Progress in detector properties of heteroepitaxial diamond grown by chemical vapor deposition on Ir/YSZ/Si(001) wafers

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Abstract

The development of CVD grown single-crystal Diamond-on-Iridium (DOI) sensors for charged-particle detection in hadrons and nuclei physics research is reviewed. A variety of samples grown at the University of Augsburg has been investigated with α and β sources in the laboratory, swift ions from the heavy-ion synchrotron SIS in Darmstadt, and relativistic protons from the COoler-SYnchrotron COSY in Jülich. The results obtained by means of I-E(V) studies, transient-current techniques (TCT), α -spectroscopy, and heavy-ion time-of-flight (ToF) measurements are compared to those of commercially available polycrystalline and homoepitaxial single crystal CVD diamond sensors of electronic grade quality.

In many aspects, the performance of DOI sensors was found quiet similar to that of homoepitaxial counters, and in any case far superior to that of polycrystalline detectors. Under single-carrier drift conditions, the CCE and energy resolution ($\delta E/E$) for holes reached levels $CCE_h > 95\%$ and $\delta E/E_h \sim 0.3\%$, respectively, which correspond to values of the Schubweg $w_{h,e}$ well above the detector thickness. In contrast, the CCE_e for electrons was typically lower than $\sim 40\%$, leading to appreciable reduction of the detection efficiency in the dual-carrier drift mode ($CCE \sim 60\%$), which characterizes the experiments with swift heavy ions and high-energy particles. We measured transport parameters comparable to those of homoepitaxial devices: $\mu_0^h \sim 3080 - 1756$ and $\mu_0^e \sim 2276 - 1150$ cm²/Vs, $v_{sat}^h \sim (1.7 - 1.4) \cdot 10^7$ and $v_{sat}^e \sim (1.5 - 1.0) \cdot 10^7$ cm/s, as well as intrinsic time resolutions $\sigma_i \sim 15$ ps. It is shown, that substantial improvements have been achieved in recent years, albeit reproducibility and the understanding of the reduced electron collection remain challenging issues.

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1 Introduction

Diamond heteroepitaxy on iridium substrates (DOI) is currently one of the most promising technological approaches towards the realization of single-crystal diamond materials for large-area detector applications [1]. The present report describes the results of a first comprehensive study on the electrical properties of intrinsic DOI films for this purpose and their classification among the different commercially available electronic-grade CVD-diamond types supplied by Element Six [2]. We checked nine heteroepitaxial diamond samples grown on iridium surfaces against four homoepitaxial single-crystal diamonds (SCD) and two polycrystalline diamond (PCD) films.

In the last decades, various scientific communities worked successfully on radiation-hard diamond detectors to be deployed in nuclear and high-energy physics experiments as well as related applications [3-10]. However, the incompatibility between the high crystal quality and the large dimensions of detector areas required for the majority of such devices remained an open challenge. In a pioneering work (1996), A. Sawabe and his group reported successful heteroepitaxial diamond deposition on iridium surfaces [11]. This changed the situation fundamentally. As revealed in a series of following investigations also by several other groups [12-15], DOI combined a high structural quality of the diamond layers with the basic potential of single-crystal iridium films to be scaled to wafer dimensions. The first DOI sensors delivering encouraging results were developed and tested at MSU (Michigan State University) for heavy-ion beam tracking [16]. This initiated several joint research activities all over Europe [17]. Our collaborations focused on the solid-state ionization-chamber concept based on large-area intrinsic DOI materials, since we were convinced from investigations of silicon sensors that detectors consisting of p-doped on intrinsic layers (mainly pursued for diamond electronic devices) are potentially less radiation hard. In addition, we agreed to concentrate first our efforts on the perfection of the crystal quality and afterwards on the enlargement of the sample area. For the present study, DOI films up to (10 x 10) mm² have been explored, though meanwhile the development of larger devices is making good progress and the characterization of plates $\geq$ (20 x 20) mm² is advancing.

In 2009, we compared for the first time an early DOI sensor with state of the art SCD and PCD detectors [5]. The results of the established diamond devices confirmed our previous studies concerning CCE as well as time and energy resolution [18]. The unrivaled dominance of SCD was evidenced. Complete charge collection and excellent energy resolution facilitated particle identification via energy-loss spectroscopy. In addition, the high breakdown field enabled ultra-fast particle timing and counting, to profit from the inherent high carrier drift velocity over the whole range of available ions and energies. The high potential of PCD sensors in ToF measurements and monitoring of intense heavy-ion beams was demonstrated, while their properties concerning ion spectroscopy or any time measurement based on single-particle readout of minimum ionizing particles (MIP) turned out to be insufficient. At that time, DOI samples had even lower CCE values than PCD. This was attributed to the high dislocation density, characteristic for DOI samples at this stage of development. In addition, extremely low currents even at high electric field accompanied by ultra-short induced-current signals of impressively large amplitudes, let us speculate that “further material removal from the nucleation side would improve significantly the charge-collection properties”.

In the present article, we review the evolution of ‘diamond-on-iridium’ since 2010, when it became more and

more evident that intrinsic DOI samples are of higher detection potential than ever suspected at the beginning of the research program [19]. We start with a short description of growth and post processing of detector-grade (DG) DOI plates including sensor preparation in Section 2, and continue with test measurements and characterization results in Section 3. In each part of the study, the results obtained for DOI crystals are compared with the data of devices made from standard PCD and SCD materials. The state of the art and the perspectives for DOI materials in the field of particle detectors are described in the final summary.

2 Preparation of Diamond-on-Iridium samples

The growth substrates used in Augsburg are 4-inch wafers of the multilayer structure Ir/YSZ/Si(001) (Iridium/Yttria-Stabilized-Zirconia/Silicon(001)) [1,12,13]. The remarkable feature of the diamond films grown on iridium surfaces is a high initial particle density ($3 \cdot 10^{11} \text{ cm}^{-2}$) combined with a unique epitaxial alignment of the grains that are formed by the bias enhanced nucleation (BEN) procedure [15]. In spite of the low initial mosaic spread ($\sim 1^\circ$), the dislocation density present in the film after coalescence is $> 10^{10} \text{ cm}^{-2}$. The diamond particles are separated by grain boundaries, which accommodate the misalignment between neighboring grains. This mosaic crystal structure first coarsens and then disintegrates after several $10 \text{ }\mu\text{m}$ growth. In the subsequent stage, individual or clusters of dislocations propagate with the growth front through the crystal (threading dislocations). Interaction between dislocations causes a systematic reduction in their density by annihilation or fusion reactions [20].

Figures 1(a) and 1(d) show scanning-electron and transmission-electron microscopy (SEM and TEM, respectively) images of a DOI film with a thickness of $0.6 \text{ }\mu\text{m}$, exhibiting individual blocks with a lateral size of $\sim 300 \text{ nm}$. The grain size (mosaic blocks) has increased considerably at a film thickness of $\sim 8 \text{ }\mu\text{m}$ (Figs. 1(b); 1(e)). At this stage, typical grain dimensions are approximately $1 \text{ }\mu\text{m}$ in diameter, and nearly dislocation-free polygonised single crystal areas have been formed. They are bounded by a closed network of surrounding grain boundaries, which has already disintegrated at a thickness of $\sim 34 \text{ }\mu\text{m}$ into short defect bands consisting of clusters of individual dislocations (Figs. 1(c); 1(f)). After this transition from the mosaic crystal structure to a monocrystal, structural improvement proceeds continuously. It was carefully monitored up to a thickness of about 1 mm by dislocation etching and high resolution Raman spectroscopy experiments [21]. Within that millimeter thick heteroepitaxial diamond, the Raman-line width decreased from ~ 10 to 1.86 cm^{-1} and the corresponding dislocation density from $N_{\text{DD}} > 10^{10} \text{ cm}^{-2}$ to $N_{\text{DD}} < 10^8 \text{ cm}^{-2}$. Underpinned by the empirically found relation $N_{\text{DD}} \propto 1/d_{\text{D}}$, DOI layers for detector applications have been henceforward cut/produced from the growth side of thick single-crystal DOI substrates.

The reasons for the uniqueness of Ir(001) surfaces for epitaxial diamond growth as well as a possible explanation and modelling of the nucleation process are described in [22]. M. Schreck et al. introduced and demonstrated a new mechanism called “Ion Bombardment Induced Buried Lateral Growth (IBI-BLG) of diamond on Iridium” as “a novel process, in which the vast majority of isolated epitaxial diamond grains are formed by buried lateral growth induced by intense ion bombardment”.

