the conduction band, there is a vacancy in the valence band. The probability per unit time to generate an electron hole by thermal excitation is given by:

$$p(T) = CT^{\frac{3}{2}}exp\left(-\frac{E_{gap}}{2kT}\right)$$

where T is the absolute temperature,  $E_{gap}$  is the energy of the band gap, k the Boltzmann constant and C a proportionality constant characteristic of the material [2].

High bad-gap energy value helps to reduce the current flowing in the diamond when the external field is applied to the detector, called dark current. Diamonds provide an high breakdown voltage ( $10^7 \ V \ cm^{-1}$ ), the limit beyond which the current through the detector increases exponentially.

When a ionizing radiation crosses the material, it ionizes the semiconductor, creating electron holes pair along its trace. A mean of 13.3 eV [3,4] are needed to promote the creation of a electron-hole pair in diamonds. In comparison silicon need a mean of 3.62 eV energy, but ionization chambers needs at least 26.3 eV (Ar) to create an electron-ion pair [2]. Due to the high drift velocity of both electrons and holes the charge can be collected easily if the diamond is under an electric field. Diamonds can so operate as a semiconductor. Moreover Si and Ge need

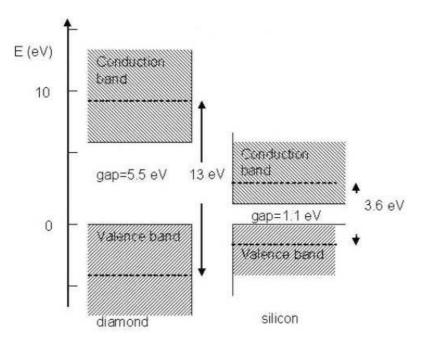


Figure 2. Valence and conduction energy values for silicon and diamond [1]

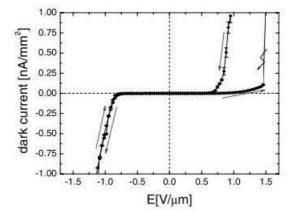


Figure 3. I-V dark current curve [3]

doping, the implantation of donor and acceptor ions in the lattice and opposite bias, to create a deplete area to avoid thermal current. Diamonds do not need to be doped, their dark current is very low at room temperature due to the high band gap energy, and the whole crystal is the active area. Dark current is on the order of nA according to [4].

### 3 Intrinsic defects

Different types of diamond are in use as nuclear detectors. In the early period researcher focused on natural IIa diamonds but the high cost, the small size and the lack of control of the crystal characteristics reduced their usage. To step forward synthetic diamonds were investigated as production techniques were developed. High Pressure-High Temperature (HP-HT) process was the first industrial technique that lead to a great production of diamonds, still impure (metal inclusions, that drastically reduces the diamond resistivity) but of adequate size and known quality to be used as detectors. With the advent of the Chemical Vapor Deposition (CVD) technique high grade diamonds were available for nuclear purposes. In the last decade CVD allowed to create single-crystal diamonds (scCVD), against the first poly-crystalline gems (pcCVD). The difference between poly crystal and single crystal is the obvious crystal form. To perform a high charge collection diamonds should avoid the presence of traps in the lattice, both for electron and holes; traps, such as boron or nitrogen inclusion in the lattice, other impurities, or grain boundaries, reduce the signal amplitude and the signal timing by collecting charge carriers and releasing them after a random period. That implies a bad signal shape and the impossibility to collect at once all the charge carriers generated by a single interaction event. A delayed release of charge leads to a reduction of the original pulse and quench the following pulses. Poly-crystal diamonds cannot perform spectrometric measurements due to their impossibility to control grain charge trapping. Single-crystal instead are not affected by this problem by definition, and the modern growing techniques lead to a negligible impurity quantities in the crystal ( $< 10^{15}$  nitrogen/cm<sup>3</sup>).

## 4 Radiation induced defects & radiation interaction

Defects in the crystals are not only intrinsic but they are created by radiation that knock off atoms from their lattice position. All charge particles act in a similar way to displace atoms at low energies: vacancies and interstitial are produced when the incident particle scatter (by Rutherford scattering) and its energy is greater than the displacement energy  $E_d$ . The values of  $E_d$  quoted in literature range from 25 eV to 80 eV, depending on the orientation of the crystal during the measurement; 40 eV is a good value to take as an approximation [5].

