

Calculation of characteristics of subnanometer delta-doped layers in diamond in the quasi-classical approach

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ABSTRACT It is shown that subnanometer boron delta-doped layers characteristics in chemically vapor deposited diamond films can be calculated in the quasi-classical approximation both in equilibrium and in an external electromagnetic field ionizing doping boron atoms. The corresponding solutions are found, in the first case – analytically, in the second – numerically. The use of the quasi-classical approximation allows one to simplify significantly the mathematical modeling of diamond structures with subnanometer delta-doped layers, necessary for their application in nanoelectronics, for example in high-speed field-effect transistors.

KEYWORDS chemically vapor deposited diamond, delta-doped layers, quasi-classical approximation

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1. Introduction

Due to its unique characteristics (high thermal conductivity and electrical strength, chemical and radiation resistance, low coefficient of thermal expansion, etc.), diamond is a promising material for the creation of electronic and optoelectronic devices with record parameters. Being an insulator in its pure state, it requires doping to ensure electrical conductivity. However, at a high degree of doping, necessary to ensure high electrical conductivity, due to scattering on ionized atoms of the doping impurity, the mobility of charge carriers and, as a consequence, the speed of electronic devices based on diamond decreases.

One of the possible solutions to this problem is well known in physics of conventional semiconductors. It reduces to the creation of so-called delta-doped layers [1]. They are thin layers (with a thickness of the order of several lattice constants) of highly doped material enclosed in a lightly (unintentionally) doped (ideally pure) surroundings. In such layers, charge carriers partially penetrate from the potential well formed by ionized impurity atoms of the delta-doped layer into the lightly doped surroundings due to the quantum-mechanical effect. As a result, when moving in the plane of the layer in an external electric field, their scattering on these atoms decreases, and the mobility, therefore, increases compared to the value that it would have in a uniformly doped material with the same concentration of doping impurity as in the delta-doped layer. By forming a sequence of such layers, it is possible to obtain a structure with sufficiently high conductivity and mobility simultaneously.

Calculation of the characteristics of delta-doped layers usually requires a joint solution of the Schrödinger equation for the quantized energy levels and corresponding wave functions of charge carriers in the potential well of such a layer and the Poisson equation for the distribution of the electrostatic potential. Therefore, it is a complex mathematical problem that can only be solved by numerical methods.

In diamond, delta-doped (as a rule, by boron) layers were obtained in chemically vapor deposited films by many researchers [2–11]. It was used in diamond electronics devices, for example in field-effect transistors. Therefore, the problem of calculating their characteristics in dependence on the control parameters, i.e. the layer thickness, the concentration of the doping impurity in its center and in the surrounding material, temperature, etc., is relevant for practice.

In this paper, it is shown that for diamond, this calculation can be performed in the quasi-classical approximation. This significantly simplifies the corresponding numerical procedure allowing one to avoid the calculation of the quantized energy levels and corresponding wave functions of charge carriers in the potential well of such a layer. Moreover, for the thinnest (i.e. of a monolayer thickness) delta-doped layer, this approximation allows one to obtain analytical (in the case

of equilibrium situation) results. The interest to this extremely thin layer is related to its promise of the hole mobility increase (needed for diamond high-speed field-effect transistors), because even in 1 nm thick layers, such an increase was not obtained [10].

In the next section, this calculation is performed for equilibrium monolayer-thick delta-doped layers at room temperature. In Section 3 – for such layers in an external electromagnetic field ionizing the doping impurity. The Discussion and conclusion section analyzes and summarizes the main results of the work.

2. Equilibrium delta-doped layers

Let us consider an ideal case when the delta-doped layer consists of only one heavily boron-doped monolayer (with a thickness of order 0.1 nm) located in a lightly (unintentionally) boron-doped diamond. Since the minimum thickness of a delta-doped layer achieved to date is of the order of 1 nm [2–11] and the maximal doping profile abruptness is 1 decade/nm [12, 13], modern technologies are close to creating such a structure. Let the two-dimensional concentration of boron atoms in such a delta-doped layer be $N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$. In the lightly (unintentionally) doped diamond surrounding the delta-doped layer, let the concentration of boron atoms be equal to $N_{bv} = 10^{15} \text{ cm}^{-3}$, which is determined by the residual boron content in the growth reactor.

