1. Introduction

As a wide band-gap semiconductor, diamond possesses excellent physical and chemical properties and shows many attractive properties [1–12]. For instance, it exhibits high electric-field breakdown strength, low dielectric constant, high thermal conductivity and high transparency conductivity. Hole-doped diamond exhibits superconductivity at high pressure and temperature [3]. Nanoscale single-crystalline and polycrystalline diamond needles exhibit ultralarge, fully reversible elastic deformation [11]. It holds potential applications in high-tech area, especially in those of high frequency, high temperature and high-power electronic devices. Despite these interesting properties, the wide band gap of 5.5 eV and low electronic conductivity also limit its applications in electronic devices. And doping is regarded as the most popular method to tune its electronic and other relative properties. In fact, diamond under doping has been studied extensively over the past decades [13–20]. P-type doping in the diamond can be easily obtained by B doping [21,22]. That is because the radius of B atom is smaller than that of other atoms, and its solubility in diamond is high. Currently, great progress has been made in the research of B doped p-type diamond. Nevertheless, it is challenging to achieve an appropriate n-type diamond through doping [23,24], because the suitable donor is difficult to find. Many elements with n-type characteristics have been studied, such as Li, Na in group I, N, P in group V and O, S in group VI [25–33]. However, interstitial Li or Na is insoluble and tends to form other impurities, which is unsuitable for realizing n-type diamond [25]. Substitutional N can denote an electron to the diamond, and then the deep donor level locate at 1.4 eV below the conduction band edge can be found, which prevents it from being used in room temperature semiconductor devices [26,27]. P is a promising shallow donor candidate with a donor level of 0.43 eV below the conduction band edge [28]. However, due to its low conductivities and carrier mobility, it is also unlikely to be used at room temperature [29]. An n-type conducting layer create in O-implanted diamond with the activation energy is about 0.32 eV. But the oxygen donors become deactivated after being annealed above 600 °C [30]. The preparation method of n-type diamond with reasonable quality is S doping, which shows a shallow donor level in the bandgap with the ionization energy of 0.38 eV and the electrons mobility of 597 cm²/Vs [24, 31]. Although good results have been obtained by S doping, it is difficult to be realized because of the quite low doping efficiency and solubility [32]. Obviously, single atom doped n-type diamond cannot meet the requirements of semiconductor fabrication. To this end, co-doping strategies have been proposed for diamond. For example, Miyazaki and his co-operator reported that N-H-N complex in diamond exhibits a much shallower donor character compared with an isolated N donor [33]. Lombardi et al. systematically studied the interaction
of H with B, P and S in diamond and found that it was energetically favorable for hydrogen to be trapped and to passivate boron and phosphorus [34]. Li et al. revealed that co-doping B could enhance S doping in the diamond [35]. The underlying physics for the B-S co-doping are also investigated based on theoretical calculations [36]. Li et al. reported that the synergistic effect of B and N atoms made a lower Fermi energy, which could improve the conductivity significantly and promote the absorption edges shift to the visible-light region [37].

The heterostructure constructed by diamond and other materials cannot only regulate the electronic properties of diamond effectively, but also provide ideal platforms for other materials [38]. In addition, diamond-composed heterostructures also hold great potential applications in photocatalysis, electronic devices and other fields [39,40]. However, few works have been reported on diamond composite systems, which calls for further study.

In this review, the recent progress of the electronic and geometric properties of diamond is summarized mainly from a theoretical (by using the first principle calculations) perspective. By introducing doping and constructing strategies, we emphasize the recent advances of the geometric and electronic properties of diamond. Then, we discuss its potential applications in electronic and optoelectronic devices. Finally, the challenges and opportunities are also provided.