Nuclei that are displaced at short distance can recombine with the vacancy, and the recombination is highly affected by the temperature. If the displaced atom has sufficient energy, it may displace further atoms, creating a damage cascade. This process is more probable with heavy or high energy particles, that can transfer to the lattice atoms a great momentum. Besides, displaced carbon have a great cross section for a carbon-carbon collision, so localized damage cascades are very probable. Nitrogen or other impurities create complex aggregates with low  $E_d$ , leading to an high probability to generate excess of damage cascade. Computer programmes as TRIM can predict accurately the distribution of the displaced atoms and the trail of damage that they leave. The damage in the lattice is therefore a consequence of the kind of particle and its energy, not only of its scattering cross section.

In the lattice charged particles loose energy by ionization; electrons can emit Bremmstrahlung radiation at high energy; gamma radiation interact via photoelectric effect, Compton scattering

or pair production. The photoelectric electrons cannot displace carbon because their low energy; on the contrary the electrons and the positron generated by Compton scattering and pair production can contribute to the lattice damages as they are high energy particles. Protons and neutrons displace atoms mainly by scattering.

Neutrons interact with carbon, and fast neutrons knock off atoms more than thermal ones. Elastic scattering has a predominant role in low energy neutrons, and the cross section ( $\approx 1b$ ) is quite constant up to 20 MeV. Carbon recoils is so produced, with 0.2 MeV residual energy if the incident neutron has 1 MeV or 4.17 MeV if it is produced by a 14 MeV fast neutron. Above 4.8 MeV inelastic scattering  $^{12}C(n,n')^{12}C^*$  occur. The excited carbon will later emit a 4.47 MeV gamma radiation. Rising with neutron energy, above 6.2 MeV the  $^{12}C(n,\alpha_0)^9Be$  reaction take place, which is the main reaction used to measure the neutron energy. At high energy, over 8.4 MeV,  $^{12}C(n,\alpha)^9Be^*$ ,  $(n,3\alpha)$  and (n,np) reactions occur. With 14 MeV neutrons the sum of the inelastic cross section and  $(n,\alpha)$  cross section is about 40% of the elastic scattering cross section [6].

**Table 1.** Neutron reactions on  $^{12}C$  [7]

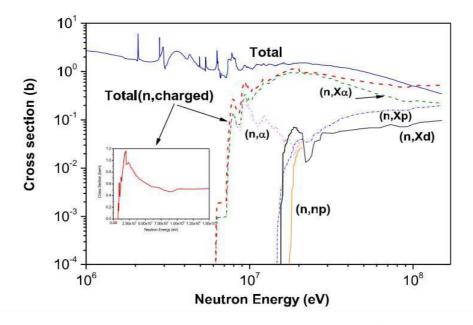
reaction	Q-value [keV]	Threshold [keV]
$12C(\mathbf{n},\gamma)^{13}C$	4946.31	0.0
$^{12}C(\mathbf{n},\alpha_0)^9Be$	-5701.25	6180.60
$^{12}C$ (n,n' $2\alpha$ ) $^4He$	-7274.75	7886.40
$^{12}C(n,n'\alpha)^8Be$	-7366.59	7985.96
$^{12}C$ (n,2 $lpha$ ) $^{5}He$	-8164.7	8851.2
$^{12}C(n,p)^{12}Be$	-12586.55	13644.81
$^{12}C(n,d)^{11}Be$	-13732.33	14886.93
$^{12}C(n,n'p)^{11}Be$	-15956.90	17298.53
$^{12}C({ m n,2n})^{11}C$	-18721.66	20295.75
$^{12}C$ (n,T) $^{10}Be$	-18929.22	20520.77

It has to be noticed that in pure diamonds the only nuclei generated by neutrons are He and Be nuclei, that generate displacement through their high ionization power. <sup>8</sup>Be can also break up producing more He nucleus. The formation of still He bubbles could affect the diamond response. Other semiconductors under neutron irradiation would have produced more extraneous atoms.

