Amorphous WO$_3$ thin films designed as gigahertz/terahertz dielectric lenses

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Abstract
Herein, tungsten oxide thin films comprising excess oxygen are treated as optical resonator suitable for gigahertz/terahertz applications. WO$_3$ thin films which are prepared by the thermal evaporation technique under a vacuum pressure of $10^{-5}$ mbar are structurally, compositionally and optically evaluated. The amorphous WO$_3$ films which showed high transparency permit electronic transitions within an indirect allowed energy band gap of 3.05 eV. The band gap comprised energy band tails of width of 190 meV. Four dominant dielectric resonators centered in the infrared (IR), visible (VIS) and ultraviolet (UV) ranges of light are detected. Analysis of the optical conductivity in accordance with the Drude-Lorentz approaches have shown that the drift mobility of free holes in this amorphous layer can be as large as 5.61 cm$^2$/Vs an as low as 1.59 cm$^2$/Vs when exposed to IR and UV light signals, respectively. In addition, the gigahertz/terahertz cutoff frequency ($f_{co}$) spectra demonstrated $f_{co}$ values in the gigahertz frequency domain when exposed to IR light. Excitations with light signals in the VIS and UV spectral ranges allow $f_{co}$ values that extends from 0.7 to 40.0 THz. The wide range of tunability of the WO$_3$ dielectric resonators nomimates them as dielectric lenses suitable for optical communications.

Keywords WO$_3$ · Optical communications · Terahertz · Dielectric resonator

1 Introduction
Tungsten trioxide thin films have captured the interest of research societies due to its remarkable signatures in optoelectronics and other sectors of technology. It have been used as electrochromic media (Rozman et al. 2020), as gas sensors (Chang et al. 2020), as super capacitors (Kariper et al. 2021) and as non-volatile random access memory devices (Lami-
In a recent work, coating stainless steel tapes with WO₃ and studying its optical and electrical properties have shown that the electrochromic properties and electrical conductivity of the films are significantly improved (Rozman et al. 2020). In addition, when Pt/WO₃ is used as hydrogen gas sensors they displayed excellent hydrogen gas sensing response of 1.41×10⁶ under a 1% H₂/air gas (Chang et al. 2020). The sensing level of the gas can be as low as 1 ppm H₂/air (Chang et al. 2020). Moreover, WO₃ films that are produced by a bio-chemical bath method showed a capacitance value of 642 F/g associated with power density of 5.6 kW/h suggesting the ability of using these materials as supercapacitors (Kariper et al. 2021). Furthermore, WO₃ films sandwiched between Au and ITO layers demonstrated metal-insulator-metal structure with appreciable resistive-switching properties (Lamichhane et al. 2020). The reset voltage for this memory device is 2.94 V.

From optical point of view, WO₃ is accounted as wide gap semiconductors usable as active media in optical devices including intelligent windows and smart displays (Garino et al. 2013). Tungsten oxide is also regarded as a media appropriate for optical diffraction gratings (Fung 2019). The optical grids which split, redirect, and disperse light are fundamental optical elements that play main role in fabricating optical devices (Fung 2019). WO₃ is a highly transparent material that allows transition of most of the visible light and infrared light spectrum (Shakoury et al. 2021). WO₃ thin films grown by the electron beam evaporation are mentioned exhibiting polycrystalline nature with extremely high resistivity (10⁸ (Ωcm))(Shakoury et al. 2021). In addition, WO₃ thin films prepared by the reactive magnetron sputter-deposition technique showed nanocrystalline nature and much lower resistivity values (10² (Ωcm)) (Vemuri et al. 2010). In general, thin films prepared by the thermal evaporation technique allow wide variety in the electrical and optical properties as well as in the crystalline nature (Henni et al. 2020; Behera et al. 2019; Chen et al. 2015; Naik et al. 2011). This technique is the most interesting technique because films produced by this economical technique show high stability, high reproducibility, high deposition rate, large area deposition, and non-expansive (Vishwakarma 2015).

