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Investigation of the optical properties of MoS_2 thin films using spectroscopic ellipsometry

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Spectroscopic ellipsometry (SE) characterization of layered transition metal dichalcogenide (TMD) thin films grown by vapor phase sulfurization is reported. By developing an optical dispersion model, the extinction coefficient and refractive index, as well as the thickness of molybdenum disulfide (MoS_2) films, were extracted. In addition, the optical band gap was obtained from SE and showed a clear dependence on the MoS_2 film thickness, with thinner films having a larger band gap energy. These results are consistent with theory and observations made on MoS_2 flakes prepared by exfoliation, showing the viability of vapor phase derived TMDs for optical applications. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4868108]

In recent years, there has been a large volume of research conducted on two dimensional (2D) materials such as graphene and transition metal dichalcogenides (TMDs) due to their suitability for future electronic/optoelectronic device applications.[1–4] In particular, molybdenum disulfide (MoS_2), a semiconducting layered TMD, has been identified as one of the most promising 2D materials for nano-electronic applications because of its properties which can be tuned by controlling the number of layers or by careful selection of substrate/gate dielectric materials.[5–8] Moreover, unlike pristine graphene which has no band gap, monolayer MoS_2 has a direct band gap of $\sim$1.8 eV and bulk MoS_2 has an indirect band gap of $\sim$1.3 eV,[5] and electronic devices based on mono- or multilayered MoS_2 films have shown good photodetection capability.[7,9] While mechanical exfoliation is a widely used method to prepare layered MoS_2 thin films, the difficulty of controlling layer thickness and the lateral size limitation have led to the development of alternative synthesis routes. Recently, vapor-phase growth methods, whereby sulfur powder is vaporized and reacted with thin molybdenum or molybdenum oxide seed layers, have been introduced.[10–12] This makes it possible to obtain large-area MoS_2 thin films with tunable thickness and such films have been employed to fabricate various types of nano-optoelectronic devices including solar cells, sensors, and phototransistors.[13–15]

For optoelectronic device applications of MoS_2 to be fully understood, it is necessary to know the refractive index (n) and the extinction coefficient (k). Spectroscopic ellipsometry (SE) is a powerful non-destructive technique to measure the optical properties of thin films. In SE, the wavelength dependent optical constants and the thickness of thin films can be determined by analyzing the change in the polarization state of the reflected light from the film surface and developing an optical dispersion model of the film material.[16] It is frequently used in industry for film characterization and in *in-situ* thickness monitoring because it is fast, cheap, and non-destructive. In recent years, SE has been employed to investigate the optical constants of graphene;[17–20] however, to date, SE characterization of 2D layered TMDs, such as MoS_2, has rarely been reported.

In this work, we report the measurement of the optical properties of MoS_2 thin films with different thicknesses using SE. The MoS_2 thin films were synthesized on substrates using a vapor phase sulfurization process and characterized by UV-visible absorption spectroscopy and Raman spectroscopy. Based on SE measurements, an optical dispersion model was developed for the MoS_2 thin films, and the optical constants (n, k) and the film thickness values were extracted. In addition, optical band gap values of the MoS_2 thin films were obtained from analysis of the k values. The values are consistent with data derived from other techniques and theoretical values. This work establishes SE as a viable tool for the characterization of TMDs. Further, our study shows that vapor phase derived MoS_2 exhibits similar optical properties to exfoliated materials, making it a strong candidate for the manufacture for optoelectronic devices.