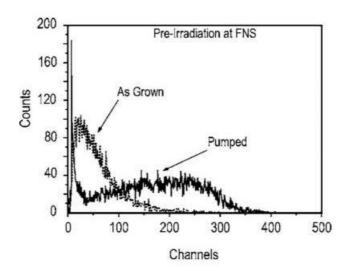
The total cross section at 1 MeV is about 2.5 b, which means that the most part of the neutrons pass through the detectors without interaction. Hence neutrons create a quite uniform damage through the whole detector. Constant damage wont happen with charge particles, such as alpha or protons. Their probability to be stopped inside the detector is relevant, and the more damaged area would be in the Bragg peak zone. Electron range in carbon is about few mm, the most of the electron pass through the detectors without stopping.

Anisotropy of the damage distribution lead to the creation of high damaged centres: by trapping charge carriers in concentrated areas they generate spatial charges that modify the external imposed electric field, an effect called polarization. Other causes generate polarization, the main in scCVD is the presence of poor contacts. Polarization affect mostly pcCVD, while single-crystal appears to be immune.

As previously reported vacancies and interstitial can recombine, reducing damage in the latter. The atoms displaced by the radiation usually have quite small energies. Therefore they do not move very far from the vacancy. Recombination can be performed and it is very affected



**Figure 4.** Cross sections for neutron interaction on  $^{12}C$  [7]



**Figure 5.** The response to  $^{241}$ Am  $\alpha$  particle irradiation by a as grown diamond and a primed one, [8]

by the temperature and the radiation flux. A way to reduce traps is the annealing process: the diamond, in high temperature environment, let the vacancies and the interstitial move and recombine. The first irradiation of the diamond can generate charge particles that fill the deep traps, provided their levels are deep enough to have long lifetimes for charges on defects, with the nett effect to improve the following responses. Priming can be reversed by temperature or UV light. To take advantage of priming effect, polycristalline diamonds are often irradiated with  $\beta$  radiation, mainly from  $^{90}$ Sr, or X-rays, in Gy order doses. The electrons colliding with the crystal passivate in-grain defects. This technique is called *priming* or *pumping*. Improvement of the response due to priming is well reported for pcCVD. Single crystal do not need any annealing or priming [9, 10].

# 5 Usefull parameters

The described defects in the microscopic scale reflect their presence in the electrical response of the detectors, quenching the ideal signal, both in amplitude and timing of the pulse. A theoretical approach is needed to describe the pulse generation and to identify the most important parameters. Charge collection efficiency, CCE, is the easiest way to determine the quote of the charge that is collected by the external circuit: it is defined as the charge measured  $Q_m$  over the total charge generated in the diamond  $Q_0$ , and it is directly related to charge collection distance, CCD:

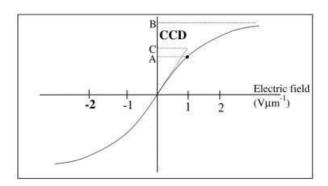
$$CCE = \frac{Q_m}{Q_0} = \frac{1}{d}CCD$$

where d is the distance between the electrodes, or the diamond thickness, as they are in contact. CCD can be written also related to the charge carriers mobility as follow:

$$CCD = (\mu_e \tau_e + \mu_h \tau_h)E$$

as  $\mu_{e,h}$  is the mobility of electrons and holes, and  $\tau_{e,h}$  their lifetime. This definition is valid under the hypothesis that the detector thickness d is greater than CCD, and the electric field E is uniform in the detector. CCD is the mean distance that electrons and holes drift before being trapped. More the crystals are wide, more the CCD is high, hence in the preparations of polycrystalline detectors the smallest crystals, that are located on growing substrate, are abraded.

Some papers quote the CCD as the asymptotic value at high fields, some quote it for a field of  $1V\mu\text{m}^{-1}$ , others quote the  $\mu\tau$  product, which is derived from the linear section of the graph at low field, Figure 6.