Figure 1. SEM ((a)-(c)) and corresponding TEM ((d)-(f)) images of the first stages of diamond growth on iridium, i.e. after 0.6 μm , 8 μm , and 34 μm , respectively. Between 8 and 34 μm , the closed polygonized grain boundary network disintegrates into individual dislocations and clusters. Plot (f) is a drawing of remaining defects observed at 34 μm film thickness. Redrawn from [1 and 15].

2.1 Differences in crystal structure of various DG CVD diamond materials

Birefringence imaging using cross polarizers is quite a simple and highly sensitive method to detect defect structures in single-crystal diamond samples. Typical images obtained from the three CVD-diamond types under test are plotted in Figure 2. Each image depicts the full area of the corresponding sample. The samples are plotted with identical length scaling. Differences in illumination and camera sensitivity impede a direct comparison of the contrasts. Distinct bright features indicate the presence of strain fields due to e.g. dislocations, grain boundaries or inclusions, while homogeneous dark areas confirm optically isotropic diamond.

The figure shows from left to right: (a) a SCD sample of $(3.5 \times 3.5 \times 0.1) \text{ mm}^3$, (b,c) two DOI films of $(5 \times 5 \times 0.435)$ and $(7 \times 7 \times 0.075) \text{ mm}^3$, respectively, and (d) one PCD of $(10 \times 10 \times 0.35) \text{ mm}^3$. The SCD reveals a homogeneous structure with some strain, indicated by the diagonal line features on the right side of the sample, and presumably also by the defective left corner area. The bright cross-like figure at bottom left was identified as potentially conductive, isolated bundles of threading dislocations [23, 24]. Both DOI films show fine-grained structure with a characteristic lateral size. The PCD film is dominated by bright and dark areas of significantly larger dimensions. In order to investigate such pattern, a micro-beam experiment with ^{12}C ions was performed at GSI. Mapping of the energy-resolution of a PCD sensor confirmed this texture as isolated single-crystal grains amongst extended defective areas [25]. The RD42 Collaboration explored the columnar growth of PCD and introduced the

‘linear model’ [26], which imply that the CCE increases with the sample thickness (i.e., decreasing defect density at increasing grain size).

Figure 2. Crossed polarizer images of CVD diamond samples: SCD (a), DOI (b,c), PCD (d), recorded with the same optical system at identical magnification. Contrast and brightness were chosen for each sample individually, while the length scale is in all plots identical. The size of the PCD sample (d) is (10 x 10) mm².

2.2 Sensor preparation

The vast majority of charged-particle diamond detectors are of parallel-plate geometry. Their final thickness and shape are defined by the requirements of the physics experiment envisaged. After chemical and mechanical removal of the substrate layers, the remaining diamond sample is sliced and laterally shaped by a pulsed IR laser. Only the slice containing the growth surface was used for the preparation of particle detectors. In contrast to high-energy physics experiments, the large range of particles and energies involved in experimental nuclear physics demand an adequate set of detector thicknesses, which have to be strictly homogeneous in order to ensure position-independent detector response. Worth to bear in mind, plane-parallel thinning of square-centimeter areas below 100 μm is an extremely challenging task.

Figure 3 illustrates the detector preparation steps. In order to link bulk and surface structure to the detector characteristics, optical inspections are routinely performed (birefringence imaging and optical microscopy, left images top to bottom). Prior to the metallization by sputter deposition of 100 – 200 nm thick metal layers, the samples are cleaned and oxidized with wet-chemistry and oxygen plasma [24, 27]. For the use as particle detectors, we mount the metallized films in massive metallic boxes (Fig. 3, right photograph, box foreseen for 16 detector channels). Each channel is wire-bonded to a 50 Ω micro-strip line soldered to a coaxial RF connector (SMA). All samples have been characterized with pad electrodes without guard rings. A minimum spacing of about 1 mm between the rim of the electrodes and the sample edges was chosen in order to avoid short circuits and breakdowns caused by currents flowing across the crystals' side faces.

2.3 Physical parameters of the samples

The parameters of the sensors discussed in this report are listed in Table 1. With the exception of Al electrodes, the contacts have been annealed at 500° C in argon atmosphere. In contrast to DOI or PCD films, which were only mechanically polished, the SCD plates have additionally been treated by reactive ion etching. It was shown [24], that SCD films brazed to the sample holder during abrasive polishing and removed chemically afterwards, remain inevitably with different morphology of the opposite surfaces: a flat terrace-like face of $\sim 1\text{-}2$ nm roughness and a

Figure 3. Detector preparation steps: Defect structures and mechanical stress analysis from birefringence images with sample orientations $0^\circ/45^\circ$ relative to the crossed polarizer filters (a,b). Growth and substrate side of an Al-metallized sample in darkfield (c,d) and brightfield (e,f) microscopy. (g) Quadrant sensor mounted in an Al-detector housing foreseen for 16 detector channels. A 4-fold glass-ceramic aperture (shown above the detector) can be fixed across the diamond to collimate α -particles onto each quadrant.

Table 1. Physical properties of the diamond films used in the following studies.

ID	Sample (year 20--)	$d_D[\mu\text{m}]$	Area [mm^2]	Electrodes [mm]	C_{pad} [pF] (calculated)
1	SC-10B50 (07)	55	3.5 x 3.5	Al dot, 2.7 $\varnothing$	5.3
2	SC-1327 (08)	480	5 x 5	Cr/Au, 4Q	4 x 0.2
3	SC-8B+ (07)	100	3.5 x 3.5	Al, 4Q	4 x 0.7
4	DOI-954 (11)	265	5 x 5	Ti/Pt/Au, 6strips ^{intercon.}	6 x 0.2
5	DOI-AIX401 (15)	75	7 x 7	Al, 4Q	4 x 2.7
6	DOI-AIX401G (15)	105	7 x 7	$^{12}\text{C}/\text{Al}$, 4Q	4 x 1.98
7	DOI-AIX334 (14)	303	5 x 5	Al, 4Q	4 x 0.2
8	DOI-963 (13)	190	7 x 7	Al, 4 x Q	4 x 1.1
9	DOI-886/2 (10)	324	3.5 x 3.5	Ti/Pt/Au, 4Q	4 x 0.2
10	DOI-1078 (13)	435	5 x 5	Al, 4 x Q	4 x 0.2
11	PC-PF350 (05)	350	10 x 10	Cr/Au, dot 8 $\varnothing$	7.3
12	PC-PF210 (05)	210	10 x 10	Al, dot 8 $\varnothing$	12.1
13	DOI-549a (09)	230	3.5 x 3.5	Ti/Pt/Au, 4Q	4 x 0.3
14	DOI-952/2 (11)	303	5 x 5	Al, dot 2.7 mm	0.96
15	SC-491/2 (13)	322	5 x 5	Cr/Au, dot 3mm	1.1

rougher one (~ 6 nm rms) without defined structural features. Differences in surface structure and roughness may result in varying contact properties, and thus, different detector operation at positive and negative bias. Systematic contact studies have been performed by other groups [28, 29]. In our own experiments, we found the clear trend that the surface treatment of the crystals is more crucial for the final detector performance than the choice of the contact metal used as electrode.

Little information is currently available about the chemical purity of present DOI, since nitrogen (N) and boron (B) concentrations have not explicitly been measured. For the growth of most of the DOI samples the hydrogen feed gas was ultra-cleaned by an AgPd purifier. The methane had a purity of 99.9995 %. We estimate the resulting nitrogen concentration in the samples to not more than few ppb. E6 specified for SCD plates N and B below 5 ppb, and for DG PCD films $N \leq 50$ ppb and $B < 10$ ppb.

3 Electronic properties

The basics of the characterization of diamond sensors at GSI have been described previously [4, 5, 18, 19, 24]. The procedures include diamond-signal processing with dedicated front-end electronics (FEE) and the realization of functional assemblies. If not mentioned otherwise, all tests except I-E(V) measurements are performed with β -primed quadrant sensors. Segmented electrodes reduce the detector-channel capacitances, thus improving the Signal-to-Noise (S/N) ratio in timing and spectroscopy applications, enabling in addition the investigation of the macroscopic lateral homogeneity of the detector properties (Sect. 3.3.3).

