An efficient ITO-free transparent electrode based on diamond-like carbon with an engineered intermediate metallic thin-film

H. Ferhati^a, F. Djeflal^a, N. Boubiche^b, F. Le Normand^b

^a LEA, Department of Electronics, University of Batna 2, Batna 05000, Algeria

^b MaCEPV/ ICube, Université de Strasbourg and CNRS, Strasbourg, France

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ABSTRACT

In this paper, a novel Diamond-Like Carbon (DLC)-based structure is proposed as an alternative material for transparent electrode applications. In this context, DLC/Ag/DLC multilayer structure was successfully fabricated by combining pulsed laser deposition (PLD) and RF magnetron sputtering techniques. DLC films were deposited at room temperature via PLD on a quartz substrate and their optical constants were extracted using ellipsometry technique. A new hybrid approach based on FDTD method supported by particle swarm optimization (PSO) technique was introduced to find out the best geometry and appropriate intermediate ultrathin metal film of the DLC/Metal/DLC multilayer structure, promoting wider opportunities to overcome the transparency-conductivity trade-off. The optimized design is then prepared by successively depositing DLC films and sputtering ultra-thin Ag layer. Characterizations showcased that the elaborated DLC/Ag/DLC tri-layered structure offers a high Haacke FoM of $55.2 \times 10^{-3} \Omega^{-1}$, emphasizing the ability of the presented systematic investigation for providing promising pathways to reach enhanced light management and reduced resistive losses. Therefore, the elaborated DLC/Ag/DLC multilayer structure opens up new perspectives towards realizing high-performance transparent electrodes based on free-critical raw materials (CRM), which are strongly desirable for optoelectronics and low cost thin film photovoltaic applications.

1. Introduction

Recently, TCEs (Transparent Conductive Electrodes) have been capturing substantial attention for various applications including solar cells, flat panel displays, optical sensors, touch screen panels and organic light emitting diodes (OLEDs) (Guillén and Herrero, 2011; Wang et al., 2019; Hssein et al., 2017; Kim and Yer, 2016). Many kinds of wide band-gap materials such as ZnO, AZO, ITO, TiO₂ and SnO₂ have confirmed their usefulness as basic building blocks for TCE applications, thanks to their fascinating optical and electrical properties (Kim and Yer, 2016; Liu et al., 2015; Seeger et al., 2016; Miao et al., 2014). It is believed that ITO films are the most commonly used material as transparent electrode in nowadays optoelectronic devices owing to their high transparency and low sheet resistance (Wei et al., 2017). Besides, ITO/Ag/ITO multilayer structures, an interesting strategy that suggests introducing Ag intermediate thin layer acting as a current path, is regarded promising for diverse optoelectronic and thin film photovoltaic applications (Lee et al., 2019).

Eco and bio-friendly compounds possessing excellent optical and electrical properties are strongly desired for future microelectronic industry (Giraldo et al., 2019; Gogova et al., 2017; Torrisi et al., 2019).

Accordingly, indium, gallium, tin, arsenic and tellurium are classified as critical raw materials (CRM) and their replacement is important to guarantee the long-term sustainability of optoelectronic and photovoltaic technologies (Giraldo et al., 2019). In this perspective, the partial or total substitution of the toxic and expensive element in ITO transparent electrodes is considered as one of the biggest challenges during the last few years (Torrisi et al., 2019). Unfortunately, the developed TCE structures based on free-CRM such as AZO and ZnO have not yet proven a noticeable improvement over ITO-based ones, making them too upstream to yet contemplate potential incorporating in future optoelectronic devices (Torrisi et al., 2019; Girtan, 2012; Hsu et al., 2017; Kim et al., 2016). Therefore, further efforts should be focused on two essential objectives: the first one relies on turning out toward exploring alternative free-CRM and the other deals with seeking novel strategies to address the transparency-conductivity trade-off associated with TCEs.

Diamond-Like Carbon (DLC) material is regarded as an efficient cost-effective material for several applications involving protective coatings, magnetic recording disks, cutting tools, biosensing, MEMS and even optical windows (Thema et al., 2015; Modabberasl et al., 2015; Robertson, 2012; Tamulevičius et al., 2018). This is mainly due to its

E-mail addresses: faycal.djeflal@univ-batna2.dz (F. Djeflal), francois.le-normand@unistra.fr (F. Le Normand).