Due to these interesting optical features of WO₃, and important features of the thermal evaporation technique, here in this work, we are motivated to explore the optical properties of WO₃ thin films coated onto glass substrates. Measurement of the optical transmittance and reflectance of thin films, provide information about the refractive index energy band tails and dielectric dispersion. These features are the key parameters for optoelectronic technology applications (Sahoo et al. 2020a, b; Naik et al. 2013; Yang et al. 2020). Thus, in our study we target exploring the features of the material as an optical device. Particularly, the necessary information including the optical transparency, reflectively, light absorbability, origin of band gap, energy band tails, dielectric dispersion and optical conductivity parameters will be reported and discussed. The optical conductivity parameters including oscillator energies, free carrier concentrations, Plasmon frequencies, scattering times at femtosecond levels and drift mobility value will be reported and discussed. The ranges of gigahertz/terahertz frequency domains that identifies the WO₃ films to be used as optical devices will also be reported and discussed.
2 Experimental details

Tungsten oxide thin films are grown by the thermal evaporation technique under a vacuum pressure of $10^{-5}$ mbar. The films are deposited onto chemically and ultrasonically cleaned glass substrates. The source material was WO$_3$ powders of high purity (99.99%, Alpha Aesar). The films were prepared in a NORM VCM – 600 vacuum evaporator equipped with Inficon STM-2 thickness monitor. Films of 1.0 μm thick were produced. The structural analyses were carried out using copper Cu$_{\text{K}a}$(λ = 1.5405 Å) Miniflex 600 X-ray diffraction (XRD) unit. The scanning speed of the XRD unit was 0.5°/min. The composition of the films was determined with the help of COXEM-200 scanning electron microscopes equipped with EDAX energy dispersive X-ray spectroscopic analyzer. The EDAX measurements were carried out at accelerating voltage of 29 kV and lifetime of 100 s per cycle. The optical transmittance and reflectance were measured with the help of Thermoscientific Evolution 300 spectrophotometer equipped with VEE MAX II Pike technology reflectometer. The $p-$ type conductivity of the WO$_3$ films was explored with the help of hot probe technique.

Fig. 1 (a) the X-ray diffraction patterns and (b) the energy dispersive X-ray spectra for WO$_3$ thin films. The inset of (a) showing the optical images for the samples under study. Inset of (b) displaying the scanning electron microscopy images for the amorphous WO$_3$ films.
3 Results and discussion

The optical images for WO\textsubscript{3} thin films coated onto glass substrates are shown in the inset of Fig. 1 (a). The inset displays wheat colored films indicating the high transparency of the films. Figure 1 (a) also shows the X-ray diffraction (XRD) patterns for the films. It is clear from the figure that WO\textsubscript{3} coated onto glass substrates by the thermal evaporation technique prefer the amorphous nature of growth. No sharp peaks can be detected in the XRD patterns. Amorphous films of WO\textsubscript{3} are mentioned beneficial for fabrication of supper capacitors (Kariper et al. 2021) and as anodes for Li-ion batteries (Yang et al. 2020). Amorphous films play important role in device fabrication because they are able to provide several advantages over crystalline ones like higher uniformity and better compatibility with flexible substrates (Vemuri et al. 2010).

On the other hand, Fig. 1 (b) show the energy dispersive X-ray spectroscopy (EDAX) for the WO\textsubscript{3} thin films coated onto glass substrates. On average, the films are composed of glass (SiO\textsubscript{2}:Na\textsubscript{2}O:MgO:CaO) and WO\textsubscript{3}. The gold appears in the spectra because it was coated to prevent electron contaminations. The stoichiometric formula of the films is WO\textsubscript{3.33}. The films contained excess oxygen. Excess oxygen is probably due to adventitious material adsorbed on the sample surface (Zubkins et al. 2020; Uner et al. 2003; Singh et al. 2021a, b). Excess oxygen in WO\textsubscript{3} is reported to play vital role in engineering the optical properties of WO\textsubscript{3} (Vemuri et al. 2012). On the other hand, as can be seen from the inset of Fig. 1 (b), the scanning electron microscopy tests did not reveal any grains assuring the amorphous nature of the films.