Vapor phase sulfurization was employed to synthesize MoS_2 thin films, as described in the previous work.[11,14] Mo (99.99%, MaTecK) films with various thicknesses (1–20 nm) were deposited by sputtering on fused quartz substrates ($\sim$10 mm $\times$ 10 mm) and on commercially available silicon dioxide (SiO_2, $\sim$290 nm thick) substrates ($\sim$10 mm $\times$ 10 mm), which were thermally grown on top of $\langle 100 \rangle$ oriented crystalline silicon (Si) wafers, using a Gatan Precision Etching and Coating System (PECS). A quartz crystal microbalance was used to monitor the Mo film deposition rate ($<$0.1 nm/s) and thickness. The Mo samples were sulfurized in a quartz tube furnace consisting of two different heating zones as shown in Figure 1(a). The samples were loaded into the hot zone which was heated to 750 °C and annealed for 30 min at a pressure of

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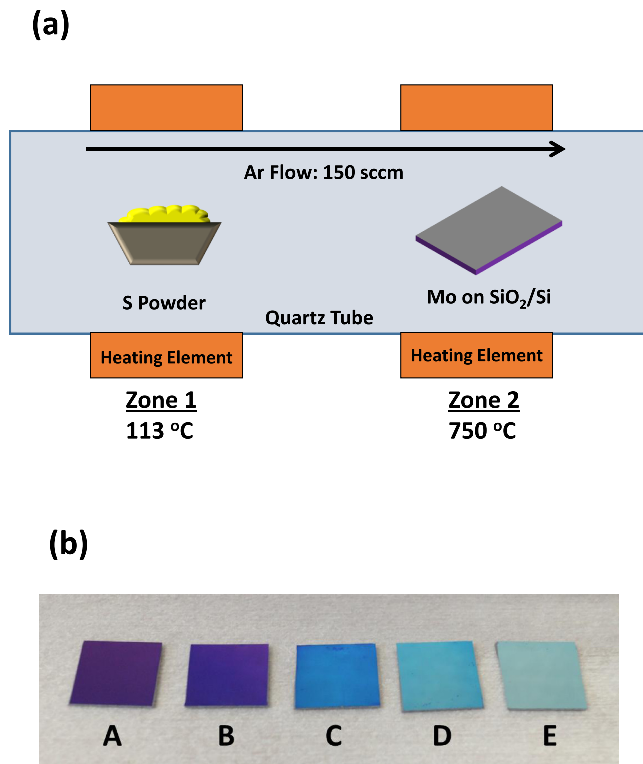

FIG. 1. (a) Schematic diagram of the vapor phase sulfurization process used for MoS_2 thin film synthesis. (b) Photograph of MoS_2 samples with increasing thickness (A–E).

~1 Torr with an argon (Ar) flow of 150 sccm (standard cubic centimeters per minute). Sulfur vapor was then produced by heating sulfur powder (MaTecK, 99%) up to its melting point (~113 °C) in the second upstream heating zone of the furnace and transported to the Mo samples using Ar gas as carrier. After sulfurization, the samples were annealed for 30 min at 750 °C and then cooled down to room temperature. Raman spectroscopy measurements were carried out with a HORIBA Jobin-Yvon Labram HR, using an excitation wavelength of 532 nm and an 1800 lines/mm grating. UV-visible absorption data were obtained with a Varian Cary 6000i double beam spectrophotometer in the spectral range of 200–800 nm. The SE characterization of the MoS_2 thin films was conducted using an Alpha SE tool (J. A. Woollam Co., Inc.) operating in the wavelength range of 380–900 nm (1.38–3.26 eV) at an angle of incidence of 70° with a beam spot size of ~40 mm^2 and a rotating compensator. SE data were analyzed using CompleteEASE 4.72 (J. A. Woollam Co., Inc.). X-ray reflectivity (XRR) measurements were conducted using a Bruker D8-Discover X-ray diffractometer which is fitted with Cu K-alpha X-ray source and Goebel mirror to produce a parallel beam. Thickness values were obtained by fitting the XRR data using a computer simulation called LEPTOS.

In our synthesis, vaporized sulfur reacts with predeposited Mo layers, yielding continuous MoS_2 thin films. An optical image of MoS_2 thin films on SiO_2/Si substrates (samples A, B, C, D, and E) is shown in Figure 1(b). The samples are clearly distinguishable from each other due to the surface color difference caused by their varying thicknesses. The films appear homogeneous over the sample area of ~10 mm × 10 mm.