2. Basic geometric and electronic properties of diamond

Diamond, displaying a completely sp³ hybridization, is a typical atomic crystal. The lattice constant is 3.57 Å, and the C-C bond is 1.54 Å. The crystal structure is shown in Figure 1. Each unit cell contains eight carbon atoms, and the C-C bond is strong covalence bond. Such strong C–C bonding within diamond ensures excellent mechanical properties. Owing to the strong sp³ hybridization, diamond is known as the hardest material in the natural environment. Besides, diamond has the highest thermal conductivity of 2 × 10³ Wm⁻¹K⁻¹ at a wide temperature range, which is 15 times higher than that of Si at room temperature. Additionally, the thermal expansion coefficient of diamond is comparable to that of invar at room temperature. Diamond is a semiconductor with a wide bandgap of 5.47 eV, which is much larger than that of other semiconductors. Hence, the corresponding electronic breakdown field (2 × 10⁷ Vcm⁻¹) is much higher than that of Si (3 × 10⁵ Vcm⁻¹). Besides these properties, diamond also possesses excellent optical properties. The corresponding optical transparency is distributed from the UV to the far IR region. These interesting properties make diamond promising for applications in high temperature, high frequency, radiation-resistant and high power devices, especially in the region of coated tools, heat sink, high fidelity speakers, field effect transistors, optical device and so on.

3. Modulate the electronic properties of diamond

The p-type material with B atom doped diamond has been widely used in industrial production. However, the high-quality n-type diamond materials have not yet been achieved, which hampers its application and exploitation in electronic devices fields. In order to develop useful electronic devices of n-type doped diamond materials, great efforts have been devoted both theoretically and experimentally.

3.1 Atom doping

The synthesis of n-type diamond via atom doping lies at the heart of diamond research in recent years. It is recognized as one of the unresolved difficult problems in the world. Various studies have proven that no donor
impurity can produce sufficiently shallow energy levels to produce electrons to conduct electricity at room temperature. The research on n-type diamond including two aspects: single element doping and multiple elements co-doping. Among them, V group elements of N, P and VI group elements of O, S are always used as the donor elements for single element doping. In the following, we will discuss them separately.

### 3.1.1 N-Doping

Nitrogen can commonly find at different concentrations in natural diamond. It can be existed by substituting or aggregating to the target diamond. The impurity level of N in diamond is quite deep, which is located at the level of 1.7 – 2.0 eV from the bottom of the conduction band.

It is to say that only little electrons can be excited into the conduction band to give appreciable conductivity. Theoretical results indicate that N atom occupied the substitutional site along the (111) direction of diamond. And the reason of the distortion is an antibonding interaction between the N long-pair orbital and a C dangling bond.

Briddon et al. [43] found that the aggregate consisting of a pair of nearest neighbor substitutional N atoms is also a deep donor, which introduced a donor level locating at the middle of the band gap. This implies that it is impossible for N element to act as a feasible donor in diamond to achieve n-type diamond with useful conductivities at room temperature.

Fortunately, the recent reported theoretical and experimental results of N-doped diamond films challenge the above traditional assumption. The diamond films is obtained by a microwave plasma-enhanced chemical-vapor-deposition (MPCVD) method with 0.2% nitrogen content using a CH$_4$ (1%)/Ar gas mixture and 1% - 20% nitrogen gas added. It is reported that the electrical conductivity of the N doped diamond films could research up to 143 Ω$^{-1}$cm$^{-1}$ [44]. What's more, Oliver et al. reported that [45] the clear n-type conductivity with very low thermal activation energy for ultra nanocrystalline diamond films grown with added nitrogen can be obtained. The mobility and sheet carrier concentration reach 1.5 cm$^2$/V·s and 2 x 10$^{17}$ cm$^{-2}$, respectively. Additionally, another high n-type conductivity (up to 150 Ω$^{-1}$cm$^{-1}$) of ultra nanocrystalline diamond film with a high N content has been achieved [46], which is higher than the n-type conductivity (143 Ω$^{-1}$cm$^{-1}$) reported in [44]. The carrier concentration and the mobility are up to 10$^{20}$ cm$^{-3}$ and above 1 cmV$^{-1}$s$^{-1}$ at room temperature, respectively [46].

Many theoretical works have been carried out to explain the mechanism of such unusual n-type conductivity in ultra nanocrystalline diamond films. It is reported that the grain-boundary (GB) conduction is responsible for the high n-type electrical conductivity [44,47,48]. Moreover, the theoretical results demonstrate that incorporation of N impurities into the GB is easier than incorporation of N into the bulk diamond [49]. Hence, it is expected that more N atoms are incorporated into the GB than that into the bulk, and the electronic structures of these N-related centers results in the high n-type conductivity in ultra nanocrystalline diamond films. Recently, Song et al. studied the effect of N on differently terminated diamond surfaces theoretically by using density functional theory (DFT). The results implied that an obvious shift of the Fermi level toward the conduction band can be found for the N-doped diamond surface. Moreover, a localized electron conductivity was observed for O on-top termination in combination with N-doping [50].

