

Systematic study of the normal and pumped state of high efficiency diamond particle detectors grown by chemical vapor deposition

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The efficiency and charge collection distance (CCD) of nuclear particle detectors based on high quality diamond films grown by chemical vapor deposition (CVD) have been systematically studied as a function of the methane content in the growth gas mixture and for varying film thickness. The effects of preirradiation with β particles (pumping) have been thoroughly studied. The results fully support a recently proposed model [Marinelli *et al.*, *Appl. Phys. Lett.* **75**, 3216 (1999)] discussing the role of in-grain defects and grain boundaries in determining the charge collection spectra of CVD diamond films both in the normal and in the pumped state. The model allows us to quantitatively explain the dependence of CCD and efficiency on film thickness, giving a microscopic picture of the effects of preirradiation with ionizing radiation in CVD diamond films. The highest average CCD obtained is 145 μm in a 160 μm thick detector (corresponding to about 50% average efficiency), while the maximum value (about 70% efficiency) is close to 370 μm . In addition, CCD is shown to be higher than film thickness and to monotonically increase with thickness, indicating margins for further improvements. © 2001 American Institute of Physics. [DOI: 10.1063/1.1332805]

INTRODUCTION

With the forthcoming advent of next generation particle accelerators, the need has arisen to replace silicon-based particle detectors, since they will not be able to sustain the high projected particle fluxes without rapid deterioration. Diamond-based detectors, because of the extremely high radiation hardness of diamond, offer an attractive alternative. In addition, diamond presents high carrier mobility, a high energy gap resulting in a very low number of free carriers, and a very high breakdown field, thus in principle making low noise, fast diamond based detectors feasible.

Because of its cost, moderate size, and lack of standardization, natural diamond is not suitable for such an application, nor is high-pressure synthesized diamond. The possibility of realizing nuclear detectors based on synthetic diamond films grown by chemical vapor deposition (CVD) is therefore actively studied in many research institutes, commercial diamond producers, and particle accelerators plants (see e.g., the RD42 activity at CERN, Ref. 1).

However, as particle detection requires high drift lengths of the carriers produced by the ionizing particle, being therefore highly sensitive to film quality, both the presence of defects and the polycrystalline nature of CVD diamond films constitute a severe limitation for this as well as for many other applications.

The priming, or pumping, effect is well known and widely used to increase CVD diamond detector perfor-

mances. It consists of a preirradiation by ionizing radiation. The increased efficiency is believed to be due to a saturation of deep traps. However, it has not been analyzed in much detail so far. The efficiency behavior of many samples before and after pumping was experimentally studied,² without however separating the effects of gas composition and thickness and without correlating the effects to the film characteristics. In the literature on the subject an analysis is particularly missing in the relationships between the macroscopic effects of priming on the detectors' performance and its microscopic effects. Such analysis must take into account the particular structure of CVD diamond films, due to their polycrystalline nature and to the columnar nature of CVD diamond growth.

A first step in this direction was recently³ made by performing a Monte Carlo simulation of pumping effects in a high quality CVD diamond film. The change in position and shape of the charge collection spectrum after irradiation were successfully explained in terms of the different behavior of in-grain defects and grain boundaries under irradiation, and their different distribution along the detector thickness due to the columnar growth of CVD diamond. Here we analyze the behavior of a wide set of samples in the framework of this model, spanning the two main parameters affecting charge collection distance (CCD), i.e., the methane content in the plasma and the film thickness. In particular, due to the different distribution of in-grain defects and grain boundaries, changing the detector thickness allows us to separate their contribution and to clarify their role in the detection process. The results support the model proposed in Ref. 3.

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TABLE I. Sample thickness and CH₄ concentration in the growth gas mixture for the CVD diamond investigated in the present work.

