

Cubic silicon carbide as a potential photovoltaic material

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Abstract

In this work we present a significant advancement in cubic silicon carbide (3C-SiC) growth in terms of crystal quality and domain size, and indicate its potential use in photovoltaics. To date, the use of 3C-SiC for photovoltaics has not been considered due to the band gap of 2.3 eV being too large for conventional solar cells. Doping of 3C-SiC with boron introduces an energy level of 0.7 eV above the valence band. Such energy level may form an intermediate band (IB) in the band gap. This IB concept has been presented in the literature to act as an energy ladder that allows absorption of sub-bandgap photons to generate extra electron-hole pairs and increase the efficiency of a solar cell. The main challenge with this concept is to find a materials system that could realize such efficient photovoltaic behavior. The 3C-SiC bandgap and boron energy level fits nicely into the concept, but has not been explored for an IB behavior.

For a long time crystalline 3C-SiC has been challenging to grow due to its metastable nature. The material mainly consists of a large number of small domains if the 3C polytype is maintained. In our work a crystal growth process was realized by a new approach that is a combination of initial nucleation and step-flow growth. In the process, the domains that form initially extend laterally to make larger 3C-SiC domains, thus leading to a pronounced improvement in crystalline quality of 3C-SiC. In order to explore the feasibility of IB in 3C-SiC using boron, we have explored two routes of introducing boron impurities; ion implantation on un-doped samples and epitaxial growth on un-doped samples using pre-doped source material. The results show that 3C-SiC doped with boron is an optically active material, and thus is interesting to be further studied for IB behavior.

For the ion implanted samples the crystal quality was maintained even after high implantation doses and subsequent annealing. The same was true for the samples grown with pre-doped source material, even with a high concentration of boron impurities.

We present optical emission and absorption properties of as-grown and boron implanted 3C-SiC. The low-temperature photoluminescence spectra indicate the formation of optically active deep boron centers, which may be utilized for achieving an IB behavior at sufficiently high dopant concentrations. We also discuss the potential of boron doped 3C-SiC base material in a broader range of applications, such as in photovoltaics, biomarkers and hydrogen generation by splitting water.

Keywords: intermediate band, silicon carbide, solar cell, photovoltaic, boron, doping, 3C-SiC, cubic

Introduction

The solar cell market is today dominated by single junction silicon solar cells with up to 91 % of the global production with respect to power output. The single junction entails that the cell can only convert a fraction of the solar spectrum into electrical current, limiting its maximum theoretical efficiency at around 29 % in the case of silicon. The efficiency can be increased by stacking several materials with increasing bandgap on top of each other to create a multiple junction solar cell. However, these combined material structures are typically too expensive for the general market and used mostly in niche markets, like space exploration. An attractive alternative concept is to create a metal-like intermediate band (IB) in the band gap (E_g) of a semiconductor by energy levels which form a band [1] or achieve extra carrier generation by impurity doping [2]. This would allow for a larger part of the solar spectrum to contribute to the electrical current since the IB would utilize photons that have lower energy than E_g . In this concept, first a sub- E_g photon generates an electron that is excited from the valence band to the IB, and a second sub- E_g photon excites the electron from the IB to the conduction band. This generates additional electron-hole pairs to the ones given by absorption of photons which generate electrons from the valence band to the conduction band. In total, the efficiency of a solar cell can increase substantially.

There are several material candidates for such IB behavior [3], but most are still struggling with growth conditions and/or finding appropriate deep levels which could create an efficient photovoltaic material. Cubic SiC (3C-SiC) is unique among other candidates as it combines nearly ideal bandgap for an IB solar cell ($E_g \sim 2.3$ eV) with excellent electronic properties and readily available deep centers. In particular, it can be doped with boron that introduces an energy level of 0.7 eV above the valence band [4]. An early attempt to use 3C-SiC for the increased carrier generation by boron doping was presented in 2003, but the material quality of 3C-SiC at that time was not sufficient [5]. To circumvent this limiting factor, we have developed a sublimation epitaxial growth approach that applies a lower ($<2000^\circ\text{C}$) growth temperature in comparison to the physical vapor transport (PVT) method that is generally used to produce hexagonal silicon carbide (6H- and 4H-SiC). The growth is carried out in vacuum and ensures an efficient transfer of vapor and dopant species from the source to the substrate. Our 3C-SiC growth approach allows us to control initial nucleation of 3C-SiC domains which laterally enlarge and completely cover the substrate surface [6]. Thereby a 3C-SiC material with very few domains is formed and a high crystalline quality is achieved.