### 3.1.2 P-Doping

Phosphorus is considered to be the most promising donor dopant due to the shallow donor energy level (0.2 – 1.09 eV). In 1997, Koizumi et al. [28] synthesized the homoepitaxial n-type diamond thin film successfully with different dopant concentrations by using phosphine (PH$_3$) as dopant source. The obtained activation energy is 0.43 eV and the Hall mobility is 23 cm$^2$/V·s at about 500 K. From then on, a plenty of works have been adopted to grow high-quality n-type P-doped diamond films [51–53]. In 1999, Koizumi [54] reported that the (111) surface of diamond was found to be the best surface as substrate for the growth of P-doped diamond. The activation energy is about 0.55 eV and the Hall mobility is 28 cm$^2$/V·s from 370 to 700 K. In the next year, Teraji et al. studied the electric contacts for P-doped n-type diamond films [55]. Their results show that Schottky barrier height of 2.4 eV could be obtained from both Au and Cu contacts, which is deposited on lightly P-doped diamond films. Ohmic contacting properties are obtained from electrodes on heavily doped layer with donor density $N_D$ of over 1 x 10$^{19}$ cm$^{-3}$. Such features imply that the characteristic of contacts formed on P-doped n-type diamond films is similar to those on typical n-type semiconductors.

Considering that carrier concentration and mobility decrease with the increase of P concentration in P-doped diamond, it is important to unveil the underlying mechanism in P-doped diamond. The lower carrier concentration in doped diamond films is related to the higher hydrogen content and vacancy number in CVD diamond films. Hydrogen in P doped diamond films may passivate the donor impurity and make the films poor conductivity. Besides PH$_3$ can act as P source during P-doped diamond films growth, other substances containing P element can also act as P source to dope diamond. Okano prepared diamond film by using P$_2$O$_5$ (dissolved in methanol and acetone solution) as doping agent and using methane and hydrogen as carbon source. The electron mobility and carrier concentration were measured as 50 cm$^2$/Vs and 10$^{13}$ cm$^{-3}$, respectively [56]. Kociniewski et al. [57] achieved n-type diamond...
films successfully by using the metal-organic chemical vapor deposition (MOCVD) technology with liquid tertiarybutylphosphine (TBP) as P source, which showed comparable conductivity to P-doped diamond from PH₃. Indeed, the organic precursors provide a great advantage over hydrides because of their low toxicity and their safe handling. Koizumi et al. investigated P-doped diamond films using a metal-chamber-type microwave plasma-assisted chemical vapor deposition (MPCVD) method over a wide temperature range [58]. The carrier activation energy is about 0.6 eV and the mobility is up to 240 cm²/V·s.

So far, P impurity has been considered as the most available donor with the activity energy of 0.6 eV in diamond. Theoretical calculation [59] indicates that P is more soluble in bulk diamond with a much shallower donor level in band gap that could be induced. It also shows that a biaxially expanded epitaxial diamond film could enhance the solubility of P and might give rise to good n-type conductivity.

3.1.3 O-Doping

Theoretical researches show that O atom can also act as donor in the form of substitution in diamond. Due to the smaller atomic radius, O atom could introduce smaller distortion into diamonds than S and P atoms, see Figure 2a. From Figure 2b, it can be seen that charges between C and O atoms are relatively larger than those between C and C atoms. This leads to relatively stronger C-O chemical bonding with respect to that of C-C bond. The larger value of charges between C and O atoms shows that the incorporation of oxygen defect can exhibit some kind of vacancy character of defect states associated with C-atom bonds [60]. As a result, it may be an ideal candidate for achieving n-type diamond. Prins et al. [30, 62, 63] doped O atoms into diamond successfully by means of implantation and following annealing at low temperature. The obtained activation energy is about 0.32 eV [30, 62], but the electron concentration and mobility are low. Hu and his co-operator also investigated the role of O atom playing in diamond [64]. They pointed out that the addition of appropriate amount of oxygen can inhibit the secondary nucleation of diamond on the growth surface, effectively. The results also indicate that too much oxygen will not only reduce the growth rate of diamond, but also increase the number of defects [65]. Anderson et al. [66] proposed that substitutional O will induce deep electronic levels in diamond. Such donors could be more complex. Consequently, the simplest alternative is an O interstitial trapped at a vacancy. Previous study by Gali et al. has shown that n-type conductivity in O-doped diamond cannot be associated with a simple substitutional neutral oxygen center. It is also suggested that the possibility of O can form combinations with other defects to give rise to n-type conductivity in the negatively charged form [67].