Sample	Set No. 1					Set No. 2			
	A	B	C	D	E	F	B	G	H
CH ₄ (%)	0.6	1.0	1.4	1.8	2.2	1.0	1.0	1.0	1.0
Thickness (μm)	30	43	44	50	53	21	43	102	161

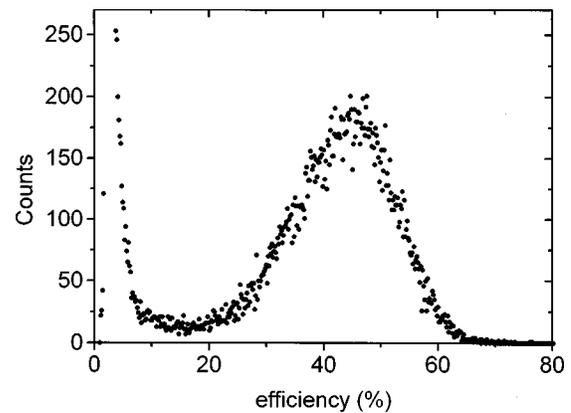
EXPERIMENT

Two sets of CVD diamond films have been deposited^{4,5} on silicon substrates in a microwave tubular reactor, modified to improve film quality.⁶ Diamond nucleation was obtained with the conventional scratching procedure.⁷ A CH₄-H₂ gas mixture was used during the deposition process, the substrate temperature, and the gas pressure being fixed to 750 °C and 110 mbar, respectively. The microwave power was 600 W. The growth parameters which were varied to realize the two sets of samples, namely the CH₄ concentration and the sample thickness, are reported in Table I for all samples.

In the first set of films (set 1) the methane concentration was varied from 0.6% to 2.2%, keeping the film thickness substantially constant and close to 40 μm . In the second set of films (set 2) the CH₄ content was fixed to 1%, while the thickness changed covering the range from about 20 μm to more than 150 μm . Film thickness was measured with a digital thickness meter, and the growth rate was estimated to be 0.9 $\mu\text{m}/\text{h}$ for all samples belonging to Set No. 2, while a monotonic increase from 0.7 to 1.1 $\mu\text{m}/\text{h}$ was observed for Set No. 1, according to the changes of the CH₄ content in the gas mixture.

Particle detectors were then realized by thermally evaporating a circular Au contact about 7 mm² in size and 100 nm thick as the top electrode, while Ag paste was used as the silicon backing contact. The detector response to 5.5 MeV α particles was studied using a ²⁴¹Am source. The irradiation was carried out through a pinhole, in the normal direction to the sample surface. The detector output has been connected, through a charge preamplifier and a shaping amplifier with a 2 μs shaping time, to a multichannel analyzer. A typical collection spectrum of our samples is reported in Fig. 1. In our experience, contact configurations others than the ones we used (e.g., Ti/Au or Cr/Au top contacts, free standing films with Au contacts on both sides) make no difference in these measurements.

We choose α particles rather than, e.g., ⁹⁰Sr β particles as the radiation probe because minimum ionizing particles, like ⁹⁰Sr β particles, would not deposit sufficient energy in our thinner films to allow a reasonably accurate measurement of efficiency and CCD. In addition, ²⁴¹Am α particles create charges only within a depth of 15 μm from the growth surface of the films, so that the effects of in-grain defects (homogeneously distributed) and grain boundaries (concentrated at the diamond-substrate interface)³ are better separated. A drawback of the high ionization produced by α particles is the occurrence of polarization effects, causing a systematic decrease of the detector response during the mea-

FIG. 1. Typical α -particle collection spectrum of our films in the pumped state.

surement. In our case this effect was minimized reducing the fluence of α particles to a few counts per second. Spectra collected immediately after applying the electric field and after 5 min (our measurement time) differed by less than 10%. In addition, samples turn back with time (or by applying reverse bias) to the “virgin” state, in the sense that their spectra revert to the ones initially measured on the same sample. The reproducibility of efficiency and CCD values extracted from spectra taken in this virgin state for a given sample is within a few percent.

All charge collection spectra have been measured at room temperature under positive bias (i.e., the growth surface electrode was positively biased, the back contact being grounded). Each measurement took 5 min. Samples were measured both in the as-grown state and after preirradiation with ⁹⁰Sr β particles for the time (typically about 60 h) necessary to drive the film in a fully pumped state, as determined by the charge collection spectrum, which was unaffected by further irradiation. The total radiation dose absorbed by the samples is about 3 krad. Charge collection spectra in the pumped state were measured within 24 h from completion of the pumping process. Measurements repeated on the same sample after several days proved that the pumped state remains stable over at least a week provided, as in our case, that the sample is not exposed to strong light sources, normal ambient light resulting in no measurable depumping on this time scale.

