Research Article

Analysis of Electrical Characteristics of Pd/n-Nanocarbon/p-Si Heterojunction Diodes: By C-V-f and G/ω-V-f

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1. Introduction

Carbon is a unique chemical element because it morphs into up to 32 allotropes, such as graphite and diamond, which represent $sp^2$- and $sp^3$-orbital hybridization, respectively. Recently, diamond has triggered many research activities for new-type optoelectronics and high temperature-high frequency devices [1, 2]. However, the formation of $n$-type diamond by nitrogen doping has always been one of the biggest struggles in diamond field, due to the deep nitrogen donor level (1.7 eV) as well as the nitrogen-associated defects that influence the carrier transport [3]. In contrast, ultrananocrystalline diamond (UNCD) can be doped with nitrogen with low activation energy wherein the $n$-type conduction primarily related to the grain boundary network [4].

It was found that nitrogen dopants create additional electric conduction paths in UNCD films [5]. Ikeda et al. [6] correlated the enhancement of electrical conductivity of nitrogen-doped UNCD films to the enlargement of ordering and fraction of $sp^3$-carbon at the grain boundary. Mertens et al. [7] have studied the electrical conductivity of nitrogen-doped UNCD films. Their results suggest that the $n$-type conduction depends on the structure and volume of the grain boundary.

Although UNCD/a-C:H:N films are mainly grown by the chemical vapor deposition (CVD) method [8, 9], the authors have previously grown them by coaxial arc plasma (CAPD) [10] and pulsed laser deposition (PLD) [11]. In our previous works, we studied $n$-type (UNCD/a-C:H:N)/$p$-type Si HJJs by the CAPD method [12–14]. The fabricated diode...
exhibited a rectifying action with $10^4$ rectification ratio in the bias range $\pm 1$ V. Additionally, the Pd/$n$-(UNCD/a-C:H:N)/$p$-Si photodiodes showed a capability of deep ultraviolet light detection under the 254 nm monochromatic light [14, 15]. In addition, we investigated the origin of dark current in Pd/$n$-(UNCD/a-C:H:N)/$p$-Si HJDs based on an established comprehensive computer device model [16].

In prior reports, we characterized the Pd/$n$-(UNCD/a-C:H:N)/$p$-Si HJDs by employing I-V measurements at room and low temperatures. However, the effects of interface state density ($N_{ss}$) and series resistance ($R_s$) on their electrical characteristics were not investigated. Practically, the $N_{ss}$ and $R_s$ can be studied by the low-frequency capacitance ($C_{1f}$), Hill-Coleman, and Nicollian and Brews techniques [17, 18]. Many researchers have proposed the frequency-dependent $C-V$ and $G/\omega-V$ method as being effective to determine the electrical characteristics of HJDs composed of various materials including diamond [19], graphene [20], ZnGaSe [21], and ZnGaTe [22]. Although $C-V$ and $G/\omega-V$ measurements in ideal diodes are frequency independent [23], $N_{ss}$ and $R_s$ significantly influence the device characteristics and cause considerable deviation from ideality [24].

The aim of this study is to investigate the influence of $R_s$ and $N_{ss}$ on the electrical characteristics of Pd/$n$-(UNCD/a-C:H:N)/$p$-Si HJDs. Therefore, frequency-dependent $C-V$ and $G/\omega-V$ measurements were performed over the wide-range frequency of 40 kHz-2 MHz at room temperature.

## 2. Experiments

### 2.1. Film Deposition and Structural Investigations

400 nm thin-nitrogen-doped UNCD/a-C:H:N films (undoped films for comparison) were grown in nitrogen and hydrogen gases atmosphere of 53.3 Pa at a substrate temperature of 550°C by coaxial arc plasma deposition (CAPD) using a pure graphite target. An arc plasma gun connected with (720 μF) capacitor was conducted at 5 Hz repetition rate. In order to investigate nitrogen atoms incorporation and estimate their contents into the deposited films, X-ray photoemission spectroscopy (Mg Ka line, $h\nu = 1253.6$ eV) has been employed. In this study, the nitrogen content of the film deposited at a nitrogen/hydrogen ratio ($I_{N/H}$) of 0.25 was estimated to be 3 at.%, by similar method in our previous report [25]. In addition, Fourier transform infrared (FTIR) spectra were recorded in the middle of the infrared range from 400 to 4000 cm$^{-1}$ for deposited films, and the CH band spectra were deconvoluted into the Gaussian peaks.

