Optical Properties of PLD-Grown MoS_2 Thin Films on Different Substrates Studied by Spectroscopic Ellipsometry

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ABSTRACT

Atomically thin transition metal dichalcogenides (TMDs), especially MoS_2, have attracted significant interest for their unique electronic and optical properties and potential applications in optoelectronic and nanoelectronic devices. In this study, MoS_2 thin films were deposited on three substrates: Sapphire (0001), GaN/Sapphire, and SiO_2/Si using pulsed laser deposition (PLD), with various growth parameters optimized. The structural and optical characteristics of the films were analyzed through Raman spectroscopy and spectroscopic ellipsometry (SE). Raman results confirmed the formation of MoS_2 and showed that the number of layers varied with the substrate: bulk-like on sapphire, trilayer on GaN/sapphire, and bilayer on SiO_2/Si. SE measurements in the range of 350–900 nm yielded optical constants and film thickness using the Tauc–Lorentz model. Thicknesses were estimated at approximately 90 nm, 2.7 nm, and 1.9 nm for sapphire, GaN/sapphire, and SiO_2/Si substrates, respectively. Tauc analysis indicated optical band gaps of about 1.6 eV for the bulk-like film and approximately 1.9 eV for the thinner films. The refractive index (n) and extinction coefficient (k) strongly depended on photon energy and substrate type, with higher n values observed in the visible spectrum. These results highlight the substrate-dependent optical properties of PLD-grown MoS_2 films, emphasizing their potential for use in optoelectronic devices, antireflection coatings, and photovoltaic applications.

Index Terms: MoS_2 thin films, pulsed laser deposition, spectroscopic ellipsometry, Raman spectroscopy, optical properties.

I. INTRODUCTION

In recent years, atomically thin layers of transition metal dichalcogenides (TMDs), such as MoS_2, have been widely studied for their potential in optoelectronic, electronic, and

nanoelectronic applications. [1, 2]. MoS_2 is a two-dimensional layered semiconductor material, where each layer consists of a single plane of hexagonally arranged Mo atoms sandwiched between two hexagonal planes of S atoms, connected by strong covalent bonds in a trigonal prismatic arrangement. Weak van der Waals forces hold the layers together [3, 4]. Because of quantum confinement, bulk MoS_2 is an indirect-band-gap semiconductor with a band gap of 1.2 eV. In contrast, monolayer MoS_2 is a direct band-gap semiconductor with an energy gap of 1.9 eV [5, 6]. Therefore, monolayer MoS_2 is more promising for future transistor and logic circuit applications due to its high on/off current ratio. Notably, due to its direct band gap and position in the visible spectrum, monolayer MoS_2 could be used in solar cells [7], light-emitting diodes [8], photodetectors [9], and flexible devices [10]. Several methods have been used to synthesize MoS_2 thin films, including mechanical exfoliation [11], chemical exfoliation [12], liquid phase exfoliation [13], sputtering [14], and pulsed laser deposition (PLD) [15, 16].

In this paper, we grew high-quality, continuous, large-area MoS_2 thin films on three different substrates—Sapphire (0001), GaN/Sapphire, and SiO_2/Si using PLD with optimized parameters. For optoelectronic device applications, understanding the optical properties of MoS_2 thin films, such as the refractive index (n) and extinction coefficient (k), is essential. These optical properties can be determined using the powerful, non-destructive spectroscopic ellipsometry (SE) technique [17, 18]. In SE, we analyze changes in the polarization state of reflected light from the film surface by developing optical dispersion models to determine the photon-energy-dependent optical constants and film thicknesses.

Recent studies have also demonstrated the use of spectroscopic ellipsometry to investigate the optical properties of thin films and nanostructured materials [22, 25]. Moreover, substrate-dependent optical properties of thin films have been reported in recent research [26]. In our work, we report SE measurements of the optical properties of PLD-grown MoS_2 thin films on various substrates. Raman spectroscopy has been used to determine the number of MoS_2 layers.