In all cases the whole film has been utilized to build the detector, i.e., without removing, as is normally done in order to increase the detector performance (see e.g., Ref. 1), the low quality layer close to the substrate interface.

Set 1 was realized to study whether the higher growth rates obtainable using more carbon-rich plasma are compatible with the production of high efficiency detectors. A decrease of the detectors' performance with increasing carbon concentration was observed for several gas mixtures,^{2,8} but only detectors in the unpumped state were considered in these studies. It is clearly of great importance to know whether the pumping process can overcome this problem, since the time needed to grow diamond samples thick enough to be used as nuclear detectors is extremely long using the CVD technique.

Set 2 was instead prepared to better clarify the role of grain boundaries and in-grain defects in the detection process of CVD diamond before and after the pumping process, also in view of the simple model recently proposed³ to explain the effects of pumping. The film thickness is varied from about 20 μm (i.e., just above the penetration depth of 5.5 MeV α particles) to about 160 μm .

RESULTS AND DISCUSSION

The main parameters assessing the performance of particle detectors are the CCD and the efficiency, defined as follows for a standard experimental setup in which a parallel plate detector is used. An electron-hole pair created by an ionizing particle in a parallel plate detector of thickness (i.e., electrode spacing) L , induces^{9,10} in the external circuit a charge $q_c = ex/L$, x being the total distance the electron and hole move apart under the action of the applied electric field. The CCD is the average distance electrons and holes would drift apart, and is given by

$$\delta = (\mu_e + \mu_h)\tau E, \quad (1)$$

where μ_e, μ_h are the electron and hole mobilities, respectively, τ is the mobility weighted lifetime of electrons and holes, and E is the applied electric field. The efficiency is the ratio of the collected charge Q_c to the total charge Q_0 generated by the ionizing particle

$$\eta = Q_c / Q_0. \quad (2)$$

The link between η and δ can be deduced from the Hecht theory¹¹

$$\eta = \frac{\delta}{L} \left[1 - \frac{\delta}{4G} (1 - e^{-(2G/\delta)}) (1 + e^{2(G-L)/\delta}) \right], \quad (3)$$

G being the penetration depth in L of the particles to be detected [in Eq. (3) we assumed $\mu_e = \mu_h$]. The lifetime τ , and therefore δ and η , are limited by the presence of trapping defects (impurities and/or grain boundaries), so that the collection efficiency is strongly correlated with crystal quality. Strictly speaking, Eq. (3) could not be used for CVD diamond detectors since it is derived for a uniform material, however it provides a useful and widely used estimate of the CCD.

In Fig. 2 the behavior of the average efficiency of the detectors of set 1 is shown as a function of the methane content in the plasma, both in the normal and in the primed state. The applied voltage is 68 V, so that the electric field is about 15 kV/cm (see below). A negative correlation can be seen in both cases, showing the progressive reduction of the quality of the films as the carbon content in the growth mixture, and therefore the growth rate, is increased. This confirms the results of Ref. 2, which anyway did not include the primed state behavior and where no indication of the thickness of the various films was given. It has been demonstrated¹² that the decrease in the detectors' efficiency is accompanied by a moderate widening of the Raman peak and by a relatively more marked increase of the photoluminescence background peaked at about 3000 cm^{-1} , and is therefore strictly connected to sample quality.

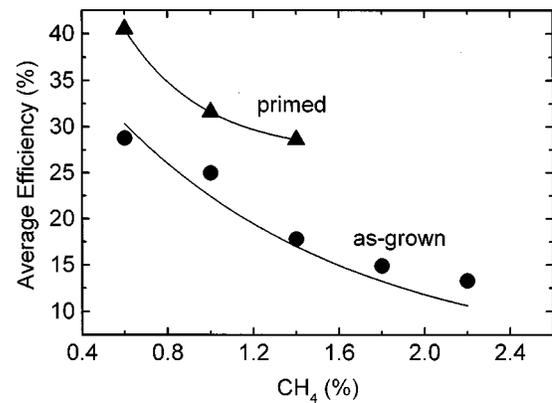


FIG. 2. Average efficiency vs methane concentration in the growth gas mixture for the detectors of set 1, in both the as-grown (circles) and pumped (triangles) states. Lines are guides to the eye.