The growth of 3C-SiC has for long been challenging due to its metastable nature. The 3C-SiC nucleation is not fully understood, but may be attributed to the supersaturation, concentration of various impurities, polarity of the substrate or influence of crystallographic defects. In general, the highest yield of 3C-SiC has been obtained by growth on nominally on-axis 6H or 4H-SiC substrates. On such substrates the growth proceeds via a spontaneous two-dimensional nucleation of 3C-SiC domains and their enlargement [7]. In this case the control of initial nucleation of 3C-SiC and polytype stability is very difficult. The 3C-SiC domains having different rotations can nucleate all over the surface leading to formation of high density of structural defects. The advantage is that 3C-SiC forms in a high yield and 100% could be obtained without any inclusions of the on-axis substrate polytype [7]. However, it is a pronounced challenge to obtain large domains, and domain sizes were less than a millimeter even though some growth parameters could influence the lateral domain expansion [8]. A new 3C-SiC growth approach on off-oriented 4H-SiC allows controlling initial nucleation of 3C-SiC and significantly reduce the density of structural defects [6]. Moreover, it has been demonstrated that this approach allows very high polytype stability and excellent reproducibility.

In this paper we have used two approaches to introduce boron into crystalline 3C-SiC: (i) boron doping during homoepitaxy on seeds; (ii) ion implantation. The materials are studied by optical methods to estimate the potential of 3C-SiC as intermediate band material.

Material and methods

The sublimation growth of 3C-SiC was performed in a graphite crucible heated by RF generator at a frequency of 46 kHz. At elevated temperatures vapour species (mainly Si, Si₂C and SiC₂) sublime from an undoped polycrystalline (ceramic) source wafer and are transferred to the substrate where they form the SiC film. The driving force for such transfer is a temperature gradient between the source and the substrate. It has been shown that this type of sublimation epitaxial configuration may reach a very high growth rate (up to 2 mm/hr at temperatures up to 2000°C) [8]. Such growth rates are attractive for a production of 3C-SiC material for photovoltaics. In our case, we have used a moderate the growth rate but high enough to prepare a free standing nominally undoped 3C-SiC substrate [6] for ion implantation or subsequent homoepitaxial growth using a boron doped source. In the latter case the boron doped polycrystalline source materials were fabricated using a PVT bulk method with a mixture of boron carbide powder and SiC carbide powder. Wafers were prepared from the boules and used as boron doped source material for the homoepitaxial growth of 3C-SiC layers on the previously grown 3C-SiC free standing material in a subsequent sublimation epitaxial process. This source preparation process is explained in detail elsewhere [9].

Transmittance measurements were performed at room temperature using UV-VIS spectrophotometer and the results were used to derive the optical absorption properties. Photoluminescence (PL) measurements were carried out by employing a continuous wave HeCd laser with a wavelength of 325nm as an excitation source. The emission was collected by a microscope and analyzed with a spectrometer system with minimal resolution 0.2 nm. PL measurements were performed at 10K temperature using closed-cycle He-refrigerator. Transmission electron Microscopy (TEM) was used for atomic scale crystallography analysis.

Results and discussion

The improvement in 3C-SiC material quality when going from on-axis growth to off-axis growth can clearly be seen from the comparison of corresponding optical micrographs in Fig. 1. On the left hand side the on-axis deposited 3C-SiC sample has formed numerous small domains that would be detrimental to any electronic devices formed on it. On the right hand side, a sample that was grown on 4° off-axis surface formed only 2-3 large domains which would expect to yield significantly improved material quality and electrical performance as was indicated in [10].

The 3C-SiC is well known for its tendency to generate stacking faults to release energy [11]. This creates a pronounced challenge in growth of material with high crystalline quality. Due to the considerable thickness of the 3C-SiC samples, there are bulk-like features of various macroscopic crystal defects. However, the TEM image in Figure 2 clearly shows that crystallographic perfection is possible to achieve. A forthcoming main challenge in the growth process is to achieve this for a large part of the sample.