3.1 Dark conductivity and I-E(V) characteristics

In order to explore both, the dark conductivity of the material and the optimum range of bias voltages for the detector operation, I-E(V) characteristics measurements have been performed at room temperature (RT) using Keithley electrometers. Each sector electrode (on the growth side) was contacted to an individual electrometer with the inputs at floating-zero potential and the high voltage on the solid contact applied to the rear side (substrate side). The setups were placed in a probe station encased in a Faraday cage made of copper-plated pcb-material, with the diamonds flushed with nitrogen in order to avoid parasitic currents due to light, electrical pickup or humidity. Typical I-E(V) curves of all three CVD-diamond types obtained over the years are shown in Figure 4. All sensors had quadrant contacts, with the exception of the two PCD samples (Fig. 4(a)) and the early DOI film (Fig. 4(c)), which consisted of circular dot electrodes (Table 1).

The first observation is that differently pronounced features appear for all diamond types: hysteretic behavior of the data, variation of the dark current between the four sectors, ‘hard’ or ‘soft’ breakdowns at relatively low fields, as well as asymmetries with respect to the bias polarity. The second observation concerns the absolute current values, which differ significantly. While the thinner PCD sample ((a), red data) exhibits about two orders of magnitude higher conductivity compared to SCD (b) and recent DOI sensors (d-f), the thicker one ((b), blue data) is comparable to both types of single-crystal counters. The extremely low current obtained in (c) was, more likely, due to slightly higher nitrogen concentration caused by the lack of a purifier in those days production. Actually, later DOI samples

revealed currents of the order 1-10 pA (Fig. 4 (d, e, f)) along with improved CCE due to reduced dislocation density (Sect. 3.3, [21]). All sensors showed varying degrees of sensitivity to white light. The lowest level was measured for SCD. The red data in Figure 4(f) illustrate the influence of white-light illumination on a high quality DOI film (ID-5, see next paragraphs), which results in current levels increased by several orders of magnitude. This behavior is clearly different from that of SCD devices and has to be investigated systematically in future studies.

Figure 4. IE(V)-characteristics of a PCD (a), a SCD (b), and a first DOI film from 2009 (c). In the bottom row are shown curves of DOI samples produced in 2010 (d), 2014 (e), and 2015 (f), respectively. The red data in (f) (right-hand Y-axis) demonstrate the tremendous sensitivity to white light, observed even for sensors of otherwise rather good detector quality (Sects. 3.2; 3.3)

3.2 Timing Properties

3.2.1 Transient Current Technique (TCT) - experimental issues

TCT is a standard technique used by different research groups to study the transport properties and the internal-field profiles of diamond sensors. Polarization and memory effects destroy the homogeneity of the externally applied electric field due to the gradual buildup of space charge generated by trapping in the sensors [24, 30-33]. Configured as real detectors, the diamond samples are mounted in metallic boxes, as shown in the right-hand image of Fig. 3. Figure 5 depicts the schematic of the TCT assemblies used. They consist of a diamond sensor (represented by the shunt circuit of the detectors capacitance C_D and a current source I_{tr}), a low-impedance low-capacitance diamond broadband amplifier (DBA, $R_i = 50 \Omega$; $C_i \approx 2$ pF) [31], and a 6 GHz Digital Sampling Oscilloscope (DSO) with a sampling rate of 20 GS/s, not included in the figure. The DBA is an inverting single-channel amplifier of gain $G \approx 130$, dedicated to heavy-ion beam monitoring and ToF experiments with diamond sensors at GSI Darmstadt [18, 24, 33].

The tests have been performed with weak radioactive ^{241}Am - α -sources (~ 1 kBq/ 2π of $E_\alpha \sim 5.5$ MeV), inducing a charge $Q_g = 68.4$ fC per α -particle in diamond, thus ensuring ‘small-signal’ measurements ($Q_g \ll C_D \cdot V_b$) in the ‘current mode’ ($RC \ll t_{tr}$) [34], with V_b the bias voltage applied and t_{tr} the corresponding charge-transit time. The

radiation entrance is through the segmented electrode on the growth side serving for both, high-voltage (HV) supply and signal readout. Positive bias enables hole drift (h-drift) and negative bias electron drift (e-drift). In contrast to laser triggered TCT setups, a slight modification of the sensors priming state during measurements has to be accepted.

Figure 5. Equivalent-circuit diagram of the diamond broadband assemblies used for TCT and heavy-ion timing measurements at GSI. (Reprinted from [18]).

3.2.2 Induced, transient-current (TC) measurements

High-quality single-crystal diamond sensors of parallel-plate geometry and homogeneous space charge respond to short-range particles impinging the sensor on arbitrary positions with identical induced TC pulses $I_{e,h}^{tr}(t)$ in the external circuit described by Equation (1) [24, 30]

$$I_{e,h}^{tr}(t) = Q_g \cdot \frac{v_{e,h}^{dr}}{d_D} \cdot e^{\frac{t}{\tau_{eff}} - \frac{t}{\tau_{e,h}}} \quad (1)$$

with Q_g the generated charge, $v_{e,h}^{dr}(E_D)$ the drift velocity of electrons or holes, d_D the sensor thickness, $\tau_{e,h}$ the deep-trapping lifetime of the charge carriers, and τ_{eff} the space-charge related lifetime contribution.

Assuming lifetimes longer than the transit times $t_{e,h}^{tr}$ ($\tau_{e,h} \gg t_{e,h}^{tr}$), the TC pulses are expected to develop flat top signals of amplitudes $I_0(E_D) = Q_g \cdot v(E_D)/d_D$, signal widths correlated to the external field, and due to charge conservation, pulses of equal areas A proportional to the collected charge Q_c at all times ($A \propto Q_c$). Steeply rising and falling edges characterize diamond sensors of negligible space charge and good quality of the contacts – both features supporting fast extraction of the particle-induced charge.

Data analysis is based on TC signals recorded and analyzed with the DSO. Figure 6 shows examples of typical ²⁴¹Am- α -induced pulses in the persistent mode (h-drift and e-drift data on the left and right-hand plots, respectively), obtained from a PCD ID-11 (a,b), two DOI sensors from different production years ID-4 (c,d) and ID-7 (e,f), and a SCD counter ID-2 (g,h). Each set of signals is measured at a constant electric field $|E_D| \geq 1.5$ V/ μ m. The colored histograms on the screens are spectra of the signal-shape parameters, i.e. rise and decay time (yellow and pink), FWHM = transit time (cyan), amplitude (red), and area (green). The width of each spectrum (which is not further discussed in this report) is given by the standard error (σ) of the corresponding shape parameter. Its value is a measure of the quality of both, charge transport and collection properties. The electronic contribution to this error is small (roughly 200 ps). Note that Figure 6 is intended to show general peculiarities of each sensor type. The detailed evaluation and discussion of charge transport and collection parameters follows in Sects. 3.2.3 and 3.3.

The triangular profiles of strongly differing amplitudes and widths measured for the PCD in (a) and (b) vary with the spots where the α -particles hit the sensor surface [5, 24]. The observed scatter confirms the inhomogeneous

structure of this detector material. It is known that, signals from low-quality sensors are often narrower than the signals of high quality devices of comparable thickness. However, an interpretation in terms of huge drift velocities is incorrect. Instead, the signals indicate a quick trapping so that actually no carriers can reach the opposite electrode. Thus, the extraction of correct transport parameters for PCD detectors is inhibited.

Figure 6. Online TC signals obtained at identical time and amplitude scales, with the DSO in persistent mode - h-drift and e-drift signals on the left and right plots, respectively: (a,b) PCD; (c,d) and (e,f) DOI, (g,h) SCD sensors. Values for the detector thickness and the strength of the applied field are given in the plots. Homogeneity of the response is demonstrated by the prominent bands of identical color comprising the most probable events.

The majority of the signals acquired for the SCD sample in both drift modes (g, h) contribute to the formation of intense bands of identical color, indicating the presence of a uniform internal field. According to Eq. (1), the trapezoidal signal shapes of constant amplitudes during charge drift, demonstrate complete charge transition as well as the absence of any significant charge trapping or space charge. Nevertheless, a certain amount of deviating signals is visible also in that case. We attribute them to events occurring near the contact borders [24]. In addition, consistently higher values of the transit times in (h) are a clear indication of lower electron drift velocity. In both drift modes, the signal areas are approximately the same, thus confirming identical efficiencies in the collection of hole and electron charge.