### 2.2. Fabrication and Characterization of the Heterojunction Diodes

In order to fabricate the heterojunction diodes, $n$-type (UNCD/a-C:H:N) films were grown onto (111) $p$-type (boron-doped) single crystal silicon (Si) by the CAPD method. $p$-Type Si substrates, having a thickness of 200 μm and 1.3-3 Ω cm resistivity, were treated by (CH3) 2CO (acetone), CH3OH (methanol), and distilled water for 5 min, sequentially. For ohmic contact, palladium (Pd) electrodes were sputtered from pure Pd target (99.99%) using a radio-frequency magnetron sputtering apparatus. Pd electrodes with a thickness of 120 nm were deposited on the back side

### 3. Results and Discussion

#### 3.1. Structural Investigations: By XPS and FTIR

XPS is a powerful tool to estimate the amount ratio between $sp^2$ and $sp^3$ bonds in their coexistence materials. Figure 1 depicts wide-scan XPS spectra of undoped and nitrogen-doped UNCD/a-C:H films with sharp C1s peaks, in addition to O1s peaks originated from the film surface. The nitrogen-doped UNCD/a-C:H:N film revealed an extra peak at 409 eV referring to N1s peak [26], which confirms the incorporation of nitrogen atoms into the UNCD/a-C:H film from the hydrogen/nitrogen ambient gases inside the CAPD chamber. To investigate the chemical structure in more detail, C1s photoemission spectrum was recorded using synchrotron radiation.

In Figure 2(a), the XPS C1s core spectra of the undoped and nitrogen-doped UNCD/a-C:H films are shown. The spectra depict slightly broader peak of the nitrogen-doped film compared with that of the undoped UNCD/a-C:H film. This observation refers to the formation of the peaks at 284.4, 285.4, 286.7, and 287.6 eV, which are assigned to $sp^2$-C bonds, $sp^3$-C bonds, Cn bonds, and C=O bonds, respectively.
Figure 2(b) illustrates the suggested mechanism of nitrogen incorporation into grain boundaries by replacing hydrogen atoms.

Figure 3 shows the FTIR spectra of undoped and nitrogen-doped UNCD/a-C:H films in the range of 2750-3100 cm⁻¹, which contain different superpositioned CHₙ (n = 1, 2, 3) vibration peaks. The spectra were deconvoluted for Gaussian peaks based on our earlier study [28], and the positions of separated peaks are summarized in Table 1. Compared to the undoped film, the FTIR spectrum of N-doped UNCD/a-C:H divulged two extra peaks at 2844 and 3049 cm⁻¹, attributable to nitrogen impurities [29] and aromatic nucleus CH stretching vibration [30], respectively. In addition, the sp²-CH and sp³-CH₂ peaks, which might originate from hydrogen atoms, are clearly weakened, which implies that hydrogen atoms at the grain boundaries are replaced by nitrogen atoms. These results indicate that the nitrogen doping not only forms carbon-nitrogen bonds (C–N and C≡N) but also expedites the sp²-CHₙ, which is

Table 1: Summary of the peak positions in FTIR spectra and assigned vibration mode of undoped and nitrogen-doped UNCD/a-C:H films.

| Wavenumber (cm⁻¹) | Vibration mode               |
|------------------|------------------------------|
| 2844             | N impurities                 |
| 2854             | sym. sp³-CH₂                 |
| 2884             | sym. sp³-CH₃                 |
| 2905             | sp²-CH                      |
| 2928             | sym. sp³-CH₂                 |
| 2955             | Asym. sp³-CH₃                |
| 2976             | Olefinic sp²-CH₂             |
| 3000             | Olefinic sp²-CH              |
| 3021             | Aromatic sp²-CH              |
| 3038             | Aromatic sp²-CH              |
| 3049             | Aromatic sp²-CH              |
| 3082             | Aromatic sp²-CH₃             |

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illustrated in Figure 2(b). The suggested mechanism and FTIR results are inconsistent with our previous study [31] which showed the shrinking optical band gap of UNCD/a-C:H films after nitrogen incorporation, due to the increase of the sp² fractions.

3.2. Capacitance (C-V-f) and Conductance (G/ω-V-f) Characteristics. It is known that p–n or the Schottky junctions containing deep impurities have significant frequency-dependent capacitance profile [20], and the capacitance vs. frequency relation can determine the characteristics of deep levels in the space-charge region (SCR) of the junction. Figures 4(a) and 4(b) illustrate the C-V and G/ω-V characteristics of Pd/n-(UNCD/a-C:H:N)/p-Si HJDs at different frequencies at room temperature. It can be seen that both C and G/ω depend on frequency and bias voltage, which refers to the influence of Nss and Rs [24]. The existence of Nss and Rs can be ascribed to the disruption of the lattice structure at the surface of the fabricated junction as well as the defects on the (UNCD/a-C:H:N)/Si interface [23]. The measured capacitance in Figure 4(a) exhibited a peak in each frequency, shifting to forward bias with increasing frequency until it vanishes at high frequencies. The observed C-V-f curves refer to the existence of different types of interface states, and that the capacitance at low and intermediate frequencies can follow the ac signal but cannot follow at high

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**Figure 4:** (a) C-V and (b) G/ω-V characteristics of Pd/n-(UNCD/a-C:H:N)/p-Si HJDs.