For long it has been unclear if 3C-SiC could be grown in similar quality like hexagonal 6H and 4H-SiC which are commercially available. In previous work, we have demonstrated that 3C-SiC of sufficient quality can be grown to obtain high carrier lifetime. The lifetime is one of the key parameters governing the electronic and optoelectronic devices, and it is very sensitive to the crystal quality. A carrier lifetime of 8.2 μs , which may be compared with 3C-SiC grown on silicon in which the carrier lifetime is ranging from a few to 120 nanoseconds, was reported [12]. The typical lifetime in as-grown 4H-SiC is around 1 μs under the low injection level, while higher values was reported to be enhanced from 0.69 to 9.5 μs after thermal treatment [13] or from 3.5 to 18-19 μs post carbon-implantation and annealing [14]. Thus a growth process that reproducibly maintains such quality could open up 3C-SiC for electronic and optoelectronic devices. In any case, there should be no fundamental limit in growth of 3C-SiC even though the stacking fault generation is higher than in 6H and 4H-SiC. It is also noteworthy at this point that presence of stacking faults in 3C-SiC is less destructive for carrier lifetimes compared to that in hexagonal 4H and 6H-SiC polytypes [15].

The 3C-SiC samples grown in this work show similar semiconductor characteristics as 6H and 4H-SiC, but have higher resistivity due to boron doping compensating the background n-type doping given by nitrogen, leading to decreased carrier concentrations.

In previous work since 1995 the nitrogen (being a shallow donor) concentration in undoped 6H and 4H-SiC, as measured using secondary ion mass spectroscopy (SIMS), was found to be from low 10^{16} cm^{-3} to mid 10^{17} cm^{-3} . Nitrogen is always present in the graphite environment that adsorbs nitrogen from air at the loading stage, as well as being present in the source material. Since the concentration of boron (a deep acceptor) in the source material is less than 10^{16} cm^{-3} , the boron is unable to overcompensate the nitrogen doping and thus the resultant nominally 3C-SiC samples are expected to be n-type. Fig. 3 shows the voltage-current curves of as-grown 3C-SiC. The current increases linearly with voltage indicating that the indium contacts used are ohmic. Hall-effect measurements reveal that the as-grown 3C-SiC samples are of n-type (the results not shown). The carrier density of the as-grown 3C-SiC is around $2 \times 10^{16} \text{ cm}^{-3}$. The resistivity of the as-grown 3C-SiC is around $\sim 18 \pm 1 \text{ } \Omega \cdot \text{cm}$.

In this work, an extended approach combining spatial, temporal and spectral features was adopted for a comprehensive optical characterization of the undoped and boron doped 3C-SiC material. The low temperature photoluminescence spectra of undoped 3C-SiC exhibit a set of intense sharp lines between 2.39 and 2.26 eV. These are assigned to recombination processes

involving annihilation of excitons bound to neutral nitrogen donor along with corresponding momentum conserving (TA, LA, TO, and LO) phonon replicas (details not shown in Fig. 4). The high crystal quality of the 3C-SiC crystal is evidenced by the sharpness and multiplicity of the observed excitonic lines. This was further validated by PL imaging (2D mapping of structural homogeneity/defects) and time-resolved PL measurements, indicating accordingly the prevalence of large domains and the high effective carrier lifetimes of the order of microseconds.

The optical activity of the incorporated B atoms into 3C-SiC host by ion implantation was deduced from the evolution in absorption and emission spectra monitored by transmittance and PL measurements of the samples before and after post-implant annealing. The PL spectra of as-implanted and post-annealed (1500°C) 3C-SiC:B are compared in Fig. 4a where, besides the typical for high-temperature treatment spectral features (D1, D2, G lines), the newly emerging emission band at around 1.6 eV is associated with activation of deep B-centers participating in donor-to-acceptor pair and free-to-bound optical transitions. The transmittance measurements of as-implanted and post-annealed 3C-SiC:B further support this assumption, as can be seen from the comparison of absorption spectra in Fig. 4b. The apparent build-up of absorption band centered at around 1.6 eV is attributed to implantation-induced deep centers, presumably caused by B atoms replacing C atoms after annealing. The results attained so far evidence the high crystallinity of 3C-SiC and clearly indicate the formation of optically active deep boron centers. The integrity of emission and absorption properties associated with the deep boron centers presented in Figures 4a and b proposes that further study of boron may show the feasibility of IB formation in 3C-SiC. It should be noted at this point that a two-photon experiment [16] is considered to be performed once boron doping of 3C-SiC is optimized to provide an unambiguous proof of the IB operation.