**Figure 6.** Schematic diagram of the variation of CCD with electric field across the detector. A, B and C show three values that may be quoted for the CCD

In a defect free semiconductor material, full charge collection is possible. Generally this does not happen, significant trapping occurs, and the induced charge becomes a function of the distance over which the charges travel. The Hecht's relation describes the effect of charge transport under a uniform electric field:

$$CCE = \frac{Q_m}{Q_0} \left\{ \frac{\nu_h \tau_h}{d} \left( 1 - exp \left[ \frac{-x_i}{\nu_h \tau_h} \right] \right) + \frac{\nu_e \tau_e}{d} \left( 1 - exp \left[ \frac{x_i - d}{\nu_e \tau_e} \right] \right) \right\}$$

where  $\nu$  is the charge carrier velocity,  $\tau$  is the charge carrier lifetime,  $x_i$  represents the radiation interaction location measured from the cathode, d the detector width. Good energy resolution can be obtained if the ratio  $\frac{\nu\tau}{d}$  (the *carrier extraction factor*) for both charge carriers

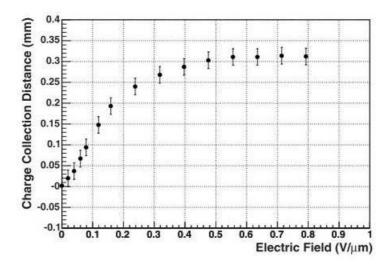
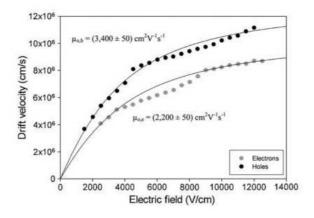


Figure 7. CCD vs electric field in a pcCVD, [11]

are greater than about 50. Because the charge lifetimes are fixed by the material, the carrier extraction factor can be risen by reducing the detector width and using higher electric fields. The charge velocity reaches a saturation value, so that the collected charge will reach the saturation. The drift velocity can be calculated by an empirical equation, with  $\nu_{sat}$  the saturation velocity:

$$\nu_{drift} = \frac{\mu E}{1 + \frac{\mu E}{\nu_{sat}}}$$



**Figure 8.** Drfit velocity for both electron and holes, [12]

In the practice the external circuit receive the current generated by the holes or the electrons moving to the grounded electrode, depending on the bias polarity. The total charge is then calculated as the integral over time of the current.

Significant informations are deduced with the analysis of the pulse shape generated by a single interaction. Particles or charge single shots are recorded on a fixed electric field and then averaged. The Full Width at Half Maximum (FWHM) of the curve gives the transition time  $t_{tr}$ . Assuming homogeneous electric field the asymptotic carrier drift velocity can be calculated as follow:

$$\nu_{dr}(E) = \frac{d}{t_{tr}}$$

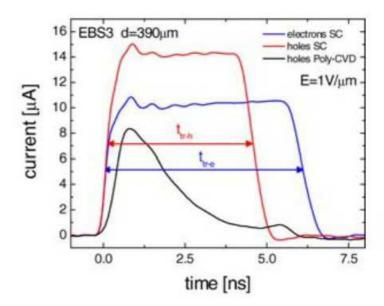


Figure 9. Current pulse shape differences between holes and electron in scCVD and a pcCVD, [13]

The current pulse shape reflects the quality of the crystal. Single crystals show a high rising time of the current signal, due to the arrival of the charge carrier cloud, and a constant current generation, because of the uniformity of the cloud. In polycrystalline detectors it can be seen that there is no current plateau. The charge carriers cloud is trapped and spread, resulting in an exponential decrease of the pulse shape.

# 6 Response after heavy irradiation

The main purpose for which diamonds has been developed is their usage under heavy irradiation fields, their response limits should be tested. Diamond shows a high hardness against gamma radiations, it can stand up to 100 kGy of 1.2MeV gamma without effects on the collection efficiency [14]. This characteristic is very useful for in-core fusion and fission reactors. Heavy particles produce more damage, leading to an efficiency reduction.

The damage created by protons and in general heavy charged particles, consequently the collection efficiency, is high influenced by the energy of the incident particle. High energy particles lead to a best efficiency. Under 2.6 MeV protons, diamonds offer 100% CCE at  $10^{12}$  proton/cm<sup>2</sup>, decreasing to 70% with  $10^{13}$  proton/cm<sup>2</sup> [15].