Figure 2. (a) Optimized crystal structure of oxygen-doped diamond, with showing equilibrium geometry of carbon-oxygen bonds around the substituted atom. (b) Electron-density distributions of oxygen doped diamond. Due to higher electronegativity of oxygen, excess negative charge is accumulated around O-atom, leading to C⁺⁻O⁻ dipole. Adapted with permission from [60]. Copyright 2015, Elsevier. (c) Low-energy structures of $S_{sub}^-$. Yellow balls represent S atoms. Red and blue balls represent C atoms, which have shorter and longer S-C distances than the others, respectively. (d) Spontaneous relaxation of a substitutional S defect in diamond to $S_{sub}^-\cdot$. Adapted with permission from [61]. Copyright 2002, WILEY-VCH.
3.1.4 S-Doping

Previous investigations reveal that S atom can also be used as donor impurity, which shows n-type doping with the donor level of about $E_C - 0.38$ eV. The relative geometry stability is also proved, see Figure 2c and d [61]. Nishitani-Gamo et al. studied the growth of S-doped diamond via microwave plasma assisted chemical vapor deposition by introducing $H_2S$ into the gas phase. They unveiled that a small amount of $H_2S$ could improve the quality of crystal effectively. The S-doped homoepitaxial (001) diamond is demonstrated with n-type conduction by Hall effect measurements under the temperature range of 250 – 550 K. The mobility of electrons and the carrier concentration are 597 cm$^2$V$^{-1}$s$^{-1}$ and 1.4$\times$10$^{13}$ cm$^{-3}$ at room temperature, respectively [68]. Garrido et al. [69] indicated that the transformation of p-type to n-type doped diamond can be observed from low temperature to high temperature. The corresponding activation energies are 0.36 and 1.55 eV, respectively. Additionally, theoretical calculations indicated that S impurity in diamond resulted in a deeper donor level than P, and also a lower bulk solid solubility than P. According to the calculation results, Lombardi [34] found that S donor level is located at $E_C - 1.1$ eV.

3.1.5 Other single element doping

Except for the doping elements mentioned above, a large amount of experimental and theoretical works have been performed to investigate other elemental dopants. For example, Tan et al. studied Ti-related point defects in diamond [70]. They found that Ti is more energetically favorable to be located adjacent to a vacancy to form a TiV structure, which introduces impurity levels into the band gap of diamond. Yan et al. have studied the electronic properties of halogen atom (X = F, Cl, Br, I) doped diamond. They reported that the F defect accepts electrons and the Cl/Br/I dopant donates electrons. Among the donors, substitutional I is found to be an ideal choice to realize n-type diamond [71]. As for arsenic doping in diamond, the calculations showed that it is also considered to be a competitive candidate for achieving n-type diamond [72]. The calculations from Yan found that the interstitial Be or Mg doped diamond which are synthesized at H-poor conditions has n-type metal conductivity. The substituted Be and Mg show acceptor characters in diamond and could compensate other interstitial donors [73].