Another important disadvantage in using high methane concentrations is the reduced ability of the detectors to withstand high applied electric fields, especially in the pumped state. In particular, efficiency measurements of films grown at high methane concentrations ($>1.4\%$) in the pumped state were not possible because of the low electric breakdown threshold observed in these films.

In addition, the CCD versus CH_4 plot in Fig. 3 shows that the CCD value slightly decreases with methane concentration. We point out that all detectors show well defined peaks in the charge collection spectra, both in the as-grown and in the pumped state.

Some considerations are necessary to correctly analyze the results shown in Figs. 2 and 3. In principle, all samples belonging to set 1 were supposed to have the same thickness (40 μm), with only the CH_4 content changed. The increase in growth rate with CH_4 concentration, however, lead to thickness increasing from 30 μm (sample A) to 53 μm (sample E), as reported in Table I. This could partly obscure the conclusions drawn upon these results. However, although the thicknesses of films belonging to set 1 are not all equal to the intended 40 μm value, it turns out that the efficiency and CCD values would not change significantly if all films were 40 μm thick, so that the previous conclusions are substan-

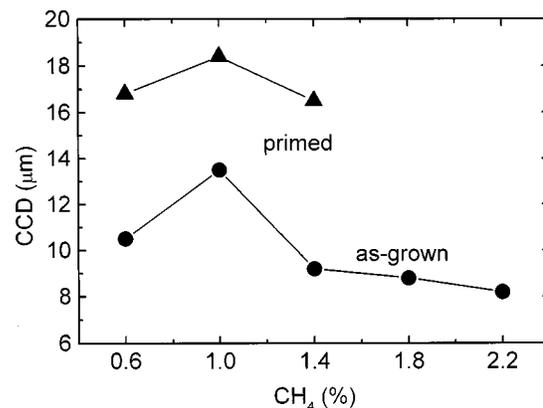


FIG. 3. Average charge collection distance vs methane concentration in the growth gas mixture for the detectors of set 1, in both the as-grown (circles) and pumped (triangles) states.

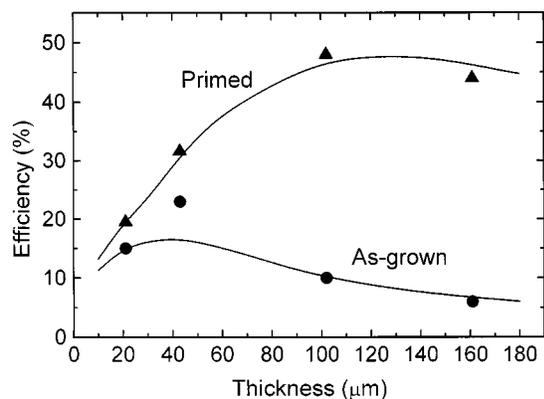


FIG. 4. Average efficiency vs film thickness for the detectors of set 2, in both the as-grown (circles) and pumped (triangles) states. Lines are simulations based on the model described.

tially correct. The reason for this is that as the film thickness varies from 30 μm (sample A) to 53 μm (sample E), two effects of opposite sign must be considered, namely (a) being the applied voltage fixed to 68 V, the thicker films (higher CH_4 concentration) were measured under a lower electric field E , leading to η and CCD values decreased with respect to those of a 40 μm film; (b) their higher thickness leads to higher efficiency values (see Fig. 4) and therefore to η , and CCD values increased with respect to those of a 40 μm film (the opposite holds for thinner films). These corrections can be calculated from the η versus E (see e.g., Ref. 3, Fig. 2) and η versus thickness (see Fig. 4) curves, and it turns out that in this thickness and electric field region, they are both rather small (less than 10%–15%) and almost exactly cancel each other. Values reported in Figs. 2 and 3 therefore represent very well the behavior of films of strictly equal thickness (40 μm) whose quality is changed by varying the methane concentration in the growth mixture.