For DOI sensors, the shape of the signals shows a significantly higher scatter as compared to SCD. The latest is particularly obvious in the case of ID-4 (c,d) because of its complex contact geometry and wiring (Table 1). As for the SCD, the signal widths of the electron pulses are wider (d). Eventually, the clear start and stop flanks of the TC signals in (c,d) and (e,f) indicate complete charge drift for the majority of the signals acquired from these samples. This property clearly discriminates between PCD and DOI sensors.

In the following, we present studies performed with early (Fig. 7) and recent (Fig. 8 and Fig. 9) diamond sensors. In Figure 7, a SCD and a PCD sample from 2004 (a, b) and 2005 (e, f), respectively, are compared to a first DOI sensor from 2010 (c, d).

Figure 7. ^{241}Am - α -TC signals of early sensors compared: (a,b) SCD CERN from 2004 (replotted from [30]); (c,d) DOI ID-9 from 2010; (e,f) PCD ID-11 from 2005. The signals of all samples are affected by trapping. The data of the SCD detector show high charge collection, but in addition, the unambiguous signature for the presence of negative space charge (see text).

A clear correlation between transit time (signal width) and external field was obtained for the SCD film [30]. The upper plateau of signals at $\pm E_D$ is characterized by a decrease of hole current (a) and an increase of electron current (b) during the charge transit. That TC behavior, combined with the absence of electron drift signals up to the critical bias voltage at which the holes start to reach the opposite electrode, has been interpreted as the “convoluted effect of negative space charge concentration N_{eff} in the bulk and trapping of charge carriers with lifetime $\tau_{e,h}$.”

The best DOI sensor of that time (ID-9) displayed a fast and a slow decay component of the signal. In addition, the duration of the transits did not correlate in a systematic way with the strength of the applied field. At the same time, the amount of collected holes was rather high, while the electron collection was quite low (Sect. 3.3.1). Moreover, the h-drift signals showed increasing signal width with increasing bias voltage, pretending a decrease of the drift velocity with increasing field. This behavior is not understandable being opposite to the trend of high-quality crystals and typical for PCD detectors. Nonetheless, in contrast to the DOI sample, the present PCD sensor showed very low CCE in both drift modes (Sect. 3.3.1). For this type of diamond sensors, it was frequently observed that signal width and area increased with increasing electric field strength. This fact was indicating improved charge collection, however without reaching real saturation even for the highest fields applied (Sect. 3.3).

In Figure 8, signals of commercial SCD sensors are compared to those of DOI samples produced more recently. Obviously, charge carriers in SCD sensors ((a, b); (e, f); (i, j)) can drift over tens of nanoseconds in low fields, while the drift time in DOI films of similar thickness ((c, d); (g, h); and (k, l), respectively) is typically limited to below 10 ns. Sample DOI ID-4 at h-drift (k) constitutes an exception to this rule. About half as thick as SCD ID-2, it exhibits low-field drift times of the same order.

Figure 8. ^{241}Am - α -TC studies of SCD and recent DOI sensors - h-drift signals on the left and e-drift signals on the right-hand graphs. For comparable detector thickness and intermediate up to highest fields, the signal shapes of both diamond types are similar, thus proving the significant improvement of the properties for DOI crystals. At very low fields, SCD is still superior.

The shape of the signals acquired for thick DOI sensors is dominated by trapping (g, h; k;l); this holds especially for electron drift (h, l). On the contrary, the thin DOI sensor (c, d) behaves quite similar to the thin SCD detector (a, b). None of the data sets displays the features typical for space charge (as seen in Fig. 7 (a,b)). However, we observe pronounced deformation of the transient-current profiles up to intermediate field strengths. It decreased and nearly completely disappeared at voltages equivalent to the typical detector operation bias. For high bias, both types of sensors show approximately rectangular shaped TC signals.

In Figure 9, priming and polarization phenomena have been studied using DOI films ID-7 and ID-5. The response of the 303 μm thick sensor (ID-7, Fig. 9 (a, b) and (c, d)) and of the 75 μm thin device (ID-5, Fig. 9 (e, f)) are shown before and after β -priming. Prior to passivation of traps, strong electric fields are apparently present near the irradiated electrode of ID-7. For both bias polarities, the induced currents vanish after short charge drift, suggesting

Figure 9. DOI ID-7 in the unprimed state (a,b) shows incomplete charge collection and strong polarization indicated by triangular signal shapes and zero-field pulses of opposite sign (red and blue lines), which disappear after β -priming (c, d). For the thin DOI ID-5 (e, f), the influence of the priming procedure appears to be negligible.

significant trapping of both charge carrier types - strongest for electrons. Moreover, zero-field pulses appeared after extended operation in both drift modes (here displayed with opposite polarity in Fig. 9 (a) and (b), red and blue signals, respectively). Conspicuously good results were obtained for h-drift after pumping (c), and even the critical e-drift (d) showed noticeable improvement. Recent thin samples like ID-5, showed little influence of priming as well as only minor differences between h and e-drift signals. This property characterizes good single-crystal sensors, which loose performance by polarization only after heavy radiation damage [24, 35].

In summary, TCT data described in this paragraph showed for the first time intrinsic DOI sensors with TC

profiles appropriate for evaluation of transport parameters for both, electrons and holes (e.g. ID-5 and ID-6). Samples ID-4, ID-7, and ID-9 demonstrated the currently most puzzling issue in the development of DOI sensors, i.e. the much poorer performance of devices thicker than 250 μm , when operated in the e-drift mode. Dedicated studies have been initiated to solve this non-trivial problem, and the data analysis is progressing (see also Sect. 3.4).

3.2.3 Charge-transport parameters $v_{dr}^{h,e}$, $\mu_0^{h,e}$, and $v_{sat}^{h,e}$

For each bias voltage, the FWHM of the average signals and the corresponding standard deviations given by the DSO are used to determine the drift velocities and the measurement errors. We studied the field dependence of the drift velocities $v_{dr}^{h,e}(E_D)$ in the $\langle 100 \rangle$ crystal direction and extracted low-field mobilities $\mu_0^{h,e}(E_D \rightarrow 0)$ and saturation drift velocities $v_{sat}^{h,e}(E_D \rightarrow \infty)$ of each sample according to the procedure illustrated in Figure 10 (red color for holes and blue for electrons). The FWHM of each average TC pulse shown in Fig. 10 (a, b) is taken as a measure of the charge-transit time $t_{tr}^{h,e}(E_D)$ (Fig. 10 (c)) as well as of the drift velocity $v_{dr}^{h,e}(E_D) = \frac{d_D}{FWHM}$ (Fig. 10 (d)). The transport parameters μ_0 and v_{sat} are derived by fitting the acquired $v(E_D)$ data sets in the range $E_D > E_c$ by Eq. (2) [36] (red and blue lines).

Figure 10. TC signals obtained by averaging of several hundred single shots at different $\pm$ HV for (a) holes and (b) electrons drift. Plots of transit times (c) and drift velocities (d) vs. the applied field strength, as derived from the TC curves in (a) and (b). The solid lines in (d) are fits for hole and electron data using Eq.(2).

$$v_{dr}^{h,e}(E_D) = \frac{\mu_0 \cdot E_D}{1 + \frac{\mu_0 \cdot E_D}{v_{sat}}} \quad (2)$$

Because this analysis procedure requires crystals for which a sufficiently high number of carriers reach the opposite electrode, DOI sample ID-9 and both PCD films had to be excluded from this evaluation. The same holds for ID-10 which showed unstable operation when higher voltage were applied, whereas DOI ID-8 has been included, despite of a slightly lower safe field $E_D = \pm 1.3 \text{ V}/\mu\text{m}$.

The initial increase of t_{tr} forming a well-defined maximum as shown in Fig. 10 (c) was routinely obtained for both charge-carrier species at low fields. It was an unexpected behavior, which is not yet understood. It coincides in a systematic way with the appearance of a cusp on the signal tail for a specific bias voltage V_c . The appearance of this feature is interpreted in the literature as the onset of charge arrival to the collecting electrode. The relevant pulses are highlighted in red and blue for holes (left) and electrons drift (right plot), respectively (Figs. 10 (a,b)).