Figure 2 (a) show the optical transmittance ($T$) and reflectance spectra ($R$) spectra for WO\textsubscript{3} thin films. The transmittance spectra show an increasing trend of variation including a shoulder and an absolute maxima at 2.76 and 1.56 eV, respectively. In addition, the reflectance spectra display one local and one absolute maxima centered at 3.43 and 2.29 eV, respectively. It is also evident from Fig. 2 (a) that WO\textsubscript{3} is a highly transparent material. The maximum transmittance reaches ~90.5% which is comparable to that of glass. On the other hand, the absorption coefficient spectra ($\alpha$) for WO\textsubscript{3} film of thicknesses of $d = 1.0$ µm which is calculated from $T$ and $R$ spectra using the Eqs. (Dresselhaus et al. 2018; Qasrawi and Zyoud 2020),

$$ T \approx (1 - R_{\text{glass}})(1 - R_{\text{WO}3}) e^{-\alpha d} \tag{1} $$

are shown in Fig. 2 (b). Interesting features of the absorption coefficient is observed. Namely, in the high absorption region (4.10–3.50 eV), the absorption coefficient values are very high and decay sharply reaching $\alpha$ values of $10^3$(cm$^{-1}$) within a short range of energy. In the incident photon energy range of 3.50–1.74 eV, $\alpha$ slowly (exponentially) decreases reaching values of ~$10^1$ (cm$^{-1}$). In the infrared (IR) energy range of 1.14–1.74 eV, the absorption coefficient increases with decreasing incident photon energies. The spectral response of $\alpha$ in that range of energy is illustrated in Fig. 2 (c). The increase in the absorption coefficient with decreasing incident photon energy is assigned to the free carrier absorption in WO\textsubscript{3}. Generally, free carrier absorption arises from the carrier movement affected by phonon scattering which transfers the energy to the random crystallites when irradiated by IR light (Zhang et al. 2012). This phenomenon was previously observed in WO\textsubscript{3} films and was also attributed to the impurity or doping agents in WO\textsubscript{3} (Rengel et al. 2019; El-Nahass et al.
Amorphous WO$_3$ thin films designed as gigahertz/terahertz dielectric materials (Charles et al. 2015). For our samples, the most reasonable factor is the existence of excess oxygen which behave as doping agent in the films.

As we mentioned here, in the moderate incident photon energy range, $\alpha - E$ variations display an exponential decay following Urbach’s rule in which (Charles et al. 2013),

$$\alpha = \alpha_0 e^{\frac{E}{E_U}} \quad (2)$$

The Urbach’s energy measures the width of the tails of the localized states in the band gap region and measures the degree of disorder in the film (Priyadarshini et al. 2021; Sahoo et al. 2020a, b, 2021a, b). The width of the band tails ($E_U$) and the pre-exponential factor ($\alpha_0$) which are determined from the slope and intercept of the $\ln(\alpha) - E$ variation illustrated in Fig. 2 (d). The width of the band tails is 190 meV. Values of 74–141 meV were previously observed for WO$_3$ thin films prepared by the sputtering technique (Charles et al. 2013). The difference in the values of $E_U$ is assigned to the structural modifications and degree of disorder (Gupta and Singh 2015). In addition, the different exponent values also refers to different transition probabilities [28–29].

On the other hand, imposing Tauc’s Eqs. (Algarni et al. 2021),

$$(\alpha E)^{1/2} \propto (E - E_g) \quad (3)$$

for indirect allowed transitions in sharp absorption region (the most appropriate form of Tauc’s equations which linearize widest range of data) results in determining the energy band gap of WO$_3$. The best fit of Tauc’s equation which crosses the $E-$ axis of the $(\alpha E)^{1/2} - E$
variation that is shown in Fig. 3 (a) is at 3.05 eV. The energy band gap value being 3.05 eV was previously observed in WO$_{3-x}$ and is assigned to the formation of non-stoichiometric tungsten oxide phases (Mohamedkhair et al. 2021). It is also reported that the energy band gap values depend on the oxygen vacancy in the films (Matsukawa and Ishigaki 2021). Films composed of WO$_3$, WO$_{2.9}$, WO$_{2.72}$, and WO$_2$ are mentioned exhibiting band gaps of 2.67 eV, 3.05 eV, 2.55 eV, and 1.86 eV, respectively (Matsukawa and Ishigaki 2021). It is believed that excess oxygen is also responsible for the observed behavior of $E_g$ values for nanocrystalline WO$_3$ films (Vemuri et al. 2012). The energy band gap in tungsten oxide corresponds to electronic transitions between the top of valence band (formed by the filled O 2p orbitals) to the conduction band (formed by the empty W 5d orbitals). Thus, reduced energy band gap values are owed to the oxygen vacancies or any other structural defects. While the increase in band gap with a progressive increase in oxygen concentration can be attributed to excess oxygen which behaves as (in the defect picture) as interstitial doping agent (Vemuri et al. 2012).