The optical properties of the MoS_2 thin films were measured by SE. The raw SE data comprise a tabulation versus wavelength of the amplitude ratio (Ψ, psi) and the phase difference (Δ, delta) between the p- and s-polarized components of the reflected light. The two parameters measured by a SE system are related to the ratio ρ, defined by the equation of $\rho = r_p/r_s = \tan(\Psi)\exp(i\Delta)$, where r_p and r_s are the amplitude reflection coefficients for the p-polarized and s-polarized light, respectively.[16] A four-layer optical model which consists of a Si substrate, an interface layer between Si and SiO_2, a SiO_2 layer, and a MoS_2 layer was built to analyze the SE spectra. Each layer of the model has several fitting variables such as thickness and optical dispersion model parameters. A value of the goodness of fit, which quantifies how well the data generated by the optical model (G) fit the experimental data (E), is derived from the root mean squared error (MSE) as defined by

$$MSE = \sqrt{\frac{1}{3m-l}\sum_{i=1}^{m}[(N_{E_i}-N_{G_i})^2+(C_{E_i}-C_{G_i})^2+(S_{E_i}-S_{G_i})^2]} \times 1000, \quad (1)$$

where m is the number of wavelengths measured, l is the number of fit parameters, and $N = \cos(2\Psi)$, $C = \sin(2\Psi)\cos(\Delta)$, $S = \sin\ (2\Psi)\sin(\Delta)$.[21] The Levenberg-Marquardt nonlinear regression algorithm is used to minimize the MSE during the fitting process.

In a multilayer structure model, it is important to define the optical response of underlying layers in order to minimize optical correlation effects between layers and find more accurate values for the optical constants and the thickness of the top layer. Therefore, SE spectra of reference SiO_2/Si substrates, where Mo layers were not deposited, were first measured and analyzed using a three-layer optical model composed of a Si substrate, an interface layer between Si and SiO_2, and a SiO_2 layer. It is assumed that the underlying layers are stable with well-established optical constants.[22] The crystalline Si substrate of the model can be presumed to have a semi-infinite thickness, so the thicknesses of the interface and SiO_2 layers were determined from the fitting process. Then, the MoS_2 layer is added on top of the SiO_2 layer in the model structure, allowing its optical properties to be characterized in the presence of well-defined underlying layers. A Tauc-Lorentz (T-L) oscillation model was used to determine the optical properties of the MoS_2 thin film. The complex dielectric function of the energy (E)

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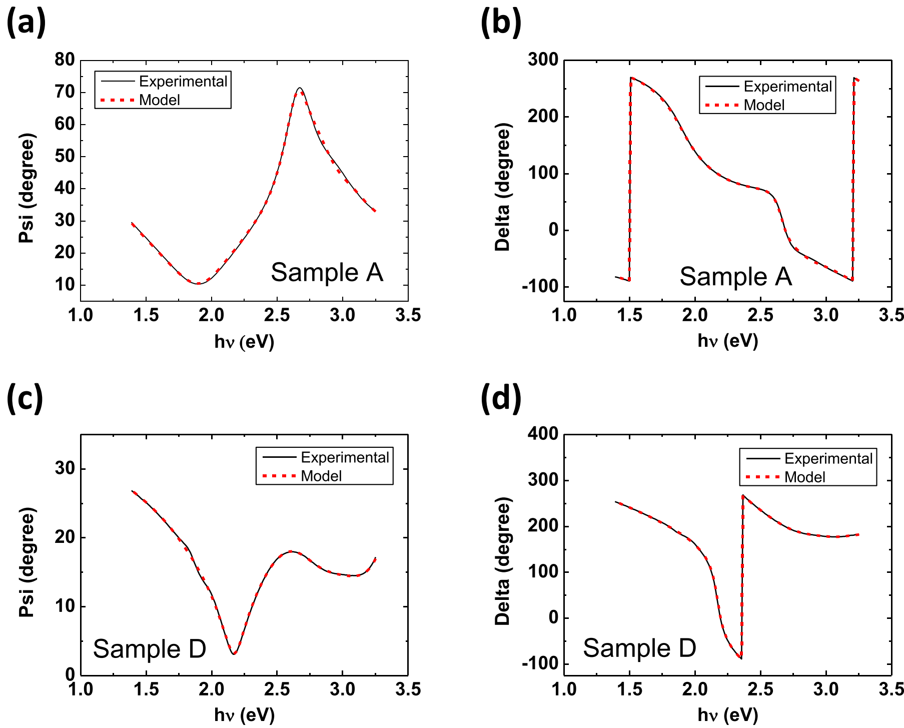

