

# Diamond in solid state electronics

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**Abstract.** Data on the physical properties of diamond as a solid state electronics material are briefly discussed with particular emphasis placed on synthetic crystals and plasma-precipitated films.

## 1. Introduction

Since the early times of human civilization, diamond has attracted a lot of attention. Isaac Newton was interested in its optical properties [1]. It was discovered in the 19th century that diamond is a modification of elementary carbon. In 1952 Custers [2] found semiconducting diamonds among crystals from South African mines. Papers describing the phenomena related to generation and transport of nonequilibrium charge carriers (NCC) were published later [3, 4]. Diamond is one of the most simple homopolar crystals, its close relatives are single crystals of germanium, silicon, and ‘gray tin’ ( $\alpha$ -Sn). By this time, the electronic spectrum of diamond has been studied in detail [3, 5, 6]. Carbon atoms in diamond are very strongly bound to one another, and the solubility of impurities in equilibrium, except nitrogen and boron, is negligible.

The issue of diamond applications in solid-state electronics emerged in the middle of the 1960s owing to the progress in the technology of growing diamond single crystals. Diamond crystals have been popular with researchers working in the fundamental solid state physics for a long time. Moreover, it was discovered fairly unexpectedly in 1956 [7] that diamond films can be grown under conditions that can be realized relatively easily, namely, without the combination of extremely high pressures and high temperatures (HPHT

technique). Recent publications containing very interesting data on the crystal structure of cubic diamond† [6] indicate that, from the viewpoint of its applications in electronics, materials based on diamond can be classified with several groups, whose characteristics are listed in Table 1.

It is known that natural diamonds are characterized by considerable diversity in their individual properties. Since natural diamond crystals inevitably contain impurities, first of all nitrogen, which is present not only in the form of single atoms, but also in various clusters and inclusions, they should be carefully sorted prior to machining them. Diamond crystals grown by the HPHT technique are sometimes superior to natural diamonds in terms of structural regularity and purity [6]. But since this technique demands sophisticated and expensive apparatus, we cannot be certain at present (1996) that there is much prospect of them being extensively applied in electronics.

Diamond films grown under metastable conditions by the chemical vapor deposition (CVD) technique attracted a lot of attention of both researchers and technologists in the last decade [6, 7]. Presently diamond films can be grown on various substrates in facilities where plasma is generated through ionization on surfaces of hot metal wires, by microwave discharge, or even by a jet of flame. The film area is only limited by the dimensions of a facility. Beside genuine diamond films, many publications describe diamond-like carbon (DLC) films, which are very interesting as wear-resistant coatings.

In this brief review, we will not discuss properties of diamond-like films, primarily because the mobility of non-equilibrium charge carriers in them is lower by several orders of magnitude than that in diamond single crystals.

The differences among the properties of five groups of diamonds accessible to both experimenters and technologists are demonstrated by Tables 2–4, which look like old geographical maps with a lot of ‘white spots’ on them. Before discussing the prospect of diamond applications in electronics

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† We will not discuss the properties of hexagonal diamond (lonsdaleite), which has been studied insufficiently.

**Table 1.** Characteristics of diamond materials.

Type of diamond	Size of crystals, films, ceramics	Impurities, defects	Mobilities of electrons and holes	Lifetime of nonequilibrium charge carriers
Natural diamond	Crystals with dimensions of about 5 mm are not expensive, but hard to machine	Most common impurity is nitrogen. Type IIb crystals (semiconducting) are very rare. Impurities are distributed nonuniformly	Highest values are $2400 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $2100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	No higher than $10^{-8} \text{ s}$
Synthetic single crystals	Crystals with dimensions of about 6 mm may be grown reproducibly. There are reports on very large crystals. The cost is very high	Impurity distribution in crystals doped during growth is nonuniform. Structure is very perfect sometimes	Up to $100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	
Epitaxial diamond films	Only films on oriented diamond substrates have been grown, hence the area is limited	Films doped with boron have been described	$100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	
Polycrystalline diamond films	Grown on various substrates (Si, metals). Areas of 50 to $90 \text{ cm}^2$	Films with <i>n</i> -type conductivity doped with phosphorus have been described	$50 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	
Diamond ceramic	Dimensions are controlled by those of the press chamber	Porosity of 1 to 8%. Sometimes inclusions of conducting phases are present		
Diamond-like carbon films	Areas are unlimited			Mobility of carriers is low, energy gap width is less than 5.5 eV