Figure 11 shows $v_{e,h}^{dr}(E_D)$ curves acquired from samples ID 1-8 for fields $|E_D| > |E_c|$; hole velocities are displayed on the left-hand graph (a) and electron velocities on the right-hand graph (b). Homo and heteroepitaxial sensors show similar general trends with respect to the curve shapes, absolute parameter values, fit quality, as well as the larger dispersion and lower drift velocity of the electrons. However, the data points derived for the DOI samples are associated with greater error bars while the quality of the fits (colored lines) is described by $R_h^2 \sim 0.990$ and $R_e^2 \sim 0.959$. The obtained transport parameters are summarized in Figure 12. They show $\sim 30\%$ variation of the low-field mobility values (left axis, red symbols) and $\sim 6\%$ of the mean saturation velocities (right axis, black symbols).

Figure 11. Charge-carrier drift velocities vs. applied field strength of holes (a) and electrons (b), derived for the samples studied in this work.

Figure 12. Transport parameters μ_0 (left axis) and v_{sat} (right axis) derived for 8 different samples.

There was a common agreement in the past that both, PCD and SCD sensors are suitable for particle timing in relativistic heavy-ion experiments. In contrast, the material of choice for single-particle beam monitoring and ToF in MIP experiments was still SCD [18,19,24,31,33]. We repeated heavy-ion ToF experiments with DOI sensors and

obtained similar values for σ_{ToF} (≈ 46 ps), defining an intrinsic time-resolution equal to $\sigma_i \approx 18$ ps. Note that these values are still limited by the bandwidth of the available FEE [31, 33].

3.3 Charge-collection properties at single-carrier drift

^{241}Am - α -spectra were measured in oil-free vacuum of $p \sim 10^{-5}$ mbar for samples ID 1-12 (Table 1), in order to evaluate the $\text{CCE}_{\text{h,e}}$, the corresponding Schubweg in the direction of the electric field $w_{\text{h,e}} < 100 \text{ \AA}$, as well as the energy resolution $\delta E/E_{\text{h,e}}$ of the diamonds. The parameters have been determined for $E_D = \pm 1.5 \text{ V}/\mu\text{m}$ and for the highest possible individual bias. Modified silicon preamplifiers (CSTA2 [37]) and high-resolution spectroscopy electronics were used for the measurements. Weak ($\sim 1 \text{ kBq}$) slightly collimated α -sources irradiating the sensors with spot radii of $\sim 1.5 \text{ mm}$ guaranteed single-particle measurements without distortion of the internal sensor field. The asymmetric spectra obtained for the main ^{241}Am - α -line of $E_\alpha = 5.486 \text{ MeV}$ (Fig. 13) were fitted by Gaussians in an energy range comprising the peak and the high-energy part of the line. This procedure provides results that are more systematic by excluding the non-Gaussian low-energy tail of the ^{241}Am - α -spectrum, which is a superposition of unsolved α -lines of low intensity originating from α -transitions to excited states of ^{237}Np , signals of inelastically scattered α -particles within and outside the probe, and particles impinging close to the edge of the electrodes [24].

3.3.1 ^{241}Am - α -spectra, Charge-Collection Efficiency (CCE), and relative energy resolution ($\delta E/E$)

In this part of our study, we consider the general shape of the pulse-height distributions acquired under alpha-particle irradiation at optimized electric-field conditions, which correspond to the highest possible bias at best S/N ratio. Spectra from samples representing different states of material development are summarized in Figure 13. The h- and e-drift distributions of an early DOI sample (ID-9, $324 \mu\text{m}$, 2010) are directly compared with a PCD (ID-12, $210 \mu\text{m}$, 2005) and a SCD film (ID-2, $480 \mu\text{m}$, 2008) in (a) and (b), respectively. The graphs (c) and (d) show again the same SCD counter now in direct comparison with ID-9 and two later DOI sensors ID-7 (2014, $303 \mu\text{m}$) and ID-5 (2015, $75 \mu\text{m}$).

It was shown [34] that, the energy resolution $\delta E/E$ of diamond sensors measured with monoenergetic charged particles is the most meaningful parameter to test the homogeneity of the crystal structure with simple detector characterization methods. Accordingly, $\delta E/E$ is expected strongly related to the Schubweg and to the charge-carrier lifetime (Sects. 3.3.2, 3.4). In both drift modes, the PCD data are dominated by very broad energy distributions, which show an average collection efficiency below 50% and $\delta E/E$ of about 40%. Their shape is a direct consequence of the heterogeneity, typical for polycrystalline films. As mentioned in Sect. 3.2.2, the behavior of the early DOI film ID-9 is still not well understood. It was for the first time that a high CCE and promising energy resolution were measured for h-drift in DOI crystals - in fact, $\text{CCE}_{\text{h}} \sim 92.5\%$ and $\delta E/E_{\text{h}} \sim 2\%$. Both values were unexpectedly good in light of the strange shape of the corresponding TC profiles, even at highest bias voltage (Fig. 7(c)). On the other hand, the broad energy spectrum at e-drift (Fig. 13(b), black line) is not in contradiction to the almost equal-narrow triangular TC signals of different amplitudes presented in Fig. 7(d). It shows a rather uniform distribution starting at $\sim 1 \text{ MeV}$ and extending even beyond the energy of the ^{241}Am - α -line. In contrast, the SCD sensor shows the

characteristics known from spectroscopic-grade diamonds supplied by [2], i.e. $CCE_{e,h} > 96\%$ and an energy width of $\delta E/E_{e,h} \sim 0.4 - 0.1\%$ [5, 24, 34]. While the performance of the thin DOI device ID-5 resembles that of SCD ID-2 ($CCE_h \sim 95\%$; $\delta E/E_h \sim 0.4\%$), the thicker detector ID-7 is affected by trapping and polarization, again particularly at e-drift (compare TCT in Fig. 9 (c,d)).

Figure 13. ^{241}Am - α -spectra acquired for hole (a,c) and electron (b,d) drift mode from different CVD-diamond samples. The field strength that yielded the optimum results was slightly different for each sample.

In summary, we conclude that improved growth processes led to enhanced performance of DOI detectors. The $CCE_h \sim 94\%$ and $\delta E/E_h \sim 0.6\%$ values achieved for the collection of holes in thin state-of-the-art DOI samples (like ID-5) is close to the standards required for spectroscopic grade sensors. In these devices, also the e-drift showed promising results ($CCE_e \sim 78\%$ and $\delta E/E_e \sim 4.7\%$). However, the striking difference between the h- and e-drift data of thicker DOI samples discovered by TCT, is now been confirmed also with integrating spectroscopy electronics. That means that the ‘missing-electrons’ puzzle remains still non understood, since all particle-induced charge which has been trapped but released again within the shaping time of $\sim 1 \mu\text{s}$ has been collected, and contributes to the average signal.

3.3.2 CCE and Schubweg $w < 100$

The electric-field dependence of the CCE measured for short-range particles stopped within a distance, which is small compared to the detector thickness (single-carrier drift), is described by the Hecht formalism [38] Eq. (4)

$$CCE(E_D) = \frac{w_x}{d_D} \left(1 - e^{-\frac{d_D}{w_x}} \right) \quad (4)$$

with $w_x(E_D)$ the average drift distance (also called ‘Schubweg’) of charge carriers in field direction away from the electrode hit by the α -particles. The thickness of the diamond sensors is d_D and the strength of the applied field E_D .

For particle detectors, the course of $CCE_{h,e}(E_D)$ allows to define the optimum bias-voltage range, for which the charge collection has reached saturation and the energy resolution $\delta E/E_{h,e}$ shows a minimum. Figure 14 depicts plateau characteristics obtained by plotting the peak values of the main ^{241}Am - α -line acquired from Gauss fits as a function of the electric field. Steep rise of CCE_h to levels higher than 98% was measured at low fields with both SCD sensors.

Figure 14. CCE at hole (+ E_D) and electron (- E_D) drift mode, as a function of the electric field in $\langle 100 \rangle$ direction. DOI samples are compared to a PCD film and two SCD sensors.

However, current DOI samples require roughly 1.5 times higher bias voltages than SCD sensors to reach the saturation level. The increase in electron collection efficiency $CCE_e(E_D)$ is slower, typically reaching maximum values well below 100%. Moreover, extremely broad-shaped α -spectra at e-drift prevented any further analysis of these profiles (e.g. ID-9, Fig. 13 (b), black data).