FIG. 2. Plots of the experimental and simulated (model) SE spectra (psi and delta) of MoS_2 samples A and D: (a) Ψ and (b) Δ data of sample A, and (c) Ψ and (d) Δ data of sample D.

is defined as $\varepsilon(E) = \varepsilon_1 + i\varepsilon_2$. In the T-L model, ε_1 and ε_2 are defined as

$$\varepsilon_2 = \left[\frac{AE_0C(E-E_b)^2}{(E^2-E_0^2)^2+C^2E^2} \times \frac{1}{E}\right] \text{ for } \quad E > E_b, \qquad (2)$$
$$\varepsilon_2 = 0 \quad \text{for} \quad E \le E_b,$$

$$\varepsilon_1 = \varepsilon_1(\infty) + \frac{2}{\pi}P\int_{E_b}^{\infty}\frac{\xi\varepsilon_2(\xi)}{\xi^2-E^2}d\xi, \qquad (3)$$

where A, E_0, C, and E_b represent the amplitude, center energy, broadening, and band gap of the oscillator, respectively, which all have units of energy (eV), and P stands for the Cauchy principal part of the integral.[23] The thickness of MoS_2 and the fitting parameters of the T-L oscillation model are varied, until the best fit between experimental spectra and simulated data from the optical model is achieved with the lowest MSE values. The experimental and simulated spectra (Ψ and Δ) for MoS_2 samples A and D are depicted in Figure 2, showing a good match between them. Additional spectral data for other samples (B, C, and E) are presented in Figure S1 of the supplementary material.[28] Extracted MoS_2 thickness values of the five samples (A, B, C, D, and E) are 1.99 ± 0.01 nm, 3.01 ± 0.07 nm, 5.53 ± 0.08 nm, 9.83 ± 0.04 nm, and 19.88 ± 0.05 nm, respectively. More details on fitting parameter values of the T-L oscillation model for the best fit are listed in the supplementary material (Table S1).[28] In addition, XRR measurements of the samples were conducted to verify the MoS_2 film thickness, and the measured values were consistent with those extracted from SE measurements. The thickness values of the MoS_2 thin films from SE and XRR measurements are summarized in Table I. More details on XRR measurements are presented in the supplementary material.[28] Figures 3(a) and 3(b) show n and k values of the MoS_2 layer for each sample derived from the optical model with the best fits. Two dominant peaks are observed at the energy of ∼2.0 eV and ∼2.9 eV in the k plot. In addition, an absorbance spectrum of a MoS_2 thin film (∼20 nm) on a quartz substrate was measured, as shown in the inset of Figure 3(b). Two spectral regions with dominant absorption variations are observed in the photon energy range of 1.5 eV–3.8 eV, which are related to A/B excitonic peaks in the range of 1.8–2.0 eV, and C/D excitonic peaks in the range of 2.7–3.1 eV, respectively.[24] This shows good agreement with the two k peaks observed at energies of ∼2.0 eV and ∼2.9 eV in the k plot.

Raman spectra were measured from the MoS_2 thin films on the SiO_2/Si substrates. When probed with a 532 nm excitation laser, bulk MoS_2 has two well-known Raman bands at ∼383 cm^{-1} and ∼408 cm^{-1} which correspond to E^1_{2g} and A_{1g} vibrational modes, respectively.[25] Raman spectra of five films, with different thicknesses, are shown in Figure 4. It is evident that the E^1_{2g} and A_{1g} peaks shift closer to one another with decreasing film thickness. This is consistent with the

TABLE I. Summary of the MoS_2 thickness values from SE data and XRR measurements for the five MoS_2 samples (samples A, B, C, D, and E). Thickness values of samples A and B from XRR measurements are not available due to the thickness resolution limit of XRR.