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attractive optical and structural properties as well as its outstanding biocompatibility (Tamulevičius et al., 2018). Basically, DLC is a metastable form of amorphous carbon composed of a mixed sp^2 and sp^3 bonded states, in which the elaboration methods and the processing conditions determinate the amount ratio of carbon hybridization and thereby illustrates its electrical and optical characteristics (Santra et al., 2010). Various techniques have been used to synthesize the DLC such as Chemical Vapor Deposition (CVD), plasma deposition, ion beam deposition, sputtering and Pulse Laser Deposition (PLD) (Santra et al., 2010; Bouabibsa et al., 2018; Kumar, 2009; Wu et al., 2016; Seker et al., 2014). The latter method is considered as the most effective, being able to deposit oxygen and hydrogen-free DLC films with high purity. In fact, it is confirmed that the DLC can offer a high visible transparency, demonstrating its potential use as an optical window in optoelectronic devices (Wu et al., 2016; Seker et al., 2014; Stock et al., 2019). However, its insulating nature inherited from diamond material can prevent the development of competitive TCEs based on DLC films (Boubiche et al., 2019). Motivated by this drawback, many attempts have been conducted to enhance the DLC conductivity using new approaches like deposition of metallic nanoparticles and UV laser surface annealing (Stock et al., 2019; Boubiche et al., 2019). Although this, further improvements are still required to eventually envisage the substitution of the more mature technology based on ITO compound. To the best of our knowledge, there are no theoretical and experimental investigations based on an engineered multilayer structure were proposed to develop high-performance transparent electrodes based on free-CRM film. In this work, a new high-performance DLC/Ag/DLC structure was successfully elaborated using facile two-step processes incorporating PLD and RF sputtering techniques. A new approach based on FDTD modeling assisted by metaheuristic technique was adopted to engineer the DLC/Ag/DLC multilayer geometry with the aim of addressing the transparency-conductivity trade-off. Transmittance and conductivity measurements demonstrated that the elaborated multilayer structure promotes new pathways for elaborating cost-effective TCEs offering high-performances. Therefore, the prepared DLC/Ag/DLC tri-layered design opens up a new route to achieve improved transparent electrodes based on free-CRM for high-performance optoelectronic and photovoltaic applications.

2. Numerical simulations and experimental procedure

In this section, we discuss our proposed systematic investigation methodology adopted for the realization of an efficient transparent electrode based on DLC/Ag/DLC multilayer design. Three investigation frameworks are conducted to develop high-performance TCE based on free-CRM. The first one relies on the deposition and characterization of DLC films separately. The second deals with introducing a novel hybrid approach based on 2D-FDTD method combined with PSO metaheuristic technique, enabling effective engineering of the multilayer structure to reach the highest performance of DLC-based transparent electrodes. The last one consists in the preparation and characterization of the optimized DLC/Ag/DLC tri-layered based TCE.

2.1. Elaboration and characterization of DLC films

DLC films were prepared by depositing carbon on an ultrasonically cleaned quartz substrate by PLD technique at room temperature. Pyrolytic graphite target with a high purity of 99.99% was used to deposit the DLC films. The elaboration of these films was performed in vacuum, where the deposition chamber was evacuated to a very low residual pressure of 10^{-8} mbar. The distance between the substrate and graphite target was maintained at 5 cm during the deposition process. The ablation process of the graphite target was carried out by means of a KrF laser source (LAMBDA PHYSIK Compex 201, 248 nm wavelength, 1–10 Hz repetition rate), which can generate 600 mJ of energy per 25 ns duration pulse. It is important to note that the adopted

experimental parameters were carefully optimized to reach a maximum of sp^3 carbon formation, where more details concerning the optimization of the carbon hybridization ratio and the determination of the carbon density can be found in our previously published works (Stock et al., 2019; Boubiche et al., 2019).

After depositing DLC films, Atomic force microscopy (NT-MDT Stand AloneSMENA) is performed to characterize the surface roughness properties of the elaborated DLC thin-films. Measurements concerning the structural, electrical and optical properties of the prepared films, including Raman spectroscopy (LabRam/Aramis Jobin/Yvon spectrometer with 532 nm laser source), conductivity (ECOPIA HMS-5000 set up) and UV–Vis transmittance (F10-RT-UV spectrophotometer) were carried out. Then, ellipsometry technique is used to extract the film thickness and refractive indices, which subsequently can be used to model the optical behavior of the proposed transparent electrode with DLC/Ag/DLC multilayer design as it is detailed in the next sub-section.