**Table 2.** Parameters of shallow impurities in diamond (theory).

Parameters	Acceptors		Donors				
	B (subst.)	Li (interst.)	Na (interst.)	C	P (subst.)	As (subst.)	Sb (subst.)
Activation energy, eV	0.37–0.09	0.1			0.09 0.03		
Energy levels, eV	$E_v + 0.37$	$E_c - 0.1$					
Efficiency	> 10%	> 1%				Low	Low
Charge carrier mobility, $\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	> 1000	$\approx 1000$			80		
Efficiency in diamond films	> 10%						
Notes	Natural diamond of type IIb	Electrically inactive when present as a substitutional impurity (?)		Vanishes at $T > 800^\circ \text{C}$	Low solubility		

in more detail, we should notice that the overoptimism of some authors, who sometimes called diamond a ‘semiconductor of the 21st century’ in recent years, is replaced with more realistic opinions. For example, A Collins [9] concluded that diamond would be used only in quite specific fields of electronics (see below).

In what follows, we will discuss the basic parameters of various types of diamond relevant only to their applications in electronics.

## 2. Natural diamond

Electrophysical properties of natural diamonds in all their diversity have been described in detail in several books [3, 5, 6]. In many countries, including Russia, the yield of natural diamonds has reached high levels. Techniques for selecting

**Table 3.** Ion-implanted lithium as a donor.

Ion energy, keV	Dose, $10^{15} \text{ cm}^{-2}$	Hall mobility, $\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	Sheet density, $10^{10} \text{ cm}^{-2}$	Sheet conductivity, $10^{-6} \Omega^{-1}$
110	4	$870 \pm 10 \%$	5.2	7.3
310	3	650	2.4	2.6
350	3	1070	2.3	3.8
110	25	1030	2.3	3.8
80	6	400	1.1	7.1

diamonds most interesting for both physical experiments and technical applications have been developed [4, 10, 11]. Electronic and optical phenomena in diamond were described in the monograph [4]. It has been generally accepted lately that the most efficient technique is the high-spatial-resolution

**Table 4.** Applications of diamond-based devices.

Technical application	Diamond-based material	Need of implantation	Comments
Wear-resistant coatings	Diamond and diamond-like carbon films		Fast growing application
Heat sinks	Natural and synthetic diamonds, films, ceramics		Widely used
Diodes, transistors	Synthetic diamonds, epitaxial films	+	Preliminary results
Particle detectors and dosimeters	Natural and synthetic diamonds, films in prospect	+	Used successfully
Impact-avalanche transit-time diodes	Highly perfect diamond crystals are needed	+	
UV light emitters	Single-crystal diamonds and diamond films	+	
IR light detectors	All types	+	
Thermistors	Small synthetic diamond single crystals		

spectroscopy of luminescence excited by a scanning, sharply focused electron beam. The technique allows one to plot a map of the distribution of native impurities or those introduced, for example, by ion implantation [10]. The price of natural diamonds with dimensions of two to four millimeters is not high, therefore they can be used in instruments such as dosimeters [11] or light detectors for recording ultraviolet solar radiation. In recent years, A M Zaitsev and his colleagues [13] in Minsk used the ion implantation technique to produce arrays of unipolar transistors and diodes on natural-diamond plates. It turned out, however, that some elements of such arrays were disabled by microdefects present in the initial material. It is known that for many years similar defects in GaAs single crystals drastically reduced the output of good planar integrated circuits in microelectronic industry [14].

Nonetheless, many researchers successfully continued their experiments with natural crystals, including South African type IIb diamonds.

Some Russian authors [10] suggest that applied research using samples of natural diamond should continue in the next few years. Besides, it follows from the recent progress in the growth of epitaxial diamond films that almost all of such films are grown on oriented substrates prepared of natural diamonds.

### 3. Epitaxial and polycrystalline diamond films

#### 3.1 Epitaxial films

Diamond films can be fabricated only under the conditions where the diamond growth rate is much higher than that of graphite. This is feasible in the presence of an etching agent that does not react with diamond, but evaporates graphite nuclei. It was demonstrated that atomic hydrogen can serve as such an agent.

In 1977, Alekseenko et al. [15] published their results on the growth of semiconducting epitaxial diamond layers on insulating diamond substrates. A boron-containing compound was added to the plasma during the deposition of the diamond film. The resulting layers were *p*-type, as was demonstrated by Hall measurements. The hole mobility was within  $80 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ , which was ascribed to scattering of carriers by the free surface of the film and by the interface between the film and the insulating

substrate. In later experiments, similar results were obtained many times.