3.1.6 Co-doping

Obviously, the limited source of dopants cannot meet the requirements of diamond-based semiconductor devices fabrication. Researchers then focus on the co-doping approach to achieve appropriate diamond devices. Normally, [74] for direct band gap semiconductors, the effect of co-doping on the ionization energy is rather small, less than 0.05 eV. For the wide and indirect band gap semiconductor diamond, the co-doping can overcome the high ionization energy problem faced in the n-type diamond. Based on first-principles calculations, Tang et al. studied B-S co-doped diamond at different concentrations to achieve effective and shallow donors in diamond. They found that the introduced B impurity could help to improve the solubility of S in diamond. In addition, the electron effective mass of the intermediate band of B-S co-doped diamond is much smaller than that of S-doped, which might give rise to an increase in the conductivity of diamond. Therefore, they believed that with the help of B doping, S became a suitable shallow donor impurity in diamond (Figure 3) [36]. The calculation of the diamond co-doping with B, P, and As impurities from Shen et al. revealed that solubility of P and As impurities in diamond could be improved by introducing B dopants, and the obtained band gap is smaller than pure diamond. The ionization energies of diamond with double-acceptor-donor center complexes are sharply decreased as compared with the B-doping diamond. This demonstrated that B-B-As and B-B-P co-doping impurities structures were more suitable for achieving p-type diamond [41]. Li et al. [37] reported that the synergistic effects of B and N atoms could improve the conductivity significantly and promoted the absorption edges shift to the visible-light region. The co-doping of B and N is also suggested to be an effective approach to obtain high quality B-doped diamond substrates [75]. Li et al. [76] reported that diamonds co-doped with B and H showed p-type semiconducting behavior, and the co-doped diamond has a higher carrier concentration, conductivity and a more compatible lattice structure than B-doped diamond. Miyazaki and his co-operator reported that N-H-N complex in diamond exhibited a much shallower donor character compared with an isolated N donor [33]. Lombardi et al. systematically studied the interaction of H with B, P and S in diamond. They showed that in the band gap of diamond, sulfur with one H atom produces shallow donor levels. Further, both P-H and P-H$_2$ could dissociate to form H$_2$, which is in contrast to B-doped diamond [34].

3.2 Color Center

Diamond color centers aroused tremendous interests both theoretically and experimentally since the first report has been published in the field of quantum optics in 1997 [77]. Due to the unique features, the nitrogen-vacancy (NV) color center in diamond hold promise for a wide range of applications in quantum technology [78–82]. Group-IV vacancy color centers in diamond are fast emerging qubits that can be harnessed in quantum communication and sensor applications [83–86]. By using the cutting-edge \textit{ab initio} calculations, Thiering et al. developed a novel spin Hamiltonian for these
qubits which incorporates the interaction of the electron spin and orbit coupled with phonons beyond perturbation theory [87]. Hepp et al. [88] explained both the SiV structure splitting in magnetic fields and the polarization of the zero field fine structure components by exposing single SiV defects to a magnetic field. The theoretical model also provided a qualitative explanation of the first spin-related experiments of the negatively charged SiV.

Moreover, SiV color center with modification diamond has also been investigated. For example, Flatae et al. [89] reported that the SiV color centers are promising candidates for single-photon emission in phosphorus-doped diamond. Iwasaki et al. demonstrated that the GeV color center in diamond showed a sharp and strong luminescence band with a zero phonon line at around 602 nm. By using the first-principles calculation, they also predicted that the GeV center has the same split-vacancy crystal structure as the SiV center, but showed the emission with the shorter wavelength resulting from the higher, e.g. state in the energy level in the GeV center [90]. Chen et al. [91] demonstrated that the shelving effect induced by the resonant laser in GeV centers, such phenomenon could be explained by the presence of a dark state. Furthermore, they also revealed that the

![Figure 3](image-url)
recovery and stabilization of the resonance fluorescence (RF) could be useful for quantum information science and scalable quantum photonics. Besides, NE8 (nickel-vacancy-nitrogen) and Ti-related color center are also promising candidate materials for realizing high quality single-photon sources in diamond. Although the NE8 exhibited very short lifetimes of 2-3 ns, the narrow emission of the NE8 center around 800 nm makes it promising for quantum key distribution applications [92]. Czelej et al. [93] investigated the electronic structure and magneto-optical properties of Ti-related point defects in diamond by means of the density function theory, which provided a comprehensive understanding of photoluminescence for other transition metal-related defects in diamond. The research of color center in diamond is leading a technological revolution in the field of quantum photonics, which exhibits an indispensable role for the future application in quantum information technology.