II. EXPERIMENTAL DETAILS

Pulsed laser deposition technique was used to synthesize MoS_2 thin films on Sapphire (0001), SiO2/Si, and GaN/Sapphire (0001) substrates by ablating a MoS_2 target with KrF excimer laser pulses (λ= 248 nm) inside a PLD chamber. A commercially available 1-inch MoS2 target was utilized. Before growth, the substrates were pre-cleaned following standard chemical procedures, then rinsed in DI water and dried with dry nitrogen. The cleaned substrates were immediately mounted into the PLD chamber, which was then evacuated to a base pressure of approximately 3×10^{-6} Torr. In addition to pre-cleaning, substrates were degassed in situ at 800 °C to remove residual surface contamination. Various PLD parameters were optimized for MoS_2 film growth, including the target-to-substrate distance, repetition rate, post-anneal time, substrate temperature, background pressure, laser power density, laser frequency, and number of laser shots. The resulting samples were labeled S1, S2, and S3, corresponding to substrates and laser shots: Sapphire (5000 shots), GaN/Sapphire (60 shots), and SiO_2/Si (40 shots), respectively. The substrate temperature for all samples was maintained at 700˚C. During growth, laser power density, laser frequency, and target-to-substrate distance were kept at approximately 0.5 J/cm², 5 Hz, and 5 cm, respectively. The background pressure was around 8×10^{-6} Torr during all depositions. Following film growth, the samples were annealed in situ at 700˚C for 20 minutes, then cooled to room temperature at 5˚C/min. The optical properties of the MoS_2 films were analyzed using a Spectroscopic Ellipsometer (J.A. Woollam, model: VASE32), with measurements taken across the wavelength range of 350-900 nm at an incident angle of 55°. Spectroscopic ellipsometry is regarded as a reliable and non-destructive method for investigating thin film optical properties, including thickness, refractive index, extinction coefficient, bandgap, dielectric constant, and dielectric functions, among others. Its key principle is to detect changes in light polarization as it passes through the film and reflects off the substrate surface. This model-fitting technique minimizes the differences between measured ellipsometer parameters, Ψ and Δ, and their calculated values across varying wavelengths or

photon energies [17]. To reduce optical correlation effects between layers in a multilayered structure and to achieve more precise measurements of film thickness and optical constants, it is essential to consider the optical response of each layer. Each layer in the model has several fitting variables, such as thickness and optical dispersion parameters. Different optical models were developed to analyze ellipsometer spectra for MoS_2 thin films deposited on Sapphire, GaN/Sapphire, and SiO_2/Si substrates.

Before measuring the ellipsometer spectra of MoS_2 thin films, we calibrated the system using a standard reference sample (SiO_2/Si, approximately 26 nm thick) and verified the repeatability of the data. Afterwards, we recorded spectra of the GaN/Sapphire and SiO2/Si substrates (without MoS_2 film) and analyzed the data using the Parameterized Semiconductor Oscillator Model (PSM) for GaN/Sapphire and the Tauc-Lorentz (TL) oscillator model for SiO_2/Si, respectively. The SE spectra of SiO_2/Si substrates were analyzed with a three-layer optical model consisting of a Si substrate, an interface layer between Si and SiO_2. Subsequently, we recorded spectra of MoS_2 thin films deposited on Sapphire, GaN/Sapphire, and SiO_2/Si substrates. The same fitting models described above were used for the underlying layers of GaN and SiO_2, while a well-established Tauc-Lorentz oscillator model was applied for measuring the thickness and optical constants of the MoS_2 film/layer [18]. Raman measurements were performed using a 514.5 nm excitation from an argon-ion laser with a single monochromator equipped commercial micro-Raman spectrometer in the backscattering geometry.