From Figs. 2 and 3 we see that for set 1 there is only a moderate difference between the CCD and efficiency values in the unprimed and primed state. This is due to the relatively low thickness of these detectors, leading to a grain size close to the mean free path of carriers due to in-grain defects. In Ref. 3 the defects leading to a limitation of the CCD were divided into in-grain defects and grain boundaries, the former being homogeneously distributed and saturated by the pumping process, the latter strongly concentrated toward the substrate side because of the columnar nature of CVD diamond growth, and unaffected by preirradiation. It was shown that under these assumptions it was possible to reproduce by Monte Carlo simulations the α -particle collection spectra of a high efficiency CVD diamond detector. It is then clear that when the grain size is comparable or smaller than the mean free path due to in-grain defects, only a moderate effect is to be expected from the pumping process, since grain boundaries are the main obstacle to high CCD values even in the as-grown state. As the film thickness and grain size increase, however, the saturation of in-grain defects should lead to substantial improvements in the efficiency and CCD of the detectors according to this model. This was the reason which prompted us to grow the samples of set 2, in which the

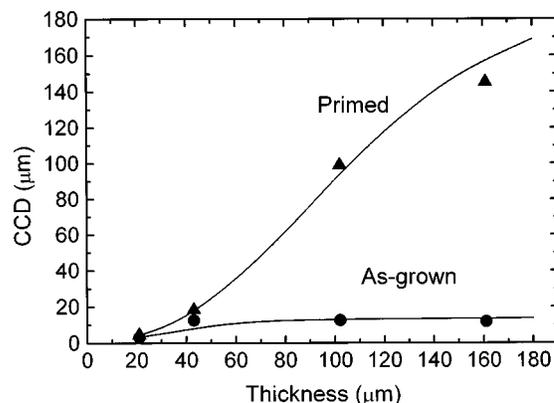


FIG. 5. Average charge collection distance vs film thickness for the detectors of set 2, in both the as-grown (circles) and pumped (triangles) states. Lines are simulations based on the model described.

growth conditions were kept constant, with only the growth time being changed in order to obtain films only differing in their thickness.

Figure 4 shows that indeed the efficiency versus thickness behavior of our detectors confirms the above reported expectations, a very high efficiency increase taking place when the thickness exceeds 40–50 μm . The curves relative to the as-grown and pumped states show a different behavior. In the former case the efficiency initially increases with thickness and then, for $d > 40 \mu\text{m}$ decreases roughly following a $1/d$ law, while in the latter case the efficiency steadily increases up to a thickness of 100 μm , and then slightly decreases. The applied electric field is 15 kV/cm for all films.

The results of Fig. 4 are better explained after looking at Fig. 5, where the dependence of the CCD on the detectors' thickness is plotted both in the normal and the pumped state. In the normal state the CCD value is very small for the 21 μm thick film, while for the other films ($d > 40 \mu\text{m}$) it is substantially independent from thickness, with a value close to 12 μm . This behavior is expected in the framework of the assumption made in Ref. 3 that defects limiting the collection length can be divided into in-grain defects and grain boundaries, which are distributed differently because of the columnar growth of CVD diamond. For very low thickness values the microgranularity of the substrate interface layer prevents significant drift lengths of the carriers, so that the CCD is very low. As the film thickness increases, so do grain dimensions, and the free path of carriers sharply increases until eventually grain dimensions are so large that grain boundaries play a minor role. In this case the CCD is limited by the substantially homogeneous distribution of in-grain defects and the curve of Fig. 5 flattens out. Such a view is confirmed by the behavior after pumping, which in Ref. 3 is proposed to saturate in-grain defects only. For films of low thickness, grain boundaries play a major role in limiting the drift lengths and pumping has therefore a very limited effect. However, eventually the average grain dimension becomes larger than about 12 μm , which is the mean free path due to in-grain defects (Fig. 4), and hence the saturation of such defects becomes important. As a consequence the pumped

curve instead of flattening out continues to increase, reflecting the continuously increasing grain dimensions. Also this curve is clearly expected to eventually saturate if the pumping process does not completely neutralize in grain defects, but no clear signal of such a saturation is observed from our data up to a film thickness of 160 μm , so that margins for even higher CCD values can be foreseen. We notice that the normal state and pumped curve depart when the film thickness is about 40 μm , and since grain dimensions are typically 1/3–1/4 of the film thickness this agrees well with the 12 μm CCD value.