3.3 Heterostructures of diamond and other materials

Except for doping and surface modification, heterostructures constructed by diamond and other materials also received special attention. Heterostructures constructing by diamond-based materials can also present their promising application in other aspects, such as substrate and optoelectronic materials. Ma et al. have studied the electronic and magnetic properties of graphene adsorbed on the (111) surface of diamond, see Figure 4 [38]. They unveiled that the finite band gap of graphene could be induced by diamond surface, but the linear band dispersion of graphene could be maintained well. Interestingly, the band gap depended on the stacking configuration, which showed different results for graphene adsorbed on transition metal dichalcogenide (TMD) materials. Additionally, the results also indicated that electronic spin can arise “intrinsically” in graphene because of the exchange proximity interaction between electrons in graphene and localized electrons on the surface of diamond rather than the characteristic graphene states (Figure 4b). Such investigations provide an ideal substrate material for future graphene-based electronics.

It should be noted that diamond can also make deep influence in optoelectronic applications. The type-II band alignment can be observed when diamond is constructed with TiO$_2$ and BN [39,40]. Such alignment can promote the separation of electrons and holes, which shows potential applications in photocatalytic water splitting (Figure 4c and d). Besides, diamond-based field-effect transistors and high power devices are also

![Figure 4](image-url)

**Figure 4.** (a) Band structures of graphene and diamond surface composite systems in different configurations. The orange (light gray) line and dark cyan (medium gray) line represent the spin-up and spin-down states, respectively. The horizontal dashed lines indicate the Fermi level. Insert: the gap of graphene near the K point induced by the diamond surface. The linear energy dispersion of the graphene in the corresponding composite system around the Fermi energy (blue dotted line) is also plotted for reference. (b) Side and top views of the three dimensional charge density difference plots. Green (gray) and yellow (light gray) isosurfaces represent charge depletion and accumulation in the space with respect to isolated graphene and the diamond surface, respectively. Adapted with permission from [38]. Copyright 2012, American Physical Society. (c) Schematic representation of the relative position of the valence band and conduction-band edges of different configurations of composite TiO$_2$/diamond as obtained from the “bulk plus lineup” method. Adapted with permission from [39]. Copyright 2018, AIP Publishing. (d) Schematic representation of the relative position of the valence band and conduction band edges of (111) c-BN/diamond heterostructure with C–B (left) and C–N (right) bonding configurations. Adapted with permission from [40]. Copyright 2019, Royal Society of Chemistry.
investigated both theoretically and experimentally, which provide a practical guidance for next generation electronic devices.

4. Conclusions and outlook

In conclusion, the recent advances in the structure and electronic properties of diamond are reviewed both theoretically and experimentally. Diamond is widely used in semiconductor devices due to the excellent mechanical and opto-electronic properties. Plenty of approaches have been performed to tune its electronic properties due to the large bandgap, including doping, constructing heterostructure and so on. The synthesis and application of p-type diamond with B as the impurity element are relatively mature, the impurity level and carrier transport properties can be well controlled by researches using ion implantation or CVD method. However, the lack of effective n-type conductive diamond materials limits the application of diamond in the field of electronic devices.

In recent years, a lot of theoretical and experimental works have been performed in the field of n-type diamond materials. The doping mechanism and effective doping methods of group I elements, mainly Li and Na, are seldom reported at present and need to be further discussed. Group V elements are used as donor impurities for N and P. However, diamond doped with N does not show the characteristics of N-type diamond at room temperature. Compared with N, P shows a better index and is regarded as an ideal donor impurity. Unfortunately, substances containing P are often highly toxic, which limits its further application. As for group VI elements, S impurity is more ideal than O, but the electron mobility is not high enough. Therefore, it is difficult to prepare n-type diamond films with low resistivity based on a single element. The possibility of co-doping in diamond has been preliminarily explored through different methods. Experiments have proved that the co-doping of B, S or B, P atoms can not only improve the mobility, lattice integrity and the solubility of donor impurity in diamond, but also can reduce the ionization activation energy of diamond. However, the study of electrical properties of co-doped n-type diamond semiconductor need to be further improved.

On the other hand, heterostructure materials constructing by diamond-based materials also show their great application potential in electronic devices. Such materials can not only be used as substrate materials, but also unveil promising application in field-effect transistors and high power electronic devices.

Disclosure statement

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