III. RESULTS AND DISCUSSION

Raman Spectra were measured from the MoS_2 films grown on different substrates (a) Sapphire (S1), (b) GaN/Sapphire (S2), and (c) SiO_2/Si (S3), as shown in Figure 1. All the films exhibit two prominent peaks, which correspond to the E^1_{2g} and A_{1g} in-plane opposite-vibration and out-of-plane vibration modes, respectively. The frequency difference (δ) between the E^1_{2g} and A_{1g} vibration modes can be used to identify the number of the MoS_2 layers. As the number of layers increases from monolayer to bulk, the E^1_{2g} and A_{1g} modes undergo red and blue shifts, respectively. As a result, the frequency

difference increases. Here, in our samples, the measured δ values were 21.6, 22.6, and 27.1 cm-1 for the S3, S2, and S1 samples, respectively. This suggests that δ increases from the S3 to the S2 sample, then reaches a maximum for the S1 samples. Therefore, S3 and S2 samples consist of bilayers and a tri-layer, respectively, while the S1 sample exhibits bulk MoS_2 [12].

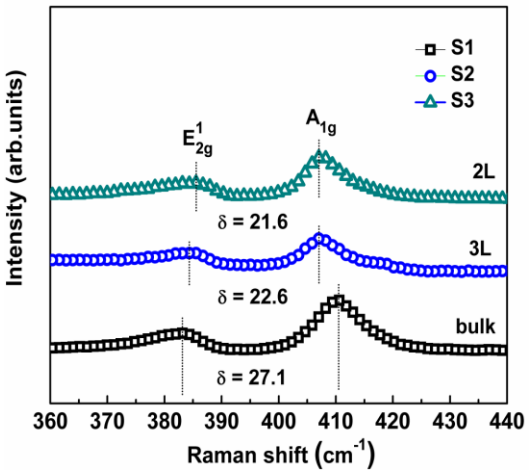


Fig. 1. Raman Spectra of MoS_2 thin films on the different substrates (a) Sapphire (S1), (b) GaN (S2), and (c) SiO_2/Si (S3), respectively.

Fig. 2. represents the Spectroscopic ellipsometer (SE) data for the ellipsometric parameter (Ψ) at an incident angle of 55 ° of MoS_2 film deposited on on different substrates, sapphire (S1), GaN/Sapphire (S2) and SiO_2/Si (S3).