Neutrons are the other high damaging particles. As previously reported their only way to interact is by scattering or by nuclear reaction that lead to a great probability to displace atoms. Damage in diamonds does not affect the response of the detector until a  $10^{14}$   $n/\text{cm}^2$  fluence, with 14.8 MeV neutrons, while silicon can not be used above  $10^{13}$   $n/\text{cm}^2$ . Besides, the fist irradiation seems to improve the subsequential resolution of the single crystal diamond [16]. In pcCVD the priming effect is well clear [6]. With 5 MeV neutrons, as in [17], the priming effect is also visible, and the CCE slightly decrease approximately by 15% up to  $6.5 \times 10^{15}$   $n/\text{cm}^2$ .

## References

[1] R. Potenza and C. Tuvé, *Measurements of defect density inside CVD diamond films through nuclear particle penetration*, Carbon, the future material for advanced technology appli-

- cations, 286 (2006) pp.267-286.
- [2] G. Knoll, *Radiation Detection and Measurement, third ed.*, New York/Chester/Weinheim/Brisbane/Toronto/Singapore: John Wiley & Sons, Inc., 2000, pp. 353-356.
- [3] M. Pomorski and E. Berdermann, *Characterisation of single crystal CVD diamond particle detectors for hadron physics experiments*, Phys. Stat. Sol. A, **202** (2005) pp.2199-2205.
- [4] H.Pernegger and S. Roe, Charge carrier properties in synthetic single-crystal diamond measured with the transient-current technique, Jour. of Appl. Phys., **97** (2005) pp. 073704.
- [5] B. Campbell and W. Choudhury, *Lattice damage caused by the irradiation of diamond*, Nucl. Instr. and Meth. A, **476** (2002) pp.680-685.
- [6] M. Pillon, M. Angelone and A.V. Krasilnikov, *14MeV neutron spectra measurements with* 4% energy resolution using a tpye IIa diamond detector, Nucl. Instr. and Meth. B, **101** (1995) pp.473-483.
- [7] M. Angelone, M. Pillon et al. *Thermal and fast neutron dosimetry using artificial single crystal diamond detectors*, Rad. Measur., **46** (2011) pp.1686-1689.
- [8] M. Angelone and M. Pillon, *Radiation hardness of a polycrystalline chemical-vapor-deposited diamond detector irradiated with 14 MeV neutrons*, Rev. of Scient. Instr., 77 (2006) pp.023505.
- [9] M. Marinelli, E. Milani et al., *Trapping and detrapping effects in high-quality chemical-vapor-deposition diamond films: Pulse shape analysis of diamond particle detectors*, Phys. Rev. B, **64** (2001) pp.195205.
- [10] C. Manfredotti, *CVD diamond detectors for nuclear and dosimetric applications*, Diamond and Rel. Mat., **14** (2005) pp.531-540.
- [11] H. Pernegger, *High mobility diamonds and particle detectors*, Phys. Stat. Sol. A, **203** (2006) pp. 3299-3314.
- [12] S. Gkoumas, A. Lohstroh et al., *Low temperature time of flight mobility measurements on synthetic single crystal diamond*, Diamond and Rel. Mat., **18** (2009) pp.1338-1342.
- [13] M. Pomorski, *Development of single crystal CVD diamond detectors for spectroscopy and timing*, Phys. Stat. Sol. A, **203** (2006) pp.3152-3160.
- [14] C. Bauer, I. Baumann et al., *Radiation hardness studies of CVD diamond detectors*, Nucl. Instr. and Meth. A, **367** (1995) pp.207-211.
- [15] A. Lohstroh, P.J. Sellin et al., *Ion beam induced charge (IBIC) irradiation damage study in synthetic single crystal diamond using 2.6 MeV protons*, Phys. Stat. Sol. A, **205** (2008) pp. 2211-2215.
- [16] M. Pillon and M. Angelone, *Radiation tolerance of a high quality synthetic single crystal chemical vapor deposition diamond detector irradiated by 14.8 MeV neutrons*, Jour. of Appl. Phys., **104** (2008) pp.054513.
- [17] A. Oh, M. Moll et al., *Neutron irradiation studies with detector grade CVD diamond*, Diamond and Rel. Mat., **9** (2000) pp.1897-1903.