**Figure 5:** The frequency-dependent (a) C-V and (b) G/ω-V characteristics of Pd/n-(UNCD/a-C:H:N)/p-Si HJDs.
frequencies [32]. Almost same trend can be observed for the \( G/\omega-V-f \) in the depletion region in the same frequency range as displayed in Figure 4(b).

To accurately analyze the influence of ac applied frequencies on the capacitance and conductance profiles, Figures 5(a) and 5(b) illustrate the measured \( C \) and \( G/\omega \) of the Pd/\( n \)-(UNCD/a-C:H:N)/p-Si HJD as a function of frequency in the range of 40 kHz to 2 MHz at different voltage biases. As mentioned earlier, \( C \) and \( G/\omega \) are ideally frequency independent, and the deviation from that ideality is mainly due to the existence of \( N_{ss} \) and \( R_s \). In Figure 5, both \( C \) and \( G/\omega \) are firmly frequency dependent, especially at low frequencies. The \( C-V-f \) profile of the Pd/\( n \)-(UNCD/a-C:H:N)/p-Si HJD is consistent with previous studies [33, 34] as follows: at low frequencies, the charges at traps or interface states can follow the ac signal, consequently, and they gain extra capacitance and conductance to the recorded \( C \) and \( G/\omega \) values. On the other hand, at sufficiently higher frequencies, interface states are not capable to follow the ac signal. This behavior is observed for the \( G/\omega-f \) profile, which indicates the presence of different time-dependent responses of \( N_{ss} \).

### 3.3. Series Resistance \((R_s)\) and Interface States \((N_{ss})\) Analysis

Practically, the \( R_s \) value of HJDs can be calculated from the capacitance and conductance values at high frequency, by utilizing Nicollian and Goetzberger method [35, 36]:

\[
R_s = \frac{G_m}{(\omega C_m)^2 + G_m^2}
\]

where \( C_m \) and \( G_m \) are the measured capacitance and conductance values in the strong accumulation region, respectively. Figure 6(a) showed the biasing voltage dependency of series resistance. \( R_s \) exhibits the peaks in the depletion region (-1 V to +1 V) at low frequencies (\( \leq 100 \) kHz), and the peak magnitude decreased from 80 \( \Omega \) to 40 \( \Omega \) when the frequency increased from 40 kHz to 100 kHz, respectively. Moreover, the peak decay accompanied by shifted positions towards the accumulation region with increasing frequency. Demirezen et al. [37] explained this observation as a result of reordering and restructuring of carrier charges in the traps under electric field or applied bias voltage. Additionally, Kaya et al. [38] suggested that the voltage-dependent \( R_s \) is ascribed to voltage-dependent charges such as interface charge, mobile oxide charge, or oxide-trapped charge. The variation of the series resistance as a function of frequency in the strong accumulation region is plotted in Figure 6(b). It can be seen that \( R_s \) has a minimum value at the highest applied frequency (2 MHz). This behavior could be attributed to the distribution density of interface states and their frequency-dependent behavior [23, 39].
Additionally, the Hill-Coleman method is adopted to estimate the $N_{ss}$ values from the measured $C-V$ and $G/\omega-V$, by equation (2) [18, 40]:

$$N_{ss} = \frac{2}{qA} \frac{(G_{ma}/\omega)_{\text{max}}}{\left(1 - C_{\text{max}}/C_{\text{ox}}\right)^2 + \left((G_{ma}/\omega)_{\text{max}}/(C_{\text{ox}})^2\right)}$$

Figure 7 represents the frequency dependency of $N_{ss}$ at room temperature. At low and intermediate frequency ranges, $N_{ss}$ has a value of $1.7 \times 10^{15}$ eV$^{-1}$ cm$^{-2}$ at 40 kHz which decreases rapidly with increasing frequency to the minimum value of $2 \times 10^{14}$ eV$^{-1}$ cm$^{-2}$ at 400 kHz. Above 500 kHz, the $N_{ss}$ reveals an almost frequency-independent behavior. The calculated $N_{ss}$ values from $C-V$ measurements are inconsistent with the values extracted from the experiment and simulation of current-voltage ($I-V$) results previously [16], which indicate a value of defect density of at least $5 \times 10^{17}$ eV$^{-1}$ cm$^{-3}$ in our Pd/n-(UNCD/a-C:H:N)/p-Si diode. The results in Figure 7 associate the high value of $C$ and $G/\omega$ at low frequency with the extra capacitance and conductance due to the $N_{ss}$. In Pd/n-(UNCD/a-C:H:N)/p-Si
HJD, the interface density can be decreased experimentally by passivation of the UNCD/a-C:H:N layer in the film and at the interface.