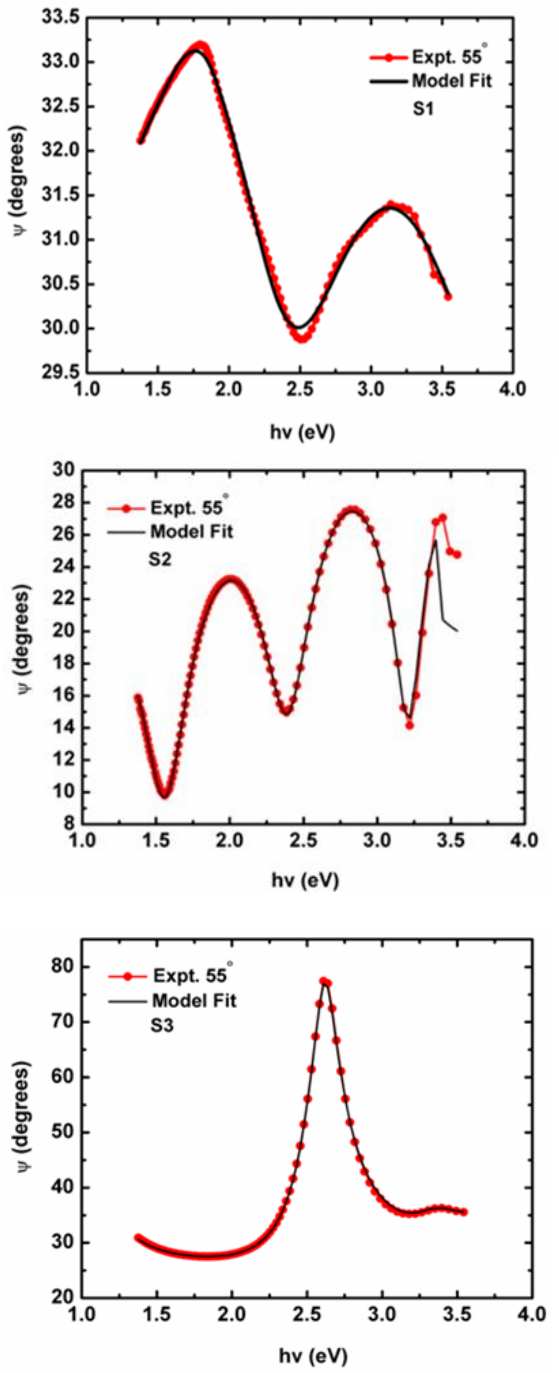


Fig. 2. Experimental and model-fitted ellipsometric parameter (ψ) of MoS_2 samples S1, S2, and S3, respectively.

The solid line in the figures represents the model-fit data and it can be seen that model fitted data very well matched with

experimental data. The experimental data has been fitted for all samples S1, S2 and S3 using Tauc-Lorentz (TL) model for the air/MoS2/substrate structure which takes into account the film thickness and contribution of the substrate. The fitting parameters within the parametric dispersion model yield thickness of MoS2 samples S1, S2 and S3 about 90 nm, 2.7 nm and 1.9 nm, respectively. According to inter-band absorption theory, the optical band gap of the films can be calculated using the following well-known Tauc relation

$$\alpha h\nu = K\,(h\nu - E_g)^n$$

where K, E_g, hν, and n are the probability parameter for the transition, band gap of the material, incident photon energy, and the transition coefficient (2 for indirect and 1/2 for direct band gap), respectively. The absorption coefficient α has been extracted from the ellipsometric data after model fitting using the equation $\alpha = 4\pi k/\lambda$, where λ is the wavelength and k is the extinction coefficient. Here, the indirect band gap of the MoS_2 films was evaluated by extrapolating the straight line part of the curves $(\alpha h\nu)^{1/2} = 0$, as shown in Fig. 2. The calculated values of the optical band gap was found to be ~1.6 eV, ~1.9 eV, and ~1.9 eV for the samples S1, S2, and S3, respectively. This suggests that bulk MoS_2 has a larger bandgap than a very thin MoS_2 film.

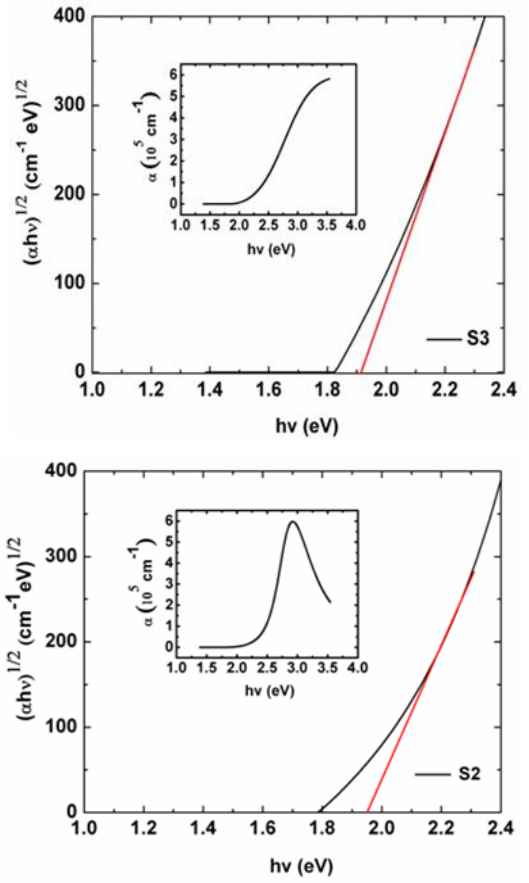
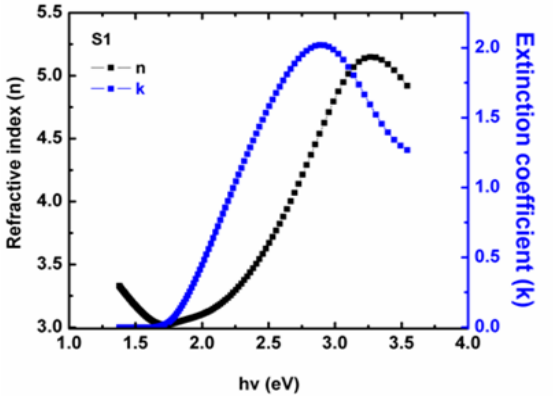
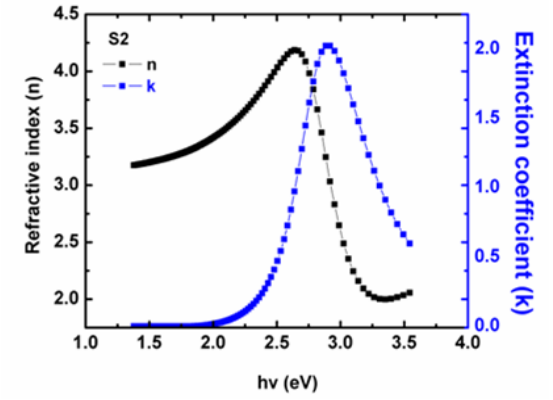