The Schubweg values have been extracted from a calculated Hecht graph (Fig. 15, black line) used as lookup table. Figure 16 summarizes the results of the above studies. The charge-collection parameters measured at $\pm 1.5 \text{ V}/\mu\text{m}$ and at maximum field $\pm E_{\text{max}}$ for holes (red dots) and electrons drift (blue squares) have been displayed in solid and open symbols, respectively. The dashed arrows in (a) and (b) highlight the huge differences of the charge-collection properties between e- and h- drift data that are found for several samples. The relationship given by Eq. (4) generally implies substantial uncertainties in the derivation of w_x -values from CCE values close to 100% - even in the case of low variance (see e.g. SCD ID 2, 3 with uncertainties of $\sim 10^{-3}$). This effect is enhanced in the rare case of samples in which high CCE values are accompanied by a poor energy resolution (e.g. DOI ID-4 and ID-9).

Noteworthy is first a significant improvement of $\delta E/E_e$ at maximum negative fields, leading to higher absolute

values and reduced scatter in CCE_e data, e.g. of ID 6 and 7. Second, the energy resolution of ID-5 in both drift modes as well as ID 4-10 at h-drift are comparable to SCD, even at $1.5 \text{ V}/\mu\text{m}$. Consistent with the explanation that attributes charge-drift limitation to the presence of grain boundaries, the PCD devices showed strongly reduced CCE resulting in Schubweg values, equivalent to only 20% of the detector thickness.

Figure 15. Plot of the Hecht relationship given by Eq. (4) (black line). The experimental CCE values measured at $\pm 1.5 \text{ V}/\mu\text{m}$ are placed on the theoretical curve. The big black diamond symbol highlights the CCE value of 63.2%, which corresponds to the special case of a Schubweg identical to the detector thickness.

Figure 16. Charge-collection parameters vs. sample ID: CCE (a), energy resolution $\delta E/E$ (b), Schubweg $w_{<100>}$ and $w_{<100>}/d_D$ (c) and (d) plots, respectively. (see text)

In summary, SCD ID 1-3 showed the best spectroscopic results in both drift modes and DOI films ID 5-7 for hole drift. In agreement with the TCT data, the charge-collection properties of DOI samples (ID-4 to 10) are ranking between the homoepitaxial diamonds (ID 1-3) and the PCD sensors (ID-11, 12), with the majority performing closer to the SCD devices. Both single-crystal types reveal worse properties at e-drift – the highest differences being observed for DOI diamond. Nevertheless, we could measure for most of the heteroepitaxial samples Schubweg values $w_{<100>} > d_D$ at both drift modes (Figs. 16 (c, d)).

The energy resolution derived for the sensors ID 1-12 has been correlated to the corresponding CCE and w -values in the Figures 17(a) and 17(b), respectively. The data are displayed in colored solid (h) and open symbols (e) for measurements at $E_D = \pm 1.5 \text{ V}/\mu\text{m}$, and in black figures for $E_D = \pm E_{\text{max}}$. We observe rough correlations of $\delta E/E$ with both quantities, yielding a monotonically slow decrease with the CCE and a steep exponential decrease with the Schubweg. As already mentioned before, the e-drift data of ID-4 (open green rhombus, bottom-left corner of the plot) deviate strongly from the general trend - a behavior, which is still not understood.

Figure 17. Correlations between energy resolution $\delta E/E$ and CCE (a) or Schubweg $w_{<100>}$ (b). Large errors in (a) are typical for samples dominated by a high density of structural defects or/and measurements far below saturation conditions.

3.3.3 Macroscopic lateral homogeneity

The position-independent response of a sensor is an important issue for the detector instrumentation in accelerator laboratories. A variety of beam monitoring applications are based on the balance between beam-induced currents in sector devices facilitating beam localization and control. We tested therefore the macroscopic lateral homogeneity of the detector response with quadrant-detectors at maximum bias. Figure 18 shows spectroscopic results of detectors from 2014 and 2015 (ID-7 and ID-5, dot and triangle symbols, respectively) along with a SCD from 2008 (ID-2, rhombi). The CCE and $\delta E/E$ values obtained in each sector are plotted versus the quadrant ID.

The striking observations in this study are on one hand the congruence of the SCD ID-2 results (found in all tests superior), and on the other, the conformity of the DOI quadrant response, which vary only within a few percent in worst case. The following results have been ascertained: in the h-drift mode, the DOI CCE_h increased by $\sim 5\%$ while $\delta E/E_h$ decreased by $\sim 50\%$. In the initially much worse e-drift mode, the DOI CCE_e increased by 15% (absolute) along with a decrease in $\delta E/E_e$ by a factor of ~ 6 . To which extent these improvements are favored by the low

thickness of the samples compared (ID-2, 480 μm ; ID-7, 303 μm ; ID-5, 75 μm) could not explicitly investigated yet.

Figure 18. CCE (left axis, solid symbols) and energy resolution (right axis, open symbols) extracted from ^{241}Am - α -spectra measured with quadrant sensors. The corresponding mean values derived for each sector are plotted against the individual quadrant number.

3.4 Deep-trapping lifetime $\tau_{DT}^{h,e}$

The standard data treatment for the estimation of lifetimes is based on the Hecht formalism in its time-dependent version given in Equation (5)

$$CCE^{h,e}(E_D) = \frac{\tau_{DT}^{h,e}}{t(E_D)_{tr}^{h,e}} \left(1 - e^{-\frac{t(E_D)_{tr}^{h,e}}{\tau_{DT}^{h,e}}} \right) \quad (5)$$

In Figure 19, the $CCE(E_D)$ measured with spectroscopy electronics is plotted as a function of the transit time $t_{tr}(E_D)$ taken from the TCT experiments. The graph comprises hole data of the currently highest quality DOI samples (ID 4, 5, and 6) and of two SCD samples (ID-1 and ID-2) fitted by Eq. (5). The quality of the current electron data does

Figure 19. CCE values of three DOI samples (ID-4,5,6) and of two SCD films (ID-1,2) plotted vs. the transit times obtained at the corresponding electric field E_D . The parameters τ_{DT} extracted by fitting Eq. (5) to the data are given in ns. The pronounced scatter in data observed for sample ID-4 could be reduced in more recent devices.

not facilitate a similar analysis. For a direct comparison, we plotted the measured data points, the fit curves, and the parameter deep trapping lifetime τ_{DT}^h as extracted from the fits in the figure.

With coefficients of determination in the range $0.757 < R^2 < 0.991$, the quality of the fits is not particularly high. In the case of the SCD sensor ID-1, the low R^2 value can be attributed to the marginal deviation of the CCE values from unity. In the case of the DOI sensors, we see a more fundamental reason: the basic assumption of a constant lifetime may not be justified. Specifically for sample ID-4, the data indicate a higher lifetime for lower transit time, i.e. for higher field strength. Closer analysis of this aspect is being performed in an ongoing study [39]. Nevertheless, lifetimes longer than the charge transit times are derived for all samples presented in Figure 19, which is consistent with values of the Schubweg higher than the detector thickness (Sect. 3.3).

3.5 Charge-collection properties at dual-carrier drift – The response to minimum-ionizing particles

For MIP detection, sensor thicknesses of about 500 μm of DG PCD and at least 300 μm of SCD are recommended. A MIP-sensor test campaign was started with a set of samples of various diamond types and thickness (ID-3, 6, 7, 11, 14, and 15). The measurements comprised investigations with ^{90}Sr - β -particles of $E_\beta > 1$ MeV in the laboratory, and beam tests with protons of $p = 1.7$ GeV/c from the Cooler Synchrotron COSY in Jülich. The ^{90}Sr -setup used at GSI has been described elsewhere [24]. In short, a ^{90}Sr source, the test diamond, and a small-area scintillator detector of 20 mm thickness are mounted in line at the smallest possible mutual distance. The scintillator thickness is chosen to stop all the electrons of the two overlapping ^{90}Sr - β -spectra including the maximum β -energy of $E_{\beta\text{-max}} = 2.234$ MeV. The output signal of the scintillator sensor is used to trigger coincident electrons traversing the diamond, and in addition, to set a high-energy condition in the scintillator pulse-height spectrum to discriminate electrons with $E_\beta > 1$ MeV (i.e. almost MIP energy for β -particles). Due to several experimental reasons (not explicitly addressed here), the precise calibration of the mean collected charge Q_c and of the CCE had to be postponed. To get an impression of the potential of DOI sensors for MIP detection, the results were normalized to the best SCD sensor used in this study (ID-15, 322 μm [40]). Similar to the experiments performed at single-carrier drift, we evaluated data obtained at $E_D = \pm 1.5$ V/ μm and at highest possible individual bias. Based on published pair creation energies ϵ_{Dia} ranging between 13.1 for PCD and 11.4 eV/e-h for best homoepitaxial materials [41], we applied $\epsilon_{\text{SCD}} = 12.86$ [24] and $\epsilon_{\text{DOI}} = \epsilon_{\text{PCD}} = 13.1$ eV/e-h, taken into account the current stage of DOI developments. Using dE/dx values from [42], charge-generation rates $N_{\text{SCD}} = 43.1$ and $N_{\text{PCD/DOI}} = 42.3$ e/ μm have been computed. Figures 20 (a-c) present a summary of the results.