Let all the energies be counted from the level of the chemical potential μ so that in what follows the latter is equal to zero. Let us denote the energy of the upper edge of the valence band as $E_v = E_{v0} - e\phi$, where E_{v0} is the energy of the upper edge of the valence band away from the delta-doped layer, e is the elementary charge ($e > 0$), ϕ is the electrostatic potential. The latter satisfies the two-dimensional Poisson equation

$$\frac{d^2\phi}{dx^2} = -\frac{4\pi e}{\varepsilon} \left[N_h - \frac{N_{bv} \exp\left(-\frac{E_{v0}-e\phi}{k_B T}\right)}{4 \exp\left(\frac{E_0}{k_B T}\right) + 2 \exp\left(\frac{E_0-E_{so}}{k_B T}\right)} \right], \quad (1)$$

where x is the Cartesian coordinate in the direction, orthogonal to the delta-doped layer plane $x = 0$, $N_h = N_v \exp\left(\frac{E_{v0}-e\phi}{k_B T}\right)$, $N_v = 0.25 [2k_B T / (\pi \hbar^2)]^{3/2} \{m_{hh}^{3/2} + m_{lh}^{3/2} + m_{so}^{3/2} \exp[-E_{so}/(k_B T)]\}$ [14], k_B is the Boltzmann constant, T is the temperature, which will henceforth be set equal to room temperature ($T = 300 \text{ K}$), $\hbar$ is the Plank constant, $m_{hh} = 0.427m$, $m_{lh} = 0.366m$, $m_{so} = 0.394m$ are the characteristic effective masses of heavy, light and spin split-off holes, respectively [15], m is the free electron mass, $E_{so} = 6 \text{ meV}$ is the energy of spin-orbital splitting [16], $E_0 = 370 \text{ meV}$ is the activation energy of a single boron atom [16], $\varepsilon \approx 5.7$ is the static permittivity of diamond [16]. The first term in the square brackets in (1) is the three-dimensional concentration of non-degenerate hole gas under the condition $-(E_{v0} - e\phi)/(k_B T) > 1$ (the validity of which is confirmed by the final result, see below), the second term is the three-dimensional concentration of negatively charged boron atoms in the lightly (unintentionally) doped diamond surrounding the delta-doped layer under the condition $(E_{v0} - e\phi + E_0 - E_{so})/(k_B T) > 1$ (the validity of which is also confirmed by the final result, see below) [17, 18].

The solution of equation (1) must be symmetrical with respect to the plane $x = 0$ and tend to zero away from the delta-doped layer. In this case, the energy of the upper edge of the valence band away from the delta-doped layer E_{v0} is found from the condition of electrical neutrality, i.e. from the condition of equality to zero of the square bracket in (1) when $\phi = 0$. This gives one that $E_{v0} = -\frac{k_B T}{2} \ln \left\{ \frac{N_v}{N_{bv}} \left[4 \exp\left(\frac{E_0}{k_B T}\right) + 2 \exp\left(\frac{E_0 - E_{so}}{k_B T}\right) \right] \right\}$. A solution of (1) with the indicated properties can be found analytically and is given by the following formula

$$\phi = \frac{4k_B T}{e} \text{Arcth} \left\{ \text{th} \left[\frac{e\phi(0)}{4k_B T} \right] \exp \left(-\frac{\sqrt{2}|x|}{r_D} \right) \right\}, \quad (2)$$

where $\phi(0)$ is the value of the electrostatic potential at $x = 0$, $r_D = \frac{\sqrt{\varepsilon k_B T}}{2e \sqrt{\pi N_v \exp[E_{v0}/(k_B T)]}}$ is the Debye radius in a non-degenerate hole gas with concentration $N_v \exp[E_{v0}/(k_B T)]$.