2.2. FDTD-PSO hybrid approach

Principally, our investigation is focused on the study of a new transparent electrode based on DLC/Ag/DLC tri-layered structure. In fact, it seems important to perform accurate numerical investigation not only to achieve an in-depth understanding of the optical and electrical theoretical mechanisms but also to optimize the structure with the objective of enhancing the transparent electrode performances. For this purpose, the extracted real and imaginary refractive indices of DLC films and that of Ag reported in (Lee et al., 2019) are used to perform an accurate numerical analysis using 2D-FDTD technique available in the ATLAS simulator, which can serve as effective predictive simulations (Atlas User's manual, 2012). In this context, a plane wave diffracted at the surface of the investigated DLC-based TCE is assumed and periodic boundary conditions are supposed to satisfy the periodicity of the structure in the x-direction. Moreover, two ports through the y-axis (1 and 2) assigned to the bottom and top of the investigated DLC/Ag/DLC design, respectively, are considered to estimate electromagnetic intensities, allowing the determination of the optical parameters. Therefore, the final formulations of these optical metrics can be computed by means of the subsequent equations, which are comprehensively elucidated in our previous works (Ferhati and Djeflal, 2016a, 2016b, 2017)

$$A_i(\lambda) = \frac{\int_V \frac{1}{2} |\vec{E}_y(\vec{r})|^2 \omega \epsilon_0 \epsilon_i''(\lambda) dV}{\int_S \frac{1}{2} \text{Re}[\vec{E}_y(\vec{r}) \times \vec{H}^*(\vec{r})] dS} \quad (1)$$

$$R_i(\lambda) = \frac{\int_{\text{port1}} (E_{ci} - E_{li}) E_{li}^* dA_i}{\int_{\text{port1}} (E_{li} E_{li}^*) dA_i} = \frac{\text{reflected power}}{\text{Incident power}} \quad (2)$$

$$T_i(\lambda) = 1 - R_i(\lambda) - A_i(\lambda) \quad (3)$$

where ϵ_0 refers to vacuum permittivity, while ϵ_i'' denotes the complex materials dielectric constant which is mainly depending on the measured refractive indices of DLC using ellipsometry technique. The main optical factors including absorbance, transmittance and reflectance are assigned as A_i , T_i and R_i . On the other hand, $\vec{H}^*$ is the conjugate of the complex magnetic field, while E_{ci} is the electric field intensity at the second suggested port and E_{li} represents the electric field pattern associated with the first one.

The electrical behavior of the proposed DLC-based transparent electrode is also investigated using SILVACO software, where it enables accurately estimating its electrical conductivity. Principally, a constant current of 1A is applied to DLC/Ag/DLC tri-layered structure, where the end of the analyzed design is grounded, while the first boundary is connected to the assumed current source. The voltage drop across the electrode allows calculating the total resistance of the suggested multilayer structure.

After calculating both transparency and sheet resistance of the DLC/Ag/DLC tri-layered-based TCE, the Haacke Figure of Merit (FoM) can be

extracted by using this equation (Haacke, 1976)

$$FoM = \frac{T_i^{10} (\lambda = 550 \text{ nm})}{R_{Sheet}} \quad (4)$$

where R_{Sheet} is the sheet resistance of the proposed *DLC*-based transparent electrode with inter-metallic layer aspect.

It is believed that *TCEs* with high transparency and conductivity are indispensable for a myriad of basic and practical optoelectronic applications including flat panel displays, solar cells and *OLEDs* (Wang et al., 2019; Hssein et al., 2017; Kim and Yer, 2016; Liu et al., 2015; Seeger et al., 2016). The structure geometry plays a crucial role in modulating the optical and conductive performances of transparent electrodes, which are strongly depending on the multilayer antireflection capabilities and conductivity behavior. In addition, designing very thin *TCE* structures can promote the fabrication of flexible optoelectronic devices like roll-to-roll solar cells and sensors for medical applications (Park et al., 2016). Nevertheless, the inverse relationship between the film thickness and its sheet resistance prevents the realization of flexible and high-performance transparent electrodes, bringing evidence of the urgent need for addressing this trade-off. In our case, the main problem associated with *DLC*-based *TCE* is related to its insulating nature as well as the relatively reduced transparency in the visible spectrum range. Intuitively, the effective engineering of the *DLC/Ag/DLC* multilayer geometry can enable making a compromise between optical and electrical characteristics. On the other hand, concerning the optical properties of the silver thin-film, it is believed that the transmittance behavior of the *Ag* thin-film decreases continuously as the *Ag* thickness increases over the entire visible-*NIR* region. As a matter of fact, the transmittance of conducting media like *Ag*, *Ni*, and *Au* are generally influenced by the absorption of the incoming light. Several recently published works emphasize that *Ag* thin-films should be sufficiently ultrathin for achieving an enhanced average transparency (Girtan, 2012; Lee et al., 2019; Hrostea et al., 2017). Basically, the transparency decreases as the wavelength increases for metallic films, which is closely related to the increase in the material extinction coefficient (k). This can cause the transparency degradation over the visible and *NIR* ranges. In addition, it is confirmed that the careful adjustment of the metal thin-films thickness and position within wide band-gap media can provide enhanced transparency behavior due to light trapping and interference effects, modulating the optical behavior over the whole solar spectrum range (Lee et al., 2019). To shed more light on this prominent topic, a global optimization approach based on *PSO* metaheuristic technique is exploited to simultaneously achieving high average transparency and low sheet resistance of the proposed *DLC/Ag/DLC* tri-layer structure.