To gain information about the WO$_3$ films as dielectric lenses or optical resonators we have calculated the effective ($\epsilon_{\text{eff}}$), real ($\epsilon_r$) and imaginary ($\epsilon_{\text{im}}$) dielectric constants ($\epsilon_{\text{eff}} = \epsilon_r + i\epsilon_{\text{im}}$), optical conductivity ($\sigma(w); w = 2\pi f$) and gigahertz/terahertz cutoff frequency ($f_{\text{co}}=\sigma(w)/\epsilon_r(w)$). The effective, real and imaginary parts of the dielectric constant are determined from the measured reflectance and transmittance using the relations (Dresselhaus et al. 2018),

$$R = \frac{(\sqrt{\epsilon_{\text{eff}}} - 1)^2 + \left(\frac{\alpha\lambda}{4\pi}\right)^2}{(\sqrt{\epsilon_{\text{eff}}} + 1)^2 + \left(\frac{\alpha\lambda}{4\pi}\right)^2}$$  \hspace{1cm} (4)

![Fig. 3](image_url)  
**Fig. 3** (a) Tauc’s equation fitting, (b) the real part of the dielectric constant, (c) the imaginary part of the dielectric constant and the optical conductivity and (d) the gigahertz/terahertz cutoff frequency spectra for WO$_3$ thin films
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\[ \epsilon_r = \epsilon_{eff} - \left( \frac{\alpha \lambda}{4\pi} \right)^2 \]  

(5) 

\[ \epsilon_{im} = 2\sqrt{\epsilon_{eff}} \left( \frac{\alpha \lambda}{4\pi} \right) \]  

(6)

. The spectra of the real and imaginary parts of the dielectric constant are displayed in Fig. 3 (b) and Fig. 3 (c), respectively. It is clear from the figure that two resonance peaks exist in the spectra. The peaks are centered at 3.43 eV and 2.27 eV. The critical energy value being 3.43 eV was previously observed and assigned to transitions from the conduction band to the valence band recombination centers (Wen et al. 2012). The critical energy value being 2.27 eV is also attributed to the presence of additional states in the energy band gap of WO$_3$ (Lagier et al. 2021). On the other hand, the optical conductivity spectra which are calculated from the imaginary part of the dielectric constant are also shown in Fig. 3 (c). The trend of variation of the optical conductivity is very similar to that of imaginary part of the dielectric constant. For both physical parameters (\( \sigma (w) = \epsilon_{im} w / (4\pi) \)) as illustrated in the figure, the higher the incident photon energy is, the larger the the value of the imaginary part (dielectric loss) and the higher the optical conductivity value. Reproducing the optical conductivity data using Drude-Lorentz approaches for optical conduction using the Eqs. (Qasrawi and Yaseen 2021; Dresselhaus et al. 2018),

\[ \sigma (w) = \sum_{i=1}^{k} \frac{w_{pi}^2 w^2}{4\pi \tau_i \left( (w_{ei}^2 - w^2)^2 + w^2 \tau_i^2 \right)} \]  

(7)

allowed determining the optical conductivity parameters for the WO$_3$ dielectric resonators. The good fitting of the \( \sigma (w) \) data which are shown by green colored circles in Fig. 3 (c) was reached by substituting the parameters shown in Table 1. In Eq. (7), \( \tau_i \) is the average scattering time at femtosecond level, \( P \) is the free hole density, \( w_{pi} = \sqrt{4\pi P e^2 / m^*} \) is the plasmon frequency, \( w_{ei} = E_{ei} / \hbar \) is the hole-plasmon coupled oscillator frequency whose energy is \( E_{ei} \), and \( m^* = 0.94m_o \) (Lin et al. 2018).

The experimental data of optical conductivity is reproduced by executing the series of Eq. (7) up to \( k = 4 \). The four oscillators (\( E_{ei} \)) are centered at 1.25 eV, 2.27 eV, 3.43 eV and 4.15 eV. One oscillator is dominant in the infrared range of light, one in the visible range and two in ultraviolet range. Among these oscillators, the mobile free carriers dominate in the IR and visible (VIS) light ranges. However, in these ranges, the Plasmon frequency values are in the radio-wave frequency domain. It indicates the suitability of the resonators to filter radio waves associated with VIS-IR light signals. Although the drift mobility (\( \mu = e\tau / m^* \))