The value of $\phi(0)$ is related to the two-dimensional concentration N_{b-} of negatively charged boron atoms in the delta-doped layer by means of the boundary condition on the jump of electric field at plane $x = 0$: $-\phi'(0+) + \phi'(0-) = -4\pi e N_{b-}/\varepsilon$. Using (2), this leads to the equation

$$\frac{8\sqrt{2} k_B T}{e r_D} \frac{\text{th}[e\phi(0)/(4k_B T)]}{1 - \text{th}^2[e\phi(0)/(4k_B T)]} = -4\pi e N_{b-}/\varepsilon. \quad (3)$$

On the other hand, the magnitude of N_{b-} can be found using the standard formula [17, 18]

$$N_{b-} = \frac{N_b}{1 + 4 \exp\left[\frac{E_0 + E_v(0)}{k_B T}\right] + 2 \exp\left[\frac{E_0 - E_{so} + E_v(0)}{k_B T}\right]}, \quad (4)$$

where $E_v(0) = E_{v0} - e\phi(0)$. Then, equation (3) becomes an equation for $\phi(0)$, from which the latter can be found numerically.

Figure 1a shows the spatial dependence of $E_v = E_{v0} - e\phi$ found in this way for $N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$. Fig. 1b shows the spatial dependence of the three-dimensional concentration of non-degenerate hole gas N_h for the same parameters.

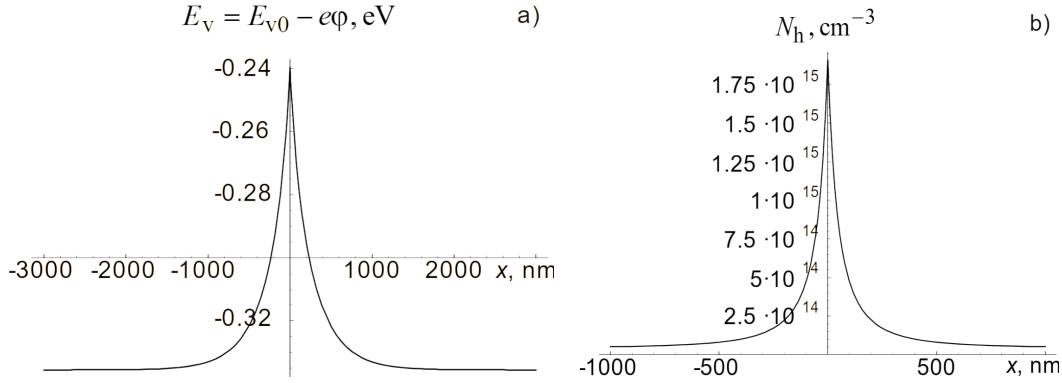


FIG. 1. (a) Energy top edge of the valence zone $E_v = E_{v0} - e\phi$ counted from chemical potential level as a function of Cartesian coordinate x along direction, orthogonal to the delta-doped layer plane $x = 0$ for $N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$. (b) The spatial dependence of the three-dimensional concentration of non-degenerate hole gas N_h for the same parameters

Let us now check the fulfillment of the above assumptions. Firstly, the condition $-(E_{v0} - e\phi)/(k_B T) > 1$ is most difficult to fulfill when $x = 0$, where the value $-(E_{v0} - e\phi)$ is minimal (see Fig. 1a), so it is necessary to demand $-(E_{v0} - e\phi(0))/(k_B T) > 1$. For $N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$, we have $-(E_{v0} - e\phi(0))/(k_B T) \approx 8.9 > 1$. Thus, the condition $-(E_{v0} - e\phi)/(k_B T) > 1$ is satisfied for all x .

Secondly, the condition $(E_{v0} - e\phi + E_0 - E_{s0})/(k_B T) > 1$ is most difficult to fulfill when $|x| \rightarrow \infty$, where the value $E_{v0} - e\phi + E_0 - E_{s0}$ is minimal and equals $E_{v0} + E_0 - E_{s0}$ regardless of the value of N_b . It is easy to check that $(E_{v0} + E_0 - E_{s0})/(k_B T) \approx 5.2 > 1$. Thus, the condition $(E_{v0} - e\phi + E_0 - E_{s0})/(k_B T) > 1$ is satisfied for all x .