Extensive limitations have been encountered to solve many real-life global optimization problems in diverse fields such as engineering, industry, science, and economics. Metaheuristic-based techniques play a central role in treating many engineering problems, which contain usually multiple-conflict goals. For this purpose, these techniques have drawn much interest in order to deal with global optimization difficulties. In general, metaheuristic-based approaches are not problem-specific and involve efficient strategies to effectively avoid being trapped in local optima. The main advantage of metaheuristic-based techniques dwells on making a good trade-off between intensification and diversification of the search mechanism. Several metaheuristic algorithms have proven their effectiveness to deal with global optimization problems such as genetic algorithms evolutionary technique, artificial immune system, scatter search and *PSO* (Holland, 1975; Farmer et al., 1986; Glover, 1977; Clerc and Kennedy, 2002). The latter approach as a nature-inspired and population-based optimization technique is considered effective for dealing with complex problems in low computation time, which explains its appropriateness for solving many optimization problems in a growing number of application paradigms involving nanoelectronics and optoelectronics (Ferhati and Djeflal,

2017; Djeflal et al., 2010; Bencherif et al., 2016). In this sense, we suggest the use of *PSO* approach to identify the best structure geometry allowing the design of high-performance transparent electrode based on *DLC* free-*CRM*. The basic working mechanism of this metaheuristic algorithm dwells on searching candidate solutions represented by populations (in our case the *DLC/Ag/DLC* multilayer geometrical parameters) in a well-specified research space using also a well-defined fitness function. More details concerning the working principle of this technique can be found in our lastly published works (Ferhati and Djeflal, 2017; Djeflal et al., 2010; Bencherif et al., 2016).

Importantly, we need to optimize the proposed *DLC/Ag/DLC* tri-layer structure properly to record a high *Haacke FoM* parameter (Eq. (4)), which is regarded as a critical metric to realize practical transparent electrodes, appropriate for solar cells and various optoelectronic devices. The adopted fitness function is given as follows

$$Fitness(X) = \frac{1}{FoM(X)} \quad (5)$$

where $X_i = (t_{DLC1}, t_{DLC2}, P_{Ag}, t_m)$ of the i^{th} generation represents the design parameter vector in which t_{DLC1} and t_{DLC2} represent the thicknesses of the top and bottom *DLC* films, respectively, t_m and P_{Ag} denotes the thickness and position of the inserted metallic layer as it is shown in the schematic depicted in Fig. 1(a) of the proposed multilayer transparent electrode design based on *DLC* material. To get reasonable values regarding these geometrical parameters, the design configuration should be confined in a given range, which are taken as a set of constraints represented by the following condition

$$\bullet x \in [x_{min}, x_{max}], x_i \in X_i$$

The stall generation and the population size concerning the *PSO*-based optimization procedure are carefully selected and their values are respectively set to 500 and 20. The flowchart of the proposed design methodology is comprehensively detailed in Fig. 1(b).

Therefore, the obtained design configuration that could allow promising pathways to achieve the highest *FoM* is then considered for elaborating high-performance transparent electrode based on *DLC* free-*CRM*, where the experimental procedure is described in the next sub-section.

2.3. Development of *DLC/Ag/DLC* multilayer structure

The first step of this work was to identify the geometrical configuration of the multilayer design, which has been done using the *FDTD-PSO* hybrid approach detailed in the preceding sub-section. Afterword, the next step relies on elaborating the optimized multilayer design yielding a high *Haacke FoM*. In this perspective, the optimized *DLC/Ag/DLC* structure was obtained by depositing *DLC* and *Ag* layers sequentially by combining *PLD* and *RF* magnetron sputtering techniques, in which the optimized thicknesses were taken into consideration. Initially, the bottom *DLC* film was deposited using a *PLD* deposition process. Thereafter, an ultrathin silver layer is sputtered onto the first *DLC* film via *RF* magnetron sputtering technique (*MOORFIELD MiniLab 060*) using silver target with a purity of 99.99%. This experimental facility is in fact efficient because of its low contamination and controllable deposition parameters (El Hajj et al., 2012; Benhaya et al., 2018). During the sputtering processes, *RF* source power and the working pressure were fixed at 250 W and 10^{-5} Pa, respectively. Besides, the deposition process of the silver intermediate thin-layer was carried out in *Ar* atmosphere with a deposition rate of 0.2 nm/s. It is important to highlight that the deposition parameters were carefully optimized through experimental testing in order to get high-quality *Ag* ultrathin-films with reduced sheet resistance. Moreover, the adopted parameters were used for depositing thin-film metals in our previously published works (Benhaya et al., 2018; Ferhati and Djeflal, 2019; Ferhati et al., 2019), where high quality metallic films were successfully