| k   | 1   | 2   | 3   | 4   |
|-----|-----|-----|-----|-----|
| \( E_{ei} \) (eV) | 1.25 | 2.27 | 3.43 | 4.15 |
| \( \tau \) (fs)    | 3.0  | 2.0  | 1.0  | 0.9  |
| \( p \) (x10$^{17}$ cm$^{-3}$) | 1.0  | 10.0 | 40.0 | 450.0 |
| \( \omega_p \) (GHz) | 0.19 | 0.61 | 1.23 | 4.11 |
| \( \mu \) (cm$^2$/Vs) | 5.61 | 3.74 | 1.87 | 1.59 |
is reduced in the UV range of light, the free carrier density and Plasmon frequency sharply increased indicating the suitability of the WO$_3$ resonators for gigahertz applications. Surface Plasmon resonance in Au/WO$_3$ was previously observed at 520 (2.39 eV) and 604 nm (2.06 eV) (Bose et al. 2016).

Figure 3 (d) show another interesting property of the WO$_3$ as dielectric resonators. Namely, the ability to filter terahertz waves. dielectric-filled parallel-plate waveguides sensitive to terahertz signals are controlled by the terahertz cutoff frequency values (Mendis 2006). It is evident from the figure that in the incident photon energy range of 1.64–1.14 eV (IR) range, $f_{co}$ display values in the gigahertz range of frequency. When light energy reaches the visible light ranges, $f_{co}$ display values in the range of 0.74–4.8 THz. Raising the energy furthers, allow $f_{co}$ to reach ~40.0 THz. This wide range of $f_{co}$ provides large scale of tunability of the dielectric resonator as terahertz filter. Hence, for our WO$_3$ dielectric resonator, the wide range of terahertz cutoff frequency that depends on exciting light signals can be regarded as THz resonator of wide range of tunability (Tepanecatl Fuentes et al. 2021).

As an additional information we have also calculated the extinction coefficient ($k = \alpha \lambda / (4\pi)$) and refractive index ($n = \sqrt{\varepsilon_{eff}}$) spectra. The respective spectra are shown in Fig. 4 (a) and (b). The extinction coefficient sharply decreases with decreasing incident photon energy in the spectra range of 4.0-2.90 eV. It then tends to remain constant in the range of 2.90–2.40 eV. It decreases again in the range of 2.40–1.70 eV and re-increases in the IR range of light (1.70–1.10 eV). The extinction coefficient provides information about how strongly a substance absorbs light at a given light energy. Thus in accordance with the illustrated spectra in Fig. 4 (a), like other glassy and/or oxide materials [42–43], WO$_3$ thin films are very appropriate material for the absorption of UV, visible and infrared light. On the other hand, the refractive index which is shown in Fig. 4 (b) exhibit the same behavior

![Fig.4](image.png) (a) the extinction coefficient and (b) the refractive index spectra for WO3 thin films
Amorphous WO$_3$ thin films designed as gigahertz/terahertz dielectric... like that of the real part of the dielectric constant. Two main resonance peaks are observed at critical energy values of 3.43 eV and 2.27 eV. In addition the numerical values of the refractive index reach maximum of 2.25 at 2.27 eV. Two local minima centered at 3.85 eV and 1.53 eV and one absolute minimum values of $n$ centered at 2.97 eV are observed. The values of the refractive index are comparable to oxide glassy system (Singh et al. 2021a, b).

4 Conclusions

Here in this work, we have explored the structural, compositional and optical properties of thin layers of WO$_3$ comprising excess oxygen in its structure. The measured optical transmittance and reflectance spectra in the spectral range of 4.1–1.15 eV indicated that the amorphous films exhibits high absorption in the ultraviolet range of light. In addition, analyses of the absorption coefficient spectra revealed band gap energy band gap value of 3.05 eV. The band gap value is suitable for visible light absorption and improves the optical performance of the film. The band gap also contained energy band tails of width of 190 meV. Moreover, getting benefit from the reflectance and absorption coefficient spectra the dielectric constant and refractive index spectra were calculated. The experimental results showed that WO$_3$ is a good dielectric resonators. The films of WO$_3$ are found attractive as Plasmon surfaces suitable for optical applications. As an optical receiver WO$_3$ films can perform in the infrared, visible light and ultraviolet ranges of frequency. The modeling of the optical conductivity parameters in accordance with the Drude-Lorentz approach revealed drift mobility values that suits optoelectronic device fabrication.

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Data availability Statement  The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declarations

Conflict of interest  The authors declare that they have no conflict of interest.

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