Thirdly, we will check the fulfillment of the condition of applicability of the quasi-classical approximation. It is known [19] that this condition means that the thermal de Broglie wavelength of a hole $\lambda \approx \frac{2\pi\hbar}{\sqrt{2m(E + E_v)}}$, where $E + E_v \approx k_B T$ (the hole energy E is measured from the chemical potential level in the direction opposite to the direction of counting the electron energy, so that the hole potential energy is $-E_v$) for any x satisfies the inequality

$$\frac{1}{2\pi} \left| \frac{d\lambda}{dx} \right| \equiv \left| \frac{d\lambda}{dx} \right| \approx \frac{\hbar m e}{(2m k_B T)^{3/2}} \left| \frac{d\phi}{dx} \right| < 1.$$

It is clear from Fig. 1 that $|d\phi/dx|$ is maximal for $x = 0$, therefore if this condition is satisfied for $x = 0$, then it is also satisfied for all other x . To calculate the derivative $|d\phi/dx|$ for $x = 0$, we can use the above boundary condition in the plane of the delta-doped layer, as a result of which the quasi-classical condition is rewritten as $\frac{2\pi\hbar m e^2 N_{b-}}{(2m k_B T)^{3/2} \epsilon} < 1$. For

$N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$, a calculation gives one $\frac{2\pi\hbar m e^2 N_{b-}}{(2m k_B T)^{3/2} \epsilon} \approx 0.02 < 1$, and the quasi-classical condition is indeed satisfied with a large margin. According to [19], the relative error of the above-obtained quasi-classical results is of the same order, i.e. less than 2 %.

The reason for this is that at $N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$, the two-dimensional concentration of negatively charged boron atoms in a delta-doped layer is as follows: $N_{b-} \approx 3.5 \cdot 10^{10} \text{ cm}^{-2}$, i.e. it is small (about 0.1 % of N_b). Accordingly, the three-dimensional concentration of free holes is also small, and therefore the electrostatic potential is a smooth function of x (Fig. 1a).

3. Delta-doped layers in an external electromagnetic field ionizing boron atoms

According to the previous section, in equilibrium even for a large two-dimensional boron atom concentration $N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$, only a small part of them is ionized. In consequence, the concentration of free holes around the doped delta-layer is also small, which leads to a small conductivity of the system. Therefore, let us consider a non-equilibrium situation when boron atoms in the delta-doped layer are fully ionized by an external electromagnetic field whereas the free hole distributions in energy and space are similar to equilibrium ones with ambient temperature. The practical implementation of such a situation is considered at the end of this section.

In this situation, all the boron atoms in the delta-doped layer are negatively charged, i.e. $N_{b-} = N_b$. Poisson's equation has the same form (1), as in the previous section, but with N_h given now by the formula (taking into account that at the considered large concentrations of free holes they will mainly be degenerate)

$$N_h = \int_{-\infty}^{+\infty} (f_{hh} + f_{lh} + f_{so}) d^3p,$$

$$f_{hh} = \frac{2}{(2\pi\hbar)^3} \frac{1}{1 + \exp \{[-E_{v0} + e\phi + p^2/(2m_{hh})]/(k_B T)\}},$$

$$f_{lh} = \frac{2}{(2\pi\hbar)^3} \frac{1}{1 + \exp \{[-E_{v0} + e\phi + p^2/(2m_{lh})]/(k_B T)\}},$$

$$f_{so} = \frac{2}{(2\pi\hbar)^3} \frac{1}{1 + \exp \{[-E_{v0} + E_{so} + e\phi + p^2/(2m_{so})]/(k_B T)\}}$$

are the quasi-classical functions of the Fermi–Dirac distributions of heavy, light and spin-orbit split off holes, respectively, the validity of the condition $(E_{v0} - e\phi + E_0 - E_{so})/(k_B T) > 1$, which allows one to write the three-dimensional concentration of negatively charged boron atoms in the lightly (unintentionally) doped diamond surrounding the delta-doped layer as in (1) [17, 18], is proved in exactly the same way as in the case of the equilibrium delta-doped layer, see the previous section.

The solution of Poisson's equation, as in the case of the equilibrium delta-doped layer above, must be symmetrical relative to the plane $x = 0$, tend to zero away from the delta-doped layer and at $x = 0$ satisfy the boundary condition $-\phi'(+0) + \phi'(-0) = -4\pi e N_b / \varepsilon$. The energy of the upper edge of the valence band away from the delta-doped layer E_{v0} is found from the condition of electrical neutrality, i.e. from the condition of equality to zero of the square bracket in (1) at $\phi = 0$. In this case, quasi-classical functions of the Fermi–Dirac hole distribution are transformed into the Maxwell functions. The result of this is that N_h takes the same form as in the case of the equilibrium delta-doped layer, i.e. $N_h = N_v \exp\left(\frac{E_{v0}}{k_B T}\right)$. Therefore, E_{v0} has the same value as in the previous section. However, due to the complex dependence of N_h on ϕ near the delta-doped layer, it is not possible to find the solution of (1) analytically.