The collection of papers [6] contains reports on three different techniques of diamond film deposition, namely, the use of chemical transport reactions, electric activation, and thermal activation of the gaseous phase. The common characteristic of these techniques is the composition of the gaseous phase, which includes both starting materials and etching agents: hydrocarbon species (molecules, ions, and radicals) and hydrogen (both atomic and molecular). The authors studied the kinetics of epitaxial film growth on various faces of natural diamonds and the growth kinetics of polycrystalline films on substrates of materials such as Mo, Ni, and Si. The substrate temperature was varied between 600 and 1200 °C, and the pressure in the gaseous phase was within 1 atm. The maximum of the film growth rate shifted with pressure toward higher temperatures. The atomic hydrogen not only evaporated non-diamond modifications of carbon, but also created favourable conditions for the formation of nucleation centers (adsorbed clusters) with tetrahedral coordination of atoms. In Ref. [6], *p*-type films doped with boron and *n*-type films doped with phosphorus were described.

At concentrations smaller than 1 at. %, boron is located predominantly at the sites of the diamond lattice, and at higher contents, at tetrahedral interstitial positions. The minimal resistivity of *p*-type films was  $10^{-3} \Omega \text{ cm}$ , and that of *n*-type films was  $10 \Omega \text{ cm}$ . The resistivity of similar undoped films was  $10^{13} \Omega \text{ cm}$  at 300 K.

Semiconducting epitaxial films [8] were also grown on substrates fabricated of synthetic diamonds by Fujimori et al. [16].

From the optimistic viewpoint, it can be anticipated that, all the difficulties resolved, synthetic diamond single crystals will be fabricated by growing films of required thicknesses on a synthesized diamond. Then the film grown will be separated from the substrate in order to re-use it in fabricating another film.

#### 3.2 Polycrystalline diamond films

Presently the possibility of growing polycrystalline diamond films on various substrates, such as silicon, is an established fact.

As to the parameters of transport and other nonequilibrium electronic processes in diamond films, specifically the

life time of nonequilibrium charge carriers, spectra of cathodoluminescence and their relation to electrically active impurities, little is known about them. Hiraki [17] reported about intense cathodoluminescence in the UV range (at 238 nm, i.e.,  $h\nu \simeq 5.2$  eV) in *p*-type polycrystalline diamond films. The authors wrote about their intention to manufacture diamond–cubic boron nitride (BN) heterojunctions with the aim to build a first diamond light-emitting diode for the UV range.

It is obvious that in the immediate future diamond films grown on various substrates will be indispensable for many applications. Let us recall that polycrystalline silicon films have become the best material for solar batteries and other applications because, as was demonstrated experimentally, interfaces between microcrystals might be transparent for nonequilibrium electrons in the processes of their transport and recombination.

#### 4. Diamond ceramics

The progress in the modern planar microelectronics depends largely on the availability of substrates, which should combine several properties, namely, a high mechanical strength at small thicknesses, stability of their dimensions at temperatures used for burning-in and soldering contacts, and high thermal conductivity. At present, the most popular substrates are glasses, quartz, sapphire, and ceramics based on aluminum oxide and beryllium oxide.

But even the beryllium oxide has thermal conductivity insufficient for efficient heat-sinking in modern miniature electronic devices. In this connection, experiments were undertaken in the Odessa State University (Ukraine) and at the Institute of Superhard Materials (Kiev, Ukraine) with a view to manufacture a diamond-based ceramic. The material that has been manufactured by this time is called OKMAL. Rotner and Rotner [18] gave a brief description of its fabrication technology, including heating to 2000 °C under a hydrostatic pressure of 7–8 GPa. The diamond-based ceramic is a three-component system consisting of diamond, a conducting phase, and pores filled with air. The porosity is 2–8%. Interesting results were obtained in experiments with laser radiation acting locally on ceramics in regimes when conducting paths of prescribed configuration and resistance (stable at temperatures used at subsequent stages of device production) were produced. The nature of such paths is still to be established. It is highly probable that they contain both amorphous carbon and nanocrystals (clusters) with diamond and graphite structures.

#### 5. Electrically active impurities in diamond. Doping and activation techniques

Those experimenters who have studied wide-bandgap semiconductors know how many difficulties and unexpected effects are caused by deep impurity centers trapping charge carriers. Diamond has a variety of such centers, which are classified in the reviews by Vavilov and Gippius [4] and Davis [6, 19]. Investigations of this problem are in progress, but we will discuss the details of such investigations only in some cases. As in other wide-bandgap semiconductors, only those samples where either the concentration of deep impurities is minimal or only deep impurities of one type dominate [20] are suitable for most practical purposes.