The notion of optically active states is further supported by 3C-SiC which has been doped during epitaxy with boron from a source material. The electron transition from boron level to conduction band has been demonstrated by the absorption spectra in boron doped epilayers. As seen in Fig. 5, the unintentionally doped 3C-SiC only shows the sharp band edge absorption. As a comparison, the intentionally boron doped 3C-SiC clearly exhibits a broad band centered around 730 nm (1.7 eV). This absorption band is attributed to the transition between the boron acceptor to the conduction band. From the boron related absorption band, the boron energy level is estimated to be localized at ~0.7 eV above the valence band. This is consistent with the previous reported boron acceptor level [4].

There is emerging interest in silicon carbide for optoelectronics. Recently, fluorescent silicon carbide using boron doped 6H-SiC was introduced for a white light emitting diode concept for general lighting [17,18]. Besides attractive to explore in photovoltaics, hydrogen generation by water splitting can be developed by using cubic silicon carbide as a photo-electrode to absorb and convert solar energy into gaseous hydrogen and oxygen via a photoelectrochemical (PEC) water-splitting cell [19]. Among all commonly used semiconductors, 3C-SiC has outstanding properties to convert visible sunlight energy and water into hydrogen fuel. The 3C-SiC has outstanding properties such as excellent mechanical strength, chemical inertness and high saturated drift velocity, and high electron mobility. Most importantly, 3C-SiC is almost a perfect material to meet all criteria for PEC water splitting. The 3C-SiC has an indirect bandgap of 2.36 eV at room temperature, which is larger than the minimum energy value of 1.9 eV required to split water and also satisfies the requirement to absorb the visible sunlight. The 3C-SiC fulfills the requirement to straddle the reduction and oxidation potentials of water splitting. The conduction band edge of 3C-SiC is located above the reduction potential of H₂/H₂O while

the valence band edge is positioned below the oxidation potential of H₂O/O₂. This results in the spontaneous (without any external electric bias) water splitting at 3C-SiC photo-electrode. Also, it is relatively easier to grown p-type 3C-SiC, which acts as photocathode to form H₂, consequently offers some protection against photo-corrosion. Lastly, 3C-SiC also enables efficient charge transport because of its high electron mobility of 1000 cm²/Vs and hole mobility of 320 cm²/Vs. A high mobility is especially desirable in photoelectrodes with an indirect bandgap, because these materials require a large thickness to absorb all the incident light, which means that photo-generated electrons and holes have to travel large distances before reaching the interface.

Finally, there is a potential approach of using graphene on the 3C-SiC. The graphene on 6H-SiC and 4H-SiC has shown prospective results in quantum Hall resistance [20]. Recently a comparison of graphene on the SiC polytypes 3C-SiC, 6H-SiC and 4H-SiC was presented [21]. The nature of the stacking sequence and surface characteristics may create another approach using 3C-SiC due to the difference between hexagonal and cubic polytypes.

Conclusions

The 3C-SiC is a potential material for a range of applications. The boron doped 3C-SiC seems promising to further explore in the intermediate band concept. We have demonstrated the possibility to achieve high crystalline quality of the material. Two approaches of boron doping indicates the potential of boron 3C-SiC as an optoelectronic material. Further research could resolve the use of solar driven water splitting to generate hydrogen, and use graphene on 3C-SiC.

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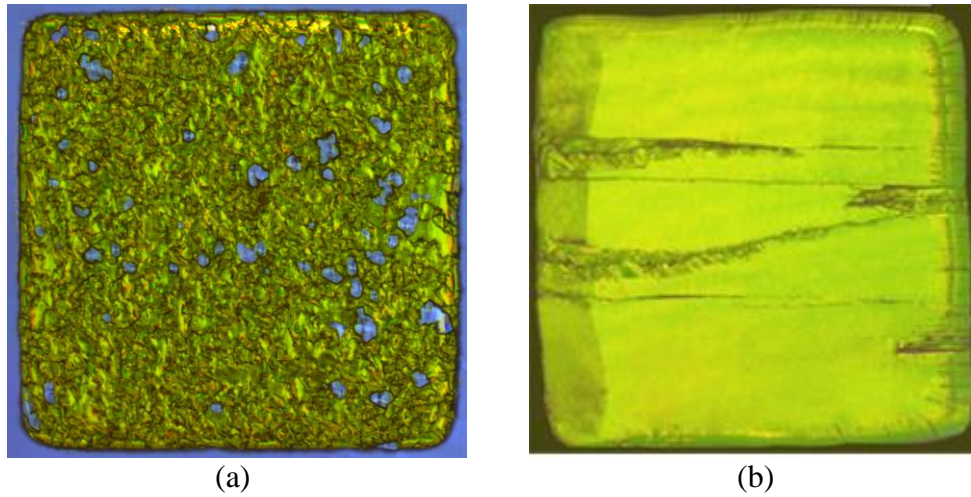