Figure 2a shows a numerical solution of (1) for $N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$, and Fig. 2b – the spatial dependence of N_h for the same parameters.

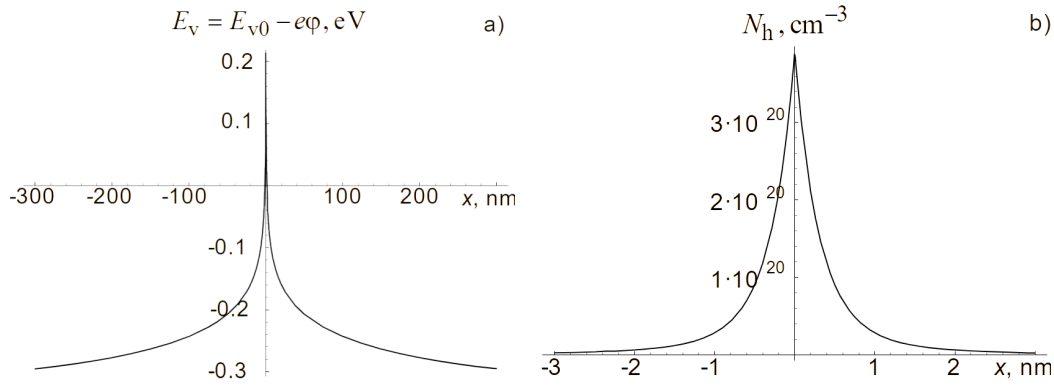


FIG. 2. (a) Energy top edge of the valence band $E_v = E_{v0} - e\phi$ counted from the level of chemical potential as a function of the Cartesian coordinate x along direction, orthogonal to the delta-doped layer plane $x = 0$ for $N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$. (b) The spatial dependence of the three-dimensional concentration of the hole gas N_h for the same parameters

At the point $x = 0$, one has $[E_{v0} - e\phi(0)]/(k_B T) \approx 8.3 \gg 1$, i.e. near the delta-doped layer, the hole gas is degenerate. At the same time, far from this layer, one has $\phi = 0$, and E_{v0} , as was said above, has the same value as in the case of the equilibrium delta-doped layer above, so that the hole gas there is non-degenerate. The transition from degenerate to non-degenerate hole gas with increasing distance from the delta-doped layer causes the complex form of the dependence of N_h on ϕ , which does not allow finding an analytical solution to (1).

Let us now check the fulfillment of the condition of applicability of the quasi-classical approximation. Now, in the formula for the characteristic de Broglie wavelength of a hole $\lambda \approx \frac{2\pi\hbar}{\sqrt{2m(E + E_v)}}$, the characteristic hole energy E is a value of the order of chemical potential, i.e. 0 (due to the above choice of the starting point for counting energies) at $E_v > k_B T$, where the hole gas is degenerate, and of order $-E_v + k_B T$ at $E_v < k_B T$, where the hole gas is non-degenerate. Using the results of the numerical solution to (1), it is easy to show that for $N_b = 3 \cdot 10^{13} \text{ cm}^{-2}$, we have

$\frac{1}{2\pi} \left| \frac{d\lambda}{dx} \right| \equiv \left| \frac{d\lambda}{dx} \right| < 0.6 < 1$ in the region near the delta-doped layer, where the bulk of the holes created by this layer resides. Thus, the condition of the validity of the quasi-classical approximation in order of magnitude is satisfied (although without a reserve). Therefore, according to [19], the relative error of the above-obtained quasi-classical results for the delta-doped layer in an ionizing external electromagnetic field is less than 60 %. So, these results can be used as estimates of the characteristics of such a layer only.

The reason for this is that although in this case the potential energy of a hole is a sharp function of the coordinate near the delta-doped layer (see Fig. 2a), but in this region, due to their high concentration, the kinetic energy of degenerate holes is also high, and therefore their characteristic de Broglie wavelength is small.