For our thicker film (161 μm) the CCD value corresponding to the average 44% efficiency is 145 μm , while the same film shows a maximum efficiency close to 70%, which corresponds to a CCD of 370 μm . The record high CCD value reported so far¹ is 280 μm , measured on 1 mm thick film produced by De Beers. This result cannot be directly compared with our values, since it was obtained with β particles, which penetrate the whole detector and therefore also sample the lower quality region close to the substrate side. It is difficult to determine the correction factor, but it can be estimated that in sufficiently thick samples, the CCD estimated using α particles is twice that measured using β particles. However, this disadvantage is at least partly compensated in Ref. 1 by the removal of about half of the detector thickness from the lower quality substrate layer and by the extremely high thickness of the detector, almost ten times higher than our thicker film. Also, our CCD value is higher than film thickness, and therefore an increase in thickness is likely to lead to higher CCDs.

Coming back to Fig. 4, its interpretation is now straightforward. The initial efficiency increase in the normal state is due to grain dimension reaching values comparable to the carriers mean free path due to in-grain defects (about 12 μm), and the following decrease takes place when grain dimensions exceed this length so that in-grain defects limit the CCD. In this case the CCD remains constant and η decreases with thickness according to Eq. (3). On the other hand, in the pumped state the efficiency almost monotonically increases because the increase in CCD compensates thickness variation in Eq. (3). Eventually, of course, the efficiency is expected to reach a limiting value. This limiting value should in principle increase by removing the low quality substrate layer of the detectors.

Since the general trends shown in Figs. 4 and 5 are well in agreement with our model, we tried to quantitatively compare them to the results of a Monte Carlo simulation based on the model. The calculated η versus thickness and CCD versus thickness curves are shown as full lines in Figs. 4 and 5, respectively. It appears that the simulated curves match the experimental data closely enough, with the only exception being the efficiency value of the 43 μm thick film, which is underestimated by our simulation. Nevertheless, considering that the model is clearly a simplified one, this agreement seems quite satisfactory. The general trends observed for η and the CCD are clearly related to the different behavior and distribution of in-grain defects and grain boundaries. Within this general concept, a more detailed analysis of features like, e.g., the number and type of in-

grain defects, the grain boundaries distribution and the different behavior of electrons and holes would naturally improve quantitative agreement with the data. However, for such an analysis to be meaningful, probably greater experimental evidence on CVD diamond physics would be required than is presently available. To calculate the simulated curves the distribution of the grain boundaries (unaffected by pumping) and the mean drift distance of carriers before trapping due to in-grain defects in both the as-grown (l_0) and pumped (l_p) states are required. Assuming an exponential density³ of grain boundaries $D(x) = A \exp(-x/b)$ to describe the columnar growth, with x the distance from the film–substrate interface, only four parameters are necessary to obtain all the curves in Figs. 4 and 5. The values we adopted for these parameters are $A = 5.3 \mu\text{m}^{-1}$, $b = 11 \mu\text{m}$, $l_0 = 10.3 \mu\text{m}$, and $l_p = 175 \mu\text{m}$. The reasonable values of these parameters constitute more evidence in favor of the proposed model. In particular, from these numbers it turns out that after pumping the concentration of active in-grain traps decreases by more than 1 order of magnitude, confirming the effectiveness of preirradiation in improving CVD diamond particle detectors quality.

CONCLUSIONS

We have presented a systematic study of the performances of particle detectors built from high quality CVD diamond films. The influence of both the growth conditions (leading to films of different quality) and of the film thickness has been carefully investigated. The increase in methane content in the growth gas mixture leads to a decrease in the detector's efficiency and in the electric field that can be applied to the films, while a higher thickness leads to higher efficiency and CCD values. Our best detector (about 160 μm thick) has an average 44% efficiency, with a maximum value close to 70%, corresponding to CCD values of 145 and 370 μm , respectively.

Particular attention has been given to the pumping effect and its influence on the detectors' performance. The results are analyzed in the framework of a recently proposed model³ taking into account the columnar growth of CVD diamond, in which in-grain defects and grain boundaries are assumed to behave differently after the pumping process. Experimental data fully support our model, which allows us to quantitatively reproduce the observed results, thus giving grounds to the proposed microscopic interpretation of the pumping process in polycrystalline CVD diamond. As a result, the successful interpretation of film's performance gives a firm basis to the expectation of even higher CCD values to be obtained by scaling up film thickness.

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