We obtained energy-loss spectra showing approximately the shape of Landau distributions (Fig. 20(a)) with different most probable (MP) values and widths, which depend primarily on diamond quality and thickness. In order to compare the average charge Q_c collected for each sample, the very high-energy tail of the Q_c distributions had to be truncated, losing about 5 % of the highest energy events. The thickness-corrected truncated-mean values normalized to ID-15 are displayed in Figures 20 (b, c) (left axes), for positive and negative bias (red and blue symbols, respectively). The same graphs show the number of charge carriers collected per micrometer (right axes, green and magenta symbols). Parameters obtained at ± 1.5 V/ μm are displayed in solid and E_{max} data in open symbols. By

increase of the bias voltage to $E_{\max}$, the DOI values measured at + HV improve roughly by 11%, indicating saturation of Q_c at $E_D > 1.5 \text{ V}/\mu\text{m}$. The other types of diamond sensors remained stable, confirming real (SCD) and pseudo saturation (PCD) at $E_D = \pm 1.5 \text{ V}/\mu\text{m}$.

Figure 20. (a) Energy-loss spectra of ^{90}Sr -electrons of $E_\beta > 1 \text{ MeV}$ measured with various samples of different quality and thickness. (b) and (c) Normalized collected charge distributions vs. the sample ID - data obtained at + HV are displayed in (b) and those at - HV in (c), respectively. Solid symbols represent the values measured at $\pm 1.5 \text{ V}/\mu\text{m}$ and open symbols data at maximum bias. The right-hand axes of the graphs display the corresponding numbers of charge carriers collected per micrometer. Data for sample DOI ID-14 at - HV have not been measured.

Finally, we compared the charge induced by 1.7 GeV/c protons with that obtained from ^{90}Sr -electrons of $E_\beta > 1$ MeV in the laboratory. These nuclear particles undergo similar specific energy loss in matter, both being slightly higher ionizing than real MIPs. This fact entails systematic uncertainties in dE/dx values utilized for the data evaluation, which cause deviations up to $\sim 20\%$ in the final data. However, the mean (MEAN) and the most probable (MP) values extracted from the corresponding Landau distributions (Fig. 20(a)), show quite a good agreement between ^{90}Sr -electrons and high-energy protons, though the data were measured with completely different setups.

Figure 21. Summary of the MIP results acquired for different diamond sensors: proton data in open and β -data in solid symbols. The MEAN and MP values of the collected-charge distributions obtained at dual-carrier drift are plotted vs. the sample ID.

Summary and conclusions

The objective of this work was the evaluation of intrinsic heteroepitaxial diamond with the potential to be synthesized in dimensions appropriate for the development of large-area detector systems, that can keep up in performance with small homoepitaxial diamond devices. As a first important conclusion, we classify the thin, highest quality DOI sensors according to their detector performance as approaching the commercial SCD devices as far as h-drift is concerned. However, e-drift is still significantly worse, particularly for thick sensors. The poorer CCE for electrons might simply indicate a higher density of electron traps. Moreover, one has to consider higher capture cross sections and a greater depth of electron traps, so that they cannot release the carriers any more within the time window of $\sim 1 \mu\text{s}$ integration time used for the measurements. We suggest positively charged (ionized) nitrogen atoms as one plausible candidate contributing to such an efficient trapping of electrons. The potential role of dislocations is currently under investigation.

The incomplete collection of electrons evolves into a serious drawback in the dual-carrier drift mode, which is typical for the detection of swift particles that traverse the whole detector volume. Nonetheless, DOI detectors are clearly outperforming standard polycrystalline counters. At $E_D \geq \pm 1.5 \text{ V}/\mu\text{m}$, the majority of the samples showed Schubweg values considerably larger than the detector thickness (actually, by factors $f \sim 2$ to 27), whereas the SCD sensors yielded unrivalled numbers $f \sim 45$. For DOI samples with high values of the Schubweg, an energy resolution in the range below 1% was measured at both drift modes. The breakthrough in charge-transport and collection

properties was achieved by growing the DG DOI layers on iridium substrates up to final thicknesses in the millimeter range.

As a final point, some of us investigated the important radiation hardness of DOI sensors [43]. Preliminary results obtained with 1 MeV reactor neutrons up to fluencies of $F \sim 10^{16} \text{ cm}^{-2}$ showed that, DOI sensors follow the damage curves of SCD and PCD counters up to $F \sim 10^{15} \text{ cm}^{-2}$. Between 10^{15} - 10^{16} cm^{-2} , the CCE decreased steeper by 15%, approaching again the level of the established diamond sensors in the damage saturation regime well beyond $F = 1 \times 10^{16} \text{ cm}^{-2}$.

For the future work, we see the highest priority in the exploration of the origins of the strong electron trapping, moreover, the search for strategies to avoid it will be pursued. At the same time, measures have to be taken to improve the synthesis and the post processing (like mechanical and plasma polishing) and to gain deeper understanding in the role of contacts for DOI sensors. We expect a final quality and size of single-crystal DOI materials enabling large-area detector applications in ion spectroscopy as well as MIP timing and tracking experiments. In the long run, single crystal DOI wafers could have the potential to eliminate a crucial bottleneck for the development and the subsequent industrial production of high power electronic devices.

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References

- [1] M. Schreck (2014). Single Crystal Diamond Growth on Iridium. In V.K. Sarin (Editor-in Chief) & C.E. Nebel (Vol.Ed.), *Comprehensive Hard Materials* (pp. 269–304). Elsevier.
- [2] *Element Six Ltd.*, Kings Ride Park, Ascot, Berkshire, SL5 8BP, UK. The denominations electronic-grade, detector-grade, spectroscopic-grade CVD diamond, have been introduced by this company.
- [3] The RD42 Collaboration (2007). Development of diamond tracking detectors for high luminosity experiments at LHC. *Report CERN-LHCC-2007-002*.
- [4] M. Pomorski, E. Berdermann, A. Caragheorgheopol, M. Ciobanu, M. Kiš, A. Martemyanov, C. Nebel, P. Moritz. Development of single-crystal CVD-diamond detectors for spectroscopy and timing. *Phys. Status Solidi (a)* 203(12) (2006) 3152–3160.
- [5] E. Berdermann, M. Pomorski, W. de Boer, M. Ciobanu, S. Dunst, C. Grah, M. Kiš, W. Koenig, W. Lange, W. Lohmann, R. Lovrinčić, P. Moritz, J. Morse, S. Mueller, A. Pucci, M. Schreck, S. Rahman, M. Träger. Diamond detectors for hadron physics research. *Diamond & Related Materials* 19 (2010) 358–367.
- [6] W. de Boer, J. Bol, A. Furgeri, S. Müller, C. Sander, E. Berdermann, M. Pomorski, M. Huhtinen. Radiation hardness of diamond and silicon sensors compared. *Phys. Status Solidi (a)* 204(9) (2007) 3004–3010.
- [7] C. Grah, K. Afanaciev, I. Emeliantchik, U. Harder, H. Henschel, A. Ignatenko. Polycrystalline CVD Diamonds for the Beam Calorimeter of the ILC. (2009) *IEEE Transactions on Nuclear Science*, 56, pp. 462–467.