Fig. 1. Cross-sectional view of the proposed *DLC/Metal/DLC* multilayer structure.

elaborated. The thickness of the deposited films was estimated by performing ellipsometry measurement. Finally, the optimized *DLC/Ag/DLC* multilayer design was obtained by depositing the top *DLC* film by means of the *PLD* method. *SEM*, transmittance and conductivity measurements were performed in order to assess the performance of the proposed transparent electrode based on *DLC/Ag/DLC* tri-layered structure.

Fig. 2. (a) AFM image (b) Raman spectra and (c) transmittance spectrum of the elaborated *DLC* thin-film on quartz substrate with 20 nm of thickness.

3. Results and discussions

3.1. Structural, optical and electrical analyses of the prepared *DLC* films

In order to show surface morphological properties of prepared *DLC* films grown on a quartz substrate, an *AFM* characterization was carried out. Fig. 2(a) illustrates the *AFM* image of the elaborated structure scanned on an area of $10 \times 10 \mu\text{m}^2$. It can be seen from this image that the deposited *DLC* film presents very good surface morphological

properties, where it clearly shows surface smooth with some clusters (see Fig. 2). Moreover, the surface roughness of the elaborated DLC film as determined from the AFM micrograph was found to be about 0.6 nm, which is considered low and appropriate for transparent electrode applications. This favorable surface roughness characteristic of the prepared DLC film can be ascribed to the high sp^3/sp^2 content, where it has been confirmed by several experimental investigations that the increase in diamond-like phase degree (sp^3 -rich carbon hybridization) can promote reduced surface roughness. The Raman spectrum associated with the elaborated DLC film, which is depicted in Fig. 2(b) can consolidate this explanation. Observing this figure, we can notice the presence of a broad complex band indicative of carbon contributions in the range of $1100\text{--}1650\text{ cm}^{-1}$. In this context, D and G characteristic bands centered respectively at wave numbers of 1360 cm^{-1} and 1565 cm^{-1} were recorded. The G band is associated with the stretching motion of sp^2 hybridized carbon atoms pair in both chains and rings, while the D gaussian peak is related to the breathing mode of sp^2 atoms in rings, demonstrating the formation of diamond-like carbon phase. Besides, the measured I_D/I_G ratio of the DLC film is about 0.7, emphasizing the presence of graphitic domains and the reduced number and size of the sp^2 clusters. This observation is found in accordance with previous investigations related to the DLC films preparation and structural characterization (Santra et al., 2010; Stock et al., 2019; Boubiche et al., 2019).

The optical performances of the prepared DLC film for TCE application were assessed by analyzing its transparency behavior over a wide spectrum range. The measured transmittance spectrum of the elaborated sample is illustrated in Fig. 2(c). This figure shows the good transparency of the prepared DLC film; where up to 64% of transmittance can be achieved in the visible range (at the wavelength of 550 nm). On the other hand, conductivity analysis of the deposited DLC film was conducted, indicating that the DLC layer shows a low electrical conductivity of $2 \times 10^{-3}\text{ S/cm}$, which is attributed to the insulating properties of the diamond-like-carbon phase. It is to note that these conductivity results are strongly dependent on the annealing temperature, as suggested in the references (Stock et al., 2019; Boubiche et al., 2019).

Accordingly, the obtained optical and electrical performances are not sufficient for the eventual emergence of the DLC low-cost material in the field of transparent electrodes. Therefore, substantial enhancements concerning the visible-light transparency of the DLC films are required to envisage the substitution of conventional TCEs.

3.2. Performance assessment of DLC/Metal/DLC transparent electrodes

The complex refractive indices associated with the deposited DLC film were measured via ellipsometry in the UV, visible and infrared spectrum ranges, and are plotted in Fig. 3. The latter figure depicts the variation of both refractive index (n) and extinction coefficient (K) as a function of the incident photon energy. This figure indicates that the refractive indices of the DLC film gradually increase with the photon energy and their maximum occurs in the UV-spectrum range. In addition, the prepared DLC film in which the thickness was measured and found about 20 nm exhibits a high extinction coefficient near the UV spectrum range, thus confirming its potential for absorbing the UV light.