	Sample	A	B	C	D	E
SE	Thickness (nm)	1.99 ± 0.01	3.01 ± 0.07	5.53 ± 0.08	9.83 ± 0.04	19.88 ± 0.05
	MSE	4.100	7.067	3.606	4.429	5.173
XRR	Thickness (nm)	N.A.	N.A.	4.4 ± 1.4	9.9 ± 1.6	19.4 ± 1.6

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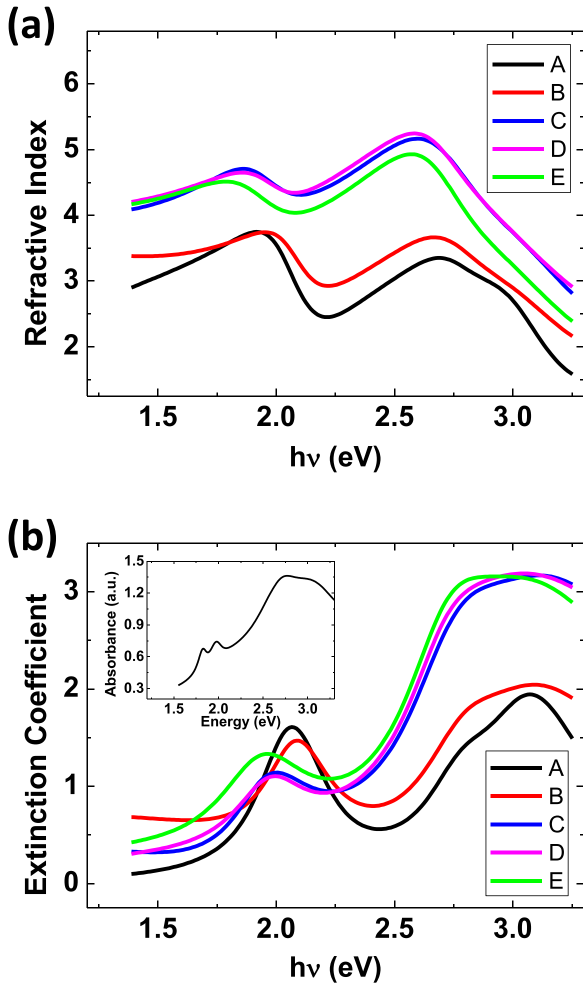

FIG. 3. Plots of (a) the refractive index n and (b) extinction coefficient k values of the MoS_2 layers for the five samples (A, B, C, D and E) derived from the optical model with the best fits. The inset of (b) shows a UV-visible absorption spectrum of a MoS_2 thin film on a quartz substrate.

report of Li *et al.*,[25] who described the layer thickness dependence of the Raman signal of mechanically exfoliated MoS_2 with decreasing layer thicknesses. In the case of sample E, the E^1_{2g} and A_{1g} peaks are seen at $384.2\,cm^{-1}$ and $409.6\,cm^{-1}$, a separation of $25.4\,cm^{-1}$, whereas for sample A, the same peaks manifest at $387.0\,cm^{-1}$ and $407.1\,cm^{-1}$, a separation of $20.1\,cm^{-1}$. This suggests that sample A consists of very few (1–3) layers, whereas sample E behaves like bulk MoS_2.