3.4. Analysis of the Impedance Spectrum for the Pd/n-(UNCD/a-C:H:N)/p-Si HJD. Impedance spectroscopy is a powerful tool for analyzing the electrical/dielectric properties and for investigating the relaxation process of the electronic devices [41]. It is employed to measure the response of the material to a small amplitude excitation over a range of frequencies (spectrum). Equation (3) describes the complex impedance:

\[ Z' + iZ'' = ReZ + iImZ, \]

where \( ReZ \) is the real part and \( ImZ \) is the imaginary part of the complex impedance. Figure 8(a) shows the frequency dependence of real part \( (ReZ) \) with the applied ac voltages at the frequency range of 10 Hz to 2 MHz. The \( ReZ \) term gradually decreases with increasing applied frequency at the lower frequency, while it revealed a plateau-type behavior at higher frequencies. The results of \( ReZ \) can be ascribed to two types of resistance existing in the frequency regions; at low frequency, the resistance is corresponding to recombination resistance \( (R_{rec}) \), while at high frequency, it is corresponds to \( R_s \).

The variation of imaginary part of impedance \( (ImZ) \) versus the applied frequency is illustrated in Figure 8(b) at different ac voltages. It is observed that the magnitudes of \( ImZ \) increase with applied frequency until they reach to a maximum peak \( (Z_{max}) \) and then decrease with a further increase of the frequency. It can be seen that the peak of \( Z_{max} \) shifts towards the higher frequency region by increasing the applied ac voltages. However, at higher frequency, the curves merge together, which may be caused by accumulation or emergence of space-charge polarization in the Pd/n-(UNCD/a-C:H:N)/p-Si HJD. This is due to the electrons injected from \( n-\)UNCD/a-C:H:N layer to space-charge region in p-Si side [42].

Impedance spectra of the Pd/n-(UNCD/a-C:H:N)/p-Si HJD can be obtained from the Cole–Cole plot of the \( ImZ \) versus the \( ReZ \) in the frequency range of 40 kHz to 2 MHz under different applied AC voltages (from 0 to 1.8 V), as shown in Figure 8(c). The figure depicts a single-semicircle, which indicates a single-carrier device [43]. The radius of the semicircle is related to the total impedance of the device and decreases as the bias increases. The semicircular shape of the impedance spectra in Figure 8(c) indicates that the Pd/n-(UNCD/a-C:H:N)/p-Si HJD can be represented by an equivalent circuit with a constant phase element (CPE). The CPE impedance \( (Z_{CPE}) \) can be given by equation (4) [44]:

\[ Z_{CPE} = \frac{1}{\sigma (j\omega)^{\alpha}}, \]

where \( j = (-1)^{1/2}, \sigma \) represents ac electrical conductivity, and \( \alpha \) represents the frequency dependence. Pleskov et al. [45] reported that a CPE (with a close to 1) is a characteristic component of the equivalent circuit of boron-doped diamond.

4. Conclusions

In this study, we have grown nitrogen-doped UNCD/a-C:H:N films by CAPD technique in ambient H2:N2 gaseous mixture. The incorporation of nitrogen into the films have been confirmed by XPS and FTIR measurements. The results indicate the incorporation of nitrogen atoms into the grain boundaries (a-C:H:N) and increase the \( sp^2 \) fractions. The constructed heterojunction diodes comprising of the deposited \( n \)-type (UNCD/a-C:H:N) films onto \( p \)-type Si substrates were examined by room-temperature \( C-V-f \) and \( G/\omega-V-f \) in the frequency range of 40 kHz-2 MHz. At low frequencies, the results indicated that the charges at traps could easily follow the ac signal. The bias-dependent profiles \( R \) exhibited peaks in the depletion region (-1 V to +1 V), which decreased and shifted towards the accumulation region with increasing frequency. This is thought to be due to the restructuring of carrier charges in traps. The high values of \( C \) and \( G/\omega \) were ascribed to the increase in the value of capacitance and conductance that resulted from the \( N_{at} \), which can follow the ac signal at low frequency, to be almost frequency independent at sufficient high-applied frequencies. Results of frequency-dependent impedance spectroscopy exhibited a plateau-type behavior at high frequencies, corresponding to the series resistance. These results show that nitrogen-doped \( n \)-type (UNCD/a-C:H:N) thin films are applicable semiconductors in diamond-based electronic devices.

Data Availability

The data used to support the findings of this study are available from the corresponding authors upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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