Fig. 3. $(\alpha h\nu)^{½}$ vs hν plots of MoS_2 sample S1, S2 and S3, respectively. The Extrapolation of the linear region to $(\alpha h\nu)^{½} = 0$ (red line) gives the value of the optical band gap (E_g). The inset shows the absorption spectra of MoS_2 samples.

Fig. 4. shows the refractive index (n) and extinction coefficient (k) of MoS2 thin films as a function of photon energy in the range 1.36 eV to 3.55 eV. For all samples,

the value of n for the MoS2 film increases with photon energy up to a certain threshold, then decreases again with increasing photon energy up to 3.55 eV. The maximum value of n is observed at different photon energies for MoS_2 films deposited on different substrates; i.e., for samples S1, S2, and S3, the maximum values of n are observed at photon energies of 3.26 eV (380 nm), 2.64 eV (467 nm), and 2.61 eV (475 nm), respectively.





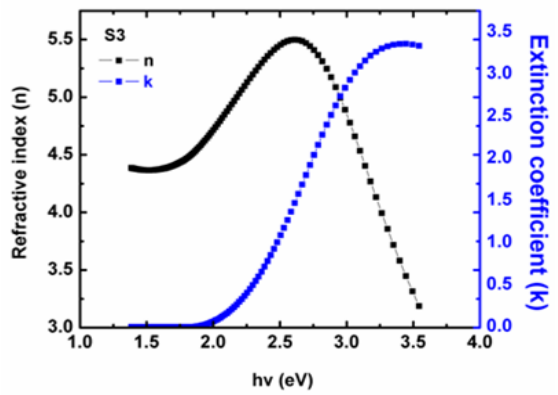


Fig. 4. Plots of the refractive index (n) and extinction coefficient (k) of MoS_2 samples S1, S2, and S3, respectively.

Also, it was observed that the value of n depends on the substrate type used to deposit MoS2 films. The maximum values of n are 5.15, 4.18, and 5.49 for the samples S1, S2, and S3, respectively. Therefore, across all samples, we observed a high refractive index in the visible range, with many applications, such as good antireflection coatings for optoelectronics [19], photovoltaic device encapsulation [20], and high-performance substrates for advanced display devices [21]. The extinction coefficient shows strong absorption peaks at 2.88 eV (430 nm), 2.89 eV (429 nm), and 3.44 eV (360 nm) for the S1, S2, and S3 samples, respectively. In all samples, we observed a single dominant peak in the k plot.

IV. CONCLUSION

High-quality MoS_2 thin films were successfully deposited on multiple substrates using pulsed laser deposition. Raman and spectroscopic ellipsometry measurements confirmed the structural and optical properties of the films. The optical constants and band gap values showed clear dependence on film thickness and substrate type, indicating the potential of PLD-grown MoS_2 thin films for optoelectronic and photonic device applications.

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