The obtained wave-dependent complex refractive indices of the elaborated DLC film were used to perform an accurate numerical investigation concerning the optical behavior of the DLC-based transparent electrode as it is detailed in the second section of this work. In this context, to evaluate the accurateness of the developed numerical model based on 2D-FDTD method, Fig. 4 compares the transmittance spectra associated with the experimental results and that of numerical data obtained for a DLC film with 20 nm of thickness. It is clearly shown from this figure that the developed model can reproduce efficiently the experimental data with a good agreement, thus confirming the accuracy

Fig. 3. Spectral dependence of the indices of refraction for the elaborated DLC film on quartz substrate with $t_{DLC1} = 20\text{ nm}$.

Fig. 4. Measured and simulated transmittance spectra of the DLC film on quartz substrate with $t_{DLC1} = 20\text{ nm}$.

of the adopted modeling methodology. This could allow the performance assessment of the proposed DLC/Metal/DLC multilayer design. It is believed that the embedded Ag, the DLC thicknesses and the metal film position within the transparent electrode structure have a profound implication and plays a crucial role in modulating the optical and electrical properties of the TCE. To address this complex behavior, a new FDTD-PSO hybrid approach, as it is explained in the second section, is used for selecting the appropriate geometry, enabling improved electrical and optical performances of the DLC-based transparent electrode. In this context, the optimization procedure has enabled identifying the optimized DLC/Metal/DLC configuration. It is found that taking the DLC/Ag/DLC with an optimized multilayer geometry of 15 nm/7 nm/5 nm, we can achieve the best Haacke FoM. Fig. 5 shows the transmittance curves of the optimized DLC/Metal/DLC multilayer with dissimilar intermediate metal film (Ni, Au and Ag) in the wavelength region from 300 nm to 1000 nm. It is observed that the transmittance spectra of multilayer structures show Vis-IR shielding effect and have relatively large transmission windows in the visible region, which are different from the transmittance curve of DLC layer which showed moderately transparent in Vis-IR region. Moreover, the optimized DLC/Metal/DLC tri-layered structures demonstrate a high transparency in the visible range, where a maximum transmittance exceeding 88% is reached for the proposed DLC-based transparent electrode with Ag as intermediate metal layer. It can be seen also that the transmittance decreases near the IR spectrum band, which is mainly

Fig. 5. (a) Transmittance spectra associated with the optimized *DLC/Metal/DLC* structures with dissimilar intermediate metal layers (Ag, Au and Ni). (b) SEM image of the prepared transparent electrode design with the optimized multilayer geometry of *DLC*(15 nm)/*Ag*(7 nm)/*DLC*(5 nm).

due to the presence of Ag conducting media leading to the absorption of the incoming electromagnetic flux and thereby affecting the transparency of the multilayered structure in the corresponding wavelength range. On the other hand, this figure indicates the outstanding capability of the proposed multilayer design based on *DLC/Ag/DLC* structure for providing larger transmission window of the incident light, yielding an improved average transparency more than 75%. This can be compared to the structures with Ni and Au intermediate metal layers, which exhibit narrow optical windows. This phenomenon is correlated to the low carrier density of the Ag metal as compared to Ni and Au. In other words, it is clear to see that in *DLC* multilayer, the cut off wavelength systematically shifted to the short wavelength region as inserting Ni or Au intermediate metallic layer, which strongly indicates that the optical properties in the Vis-IR long wavelength region are largely depending on the carrier density. The obtained encouraging results concerning the improved transparency of the *DLC*-based

transparent electrode are mainly attributed to a mixture of two effects. Firstly, insertion of a conducting medium promotes an effective light scattering within the *DLC* film. Secondly, the adopted hybrid approach based on accurate numerical modeling assisted by PSO metaheuristic technique has enabled distinctively engineering the multilayer structure to modulate the optical behavior of the *DLC/Metal/DLC* tri-layer, leading to outstandingly enhancing the light transmission of the transparent electrode. Besides, this approach has allowed also enhancing the electrical conductivity of the *DLC*-based electrodes while maintaining high-transparency in a wide spectrum range.