- [8] P. Bergonzo, D. Tromson, C. Descamps, H. Hamrita, C. Mer, N Tranchant, M Nesladek. Improving diamond detectors: A device case. *Diamond & Related Materials* 16 (4-7) (2007) 1038-1043.
- [9] J. Morse, B. Solar, H. Graafsma. Diamond X-ray beam-position monitoring using signal readout at the synchrotron radiofrequency. *Journal of Synchrotron Radiation* 17(4) (2010) 456–464.
- [10] E. Berdermann (2014). Diamond for Particle and Photon Detection in Extreme Conditions. In V.K. Sarin (Editor-in-Chief) & C.E. Nebel (Vol.Ed.), *Comprehensive Hard Materials* (pp. 407–467). Elsevier.
- [11] K. Ohtsuka, K. Suzuki, A. Sawabe, and T. Inuzuka. Epitaxial Growth of Diamond on Iridium. *Japanese Journal of Applied Physics* 35 (1996) L1072–L1074.
- [12] M. Schreck, F. Hörmann, H. Roll, J. K. N. Lindner, & B. Stritzker. Diamond nucleation on iridium buffer layers and subsequent textured growth: a route for the realization of single-crystal diamond films. *Appl. Phys. Lett.* 78 (2001) 192-194.
- [13] S. Gsell, T. Bauer, J. Goldfuß, M. Schreck & B. Stritzker. A route to diamond wafers by epitaxial deposition on silicon via iridium / yttria stabilized zirconia buffer layers. *Appl. Phys. Lett.* 84 (2004) 4541-4543.
- [14] B. Golding, C. Bednarski-Meinke, Z. Dai. Diamond heteroepitaxy: pattern formation and mechanisms. *Diamond & Related Materials* 13 (4-8) (2004) 545-551.
- [15] R. Brescia. Transmission electron microscopy studies and simulations of heteroepitaxial diamond nucleation on Ir/YSZ/Si. *PhD thesis* (2013) *University of Augsburg*.
- [16] A. Stolz, M. Behravan, M. Regmi & B. Golding. Heteroepitaxial diamond detectors for heavy ion beam tracking. *Diamond & Related Materials* 15 (2006) 807–810.
- [17] CARAT & ADAMAS, EC FP7 Projects *HadronPhysics2* & *HadronPhysics3*, Proj.Nrs.: RII3-CT-22743 & 283286.
- [18] E. Berdermann and M. Ciobanu (2009).CVD-Diamond Detectors for Experiments with Hadrons, Nuclei, and Atoms. In R. S. Sussmann (Ed.) *CVD diamond for electronic devices and sensors* (pp. 227–256). John Wiley & Sons Ltd.
- [19] E. Berdermann, W. de Boer, M. Ciobanu, S. Dunst, M. Kiš, W. Koenig, P. Moritz, J. Morse, S. Muller, C. Nociforo, M. Pomorski, M. Schreck, M.S. Rahman, M. Traeger, H. Weick. CVD Diamond Detectors - R&D Status and New Results. *XLVIII Int. Winter Meeting on Nuclear Physics - BORMIO2010*. PoS (BORMIO2010) 040.
- [20] A. E. Romanov, W. Pompe, G. Beltz, J. S. Speck. Modeling of Threading Dislocation Density Reduction in Heteroepitaxial Layers I. Geometry and Crystallography. *Phys. Status Solidi (B)* 198 (1996) 599-613.
- [21] C. Stehl, M. Fischer, S. Gsell, E. Berdermann, M. S. Rahman, M. Traeger, O. Klein, and M. Schreck. Efficiency of dislocation density reduction during heteroepitaxial growth of diamond for detector applications. *Appl. Phys. Lett.* 103 (2013) 151905.
- [22] M. Schreck, S. Gsell, R. Brescia, M. Fischer. Ion bombardment induced buried lateral growth: the key mechanism for the synthesis of single crystal diamond wafers. *Scientific Reports* 7 (2017) 44462, doi:10.1038.
- [23] H. Pinto and R. Jones. Theory of the birefringence due to dislocations in single crystal CVD diamond. *Journal of Physics: Condensed Matter* Vol. 21, Nr. 36 (2009) 364220 (7pp).
- [24] M. Pomorski. Electronic Properties of single crystal CVD Diamond and its suitability for Particle Detection in Hadron Physics Experiments. *PhD thesis* (2008) *Wolfgang Goethe University of Frankfurt*.
- [25] E. Berdermann, K. Blasche, P. Moritz, H. Stelzer, B. Voss. The use of CVD-diamond for heavy-ion detection. *Diamond & Related Materials* 10 (2001) 1770-1777.
- [26] The RD42 Collaboration (2003). The development of diamond tracking detectors for the LHC. *Nuclear Instruments and Methods in Physics Research A*, 514, Issues 1-3 (2003) 79-86.
- [27] B. Kindler, W. Hartmann, A. Hübner, B. Lommel, J. Steiner, V. Yakusheva. Coating of diamonds for detector application. *J. Radioanal. Nucl. Chem.*, Vol. 303, No 1 (2014) Springer, doi:10.1007/s10967-014-3912-5.
- [28] A. Galbiati, S. Lynn, K. Oliver, F. Schirru, T. Nowak, B. Marczevska, J. A. Dueñas, R. Berjillos, I. Martel, L. Lavergne. Performance of Monocrystalline Diamond Radiation Detectors Fabricated Using TiW, Cr/Au and a Novel Ohmic DLC/Pt/Au-Electrical Contact. (2009) *IEEE Transactions on Nuclear Science*, 56 (4), pp. 1863-1874.

- [29] W. Deferme, G. Tanasa, J. Amir, K. Haenen, M. Nesládek, C. F. J. Flipse. The role of (sub)-surface oxygen on the surface electronic structure of hydrogen terminated (100) CVD diamond. *Diamond & Related Materials* 15 (4-8) (2006) 687–69.
- [30] H. Pernegger, S. Roe, P. Weilhammer, V. Eremin, H. Fraiss-Kolbl, E. Griesmayer, H. Kagan, S. Schnetzer, R. Stone, W. Trischuk, D. Twitchen and A. Whitehead. Measurement of Charge Carrier Properties in Single Crystal CVD Diamond using the Transient-Current Technique. *Journal of Applied Physics*, 97, 073704–1-9 (2005).
- [31] P. Moritz, E. Berdermann, K. Blasche, H. Stelzer, B. Voss. *Diamond & Related Materials* 10 (2001) 1765–1769.
- [32] J. Isberg, M. Gabrysch, A. Tajani, D. J. Twitchen. *Semicond. Sci. Technol.* 21 (2006) 1193–1195.
- [33] M. Ciobanu, E. Berdermann, N. Herrmann, K. D. Hildenbrand, M. Kis, W. Koenig, J. Pietraszko, M. Pomorski, M. Rebisz-Pomorska, A. Schuttauf. In-Beam Diamond Start Detectors. (2011) *IEEE Transactions on Nuclear Science*, 58 (4), pp. 2073-2083.
- [34] E. Berdermann, A. Caragheorgheopol, M. Ciobanu, M. Pomorski, A. Pullia, S. Riboldi, M. Traeger, H. Weick. Ion spectroscopy – A diamond characterization tool. *Diamond & Related Materials* 17 (2008) 1159-1163.
- [35] Guthoff, M., Afanaciev, K., Dabrowski, A., De Boer, W., Lange, W., Lohmann, W., Stickland, D., *Nuclear Instruments and Methods in Physics Research A* 730 (2013) 168-173.
- [36] C. Jacoboni, C. Canali, G. Ottaviani, and A. A. Quaranta. A review of some charge transport properties of silicon. *Solid State Electronics* 20 (1977) pp. 77-89.
- [37] CSTA2 Charge-Sensitive-Timing Amplifiers, Coarse Gain ~ 20 mV/MeVSi; Eq. Noise ≈ 1.1 keVSi, *TU Darmstadt*
- [38] K. Hecht. Zum Mechanismus des lichtelektrischen Primärstromes in isolierenden Kristallen. *Zeitschrift für Physik* 77 (1932) 235-246.
- [39] M. Mayr, to be published.
- [40] Courtesy of the HADES Collaboration
- [41] M. Gabrysch. Charge Transport in Single-crystalline CVD diamond. *PhD Thesis* (2010) *University of Uppsala*
- [42] *ESTAR*-Stopping Power and Range Tables for Electrons.
- [43] S. Sciortino, S. Lagomarsino, M. Bellini, V. Cindro, C. Corsi, K. Kanxheri, et al. Radiation hardness of three-dimensional sensors fabricated on different CVD diamond materials, to be published.