Furthermore, studies of the optical band gap energy of the MoS_2 thin films were conducted. The optical absorption coefficient (α) of MoS_2 thin films can be calculated from the extinction coefficient k and wavelength λ by means of $\alpha = 4\pi k/\lambda$. The relationship between absorption coefficient α and the photon energy can be expressed by

$$\alpha = \frac{K(h\nu - E_g)^m}{h\nu}, \qquad (4)$$

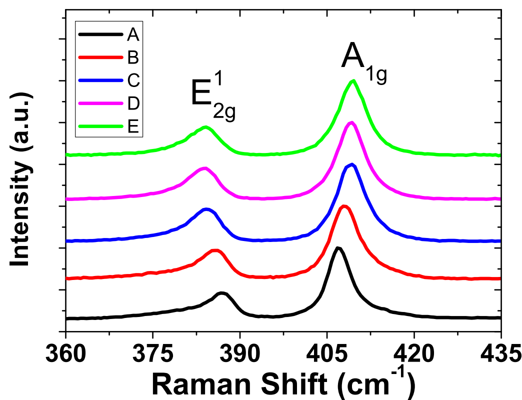

FIG. 4. Raman spectra of the MoS_2 thin films on the SiO_2/Si substrates (samples A, B, C, D, and E).

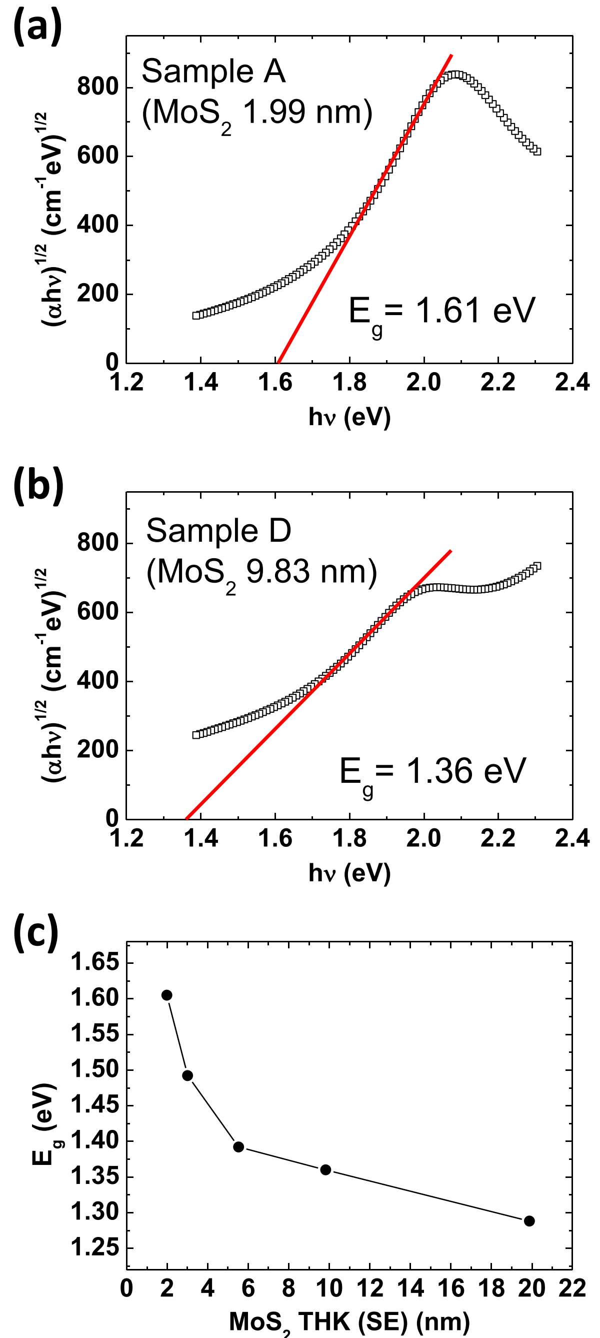

FIG. 5. (a) Plot of $(\alpha h\nu)^{1/2}$ vs. $h\nu$ for sample A. (b) Plot of $(\alpha h\nu)^{1/2}$ vs. $h\nu$ for sample D. The extrapolation of the linear region to $(\alpha h\nu)^{1/2} = 0$ (red line) gives the E_g value. (c) Plot of E_g vs. MoS_2 thickness for samples A, B, C, D, and E.