After estimating the optimized geometry of the investigated *DLC/Metal/DLC* multilayer design offering new pathways for realizing high-performance TCEs, the optimized structure is subsequently elaborated by using facile two-step processes involving PLD and RF magnetron sputtering techniques as it is explained in Section 2.3. The surface morphology of the top *DLC* layer in the *DLC/Ag/DLC*-based multilayer design was analyzed by FESEM analysis. Accordingly, Fig. 5(b) shows the SEM micrograph of the prepared tri-layered structure with the optimized geometry. It can be noticed from this figure that the *DLC* surface morphology is homogeneous and is not affected by inserting an ultrathin Ag metallic film, which may indicate the high level of sp^3 content and the reduced number and size of the sp^2 clusters in *DLC* film. The top surface of the *DLC* film illustrates regularly arranged particles with smooth surface. Several published works have investigated the surface roughness of Ag-*DLC* films using FESEM technique, where it is found that good surface properties can be reached (Gayathri et al., 2012; Písařík, 2017). In fact, using the appropriate deposition parameters of RF sputtering, we have succeeded in achieving high-quality surface as it is illustrated in Fig. 5(b), which has led to reach improved optical and electrical performances of *DLC/Ag/DLC* tri-layered transparent electrode. This achievement is mainly attributed to high-performance cathodic RF magnetron sputtering technique, which is considered as the most used method for depositing high-quality ultrathin-films with superior crystallinity and reduced defect interfaces. This is because of its controllable deposition parameters as well as the low contamination advantage as it is above outlined. Moreover, several experimental investigations have confirmed the outstanding capability of the RF sputtering method for depositing metallic films with improved structural characteristics and drastically reduced defects (El Hajj et al., 2012; Benhaya et al., 2018; Ferhati and Djefal, 2019; Ferhati et al., 2019). It is to note that the Point 1 illustrated in Fig. 5(b) represents the center of the image captured by SEM technique. This point is provided by the SEM facility during the characterization procedure.

Electrical and optical characterizations of the optimized structure were performed and the results are summarized in Table 1. This latter table recapitulates a performance comparison between the proposed transparent electrodes based on *DLC* free-CRM, those of previous experimental results associated with *DLC*, and *ITO*-based TCEs provided in Girtan (2012), Stock et al. (2019), Boubiche et al. (2019), Park et al. (2016), and Guillén and Herrero (2008). This comprehensive comparison could give a global insight concerning the ability of the optimized *DLC/Metal/DLC* tri-layered design for potentially replacing the conventional TCOs based on CRMs like *ITO* in various optoelectronic applications. It is demonstrated from this table that the elaborated *DLC/Ag/DLC*-based transparent electrode with an optimized geometry of 15 nm/7 nm/5 nm offers a high Haacke FoM of $55.2 \times 10^{-3} \Omega^{-1}$, superior maximum transparency of 87%, high electrical conductivity exceeding 10^5 S/cm and low sheet resistance of $4.5 \Omega \times sq^{-1}$. While, the calculated FoM parameter using the FDTD/PSO modeling is found to be equal to $59.3 \times 10^{-3} \Omega^{-1}$, which indicates that The measured results are in good agreement with the simulated ones associated with the optimized *DLC*-based transparent electrode. This accordance can be explained by the effectiveness of the adopted modeling methodology as it is confirmed by Fig. 4. More precisely, the measured optical constants associated with Ag and *DLC* films, which are obtained under the same deposition conditions were introduced in our FDTD modeling in order

Table 1
Performance metrics comparison between the proposed *DLC/Metal/DLC* multilayer designs and that of various reported *DLC* and *ITO* based transparent electrode structures.

Transparent electrode structures	TCE geometrical configuration	Electrical Conductivity (S/cm)	Sheet-resistance ($\Omega \times \text{sq}^{-1}$)	Transmittance at $\lambda = 550 \text{ nm}$ (%)	Average Transparency	HaackeFoM $\times 10^{-3} \Omega^{-1}$	Average Haacke FoM $\times 10^{-3} \Omega^{-1}$
This work:							
<i>DLC/Ag/DLC</i>	15 nm/7nm/5nm	1.7×10^5	4.5	87	74.5	55.2	14.8
<i>DLC/Au/DLC</i>	5 nm/7nm/5nm	1.6×10^5	5.48	83	67	30.6	3.7
<i>DLC/Ni/DLC</i>	8 nm/2nm/4nm	3.2×10^4	21	75	56	2.8	0.6
Published results:							
<i>DLC</i> film annealed by UV laser (Boubiche et al., 2019)	20 nm	5×10^4	17	63	65	1.61	2.7
<i>DLC</i> decorated with Ni nanoparticles and annealed (Stock et al., 2019)	20 nm	5×10^3	23	67	64.7	1.83	2.4
<i>ITO/Au/ITO</i> (Guillén and Herrero, 2008)	20 nm/8nm/20 nm	/	8.1	82	80	17	14.2
<i>ITO/Ag/ITO</i> (Girtan, 2012)	35 nm/10 nm/20 nm	/	6	90	76.6	85	12.1
<i>ITO/Cu/ITO</i> (Park et al., 2016)	35 nm/6nm/35 nm	/	11.8	74	72	4.12	3.1

to be very close to the realistic behavior of the investigated multilayer electrodes. Moreover, *FDTD* modeling method is considered effective, been able to provide an accurate solution of Maxwell equation even for more complicated structures as it is illustrated by various published works (Lee et al., 2019; Stelling et al., 2017; Abdullayeva et al., 2019). Therefore, the use of accurate *FDTD*-based numerical modeling supported by ellipsometry technique has enabled exciting opportunities to efficiently predict the performance of the investigated *DLC/Metal/DLC* structure and has contributed to reach the highest Haacke FoM parameter for high-performance thin-film solar cell and optoelectronic applications.