Figure 1. A 3C-SiC layer grown on: (a) nominally on-axis 4H-SiC substrate with a large number of small domains, (b) 4 degrees off-oriented 4H-SiC substrate with only 2-3 large domains. The size of both layers is $7 \times 7 \text{ mm}^2$.

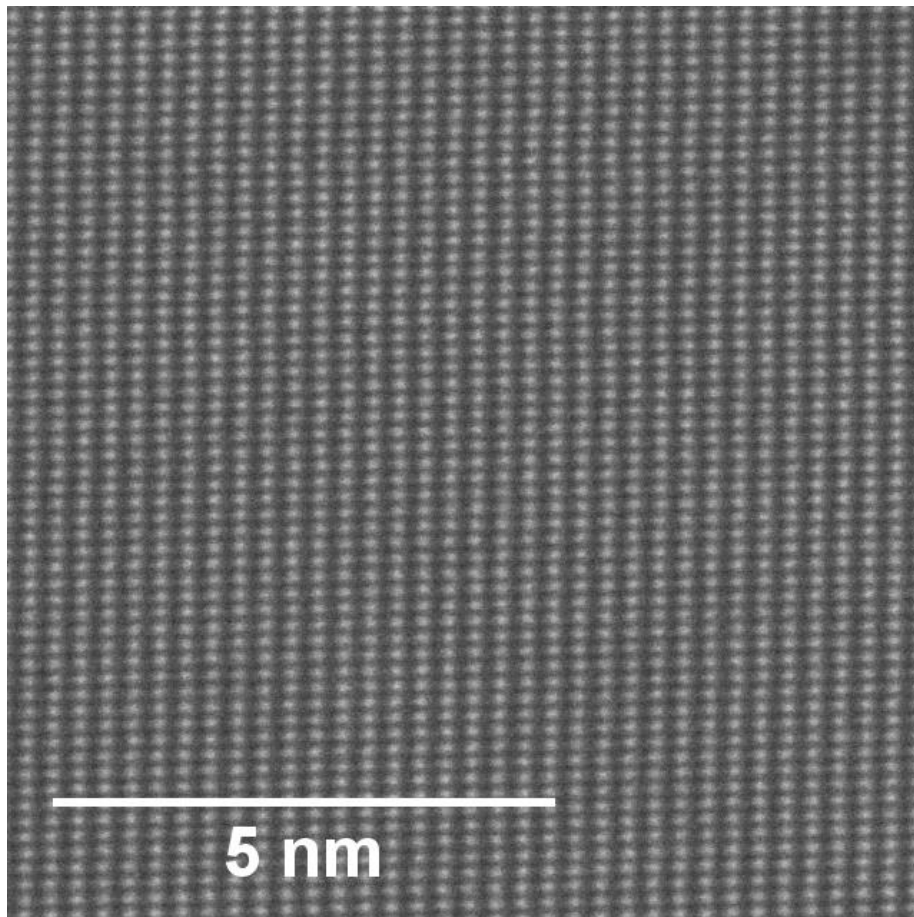


Fig. 2. TEM images showing a perfect alignment of atoms in 3C-SiC showing that areas of high perfection are possible to achieve.

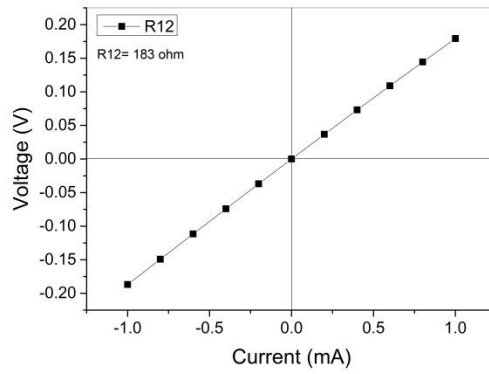


Fig. 3. I-V measurement of 3C-SiC using ohmic contact scheme as applied for 4H-SiC.