Now, let us consider the practical realization of the situation when boron atoms in the delta-doped layer are fully ionized by an external electromagnetic field. First, the photon energy $\hbar\omega$ of this field should be larger than the difference between the chemical potential $\mu = 0$ and the initial energy of a hole, localized on a boron atom, $-E_{v0} + e\phi(0) - E_0 = -584$ meV (see Fig. 2a), to ensure that the final free hole state is at least half-empty. It means that its wavelength should be shorter than $2.1 \mu\text{m}$. Second, the ionization rate of a neutral boron atom should be larger than the recombination rate of free holes with a negatively charged boron ion. The former is estimated as the product of the photon flow $\frac{I}{\hbar\omega}$ (I is the electromagnetic field intensity) by the ionization cross-section, which is, according to [20], is of order 10^{-16} cm^2 . To estimate the latter, let us take a value $1.77 \cdot 10^{-13} \text{ cm}^2$ for the recombination cross-section [21] and multiply it by the free hole Fermi velocity $\sim \sqrt{\frac{E_{v0} - e\phi(0)}{m}} \approx 2 \cdot 10^7 \text{ cm/s}$ and concentration $N_h(0) \approx 3.9 \cdot 10^{20} \text{ cm}^{-3}$ near the delta-doped layer (see Fig. 2) to obtain $1.4 \cdot 10^{15} \text{ s}^{-1}$. For $\hbar\omega = 584$ meV, the equality of these two rates means that I should be larger than $1.3 \cdot 10^{12} \text{ W/cm}^2$. For a focal spot with a diameter of several microns, such intensity can be supplied by a fiber laser. To avoid the diamond damage, the laser should work in a pulsed mode.

Certainly, electromagnetic field with such a large intensity will cause the hole heating. To estimate its rate, let us take a value smaller than 3 fs for the relaxation time of the hole quasimomentum and energy as it follows from the results of [22] for the hole energies more than by 2 times exceeding 165 meV – the energy of diamond optical phonon [16]. A simple estimate by Drude's formula for the hole conductivity [14] gives one that for such energies, a hole energy increase rate is smaller than 90 erg/s. On the other hand, for such energies, a hole energy decrease rate due to optical phonon emission is larger than $165 \text{ meV}/3 \text{ fs} = 90 \text{ erg/s}$, i.e. just the same as the increase rate. So, the hole kinetic energy cannot exceed 2 optical photon energies. As this value is of order $E_{v0} - e\phi(0) = 214$ meV (the maximal kinetic energy of degenerate holes according to the results of the above calculations, see Fig. 2a), then the hole gas will be in a state with energy and space distributions qualitatively similar to those found in this section.

4. Discussion and conclusion

The paper shows that the characteristics of delta-doped layers in lightly (unintentionally) doped diamond can be calculated in the quasi-classical approximation for both equilibrium and non-equilibrium situations. In the first case, the electrostatic potential is a smooth function of the coordinates, which ensures the applicability of the quasi-classical approximation. In the second case, the electrostatic potential is a sharp function of the coordinates, but, due to their degeneracy, the characteristic kinetic energy of holes is also large, which leads to the smallness of their de Broglie wavelengths and, as a consequence, the validity of the quasi-classical approximation. In ordinary semiconductors, such a situation does not occur, since the activation energies of impurities in them are significantly smaller than in diamond.

This result allows one to significantly simplify the calculation of the characteristics of the delta-doped layer in a lightly (unintentionally) doped diamond allowing to avoid the Schrödinger equation solution and finding the quantized energy levels and the corresponding wave functions of holes in a potential well formed by this layer. As a result, analytical formulas were found for the distribution of the electrostatic potential and the three-dimensional concentration of holes in the vicinity of such a layer in a lightly (unintentionally) doped diamond in the equilibrium case. Also, using a simple scheme, a numerical solution was constructed in the non-equilibrium when doping boron atoms in this layer are ionized by an external electromagnetic field.

Because the above quasi-classical solutions are found for a extremely thin (of thickness of 1 monolayer) delta-doped layer which is not created yet, they (e.g. the hole concentration distribution, please see Fig. 1b and Fig. 2b) cannot be compared with existing experimental data (e.g. capacitance-voltage measurements of hole concentration profile [9]). But as the modern technology approaches to the creation of such layers, this comparison is hoped to become possible soon.

The obtained results can be used to calculate the characteristics of diamond nanoelectronic devices with subnanometer delta-doped layers.

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