More importantly, it is clearly shown that the elaborated *DLC/Ag/DLC* multilayer outperforms strongly the conventional *DLC* film, which is not conductive if unannealed, and the reported transparent electrode based on decorating the *DLC* surface with Ni nanoparticles, annealed at 550 °C, in terms of optical and electrical properties. These improvements are attributed to the role of inserting an engineered an Ag interlayer, acting as current path and promoting effective light management. In other words, the introduction of Ag thin-film can enhance the transparency behavior and enable getting rid of the insulating properties of *DLC* film, which leads to achieving an excellent compromise between both transparency and electrical conductivity properties. These benefits have enabled reaching encouraging FoM parameter comparable with that provided by *ITO/Ag/ITO* structure and even better than other *ITO*-based transparent electrodes with Au and Cu intermediate metallic layer as it can be deduced from Table.1. Although this improvement, the optimized design shows relatively lower transparency over the NIR range as it is shown in Fig. 5 (a), which is quite logical because of the absorption losses of the inserted conductive media. This behavior is observed in the majority of multilayered TCE based on intermediate metal film engineering aspect (Guillén and Herrero, 2008; Girtan, 2012; Lee et al., 2019; Haacke, 1976; Park et al., 2016; Hrostea et al., 2017; Guillén and Herrero, 2008; Kim et al., 2015; Jo, 2019; Kim, 2016). To address this challenge, researchers have focused on investigating the performance of multilayer transparent electrodes using Average FoM parameters, which is defined as the ratio between the average transmittance over the visible-NIR spectrum band and the sheet resistance. The latter parameter can be expressed as follows (Kim et al., 2015; Jo, 2019)

$$FoM_{Avg} = \frac{T_{Avg}^{10}}{R_{Sheet}} \quad (6)$$

where T_{Avg} represents the average transmittance in the wavelength range of [350 nm, 850 nm].

It is found that Average FoM is always found less than the Haacke FoM for a variety of tri-layered designs. In our case, the proposed *DLC/Ag/DLC* design exhibits a high Average FoM of $14.8 \times 10^{-3} \Omega^{-1}$, which is superior to the *ITO/Me/ITO*-based transparent electrodes as it is illustrated in Table 1. Moreover, the optimized design shows a favorable transparency at the NIR range (75%), which is higher than that of *ITO/Ag/ITO* multilayer TCE (71%). This makes the proposed TCM appropriate for replacing *ITO*-based TCE for organic and thin-film photovoltaics including amorphous Si, CdTe, perovskite and CZTS-based solar cells for which transparent electrodes possessing high conductivity and improved transparency are highly required.

Ultimately, the presented investigation methodology has not only allowed the better understanding of the influence of inserting ultrathin metallic layer on the electrical and optical properties of *DLC*-based transparent electrodes but also promoted exciting opportunities for realizing potential alternative TCEs based on low-cost and free-CRM, which are strongly desired in future flexible optoelectronic devices.

4. Conclusion

In summary, a new *DLC/Ag/DLC* multilayer structure was

elaborated by combining *PLD* and *RF* magnetron sputtering techniques, which can serve as an efficient transparent electrode. *PLD* technique was used to elaborate *DLC* films on a quartz substrate. Structural, optical and electrical properties of the prepared *DLC* films were performed using Raman spectroscopy, *AFM* microscopy, ellipsometry and *UV-Vis* spectrometry and then thoroughly discussed. Before the elaboration of the proposed *DLC*-based multilayer design, a new hybrid approach based on *FDTD* and *PSO* techniques was introduced to engineer the multilayered structure, aiming at overcoming the transparency-conductivity trade-off. Optical and electrical characterizations demonstrated that the optimized *DLC/Ag/DLC* tri-layered design exhibits superior *FoM* of $55.2 \times 10^{-3} \Omega^{-1}$, far surpassing those found up to now for *DLC*-based transparent electrodes. These outstanding results can be attributed to the proposed systematic investigation for promoting the possibility to reach enhanced light-matter interaction and superior conductivity. This emphasizes that appropriate engineering of the *DLC/Ag/DLC* multilayer can enable bridging the gap between the transparency and conductivity properties and could eventually pave the way to elaborate efficient low-cost transparent electrodes based on free-CRM with uncomplicated fabrication processes.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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