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where K is a constant, $h\nu$ is the incident photon energy, and m is a number that characterizes the transition process.[26,27] Depending on the transition type, m has a value of 1/2 for direct transition and 2 for indirect transition. Since our MoS_2 thin films are considered as multilayer MoS_2 films with thicknesses of 1.99–19.88 nm, it is believed that they have primarily indirect band gap transitions.[5] Thus, the variation of α with $h\nu$ can be determined by using the relation of $(\alpha h\nu)^{1/2} \sim (h\nu - E_g)$ and the optical band gap E_g can be extracted by extrapolating the linear region of a $(\alpha h\nu)^{1/2}$ vs. $h\nu$ plot to $(\alpha h\nu)^{1/2} = 0$. Plots of $(\alpha h\nu)^{1/2}$ vs. $h\nu$ for the MoS_2 thin films (samples A and D) are shown in Figures 5(a) and 5(b). The extracted E_g values of the five samples are 1.61, 1.49, 1.45, 1.36, and 1.29 eV for samples A, B, C, D, and E, respectively. Additional plots of $(\alpha h\nu)^{1/2}$ vs. $h\nu$ for samples B, C, and E are shown in Figure S3 of the supplementary material.[28] It is evident that the thinner MoS_2 films have a larger E_g, as presented in Figure 5(c). The thinnest MoS_2 film (sample A) with a measured thickness of 1.99 nm shows the largest E_g (1.61 eV), and as the MoS_2 film thickness increases E_g decreases towards the band gap of bulk MoS_2 ($\sim$1.3 eV). These first ellipsometric measurements of layered MoS_2 films are comparable with the previous reports on E_g of single and multilayer MoS_2 derived by exfoliation. Mak *et al.*[5] and Lee *et al.*[7] characterized the optical band gap energy of single and few layer mechanically exfoliated MoS_2 from photoluminescence (PL) spectra and electrical measurements of MoS_2 phototransistors, respectively, and found that the band gap decreased in energy with increasing MoS_2 thickness. Eda *et al.*[24] also observed a similar trend in E_g from chemically exfoliated MoS_2 using PL spectra measurements. This consistency is notable, since it implies that our vapor phase derived MoS_2 has comparable optical characteristics to those of exfoliated materials. Our study further establishes SE as a viable tool for the characterization of 2D materials as it can be used to investigate E_g of MoS_2, or indeed other TMDs (e.g., WTe_2, $MoSe_2$, etc.), while also giving information on optical constants and thicknesses. Such measurements are fast, non-destructive, and relatively easy to perform.

In summary, SE measurements have been carried out on MoS_2 thin films with different thicknesses synthesized by vapor phase sulfurization. In addition, they were characterized by XRR, UV-visible absorption spectroscopy, and Raman spectroscopy. An optical dispersion model of MoS_2 was developed for SE data analysis. Values of optical constants (n, k) and the thickness were determined by fitting the optical model function to the experimental data. The optical band gap energy E_g of the MoS_2 thin films was also investigated. Using the absorption coefficients of MoS_2 calculated from the k values, E_g of each MoS_2 film was extracted. The E_g values of the MoS_2 films show a clear dependence on the MoS_2 film thickness, indicating that thinner MoS_2 films have a larger E_g. This study establishes SE as a promising technique for optical characterization of TMD films.

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[1]K. S. Novoselov, D. Jiang, F. Schedin, T. J. Booth, V. V. Khotkevich, S. V. Morozov, and A. K. Geim, Proc. Natl. Acad. Sci. U.S.A. **102**, 10451 (2005).

[2]A. H. C. Neto and K. Novoselov, Mater. Express **1**, 10 (2011).

[3]K. S. Novoselov, Rev. Mod. Phys. **83**, 837 (2011).

[4]Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, and M. S. Strano, Nat. Nanotechnol. **7**, 699 (2012).

[5]K. F. Mak, C. Lee, J. Hone, J. Shan, and T. F. Heinz, Phys. Rev. Lett. **105**, 136805 (2010).

[6]B. Radisavljevic, A. Radenovic, J. Brivio, V. Giacometti, and A. Kis, Nat. Nanotechnol. **6**, 147 (2011).