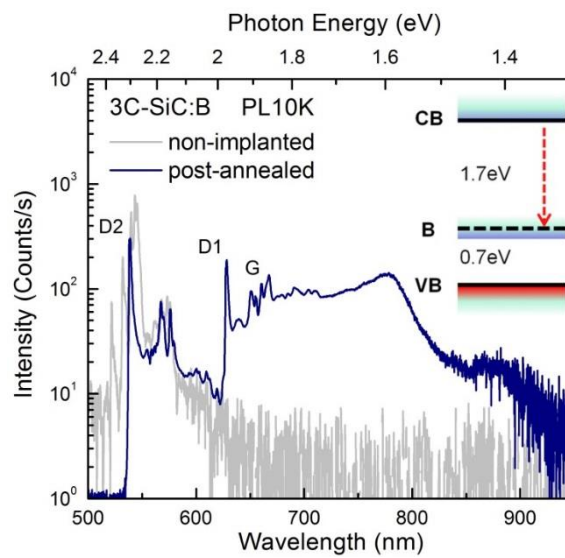


Fig 4a. PL spectra obtained at 10K of B-implanted and post-annealed 3C-SiC:B compared with a virgin material. The emerging emission at around 1.6 eV after annealing is associated with activation of deep B-centers ($E_A(B) \sim E_V + 0.7$ eV) participating in donor-to-acceptor pair and free-to-bound optical transitions.

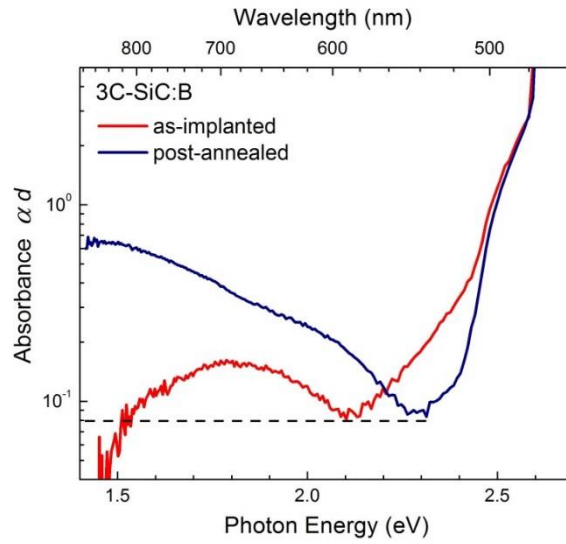


Fig 4b. Room-temperature absorbance spectra of as-implanted and post-annealed (1500C) 3C-SiC:B. Note a considerable, by an order of magnitude, build-up of absorption band centered at around 1.5 eV attributed to activation of deep B-centers upon annealing.

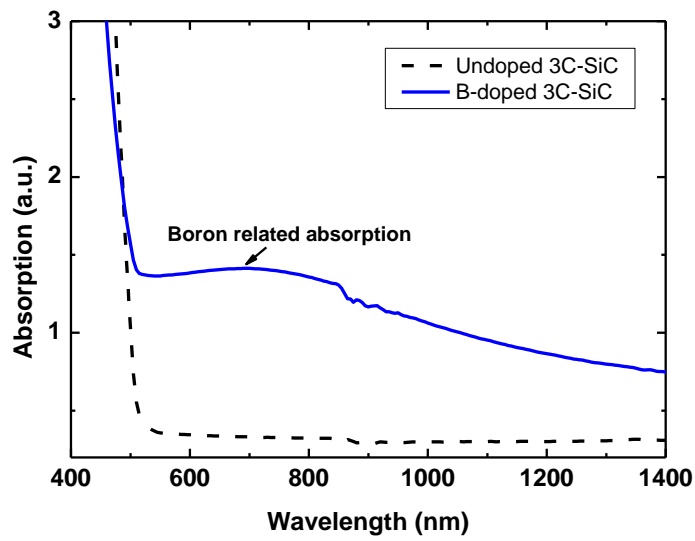


Figure 5. Absorption spectra of the unintentionally doped and boron doped 3C-SiC.