As early as 1979, convincing evidence was obtained in favor of the relation between relatively shallow acceptors ( $E_v - 0.37$  eV) in diamond and substitutional boron impurities [3]. At present, it is generally accepted, especially after efficient techniques for numerical computer calculations of energy spectra of local centers based on ‘cluster’ models were developed, that substitutional boron impurities are stable acceptors. There is a lot of evidence that introduction of boron in the process of diamond growth or by other methods, such as ion implantation, allows one to dope diamond with boron to a concentration at which hopping conductivity dominates [21, 22]. Thus, the problem of manufacturing *p*-type diamond doped with boron is as good as resolved, given that the effect of deeper impurities is not too large. Presently there is no available information on acceptors with lower ionization energies.

#### 5.1 Donors

It is known [3, 6] that substitutional nitrogen atoms, as well as pairs of such atoms, form very deep donor levels ( $E_c - 1.5$  eV,  $E_c - 4$  eV), which cannot be efficiently employed in electronic devices.

The leading specialists in manufacturing integrated circuits assume that the problem of fabricating *n*-type diamond, which has not been resolved so far, is one of the major obstacles on the way to commercial manufacture of diamond-based devices.

A considerable number of publications based on experimental results report on the fabrication of *n*-type diamond (see, for example, Ref. [23]). Most of these reports were reviewed in Refs [5, 6]. These papers, however, except the publications about doping diamond with lithium [24, 25], do not contain any data about large mobilities of electrons, which should have been expected in the case of electron transport via the conduction band.

According to the theoretical estimates by Kajihara et al. [26], Li, Na, and P atoms should generate donor levels in diamond. In the case of nitrogen, the energy level lies deep in the band gap [27]. The calculations of Ref. [26] are based on the available data on the band structure of diamond and molecular quantum dynamics techniques applied to atomic configurations of an impurity and surrounding carbon atoms in ‘supercells’ containing up to 64 atoms. According to these calculations, a phosphorus atom should substitute for a carbon atom, whereas Li and Na atoms should be interstitial donors.

In all the three cases, the donor levels must be shallow. The solubilities of these impurities, however, are low, hence their concentrations in diamond obtained using thermal diffusion could only be very low. According to Ref. [26], the kinetic capture in the process of growth or ion implantation should be used instead [21, 22]. Ion implantation was used in many experiments. Phosphorus atoms should be most stable at carbon sites [26], i.e., they should largely penetrate via vacancy diffusion, which is characterized by a high activation energy. According to calculations, the activation energy for diffusion of interstitial Li and Na is 0.85 and 1.6 eV, respectively, i.e., interstitial lithium atoms should migrate relatively easily. Experiments on ion implantation of Na is likely to be reported in the not-too-distant future. It is should be remembered that diffusional doping of silicon with Na does not generate donors because Na atoms coalesce, probably, in neutral complexes.

## 5.2 Introduction of electrically active impurities in the process of growth of diamond single crystals and films

**5.2.1 Diamond single crystals.** According to Refs [5, 6], the largest and most perfect single crystals of synthetic diamond were grown by the recrystallization technique, which uses a fine diamond powder rather than graphite as a starting material in the high-pressure chamber. Some crystals grown from the material containing boron had electrical parameters identical to those of South-African type-IIb natural diamonds. At the same time, workers of the General Electric, and other authors after them, noted that boron is inhomogeneously distributed in synthetic diamonds. In addition, as Gontar' et al. noted [10], the differences between the properties of the growth pyramids of the (111), (113), and (100) faces are due to the predominant unintentional doping of <111> growth pyramids with nitrogen and <111> and <113> pyramids with nickel.

These features of the growth of doped diamond along specific crystallographic directions may prove useful for practical purposes in the future, when the technology is improved. Since the designs and volumes of high-pressure chambers and temperature regimes of growth are different, it is too early to compare results obtained by different researchers. In recent reports on large single crystals that appear to be produced on a commercial scale in the Sumitomo laboratory, no data about the conductivity and distribution of impurities were given.

In 1985–1991, papers were published in the Soviet Union that concern practical applications of relatively small synthesized semiconducting *p*-type diamonds doped with boron in the process of growth [4, 6]. Synthesis of *n*-type diamonds doped with arsenic was also described in these papers. They contain no detailed information on the concentration of effective acceptors and donors, and we will turn back to them in the following sections when discussing applications of diamond in electronic devices.