[7]H. S. Lee, S.-W. Min, Y.-G. Chang, M. K. Park, T. Nam, H. Kim, J. H. Kim, S. Ryu, and S. Im, Nano Lett. **12**, 3695 (2012).

[8]B. Radisavljevic and A. Kis, Nature Mater. **12**, 815 (2013).

[9]O. Lopez-Sanchez, D. Lembke, M. Kayci, A. Radenovic, and A. Kis, Nat. Nanotechnol. **8**, 497 (2013).

[10]Y.-H. Lee, X.-Q. Zhang, W. Zhang, M.-T. Chang, C.-T. Lin, K.-D. Chang, Y.-C. Yu, J. T.-W. Wang, C.-S. Chang, L.-J. Li, and T.-W. Lin, Adv. Mater. **24**, 2320 (2012).

[11]Y. Zhan, Z. Liu, S. Najmaei, P. M. Ajayan, and J. Lou, Small **8**, 966 (2012).

[12]S. Najmaei, Z. Liu, W. Zhou, X. Zou, G. Shi, S. Lei, B. I. Yakobson, J.-C. Idrobo, P. M. Ajayan, and J. Lou, Nature Mater. **12**, 754 (2013).

[13]M. Shanmugam, C. A. Durcan, and B. Yu, Nanoscale **4**, 7399 (2012).

[14]K. Lee, R. Gatensby, N. McEvoy, T. Hallam, and G. S. Duesberg, Adv. Mater. **25**, 6699 (2013).

[15]W. Zhang, J.-K. Huang, C.-H. Chen, Y.-H. Chang, Y.-J. Cheng, and L.-J. Li, Adv. Mater. **25**, 3456 (2013).

[16]H. Fujiwara, *Spectroscopic Ellipsometry: Principles and Applications* (John Wiley & Sons Ltd, Chichester, 2007), p. 81.

[17]V. G. Kravets, A. N. Grigorenko, R. R. Nair, P. Blake, S. Anissimova, K. S. Novoselov, and A. K. Geim, Phys. Rev. B **81**, 155413 (2010).

[18]F. J. Nelson, V. K. Kamineni, T. Zhang, E. S. Comfort, J. U. Lee, and A. C. Diebold, Appl. Phys. Lett. **97**, 253110 (2010).

[19]J. W. Weber, V. E. Calado, and M. C. M. van de Sanden, Appl. Phys. Lett. **97**, 091904 (2010).

[20]A. Matkovic, U. Ralevic, M. Chhikara, M. M. Jakovljevic, D. Jovanovic, G. Bratina, and R. Gajic, J. Appl. Phys. **114**, 093505 (2013).

[21]*CompleteEASE™ Data Analysis Manual Version 4.63*, J. A. Woollam Co., Inc., Lincoln, NE, 2011, p. 45.

[22]C. M. Herzinger, B. Johs, W. A. McGahan, J. A. Woollam, and W. Paulson, J. Appl. Phys. **83**, 3323 (1998).

[23]G. E. Jellison and F. A. Modine, Appl. Phys. Lett. **69**, 371 (1996).

[24]G. Eda, H. Yamaguchi, D. Voiry, T. Fujita, M. Chen, and M. Chhowalla, Nano Lett. **11**, 5111 (2011).

[25]H. Li, Q. Zhang, C. C. R. Yap, B. K. Tay, T. H. T. Edwin, A. Olivier, and D. Baillargeat, Adv. Funct. Mater. **22**, 1385 (2012).

[26]F. P. Koffyberg, K. Dwight, and A. Wold, Solid State Commun. **30**, 433 (1979).

[27]E. R. Shaaban, M. S. Abd El-Sadek, M. El-Hagary, and I. S. Yahia, Phys. Scr. **86**, 015702 (2012).

[28]See supplementary material at http://dx.doi.org/10.1063/1.4868108 for additional SE spectral data, fitting parameter values of the T-L oscillation model, XRR details, and plots on the optical band gap extraction.

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