**5.2.2 Epitaxial and polycrystalline films.** As was noted above, Alekseenko et al. [15] identified in 1977, using conductivity and Hall measurements, the mechanism of hole transport in *p*-type diamond layers grown at the Institute of Chemical Physics, USSR Academy of Sciences. Those experiments were continued later by other authors. Spitsyn et al. [28] reported on the growth of epitaxial layers on synthesized diamond single crystals, both insulating and semiconducting. The single crystals were about 0.8 mm in size and had a resistivity of  $10^2$  to  $10^{14}$   $\Omega$ m. The growth of diamond layers with thicknesses of 1–10  $\mu$ m was controlled by the homoepitaxy mechanism. Given the low concentration of defects at the substrate–film interface, the resulting structures could be heated to temperatures of up to 1500 °C. The films were grown on the (100) and (111) faces.

Among numerous recent, publications about the growth of polycrystalline diamond films by the plasma-assisted decomposition of hydrocarbons [6], there is a remarkable paper by Okano et al. [30]. Theoretical considerations [25] concerning the mechanism of film growth under such conditions and the feasibility of introducing impurities with low solubility (in this case, phosphorus) into growing films are also interesting.

Okano et al. [30] used  $P_2O_5$  as a dopant and obtained polycrystalline *n*-type diamond films with a resistivity of about 100  $\Omega$  cm on silicon substrates with (100) orientation. They gave evidence that the conductivity was *n*-type. The

electron mobility derived from Hall-effect measurements was  $50 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ . This result is quite interesting, as well as the fact that they produced ohmic contacts using the common silver paste. The activation energy of conductivity was as low as 0.05 eV. This indicated that grain boundaries represent no high potential barriers. The paper also contained data about Raman spectra indicating that most of the material had the diamond structure, although a small fraction of it had  $sp^2$  bonds. The interplanar spacings derived from high-energy electron diffraction (HEED) data were almost identical to the standard parameters of diamond single crystals.

**5.2.3 Ion implantation.** The advantages of the ion implantation technique, as well as the inevitable difficulties associated with this method, are known to specialists and described in manuals and reviews [31, 32]. It seemed natural to apply the ion implantation to diamond-based materials quite long ago [4, 21]. The first publications contained reports on conducting layers on surfaces of samples exposed to ion beams. The interpretation of results, however, was not unambiguous, because phase transitions of carbon to other modifications could occur. Later, conditions were found under which the diamond lattice, strongly damaged by implanted ions, was almost fully restored. Presently, it is possible to produce areas of a required configuration with the *n*-type conductivity through implantation of boron ions according to a prescribed program. Ion implantation at high ion energies of up to several megaelectronvolts attracts special attention [8].

## 6. Conclusions

As was noted in Introduction, the real state of affairs concerning wide applications of diamond in electronics has fallen short of the overoptimistic predictions by some authors. In commercially produced devices, natural and synthesized diamonds have been used mostly as heat sinks [28, 33]. According to the reports in Ref. [6], diamond films produced by the CVD technique are still inferior in thermal conductivity to bulk crystals. There is an optimal area for devices operating in the continuous mode. In the pulse mode, the use of diamond heat sinks makes it possible to increase the repetition rate of the device operation [34].

The difficulties on the way to commercial manufacture of diamond transistors are related to the fairly large depth of acceptor levels and the instability of relatively shallow donors (interstitial lithium with the ionization energy  $E_C - 0.1-0.15$  eV) at high temperatures, as follows from theoretical calculations. This property was confirmed by direct experiments using the secondary ion mass spectrometry (SIMS). As concerns interstitial phosphorus donors, which are expected to be more stable, no conclusive experimental data have been obtained by this day. Theoretical estimates indicate that phosphorus atoms substituting carbon atoms at lattice sites should have a very low solubility in diamond. We cannot exclude, however, the possibility that the regimes of the phosphorus doping in the process of diamond-film growth and upon ion implantation could be optimized.

For the reasons discussed above, only experimental specimens of low-power, high-temperature diamond-based transistors have been produced. Their service life is much shorter than the usually required time (15 000 h). The contacts of such alloys as Cr–Au and Cr–Pt with diamond have the activation energy for failure of about 1 eV and 1.4–1.5 eV [34]. The

degradation of such contacts at temperatures above 500 °C has not been studied so far.

Experimental specimens of Schottky diodes have been manufactured on the base of natural diamonds, synthesized crystals [5, 6], and recently on diamond films [35].

It seems that the development of integrated planar technology will pay its way only with diamond films, which nearly always have, as was mentioned above, a polycrystalline structure and electrical parameters inferior to those of diamond single crystals. Presently there is a considerable demand for diamond dosimeters invented by Konorova and Kozlov in 1968 [12]. These devices have a high radiation stability and are convenient for research in radiobiology.

Diamond-like carbon films are used in optics as protective layers, antireflection coatings, and heat sinks. Since they can be produced at ambient temperatures, they may prove convenient for many technical applications.

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